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COMPUTER-AIDED SIMULATION MODEL
FOR HYDROGEN ISOTOPE SEPARATING
COLUMN BY CRYOGENIC DISTILLATION

August 1982

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Computer-Aided Simulation Model for Hydrogen Isotope
Separating Column by Cryogenic Distillation

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This paper complements and extends the previously reported works by the same author on the mathematical simulation model for hydrogen isotope separating column by cryogenic distillation.

The convergence features of the proposed mathematical simulation procedure are investigated in detail and several significant estimation methods for initial values of a set of temperatures and phase flows are developed for ensurance of high efficiency of the iterative calculation as well as achievement of convergence.

Discussions are made on the effects of decay heat of tritium and nonideality of the hydrogen isotope solutions on steady state column performance.

KEYWORDS : Isotope Separation, Hydrogen Isotopes, Cryogenic
Distillation, Mathematical Model, Convergence,
Decay Heat, Tritium, Nonideality, Column Performance

深冷蒸留法による水素同位体分離塔の
コンピューターシミュレーションモデル

日本原子力研究所東海研究所核融合研究部

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(1982年7月30日受理)

本報は、深冷蒸留法による水素同位体分離塔に関する著者の現在までの研究の拡充を図ったものである。提案された数学的シミュレーション手法の収束性が詳細に調べられ、より優れた収束性を保証するために、塔内の気液流量分布及び温度分布の初期仮定の設定法に対するいくつかの重要な手法が開発されている。トリチウムの崩壊熱や溶液の非理想性が、塔の定常分離特性に及ぼす影響を及ぼすかについても、再度検討がなされている。

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1. Introduction

The estimated fuel (equimolar mixture of deuterium and tritium) burnup rate during a typical fueling cycle for presently-conceived tokamak-type fusion reactors is of the order of a few percent of the injected species. Hence, it is essential that the spent fuel mixture be recycled. However, the contaminants produced in the plasma chamber (He, H, C, N, O, Ar, etc.) must be removed from the mixture before the fuel can be reinjected. After removal of the high molecular weight atoms and He, the hydrogen isotopes (H_2 , HD, HT, D_2 , DT and T_2) is separated into reinjectable forms. The mainstream fuel circulation system is illustrated in Fig. 1.

Although there are a number of separation methods which could be applicable, cryogenic distillation appears to be most promising and attractive. The required purity or composition of the output streams from the hydrogen isotope separation process shows that a rather complex separation scheme involving several distillation columns, arranged in cascade, would be needed.

As the first step of research and development related to this process, investigation of the number of columns needed and the pertinent cascade configuration which meet the separation requirements under the assumed input specifications, is of vital importance. This investigation has been made by several workers.^{(1)~(3)} Among the proposals made by these workers, the column cascade developed by Sherman et al.⁽³⁾ seems most promising because of no feedback stream and its high separating performance. The configuration of this cryogenic distillation column cascade is shown in Fig. 2. The cascade is composed of the four-interlinked columns and the catalytic

equilibrators for promoting $HT + D_2 = HD + DT$. The 50-50 D-T mixture containing 1 % H is continuously fed to Column(1). The deuterium gas stream of the NBI (Neutral Beam Injector) recycle, containing H and T is mixed with the top product from Column(1). The two streams are separated into four product streams : an essentially tritium-free stream of H_2 -HD ; a high-purity stream of D_2 for the neutral beam injection ; a basically protium-free stream of D-T for refueling ; and a high-purity stream of T_2 for refueling. The raw deuterium gas of the NBI recycle in sufficient flow rate allows the percentage of HT in the feed stream to Column(2) to be decreased by almost two orders of magnitude, and this ensures both the sufficiently low tritium concentration in the H_2 -HD waste gas and the high-purity of the D_2 stream. If the flow rate of the neutral beam recycle is considerably small, it is mixed with the main stream D-T mixture fed to Column(1), and a certain percentage of the top product from Column(4) is recycled to the top product from Column(1) (Fig. 3).

In addition to the mainstream fuel circulation for fusion reactors, there are a number of other situations where cryogenic distillation could be useful as a process of hydrogen isotope separation. These situations are described elsewhere⁽⁴⁾⁽⁵⁾ and need not be repeated here.

To ensure the long-term smooth operation of the cryogenic distillation columns, detailed computer simulation studies as well as experimental works are needed. However, because of the cryogenic temperature, accumulation of reliable experimental data observed under a wide range of operational conditions is extremely formidable. For this reason, prediction of column behavior by use of the computer making the most of limited experimental data, plays a very important role. Development of

mathematical simulation models and computer-aided simulation procedures is an essential step for investigation of the column behavior.

Although there are a number of simulation procedures available which were developed for multicomponent distillation processes, they cannot directly be applied to hydrogen isotope distillation because they were developed for normal chemical engineering processes such as hydrocarbon processing. Hydrogen isotope separation processes have several specific features to be considered in the simulation study : 1) the separation requirements are usually very strict and high column performance is required ; 2) the columns need to be complex with a feedback stream in several situations⁽⁵⁾⁽⁶⁾; and 3) prediction of the level of infinitesimal amount of elements in product streams (e.g. tritium concentration in the waste stream discarded to the environment) need to be accurately made. Although rather simplified simulation models neglecting the heat balance equations and assuming the ideal form of the Raoult's law for the vapor-liquid equilibrium seem quite acceptable in the first step previously described, more complex models may be needed in the detailed analyses of the column behavior. Especially, the third feature listed above may require incorporation of heat balances and nonideality of the hydrogen isotope solutions into the simulation models. The heat balance equations can be derived considering appreciable differences in latent heat of vaporization among the six isotopic species of molecular hydrogen and decay heat of tritium (0.325 W/g-T), and if necessary, heat transfer through the column wall. Additionally, the first feature requires the large number of the total theoretical stages and large reflux ratio with the result that achievement of convergence of the iterative distillation calculation is a cumbersome task.

The present paper complements and extends several works previously reported by Kinoshita.^{(5)~(8)} These works were made especially for developing a rigorous mathematical simulation procedure which incorporates all the factors inherent in hydrogen isotope distillation (e.g. heat balances and nonideality) and nevertheless presents rapid achievement of convergence under a wide range of conditions. The present paper also includes sufficient amount of information on convergence characteristics of the procedure, effects of the factors on the column performance and column behavior, and specification of each column in Fig. 2.

2. Multi-dimensional Newton-Raphson Method

The solution of a set of nonlinear simultaneous equations is the final step in the solution of simulation problems of the equilibrium stage processes such as cryogenic distillation columns, falling liquid film condensers and multistage-type water/hydrogen-exchange columns. These equations are expressed as the simultaneous zeroing of a set of functions (residual functions), where the number of functions to be zeroed is equal to the number of the independent variables.

Consider the following set of n nonlinear equations :

$$\left. \begin{array}{l} f_1(x_1, \dots, x_n) = 0 \\ \vdots \\ f_n(x_1, \dots, x_n) = 0 \end{array} \right\} \quad (1)$$

By using the Taylor series expansion, we obtain

$$\begin{aligned} f_j(x_1 + \Delta x_1, \dots, x_n + \Delta x_n) &= f_j(x_1, \dots, x_n) + \\ &(\partial f_j / \partial x_1) \Delta x_1 + \dots + (\partial f_j / \partial x_n) \Delta x_n + \dots, \\ (j = 1, \dots, n) &. \end{aligned} \quad (2)$$

Assuming that the second and higher-order terms in the Taylor series expansion are sufficiently small, and $f_j(x_1 + \Delta x_1, \dots, x_n + \Delta x_n)$ is equal to zero, we obtain the following linear simultaneous equations with respect to Δx_j 's :

$$\left. \begin{aligned} (\partial f_1 / \partial x_1) \Delta x_1 + \cdots + (\partial f_1 / \partial x_n) \Delta x_n &= -f_1 \\ &\vdots \\ (\partial f_n / \partial x_1) \Delta x_1 + \cdots + (\partial f_n / \partial x_n) \Delta x_n &= -f_n \end{aligned} \right\} \quad (3)$$

The above equations can be written by

$$\Delta \vec{x} = -(\bar{G})^{-1} \vec{f} \quad , \quad (4)$$

where \vec{x} , $\Delta \vec{x}$ and \vec{f} are n-dimensional vectors defined by

$$\vec{x} = \begin{bmatrix} x_1 \\ \vdots \\ x_n \end{bmatrix} \quad , \quad \Delta \vec{x} = \begin{bmatrix} \Delta x_1 \\ \vdots \\ \Delta x_n \end{bmatrix} \quad \text{and} \quad \vec{f} = \begin{bmatrix} f_1 \\ \vdots \\ f_n \end{bmatrix} \quad , \quad (5)$$

respectively.

\bar{G} is the Jacobian matrix expressed by

$$\bar{G} = \begin{bmatrix} \partial f_1 / \partial x_1 & \cdots & \partial f_1 / \partial x_n \\ \vdots & \ddots & \vdots \\ \partial f_n / \partial x_1 & \cdots & \partial f_n / \partial x_n \end{bmatrix} \quad , \quad (6)$$

and $(\bar{G})^{-1}$ denotes the inverse matrix of \bar{G} .

