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A SIMULATION CODE TREATING ALL TWELVE ISOTOPIC SPECIES
OF HYDROGEN GAS AND WATER FOR MULTISTAGE
CHEMICAL EXCHANGE COLUMN

December 1994

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A Simulation Code Treating All Twelve Isotopic Species of Hydrogen Gas
and Water for Multistage Chemical Exchange Column

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A simulation code of the multistage chemical exchange column has been developed. The column has an electrolysis cell, a section for the liquid phase catalytic exchange, and a recombiner. The sieve trays and the catalyst beds are separated in the section for the liquid-vapor scrubbing steps and for the vapor-hydrogen gas exchange steps. This type of column is a promising system for the tritiated water processing.

The code can deal with all the twelve molecular species of the hydrogen gas and the water. The equilibrium of atomic elements of H, D and T is also considered in the liquid phase. The Murphree-type factors are introduced in the code to evaluate the efficiencies for the sieve trays and catalyst beds. The solution of basic equations can be found out by the Newton-Raphson method. The atom fractions of D and T on the scrubbing trays are the independent variables of the equations: The order of the Jacobian matrix is only twice the number of sieve trays. The solution of the basic equations could be obtained for several example cases; and no difficulty was observed for the convergence of the calculations. Broyden's method was quite effective to reduce computation time of the code.

Keywords: Fusion Reactor, Isotope Separation, Water Detritiation, Tritium, Chemical Exchange, Multistage, Murphree Efficiency, Newton-raphson, Convergence, Broyden Method

水素ガス及び水における12のすべての同位体成分を取り扱う
多段型化学交換塔のためのシミュレーションコード

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(1994年11月14日受理)

多段型化学交換塔のためのシミュレーションコードを開発した。塔は、電解セル、液相化学交換部、再結合器からなり、液相化学交換部は、気液平衡のための充填部と水素-水蒸気化学交換のための触媒層が分離されている。このタイプの塔は、核融合炉の水処理系システムとして1つの有望なシステムである。

コードは、水素ガス及び水におけるすべての12の同位体成分を取り扱うことが可能である。液相における3つの原子種、H、D、Tの平衡もまた考慮されている。充填部と触媒層の効率を評価するために、マーフリー型の段効率係数を導入した。導かれた基礎式の解は、ニュートンラフソン法で求めることができる。式の独立変数は、充填部各段のDとTの原子分率であり、ヤコビアン行列の階数は充填部段数の2倍にすぎない。数列のシミュレーションに対し解を得ることができ、ニュートンラフソン法の収束困難性は認められなかった。ブロイデンの方法を試みたところ、本コードの計算時間短縮に極めて有効であることが確認できた。

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1. Introduction

In the fusion reactors, large amount of tritium-contaminated water is expected to be produced: cooling water for plasma facing components; and waste water from the tritium systems. The tritium concentration in the water is too high to discard directly to the environment, so that it is required to recover tritium from the water¹. For this situation²⁻⁴, a chemical exchange column in liquid phase is one of the most attractive process. The column is composed of an electrolysis cell, a liquid phase catalytic exchange section, and a reactor^{3,4}. The reactor is called as a recombiner. The hydrogen gas produced by the electrolysis cell at the bottom of the column flows in the liquid phase catalytic exchange section, and is oxidized to water by the recombiner. The water drops to the electrolysis cell again. The multistage separation can thus be obtained by a single column in contrast to vapor phase exchange process⁵. However, it has been known that the catalysts that cause the water/hydrogen exchange reactions often lose the activity; where the surface of the catalyst is covered with water^{6,7}. The multistage type chemical exchange column has been proposed to overcome this problem. The liquid-vapor scrubbing step proceeds in sieve trays; while vapor-hydrogen exchange step is separately caused in hydrophobic catalyst beds (See Fig. 1)^{5,8}. This type of column is expected to be promising process for the system of tritium recovery from the water for the fusion reactors.