In the Newton-Raphson iterative calculation, the initial values of the independent variables, \vec{x}^1 , are assumed before proceeding with the calculation. If \vec{f}^i is written for $\vec{f}(\vec{x}^i)$ and \bar{G}^i for the Jacobian matrix evaluated at \vec{x}^i , the values of the independent variables are repeatedly improved by

$$\begin{aligned} \vec{x}^{i+1} &= \vec{x}^i + \Delta \vec{x}^i \\ &= \vec{x}^i - (\bar{G}^i)^{-1} \vec{f}^i \quad , \quad (i = 1, 2, \dots ; i \text{ denotes the} \\ &\quad \text{iteration number}) , \end{aligned} \quad (7)$$

until the following convergence criterion is satisfied :

$$J = \sum_{j=1}^n |f_j|/n < \text{convergence tolerance } (\epsilon) . \quad (8)$$

In the Newton-Raphson method, initial estimation of the independent variables is a key step. If the initial estimates are far from the solutions, the iterative calculation usually fails to converge. On the other hand, even with fairly good estimates, divergence or great difficulty in achievement of convergence is often observed. In these cases, Eq.(7) must be replaced by the following special procedures.

2.1 Convergence Method 1

A coefficient, α^i ($0 < \alpha^i \leq 1$), is introduced and Eq.(7) is replaced by

$$\vec{x}^{i+1} = \vec{x}^i + \alpha^i \Delta \vec{x}^i . \quad (9)$$

α^i is determined so that J^{i+1} ($= \sum_{j=1}^n |f_j^{i+1}|/n$) is minimized at each iterative step. However, there is no single-variable optimization technique applicable to the cases where the unimodality is not inherent in J . For this reason, J^{i+1} is evaluated at a total of twenty different values of the coefficient :

$$\alpha_k^i = 0.05k , \quad (k = 1, \dots, 20) , \quad (10)$$

and α_q^i which minimizes J^{i+1} is chosen and a set of the independent variables is determined for the next iterative step by

$$\vec{x}^{i+1} = \vec{x}^i + \alpha_q^i \Delta \vec{x}^i . \quad (11)$$

2.2 Convergence Method 2

In this method, the maximum values of $|\Delta x_j|$ which are allowable at the iterative steps, δ_j , are prescribed before proceeding with the calculation. At every step, $|\Delta x_j^i|$ is checked if it exceeds δ_j or not. If $|\Delta x_j^i|$ is smaller than δ_j , the following is used :

$$x_j^{i+1} = x_j^i + \Delta x_j^i, \quad (12)$$

but if it exceeds δ_j , the above equation is replaced by

$$x_j^{i+1} = x_j^i + \text{SIGN}(\delta_j, \Delta x_j^i), \quad (13)$$

where

$$\text{SIGN}(a, b) = \begin{cases} a & (b > 0) \\ -a & (b < 0) \end{cases}, \quad (a > 0). \quad (14)$$

More difficulty in achievement of convergence requires smaller δ_j . In many cases, δ_j is required to be less than 10 % of x_j^1 .

2.3 Convergence Method 3

Although application of the previously described convergence methods are often successful, we still come across the cases where convergence is greatly difficult to achieve. In the third method, the procedure for determination of a new set of the independent variables is the same as that in the first one, with the exception of the following special modification. A set of coefficients, β_j 's ($0 < \beta_j \leq 1$), are prescribed in advance. At every iterative step, J^{i+1} is evaluated at α_k^i 's ($k = 1, \dots, 20$), but $|\alpha_k^i \Delta x_j^i|$ is checked before the evaluation if it

exceeds $\beta_j x_j^i$ or not. If it is smaller than $\beta_j x_j^i$, the evaluation is made by using

$$x_j^{i+1} = x_j^i + \alpha_k^i \Delta x_j^i \quad (15)$$

If it exceeds $\beta_j x_j^i$, the following is used instead :

$$x_j^{i+1} = x_j^i + \text{SIGN}(\beta_j x_j^i, \alpha_k^i \Delta x_j^i) \quad (16)$$

α_q^i minimizing J^{i+1} and the corresponding \vec{x}^{i+1} are thus chosen for the next iterative step*. More difficulty in achievement of convergence requires smaller β_j . In many cases, β_j is required to be smaller than 0.1.

* x_j^{i+1} 's are determined by using Eqs.(15) and (16) where α_k^i is replaced by α_q^i .

3. Mathematical Simulation Procedure

The basic idea developed by Tomich⁽⁹⁾ is used in the simulation procedure, while several specific modifications are made.

The model column for mathematical simulation is illustrated in Fig. 4. Basic equations are derived from the requirements for conservation of material and heat, and for phase equilibrium on any stage.

Component material balances:

$$V_2 y_{i,2} - V_1 y_{i,1} - L_1 x_{i,1} = 0, \quad (17)$$

$$L_{j-1} x_{i,j-1} + V_{j+1} y_{i,j+1} + F_j z_{i,j} - (V_j + W_j) y_{i,j} - (L_j + U_j) x_{i,j} = 0, \quad (j=2, \dots, N-1), \quad (18)$$

$$L_{N-1} x_{i,N-1} + F_N z_{i,N} - V_N y_{i,N} - L_N x_{i,N} = 0. \quad (19)$$

Heat balances:

$$E_{j,in} = L_{j-1} h_{j-1} + V_{j+1} H_{j+1} + F_j H_{Fj} + H_{Lj} (x_{3,j} + x_{5,j} + 2x_{6,j}) a\lambda + q_j, \quad (20)$$

$$E_{j,out} = (L_j + U_j) h_j + (V_j + W_j) H_j + Q_j, \quad (21)$$

$$E_j = 1 - E_{j,out}/E_{j,in} = 0, \quad (j=2, \dots, N-1). \quad (22)$$

Vapor-liquid equilibrium:

$$y_{i,j} = K_{eqi,j} x_{i,j}, \quad K_{eqi,j} = \xi_{i,j} p_i^o(T_j)/P_j. \quad (23)$$

Overall material balances:

$$L_j + V_1 - V_{j+1} - \sum_{k=2}^j (F_k - W_k - U_k) = 0, \quad (j=2, \dots, N-1). \quad (24)$$

Flow rates of the top and bottom products, D and B, are expressed by the following equations :

$$E_1 = 1 - RD/L_1 = 0, \quad (D=V_1), \quad (25)$$

$$E_N = 1 - B/L_N = 0, \quad (26)$$

respectively.

Substituting Eq.(23) in Eqs.(17) ~ (19) yields the following tridiagonal matrix equations :

$$\begin{pmatrix} B_{i,1} & C_{i,1} & 0 & \dots & 0 \\ A_{i,2} & B_{i,2} & C_{i,2} & & \\ \dots & \dots & \dots & & \\ & A_{i,j} & B_{i,j} & C_{i,j} & \\ & \dots & \dots & \dots & \\ & & A_{i,N-1} & B_{i,N-1} & C_{i,N-1} \\ 0 & \dots & 0 & A_{i,N} & B_{i,N} \end{pmatrix} \begin{pmatrix} x_{i,1} \\ x_{i,2} \\ \vdots \\ x_{i,j} \\ \vdots \\ x_{i,N-1} \\ x_{i,N} \end{pmatrix} = \begin{pmatrix} D_{i,1} \\ D_{i,2} \\ \vdots \\ D_{i,j} \\ \vdots \\ D_{i,N-1} \\ D_{i,N} \end{pmatrix}, \quad (27)$$

where

$$\begin{aligned} B_{i,1} &= -(L_1 + V_1 K_{eqi,1}), \quad C_{i,1} = V_2 K_{eqi,2}, \quad D_{i,1} = 0, \\ A_{i,j} &= L_{j-1}, \quad B_{i,j} = -(V_j + W_j) K_{eqi,j} - (L_j + U_j), \\ C_j &= V_{j+1} K_{eqi,j+1}, \quad D_j = -F_j z_{i,j}, \quad (j=2, \dots, N-1), \\ A_{i,N} &= L_{N-1}, \quad B_{i,N} = V_N K_{eqi,N} - L_N, \quad D_N = -F_N z_{i,N}. \end{aligned}$$

If L_j and T_j ($j=1, \dots, N$) are known, V_j and $K_{eqi,j}$ can be calculated in cases of $\xi_{i,j}=1$, and the liquid composition distribution is determined by solving Eq.(27) m times, followed by normalization of $x_{i,j}$:

$$x_{i,j} := x_{i,j} / \sum_{i=1}^m x_{i,j}. \quad (28)$$

The main distillation calculation is performed by finding out the solutions of the following $2N$ -dimensional nonlinear simultaneous equations :

$$\left. \begin{aligned} S_1(T_1, \dots, T_N, L_1, \dots, L_N) &= 0 \\ &\vdots \\ S_N(T_1, \dots, T_N, L_1, \dots, L_N) &= 0 \\ E_1(T_1, \dots, T_N, L_1, \dots, L_N) &= 0 \\ &\vdots \\ E_N(T_1, \dots, T_N, L_1, \dots, L_N) &= 0 \end{aligned} \right\}, \quad (29)$$

where S_j is defined by

$$S_j = \sum_{i=1}^m K_{eqi,j} x_{i,j} - 1. \quad (30)$$

Equation (29) is solved by application of the 2N-dimensional Newton-Raphson method described in the previous chapter.

The new values of the independent variables for the $(\beta+1)$ 'th iteration are calculated from

$$\left. \begin{aligned} T_j^{\beta+1} &= T_j^{\beta} + \Delta T_j^{\beta} \\ L_j^{\beta+1} &= L_j^{\beta} + \Delta L_j^{\beta} \end{aligned} \right\}, \quad (31)$$

where T_j^{β} 's and L_j^{β} 's are calculated by solving the following matrix equation :

$$\begin{pmatrix} \partial S_1 / \partial T_1 & \dots & \partial S_1 / \partial L_N \\ \vdots & & \vdots \\ \partial E_N / \partial T_1 & \dots & \partial E_N / \partial L_N \end{pmatrix} \begin{pmatrix} \Delta T_1^{\beta} \\ \vdots \\ \Delta L_N^{\beta} \end{pmatrix} = - \begin{pmatrix} S_1 \\ \vdots \\ E_N \end{pmatrix} \quad (32)$$

If Eq.(31) results in any difficulty in achievement of convergence, the previously described convergence methods need to be applied.

The iterative calculation is repeated until the following convergence criterion is satisfied :

$$J = \left\{ \sum_{j=1}^N (|S_j| + |E_j|) \right\} / (2N) < \epsilon, \quad 0 < \epsilon \ll 1. \quad (33)$$

When the nonideality of the hydrogen isotope solutions is incorporated, an additional iterative loop is needed as illustrated in Fig. 5.

Before proceeding with the calculation, initial estimates of the independent variables must be made in some way. In the present study, for initial estimation of temperatures, one of the following two methods is selected :

Method A ——— The two end temperatures, T_1 and T_N , are first assumed, and then the other temperatures are determined by interpolation by using the assumption of linear functions of the number of the theoretical stages.

Method B ——— The three temperatures, T_1 , T_M and T_N ($1 < M < N$), are first assumed, and an initial set of temperatures are determined in a similar manner.

The initial temperature profiles by these two methods are compared in Fig. 6.

The conventional procedures use the assumption of equal molal heats and equal molal overflows within the column for estimation of an initial set of phase flows. In some cases, this assumption presents stable progress of the iterative calculation and is quite successful. However, in cases of large amount of liquid holdups and high tritium concentration within the column, phase flows estimated by using this simple assumption are far from the solutions and often fail to present convergence. Initial estimates for the liquid flows originally developed by Kinoshita⁽⁵⁾⁽⁷⁾ summarized as follows :

- 1) Assume the values of L_j 's and V_j 's on the usual assumption of equal molal heats and equal molal overflows within the column.
- 2) Solve Eq.(27) and normalize $x_{i,j}$ by the procedure expressed by Eq.(28).
- 3) Calculate $y_{i,j}$ from Eq.(23), and normalize $y_{i,j}$.
- 4) Calculate molal enthalpies of liquid and vapor flows.
- 5) Recalculate L_j ($j=2, \dots, N-1$) from the following equation⁽⁶⁾:

$$L_j = [L_1 h_1 - \sum_{k=2}^j (Q_k - q_k) - (L_1 + D)H_2 + H_{j+1}D - H_{j+1} \sum_{k=2}^j (F_k - W_k - U_k) + \sum_{k=2}^j (F_k H_{Fk} - H_k W_k - h_k U_k) + \sum_{k=2}^j \{H_{Lk} (x_{3,k} + x_{5,k} + 2x_{6,k}) a\lambda\}] / (h_j - H_{j+1}),$$

($j=2, \dots, N-1$). (34)

- 6) Recalculate V_j ($j=2, \dots, N$) from Eq.(24).

This special method for initial estimates of liquid flows ensure fast convergence even in cases of large amount of liquid holdups resulting in large decay heat effects and any values of Q_j 's and q_j 's.

The partial derivatives of elements of the Jacobian matrix are numerically calculated by

$$\partial S_j / \partial L_k \approx (S_j(L_k + \delta L_k) - S_j(L_k - \delta L_k)) / (2\delta L_k),$$

($k = 1, \dots, N-1$), (35)

for instance. Equation (35) must be replaced by

$$\partial S_j / \partial L_k \approx (3S_j(L_k) + S_j(L_k - 2\delta L_k) - 4S_j(L_k - \delta L_k)) / (2\delta L_k),$$

(36)

in the case of $k = N$, to prevent V_1 from being minus when the total condenser is used. In calculation of these derivatives, the following two significant matters should be noted : 1) L_k 's must be sufficiently small to correctly evaluate the Jacobian matrix and achieve convergence ; and 2) since the vapor flows, V_j 's, are dependent on the liquid flows, L_j 's, V_j 's need to also be changed by using the overall material balances expressed by Eq.(24) whenever L_k is increased or decreased by δL_k (or $2\delta L_k$ in Eq.(36)).

4. Computer Simulation Study

4.1 Convergence Characteristics of Simulation Procedure

The principal objective of this section is exploration of convergence characteristics of the Newton-Raphson iterative calculation, such as stability of the iterative calculation, rapidity in achievement of convergence, and effects of initial estimates of the independent variables. The design of the fusion reactor and its associated systems is still in a very early stage. For this reason, the input specifications for the hydrogen isotope separation process and separation requirements have not been determined yet. However, the probable values of the input variables are assumed for each column as given in Table 1. These values stand for the specific feature of each column, although they are arbitrarily assumed.

A total of seven calculations are first made for Columns(1), (2) and (4) to explore the convergence features. The parameters which could have profound effects on the convergence features are established for each run as shown in Table 2. In these runs, neither decay heat of tritium nor nonideality of the hydrogen isotope solutions is incorporated in the simulation model. Comparatively low tritium percentage within these three columns and reasonable neglect of decay heat of tritium eliminate the need for introduction of the previously described special estimation method of an initial set of phase flows. The nonideality is neglected because the outer successive iteration loop is not of interest to us in this section. The convergence features are observed from Fig. 7 where the average

value of the residual functions (J defined by Eq.(33)) is plotted against the iteration number for each run. It is observed that the estimation method of an initial set of temperatures may be more essential than the special convergence technique, for ensurance of achievement of convergence or small number of the total iterations needed. This fact is appreciated in the test runs made for Column(2). For this column, the high percentage of HD is expected on the first stage through the stage the number of which is fairly large. Hence, the temperature drop across the column is considerably small for these stages (especially in the enriching section of the column) and this requires a special method (Method B) for initial estimation of temperatures.

In order to appreciate importance of the special initial estimation method for phase flows in cases of large amount of liquid holdups and high tritium percentage within the column, four test runs are made for Column(3) which has the highest tritium inventory among the four columns in Fig. 2. The calculational conditions of these four test runs are given in Table 3. Information concerning the convergence features observed is illustrated in Fig. 8 as well as in Table 3. It indicates that in cases of comparatively small heat generation rate (due to decay of tritium), the usual assumption of equal molal overflows cause no serious problem although the number of total iterations needed may be larger. However, in cases of considerably large heat generation rate, the usual assumption results in divergence of the iterative calculation (the phase flows become minus). The divergence could be avoided by application of the special convergence techniques, but unacceptably many iterations would be needed. It is desired that the special estimation method of an initial set of phase flows be used in the

simulation model regardless of liquid holdups and tritium concentration within the column.

In order to extend the understanding of the features of the four distillation columns, the liquid composition profiles calculated for these columns are shown in a series of figures (Figs. 9 ~ 12). Although they are calculated neglecting decay heat of tritium and nonideality, they never fail to provide appropriate information concerning the column features.

4.2 Effects of Major Factors on Column Behavior

There are three major factors inherent in hydrogen isotope separation by cryogenic distillation which could have substantial effects on column behavior : 1) differences in latent heat of vaporization among the six molecular species ; 2) decay heat of tritium ; and 3) nonideality of the hydrogen isotope solutions. In order to investigate the extents of the effects of these factors, calculations are made for Column(3) using the following five different models :

Model I — All the three factors are neglected.

Model II — Only Factor 1) is incorporated.

Model III — Factors 1) and 2) are incorporated neglecting Factor 3).

Model IV — Factors 1) and 3) are incorporated neglecting Factor 2).

Model V — All the three factors are incorporated.

The calculation which uses Model I is performed by use of the tridiagonal matrix method developed by Amundsen.⁽¹⁰⁾

The extent of the effects of decay heat of tritium largely depends upon the amount of liquid held within the column. Liquid holdups can be estimated only if the column dimensions are specified. It was reported that the liquid holdup in the packed section of the column was approximately 15 % of the packed volume.⁽¹¹⁾⁽¹²⁾ This means that the liquid holdup depends on the HETP value and the inner diameter selected in design to a large extent. If we assume the liquid holdup remains unchanged for any theoretical stage, it may be approximately estimated by the following equations :

$$d = 2 \sqrt{1000 \zeta V_2 R_g T_1 / (3600 v \pi P_1)} \quad , \quad (37)$$

$$H_{Lj} = \pi d^2 h_e \rho_{N-1} \eta / 4000 , \quad (38)$$

where

ζ = coefficient for accounting for deviation from the ideal gas law,

V_2 = vapor flow rate leaving second stage (g-mol/h),

R_g = gas constant (=0.08206 l·atom/g-mol/K),

T_1 = absolute temperature on first stage (K),

v = selected superficial vapor velocity within column (cm/sec),

d = inner diameter (cm),

h_e = HETP (cm),

H_{Lj} = liquid holdup ($j=2, \dots, N-1$) (g-mol/stage),

ρ_{N-1} = molal density of liquid on (N-1)'th stage (g-mol/l),

η = ratio of liquid volume to packed volume (=0.15) .