The multistage chemical exchange column has been designed and operated for enrichment of D_2O for a fission reactor in Japan⁸. However, the characteristics of the column have not yet been understood especially for the behavior of tritium in the column. The atom fractions of tritium in the cooling and waste water of the fusion reactors are expected to be $10^{-6} \sim 10^{-8}$ ¹. The atom fractions of deuterium naturally contained in the water is $\sim 10^{-4}$. The concentrations of HDO, D_2O , and DTO are $\sim 10^{-4}$, $\sim 10^{-7}$, and $\sim 10^{-10}$. Hence, the effect of presence of deuterium in the water is not negligible. This

means that at least ten-component system ($H_2 - HD - HT - D_2 - DT - H_2O - HDO - HTO - D_2O - DTO$) must be considered to study the characteristics of the column. It is also significant to study the dynamic behavior of the column for design of control system. There are two approaches to simulate the chemical exchange column: the model using the mass transfer coefficient⁹⁻¹¹ and the stage model¹²⁻¹⁵. The sieve trays for the scrubbing step and the catalyst beds for the exchange step are completely separated in the multistage chemical exchange column. It is reasonable to apply the stage model for simulation. In addition, the stage model has great advantages: The model is appropriate to apply for the multi-component system; and the dynamic behavior of the column can easily be simulated in comparison with the other model. Kinoshita et al. has developed the simulation code of the multistage chemical exchange column by the stage model¹²⁻¹⁴. Their simulation code can deal with H-D system^{13,14}. It is necessary to develop the code for H-D-T system.

The purpose of the present study is to develop a simulation code dealing with all the twelve molecular species of hydrogen gas and water for the multistage chemical exchange column. The equilibrium of the atomic elements in the liquid phase is also taken into account. The Murphree-type factors are also introduced in the code to consider the efficiencies of scrubbing trays and catalysts beds. The code is applicable to the simulation of the column in a wide range of operating conditions.

2. Mathematical simulation procedure

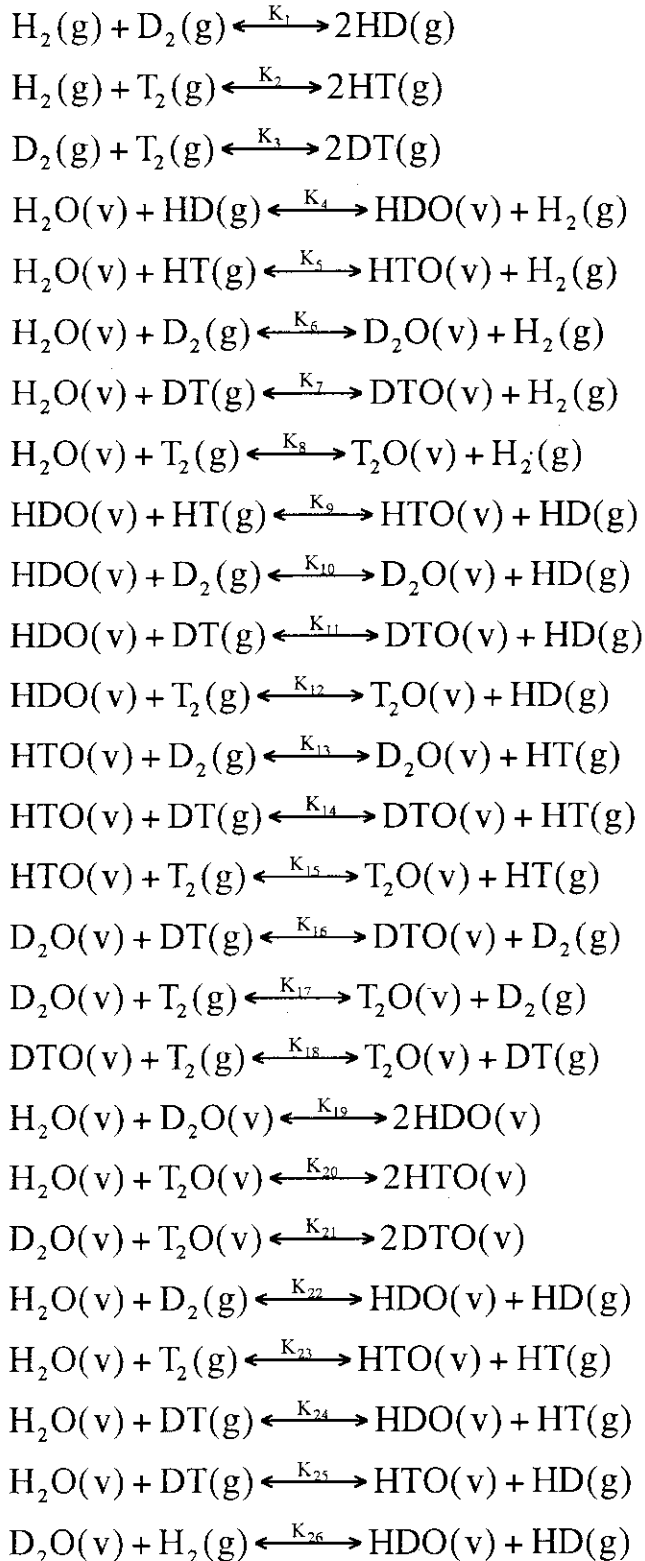
The model column is illustrated in Fig. 2. The column has the recombiner at the top and the electrolysis cell at the bottom. The liquid phase catalytic exchange section is composed of N sieve trays and $(N-1)$ catalyst beds. The model can accept multiple feed streams and multiple withdrawal streams of liquid water. All the feed streams are assumed to be supplied in liquid state. Unlike the cryogenic distillation columns within which two phases exist, the three phases must be considered in this case.

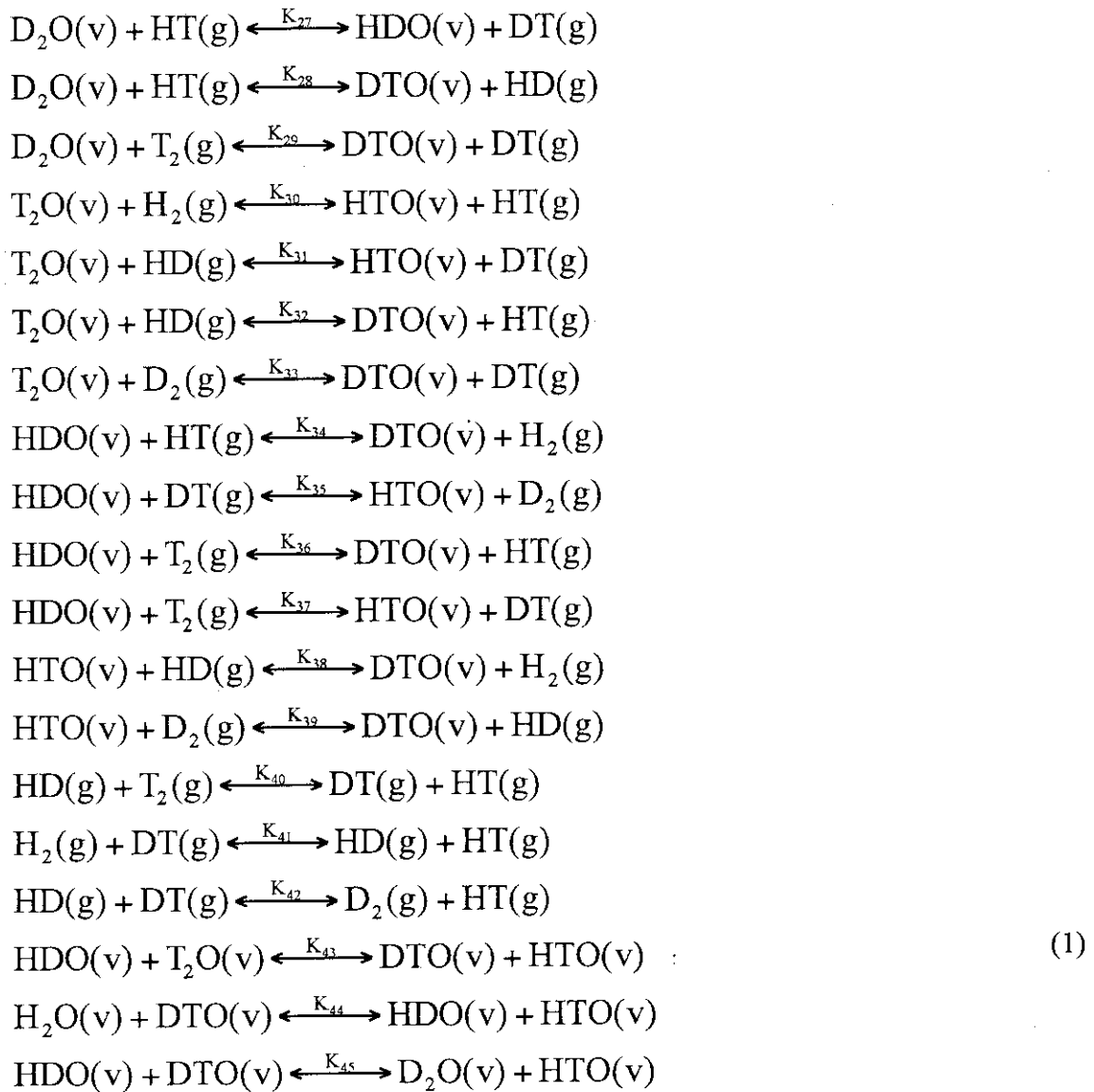
The differences in latent heat of vaporization can be neglected for the isotopes of water. An operating temperature is slightly higher than the room temperature, and a hot water jacket is attached to the column. Hence, the heat transfer through the column wall is expected to be negligible. As a consequence, the heat balance equations are not required to be taken into account, and flow rates of liquid and vapor are input variables¹².