The vapor velocity is usually chosen to be ~50 % of the flooding velocity. However, the flooding velocity is still unknown. It was reported that the columns were successfully operated using vapor velocities above 12.5 cm/sec with no sign of deterioration in separating efficiency.⁽¹²⁾ At Los Alamos National Laboratory, they used 8.9 cm/sec for vapor velocity in their design of the column cascade for the Tritium Systems Test Assembly.⁽³⁾ In the present study, v is chosen to be 10 cm/sec.

Sherman et al. reported that the HETP value was measured to be around 5 cm.⁽³⁾⁽¹²⁾ Wilkes reported HETP values much smaller than 5 cm,⁽⁴⁾⁽¹³⁾ and the problem of this discrepancy has not been solved yet. In the present study, Sherman's value is used for estimation of the liquid holdup, pending further experimental studies. The values used for parameters in Eqs.(37) and (38) are $\zeta = 0.9$; $v = 10$ cm/sec ;

$\rho_{N-1} = 42.5$ g-mol/l ; and $h_e = 5$ cm. As a consequence, H_{Lj} is 1.0 g-mol/stage for Column(3). The liquid holdups in the reboiler and the condenser are postulated to be five times more than that in one stage of the packed section and one-tenth, respectively.

Although several experimental works were reported dealing with nonideality of the hydrogen isotope solutions,^{(14)~(16)} these works do not allow us to accurately estimate the deviation coefficients, $\xi_{i,j}$'s defined by

$$y_{i,j} = \xi_{i,j} p_i^o(T_j) x_{i,j} / P_j \quad , \quad (39)$$

under any condition. In the present study, the procedure proposed by Souers⁽¹⁶⁾ is used though it does not always coincides with the experimental observations.

The specific results for the five models are summarized in Table 4. It is observed that all the three factors cause deterioration in the column performance. The effect of Factor 2) could be remarkable. Factors 1) and 2) cause decrease both in vapor flows and liquid flows. Table 5 shows comparison among the three models in the liquid flow rate distribution within the column and the reboiler load estimated. The liquid flow near the bottom decreases by ~5 % due to Factor 1) and it is further decreased by ~40 % due to Factor 2). These decreases cause deterioration of the separating efficiency. The significant result is that the reboiler load estimated by use of Model II is accompanied by a large error exceeding +80 %. As described before, the extent of the decay heat effects largely depend upon the column dimensions. If the superficial vapor velocity within the column is chosen to be 5 cm/sec (Model III'), or if this parameter and the HETP are 15 cm/sec and 3.75 cm, respectively (Model III"), for instance, the

column performance can vary as summarized in Table 4. Because of this variation and uncertainty of the experimentally determined parameters, it is not easy to draw firm conclusions, but it is needed that Factor 2) be incorporated in the simulation model, especially in the situations where large tritium decay heat effects are expected.

Table 6 shows distributions of the deviation coefficients for D_2 , DT and T_2 calculated in Model IV. It indicates that Factor 3) causes decreases in relative volatilities for D_2 -DT and DT- T_2 except the section near the bottom, and this fact is the reason for the deterioration of the column performance. Experimental observations reported by Sherman et al.⁽¹⁵⁾ indicate stronger nonideality of the D_2 -DT- T_2 system than the prediction by Souers' procedure, hence, the effect of nonideality could be larger than the results given in Table 4.

All the models except Models I and II predict that the tritium percentage in the bottom product does not attain 99 atom%, while Models I and II predict that the requirement is successfully met. This fact also points out importance of incorporation of Factors 2) and 3) in the simulation model. The convergence feature observed in simulation for Model V is illustrated in Fig. 13 for understanding of the feature of both the inner iteration loop and the outer one.

Desirability of accurate estimation of the infinitesimal amount of element is one of the most specific features of hydrogen isotope separation. In order to prove importance of incorporation of the factors from another standpoint, the similar comparisons are made among the results of performance prediction for Column(2) using the five different models. The tritium level in the top product is a most critical variable to be accurately evaluated.

The results are summarized in Table 7. Because the tritium concentration within Column(2) is considerably small, Factor 2) is not significant at all. However, on account of simultaneous effects of Factors 1) and 3), the tritium level is almost one order of magnitude higher. In other words, Model I fails to accurately estimate the tritium level.

The conclusion is that the previously described factors need to be incorporated in the simulation model, because they have comparatively large effects in some situations.

5. Conclusion

- (1) A rigorous mathematical simulation procedure previously reported by Kinoshita is fully described. Although the method proposed by Tomich is adapted to hydrogen isotope separating columns, special estimation procedures for initial values of a set of temperatures and phase flows are originally developed. These methods ensure rapid achievement of convergence even in cases where decay heat effects are remarkably large or separation requirements are considerably strict.
- (2) Importance of incorporation of the heat balance equations and nonideality of the hydrogen isotope solutions is proved by several case studies. A simple model neglecting these factors fails to correctly estimate the column performance and the reboiler load. A computer code is available to detailed analyses of column behaviour once the input specifications are given.

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The author wishes to give his sincere thanks and appreciation to Profs. T. Takamatsu and I. Hashimoto (Kyoto University), and Mr. Y. Naruse (JAERI) for their valuable comments on the present study. Acknowledgment is also due to Dr. Y. Obata (JAERI) for his continuous encouragement.

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NOMENCLATURE

- a : Atomic weight of tritium (=3.017) (g-T)
 B : Prescribed value of bottom product flow rate (g-mol/h)
 D : Prescribed value of top product flow rate (g-mol/h)
 F_j : Flow rate of feed stream supplied to j-th stage (g-mol/h)
 H_j : Molal enthalpy of vapor stream leaving j-th stage (cal/g-mol)
 h_j : Molal enthalpy of liquid stream leaving j-th stage (cal/g-mol)
 H_{Fj} : Molal enthalpy of feed stream supplied to j-th stage
 (cal/g-mol)
 H_{Lj} : Liquid holdup in j-th stage (g-mol)
 $K_{eqi,j}$: Vapor-liquid equilibrium ratio for i-th component on
 j-th stage
 L_j : Flow rate of liquid stream leaving j-th stage (g-mol/h)
 m : Number of components (=6)
 N : Number of total theoretical stages
 N_F : Feed stage number
 P_j : Total pressure on j-th stage (Torr)
 $p_{i,j}^o$: Vapor pressure for i-th pure component at T_j K (Torr)
 Q_j : Heat subtraction rate on j-th stage (cal/h)
 q_j : Heat leak rate on j-th stage (cal/h)
 R : Reflux ratio
 T_j : Absolute temperature on j-th stage (K)
 U_j : Flow rate of liquid sidestream from 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 i-th component in liquid stream
 leaving j-th stage

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

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

λ : Decay heat of tritium (=280 cal/h/g-T)

$\xi_{i,j}$: Coefficient for accounting for deviation from Raoult's
Law

(Subscript)

(Subscript)

3 : HT 5 : DT 6 : T₂

$a:=b$ means the procedure that b is used as a new value of a .

$SIGN(a,b)$ denotes the number whose absolute value is a and whose
sign is the same as that of b .

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Table 1 Input Specifications Assumed for Four Distillation Columns

	Column(1)	Column(2)	Column(3)	Column(4)
Feed Flow rate (g-mol/h)	100.0	100.0	100.0	100.0
Feed Composition	H ₂	0.1368 X 10 ⁻³	0.1000 X 10 ⁻¹⁴	0.1114 X 10 ⁻⁶
	HD	0.1048 X 10 ⁻¹	0.1000 X 10 ⁻⁸	0.5969 X 10 ⁻³
	HT	0.9248 X 10 ⁻²	0.1351 X 10 ⁻³	0.2570 X 10 ⁻⁵
	D ₂	0.2481	0.9604	0.9890
	DT	0.4832	0.9813 X 10 ⁻²	0.6450
	T ₂	0.2488	0.2650 X 10 ⁻⁴	0.3300
Feed Temperature (K)	24.30	23.62	24.57	23.67
Flow Rate of Top Product (g-mol/h)	25.0	2.9	70.0	98.5
Reflux Ratio	25.0	150.0	8.0	8.0
Number of Total Theoretical Stages	80	80	65	80
Feed Stage Number	50	55	30	40

1) The feed is supplied in saturated liquid state. All the feed streams are equilibrated at 25 °C by

H₂ + D₂ = 2HD, H₂ + T₂ = 2HT and D₂ + T₂ = 2DT, except the feed to Column(3).

2) The condenser is the partial one.

3) The operating pressure is 760 Torr and the pressure drop across the column is neglected.