The fusion reactors have a system composed of the cryogenic distillation columns for the hydrogen isotope separation¹. The bottom product of the chemical exchange column in the fusion reactors is a hydrogen gas stream. The hydrogen gas stream is sent to the system of the cryogenic distillation columns. The tritium in this stream is finally recovered by the cryogenic distillation system.

2.1 Basic equations for the catalyst bed

There are twelve molecular species of hydrogen gas and water vapor. We can describe the following 45 reactions between the water vapor and the hydrogen gas:





From the stoichiometric matrix of the reactions of (1), it can be shown that the rank of the matrix is eight. In other words, among 45 reactions, only eight reactions are independent. We choose the first eight reactions. For other reactions, the equilibrium constants can be described by using those of the eight reactions.

$$\begin{aligned}
K_9 &= K_5/K_4, & K_{10} &= K_6/K_4, & K_{11} &= K_7/K_4, & K_{12} &= K_8/K_4, \\
K_{13} &= K_6/K_5, & K_{14} &= K_7/K_5, & K_{15} &= K_8/K_5, & K_{16} &= K_7/K_6, \\
K_{17} &= K_8/K_6, & K_{18} &= K_8/K_7, & K_{19} &= K_1 K_4^2 / K_6, & K_{20} &= K_2 K_5^2 / K_8, \\
K_{21} &= K_3 K_7^2 / (K_6 K_8), & K_{22} &= K_4 K_1, & K_{23} &= K_5 K_2, \\
K_{24} &= K_4 \sqrt{K_1 K_2 / K_3}, & K_{25} &= K_5 \sqrt{K_1 K_2 / K_3}, & K_{26} &= K_1 K_4 / K_6, \\
K_{27} &= K_4 / (K_6 \sqrt{K_2 / K_1 K_3}), & K_{28} &= K_7 / (K_6 \sqrt{K_2 / K_1 K_3}), & K_{29} &= K_3 K_7 / K_6, \\
K_{30} &= K_2 K_5 / K_8, & K_{31} &= K_5 \sqrt{K_2 K_3 / K_1} / K_8, & K_{32} &= K_7 \sqrt{K_2 K_3 / K_1} / K_8, \\
K_{33} &= K_7 K_3 / K_8, & K_{34} &= K_7 \sqrt{K_1 K_2 / K_3} / K_4, & K_{35} &= K_5 \sqrt{K_2 / K_1 K_3} / K_4, \\
K_{36} &= K_7 \sqrt{K_2 K_3 / K_1} / K_4, & K_{37} &= K_5 \sqrt{K_2 K_3 / K_1} / K_4, \\
K_{38} &= K_7 / (K_5 \sqrt{K_1 K_2 / K_3}), & K_{39} &= K_7 / (K_5 \sqrt{K_2 / K_1 K_3}), & K_{40} &= \sqrt{K_2 K_3 / K_1}, \\
K_{41} &= \sqrt{K_1 K_2 / K_3}, & K_{42} &= \sqrt{K_2 / K_1 K_3}, & K_{43} &= K_5 K_7 \sqrt{K_2 K_3 / K_1} / (K_4 K_8), \\
K_{44} &= K_4 K_5 \sqrt{K_1 K_2 / K_3} / K_7, & K_{45} &= K_5 K_6 \sqrt{K_2 / K_1 K_3} / K_7
\end{aligned}$$

(2)

Let us assume that compositions of the input streams, $y_{i,j+1}^{\text{out}}$'s and $z_{i,j+1}$'s, are all known. There are ten unknown variables for the output streams: $y_{i,j}^{\text{in}}$ ($i = 1, 6, i \neq 2$), and $z_{i,j}$ ($i = 1, 6, i \neq 2$). From the mass balances for atomic elements around the catalyst bed, we can obtain

$$\begin{aligned}
Ga_1 + Vb_1 - G\left(\frac{z_{5,j}}{2} + z_{6,j} + \frac{z_{3,j}}{2}\right) - V\left(\frac{y_{5,j}^{\text{in}}}{2} + y_{6,j}^{\text{in}} + \frac{y_{3,j}^{\text{in}}}{2}\right) &= 0 \\
Ga_2 + Vb_2 - G\left(\frac{z_{1,j}}{2} + z_{4,j} + \frac{z_{5,j}}{2}\right) - V\left(\frac{y_{1,j}^{\text{in}}}{2} + y_{4,j}^{\text{in}} + \frac{y_{5,j}^{\text{in}}}{2}\right) &= 0 \\
a_1 &= \frac{z_{5,j+1}}{2} + z_{6,j+1} + \frac{z_{3,j+1}}{2} \\
a_2 &= \frac{z_{1,j+1}}{2} + z_{4,j+1} + \frac{z_{5,j+1}}{2} \\
b_1 &= \frac{y_{5,j+1}^{\text{out}}}{2} + y_{6,j+1}^{\text{out}} + \frac{y_{3,j+1}^{\text{out}}}{2} \\
b_2 &= \frac{y_{1,j+1}^{\text{out}}}{2} + y_{4,j+1}^{\text{out}} + \frac{y_{6,j+1}^{\text{out}}}{2}
\end{aligned} \tag{3}$$

Thus, we obtain ten equations.

Here, we introduce the Murphree-type factors to consider the efficiency of the catalyst bed.

$$\gamma = \frac{y_{i,j}^{\text{in}} - y_{i,j+1}^{\text{out}}}{y_{i,j}^{\text{in,eq}} - y_{i,j+1}^{\text{out}}}, \quad (i = 1, 6, i \neq 2) \tag{4}$$

It has been known that the rates of the first three reactions in Eq. (1) are very rapid¹⁴. We assume that the equilibrium for the three reactions is readily achieved at the outlet of the catalyst bed. The bed performance is not a monotone increasing function of γ ¹⁴. It may be better to introduce a factor for each hydrogen-water exchange reaction. However, there has been no experimental data for the efficiency of catalyst bed; and it is quite difficult to give a reasonable value for the factor of each reaction. For these reasons, we introduce a factor for the efficiency of the catalyst bed.

From Eqs. (1), (3), and (4), we obtain

$$\begin{aligned}
 F_1 &= z_{1,j}^2 - K_1 z_{4,j} (1.0 - z_{1,j} - z_{3,j} - z_{4,j} - \sqrt{K_3 z_{6,j} z_{5,j}} - z_{6,j}) = 0 \\
 F_2 &= z_{3,j}^2 - K_2 z_{6,j} (1.0 - z_{1,j} - z_{3,j} - z_{4,j} - \sqrt{K_3 z_{6,j} z_{5,j}} - z_{6,j}) = 0 \\
 F_3 &= Ga_2 + Vb_2 - G \left(\frac{z_{1,j}}{2} + z_{4,j} + \frac{\sqrt{K_3 z_{4,j} z_{6,j}}}{2} \right) - \\
 &V\gamma \left\{ \left(\frac{K_5 z_{1,j}}{2} + K_6 z_{4,j} + \frac{K_7 \sqrt{K_3 z_{4,j} z_{6,j}}}{2} \right) / \text{xx} \right\} - V(1-\gamma)b_2 = 0 \\
 F_4 &= Ga_1 + Vb_1 - G \left(\frac{z_{3,j}}{2} + z_{6,j} + \frac{\sqrt{K_3 z_{4,j} z_{6,j}}}{2} \right) - \\
 &V\gamma \left\{ \left(\frac{K_4 z_{3,j}}{2} + K_8 z_{6,j} + \frac{K_7 \sqrt{K_3 z_{4,j} z_{6,j}}}{2} \right) / \text{xx} \right\} - V(1-\gamma)b_1 = 0 \\
 \text{xx} &= (K_5 - 1)z_{1,j} + (K_4 - 1)z_{3,j} + (K_6 - 1)z_{4,j} + \\
 &(K_7 - 1)\sqrt{K_3 z_{4,j} z_{6,j}} + (K_8 - 1)z_{6,j} + 1.0 \tag{5}
 \end{aligned}$$