Table 2 Convergence Parameters and Methods Used in Test Runs

	δL_k (g-mol/h)	δT_k (K)	T_L (K)	T_N (K)	M	T_M (K)	Convergence Method	n
Column(1)	Run 1	0.005	23.5	24.5			0	5
Column(2)	Run 1	0.001	22.1	23.7			0	divergence
	Run 2	0.001	22.1	23.7			1	divergence
	Run 3	0.001	22.1	23.7			2*	16
	Run 4	0.001	22.1	23.7	35	22.2	0	5
Column(4)	Run 1	0.001	23.6	23.9			0	4
	Run 2	0.001	23.6	23.9	45	23.65	0	3

*) δ_j for liquid flows is 5.0 g-mol/h and that for temperatures is 0.2 K.

T_L , T_M and T_N : Temperatures initially assumed in Fig. 6.

Convergence Method 0 : Any convergence technique described in Chapter II is not used.

n : Number of total iterations needed.

divergence : Infinitely large numbers were observed in the process.

Table 3 Convergence Parameters and Methods Used in Calculations

Case	Liquid Holdup in Packed Section * (g-mol/stage)	Initial Estimation Procedure for Phase Flows	n
1	1.0	Procedure A	4
2	1.0	Procedure B	5
3	2.0	Procedure A	4
4	2.0	Procedure B	divergence

Parameters used are as follows : $\delta L_k = 0.005$ g-mol/h, $\delta T_k = 0.02$ K, $T_1 = 24.5$ K,
 $T_N = 25.0$ K and Convergence Method 0 is used.

Procedure A : Special estimation procedure developed by Kinoshita.

Procedure B : Usual assumption of equal molar overflows within the column.

*) Estimation procedure of liquid holdups is described in 4.2.

Table 4 Column Performance Predicted for Column(3) by Several Different Models

Model	Atom Fraction of Each Element in Top Product		Atom Fraction of Each Element in Bottom Product		L_{N-1} (g-mol/h)	Q_R (W)
	H	D	H	D		
I	7.144×10^{-6}	0.4937	3.822×10^{-14}	6.353×10^{-3}	660.0	242
II	7.144×10^{-6}	0.4934	4.377×10^{-14}	7.125×10^{-3}	625.4	229
III	7.144×10^{-6}	0.4889	1.380×10^{-13}	1.764×10^{-2}	385.2	127
IV	7.144×10^{-6}	0.4907	7.526×10^{-14}	1.337×10^{-2}	626.0	229
V	7.144×10^{-6}	0.4836	2.334×10^{-13}	2.989×10^{-2}	391.0	129
III'	7.144×10^{-6}	0.4729	1.135×10^{-12}	5.499×10^{-2}	152.0	28
III''	7.144×10^{-6}	0.4918	7.231×10^{-14}	1.074×10^{-2}	503.8	177

L_{N-1} : Flow rate of liquid stream leaving (N-1)'th stage

Q_R : Reboiler Load

Table 5 Comparison among Three Models in Liquid Flow Rate
Distribution within Column

	Stage	Model I	Model II	Model III
Distribution of Liquid Flow Rate (g-mol/h)	1	560.00	560.00	560.0
	5	560.00	557.85	545.92
	10	560.00	555.24	527.67
	15	560.00	552.64	508.84
	20	560.00	550.33	489.82
	25	560.00	548.50	470.92
	30	660.00	646.62	551.85
	35	660.00	642.14	530.44
	40	660.00	637.11	507.44
	45	660.00	632.54	482.97
	50	660.00	629.18	457.55
	55	660.00	627.08	431.76
	60	660.00	625.90	405.89
	65	30.00	30.00	30.00
Reboiler Load (W)		242	229	127

Table 6 Distribution of Deviation Coefficient
in Model IV

Stage	Deviation Coefficient for Components		
	D ₂	DT	T ₂
1	0.9936	0.9997	1.016
10	0.9931	0.9982	1.014
20	0.9927	0.9968	1.011
30	0.9924	0.9960	1.010
40	0.9920	0.9941	1.006
50	0.9917	0.9922	1.003
60	0.9916	0.9913	1.001
65	0.9916	0.9911	1.000

Table 7 Column Performance Predicted for Column(2)
by Five Different Models

Model	Atom Fraction of T in Top Product	Tritium Release Rate (mCi/day)
I	7.61×10^{-9}	31.1
II	1.26×10^{-8}	51.4
III	1.26×10^{-8}	51.4
IV	5.11×10^{-8}	209
V	5.12×10^{-8}	209

Flow rate of top product = 2.9 g-mol/h

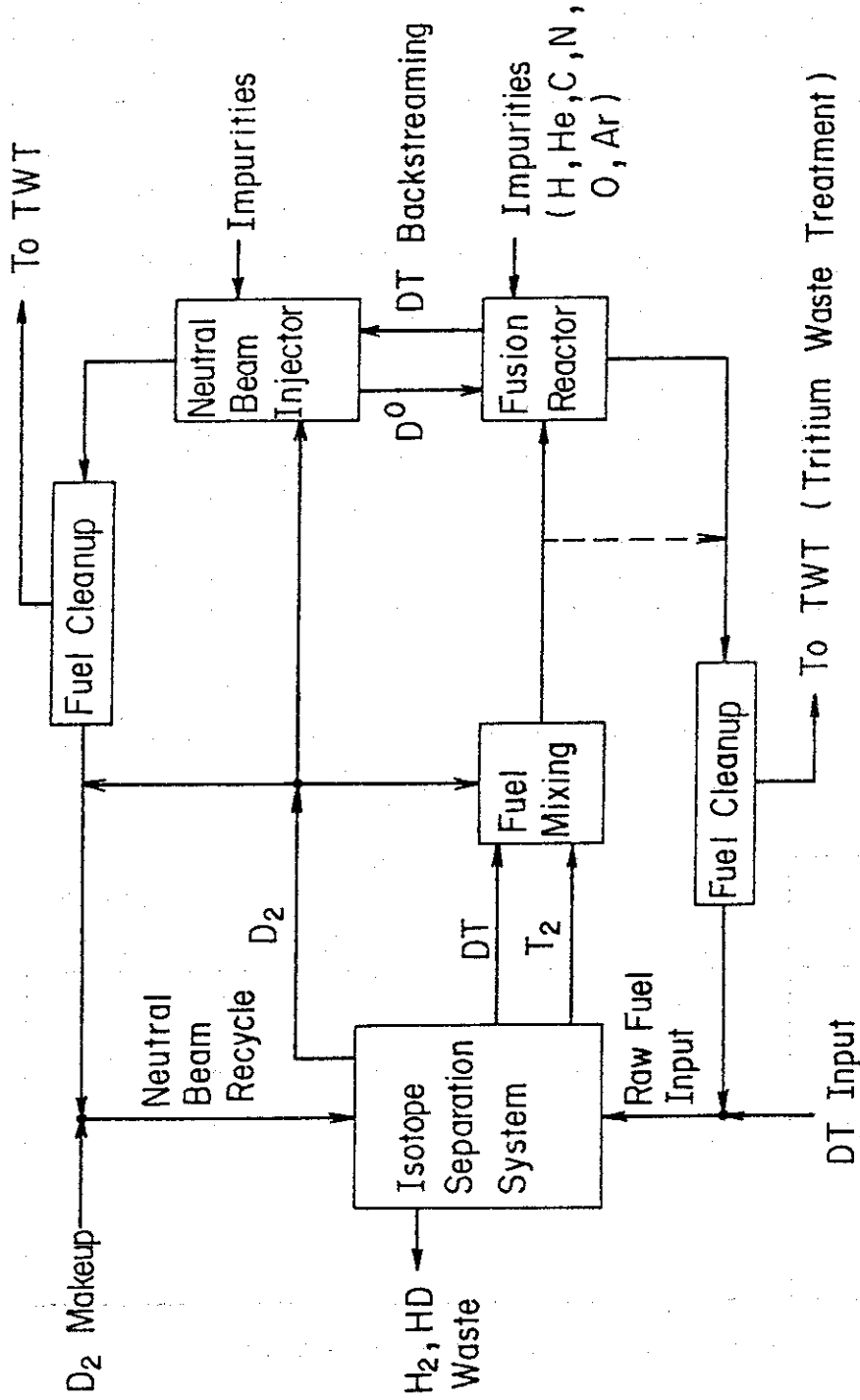


Fig. 1 Conceptual Flow Sheet of Fuel Circulation System for Fusion Reactor

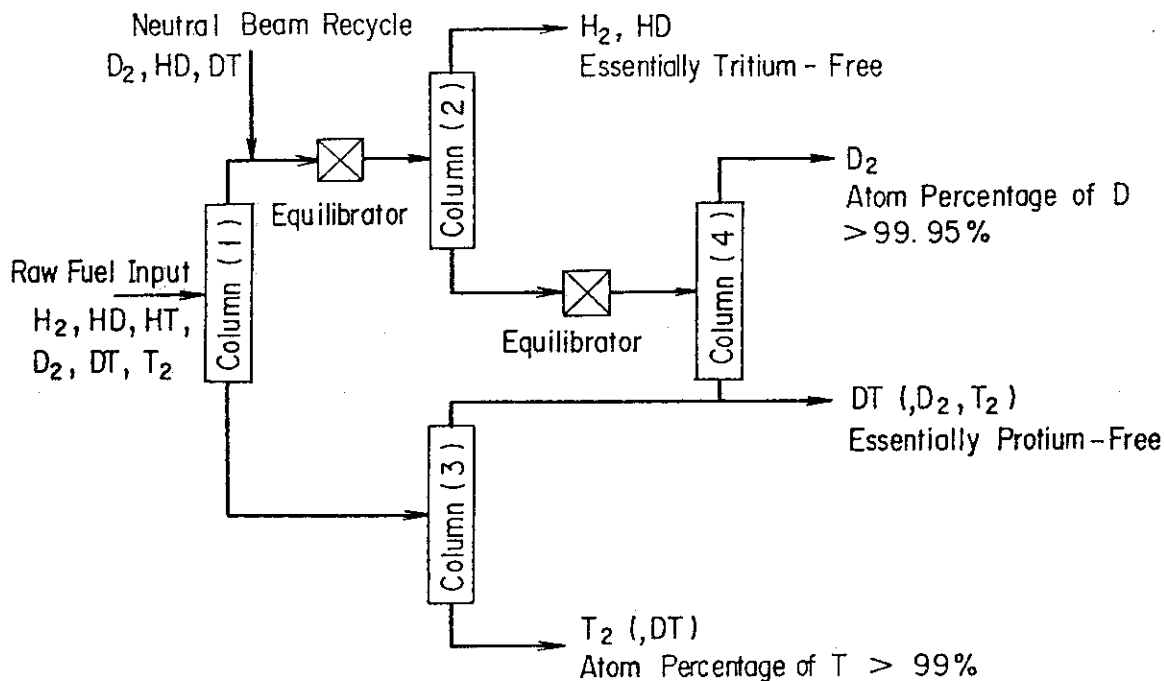


Fig. 2 Conceptual Flow Sheet of Cryogenic Distillation Column Cascade (1)

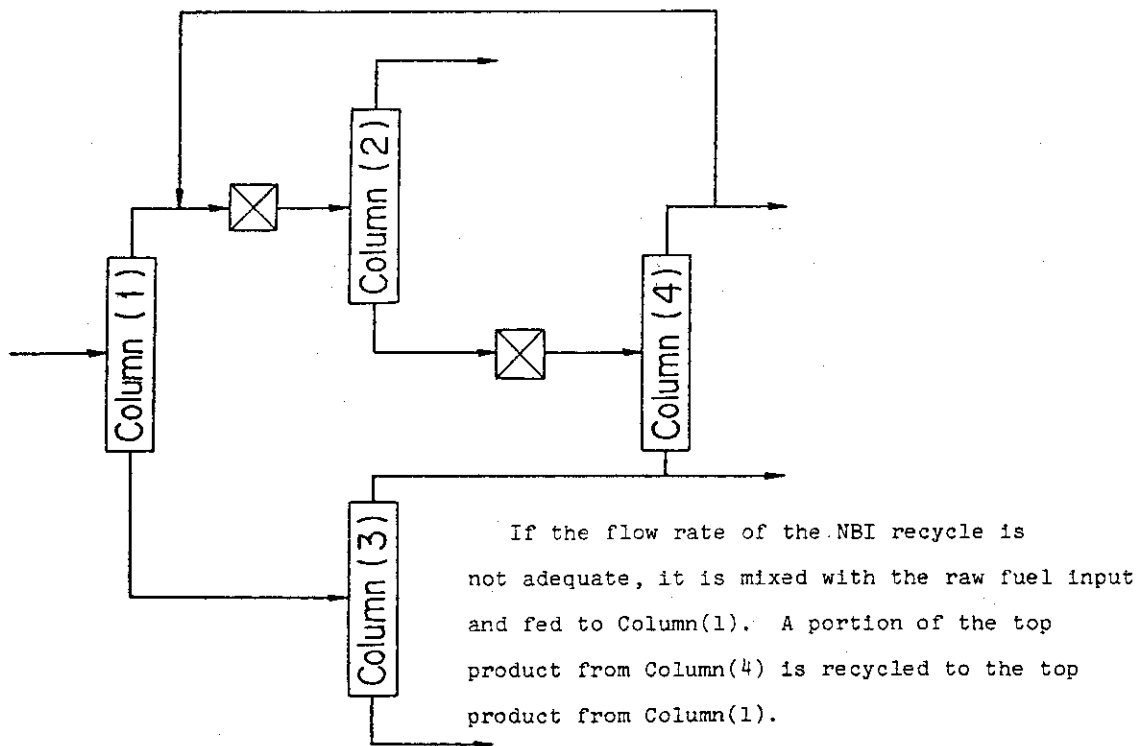


Fig. 3 Conceptual Flow Sheet of Cryogenic Distillation Column Cascade (2)

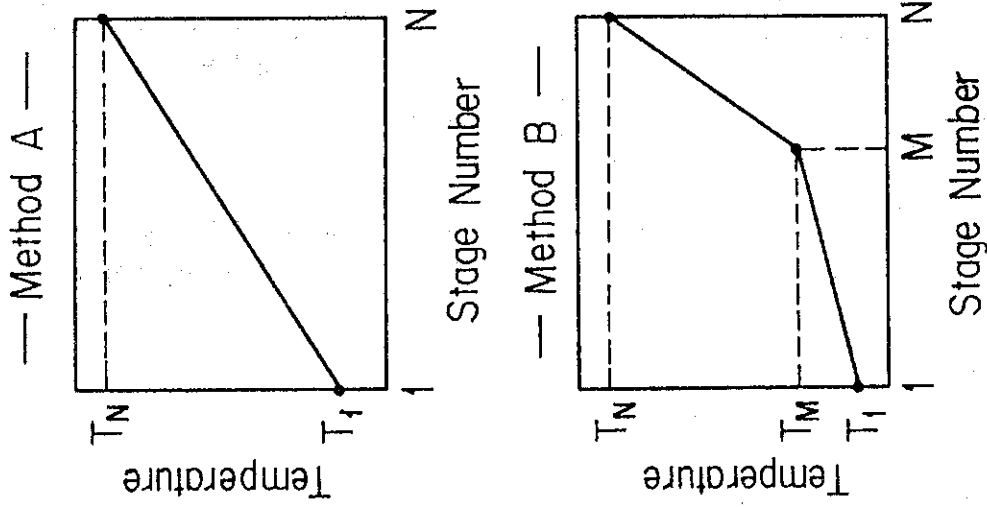


Fig. 6 Determination Methods for Initial Values of a Set of Temperatures

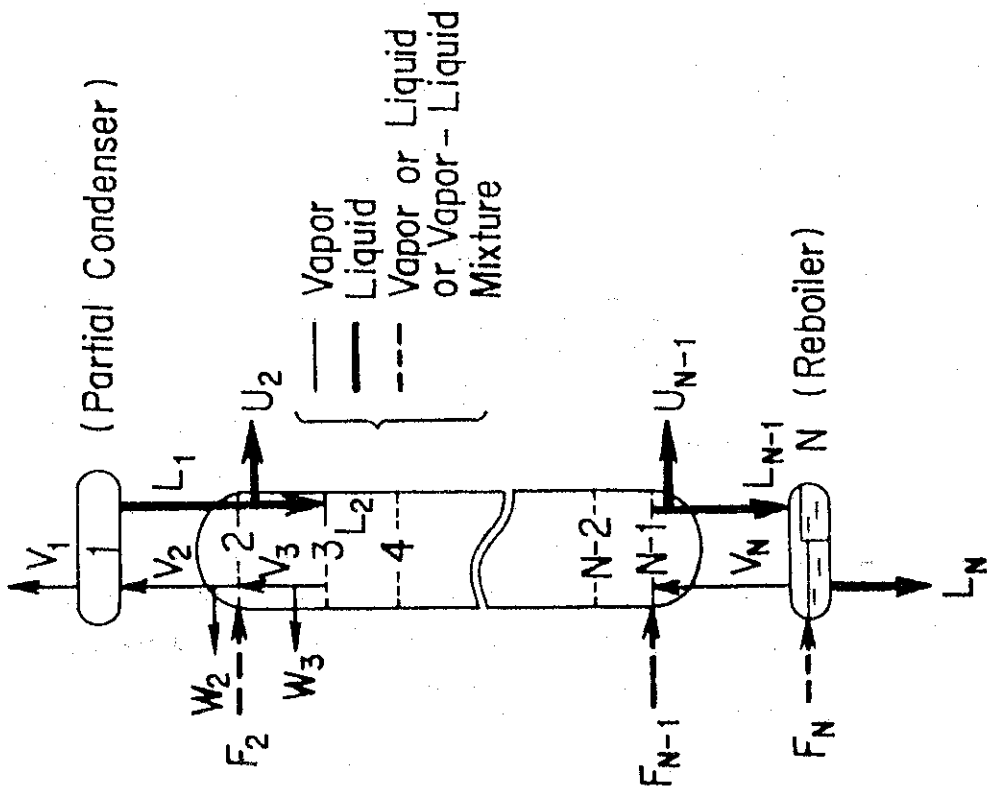


Fig. 4 Model Column for Mathematical Simulation

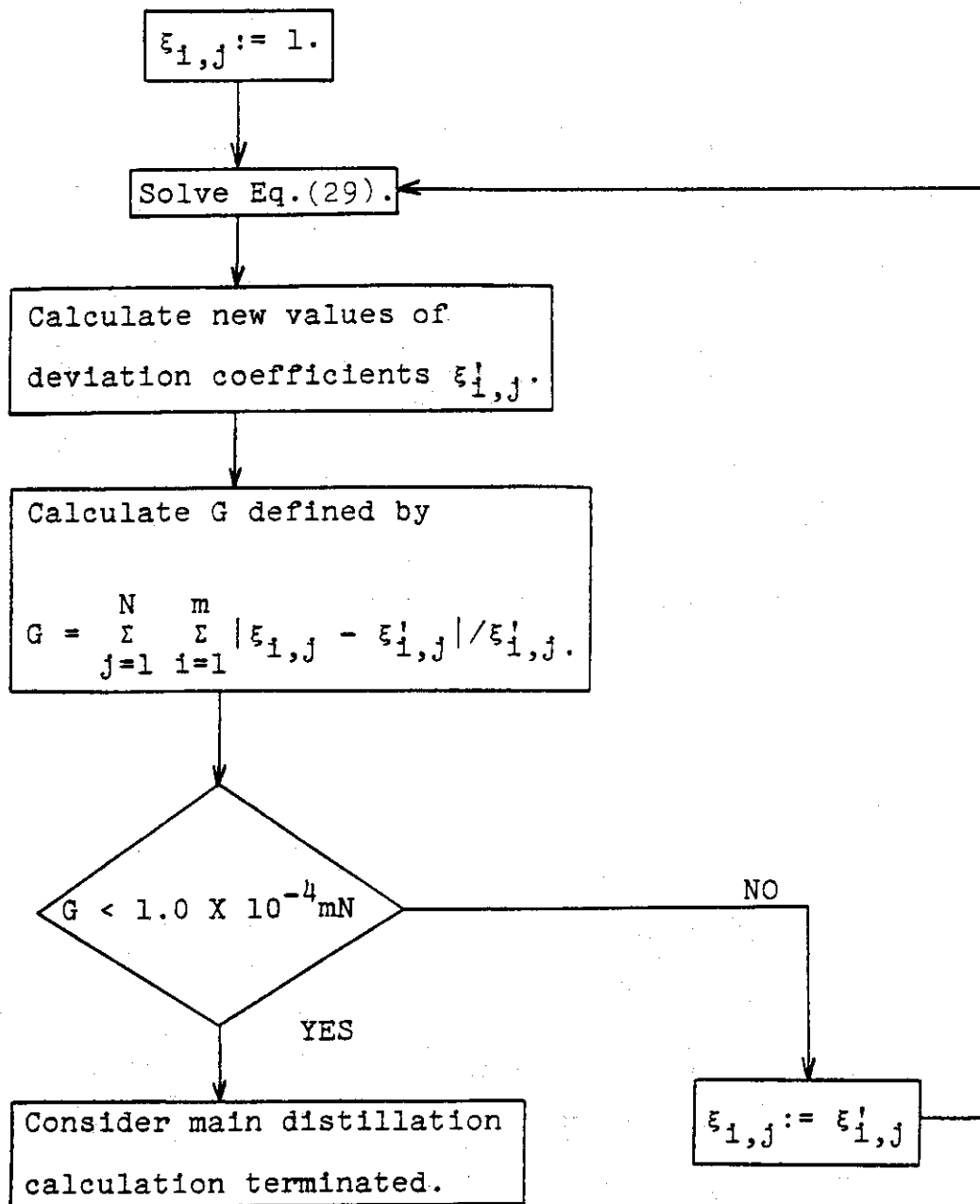


Fig. 5 Successive Iteration Loop for Incorporation of Nonideality of Hydrogen Isotope Solutions

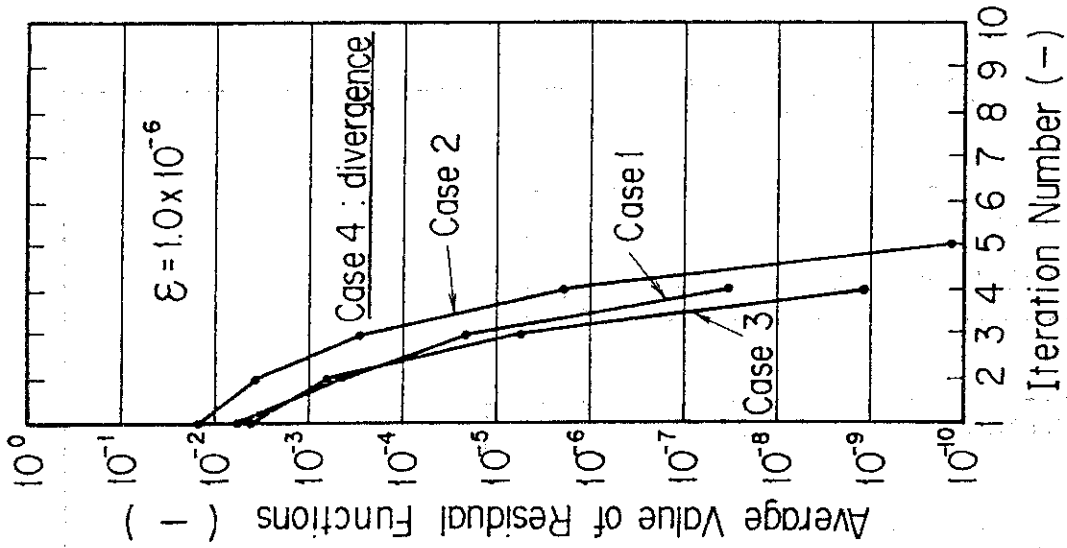


Fig. 8 Average Value of Residual Functions Plotted against Iteration Number (Column(3))

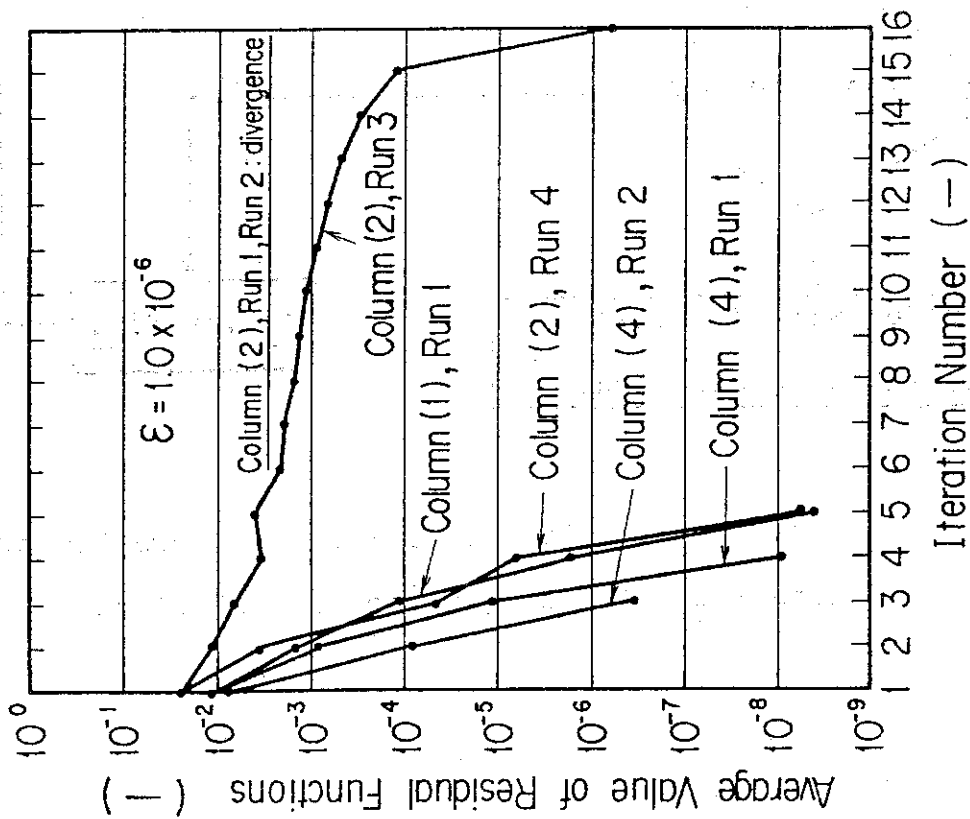


Fig. 7 Average Value of Residual Functions Plotted against Iteration Number (Columns(1), (2) and (4))

Number of Total Theoretical Stages = 80
 Feed Stage Number = 55
 Reflux Ratio = 150
 $\frac{\text{Top Product Flow Rate}}{\text{Feed Flow Rate}} = 0.029$

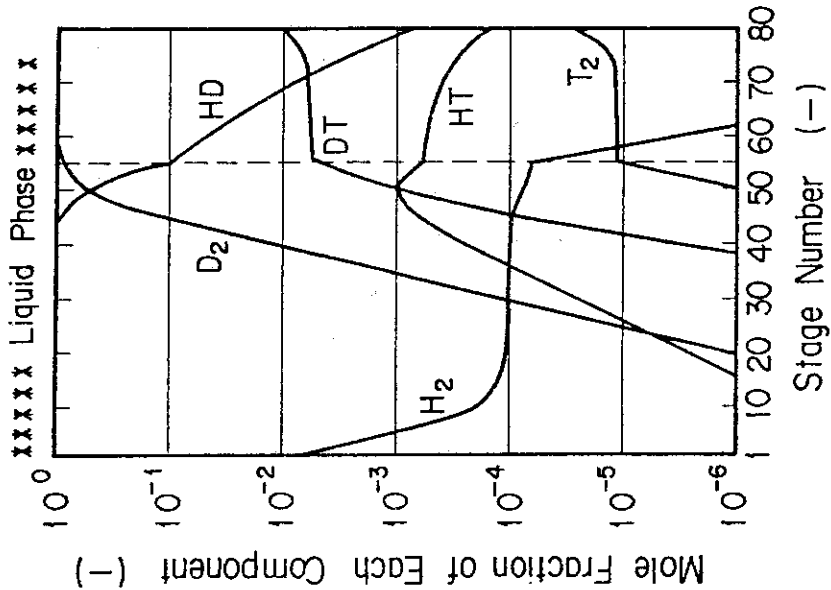


Fig. 10 Illustration of Composition Distribution within Column(2)

Number of Total Theoretical Stages = 80
 Feed Stage Number = 50
 Reflux Ratio = 25
 $\frac{\text{Top Product Flow Rate}}{\text{Feed Flow Rate}} = 0.25$

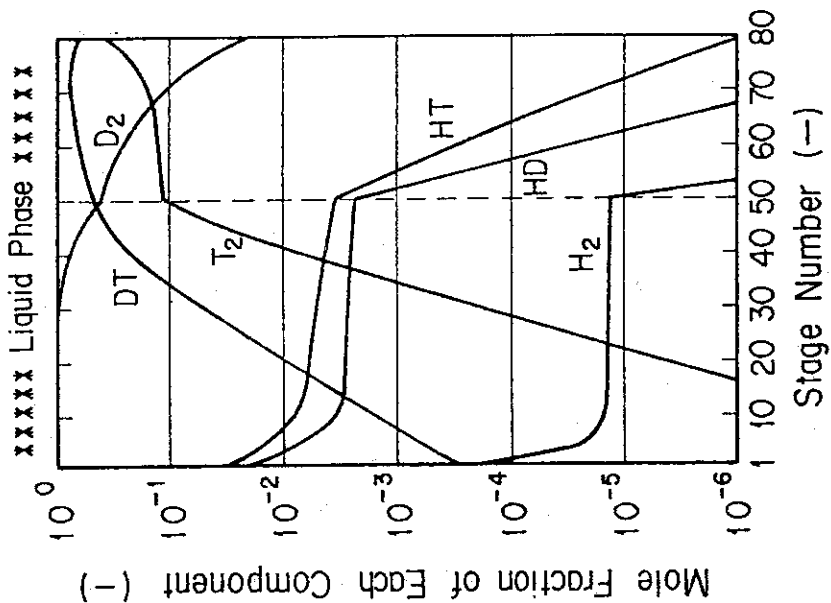


Fig. 9 Illustration of Composition Distribution within Column(1)

Number of Total Theoretical Stages = 65
 Feed Stage Number = 30
 Reflux Ratio = 8
 $\frac{\text{Top Product Flow Rate}}{\text{Feed Flow Rate}} = 0.7$

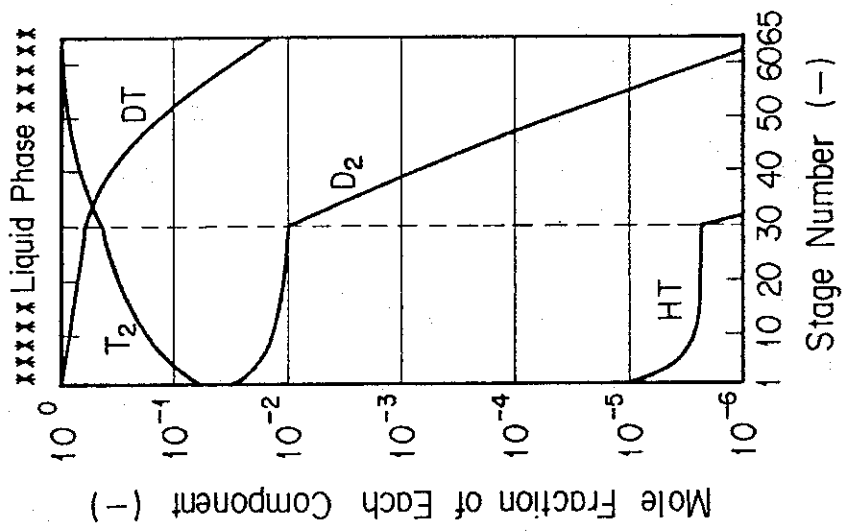


Fig. 11 Illustration of Composition Distribution within Column(3)

Number of Total Theoretical Stages = 80
 Feed Stage Number = 40
 Reflux Ratio = 8
 $\frac{\text{Top Product Flow Rate}}{\text{Feed Flow Rate}} = 0.985$

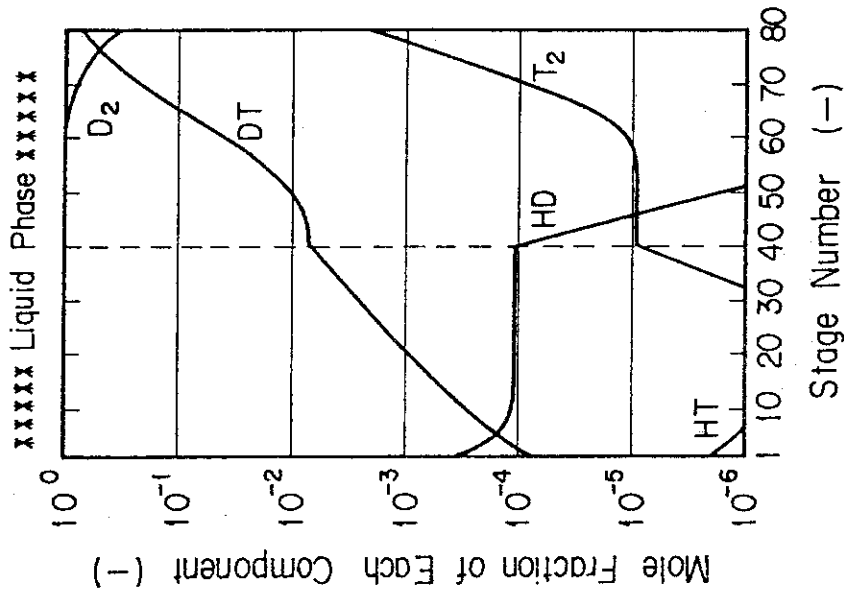


Fig. 12 Illustration of Composition Distribution within Column(4)

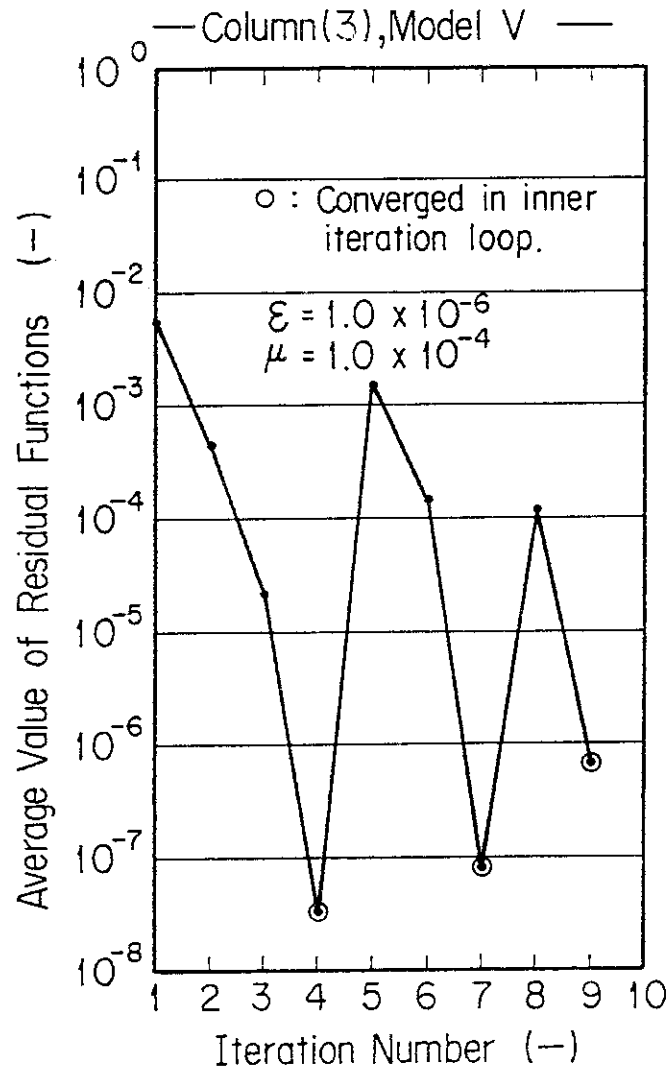


Fig. 13 Convergence Characteristics of Inner Iteration Loop and Outer Iteration Loop