Consequently, for four unknown variables of $z_{1,j}, z_{3,j}, z_{4,j}, z_{6,j}$, four equations are derived.

We can solve these equations by the Newton-Raphson method:

$$\begin{aligned}
 \mathbf{z}^{k+1} &= \mathbf{z}^k + \Delta \mathbf{z}^k, \\
 \Delta \mathbf{z}^k &= -\mathbf{J}_F^{-1} \mathbf{F}, \\
 \mathbf{z} &= \begin{pmatrix} z_{1,j} \\ z_{3,j} \\ z_{4,j} \\ z_{6,j} \end{pmatrix}, \quad \mathbf{F} = \begin{pmatrix} F_1 \\ F_2 \\ F_3 \\ F_4 \end{pmatrix}, \\
 \mathbf{J}_F &= \begin{pmatrix} \partial F_1 / \partial z_{1,j} & \cdot & \cdot & \partial F_1 / \partial z_{6,j} \\ \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot \\ \partial F_4 / \partial z_{1,j} & \cdot & \cdot & \partial F_4 / \partial z_{6,j} \end{pmatrix}
 \end{aligned}
 \tag{6}$$

2.2 Basic equations for the scrubbing trays

The mass balance equations for atomic element of H can be derived as follows:

$$\begin{pmatrix} B_1 & C_1 & \cdot & \cdot & \cdot & \cdot \\ A_2 & B_2 & C_2 & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & A_{N-1} & B_{N-1} & C_{N-1} \\ \cdot & \cdot & \cdot & \cdot & A_N & B_N \end{pmatrix} \begin{pmatrix} X_{H,1}^{\text{out}} \\ \cdot \\ \cdot \\ \cdot \\ \cdot \\ X_{H,N}^{\text{out}} \end{pmatrix} = \begin{pmatrix} E_1 \\ \cdot \\ \cdot \\ \cdot \\ \cdot \\ E_N \end{pmatrix}, \tag{7}$$

where

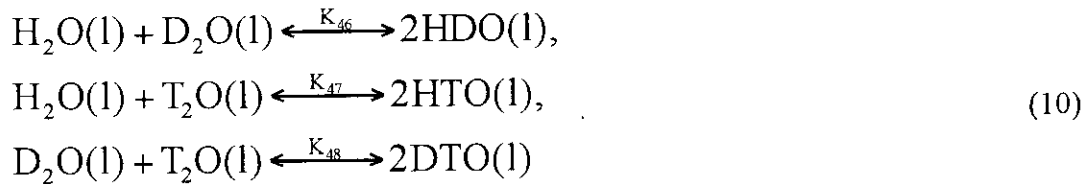
$$\begin{aligned}
 A_j &= -L_{j-1}, & B_j &= L_j + U_j + \kappa_{H,j}V, & C_j &= -V\kappa_{H,j+1}, \\
 E_1 &= (V + G - D)X_{H,1}^{\text{in}} + V\beta_{H,1} + O_1Z_{FH,1}, \\
 E_j &= V\beta_{H,j} + O_jZ_{FH,j}, \\
 E_N &= vY_{H,N}^{\text{in}} + O_NZ_{FH,N}, \\
 \beta_{H,j} &= \frac{y_{1,j}^{\text{in}} - y_{1,j+1}^{\text{out}}}{2} + y_{2,j}^{\text{in}} - y_{2,j+1}^{\text{out}} + \frac{y_{3,j}^{\text{in}} - y_{3,j+1}^{\text{out}}}{2}, \\
 \kappa_{H,j} &= \left(\frac{y_{1,j}^{\text{out}}}{2} + y_{2,j}^{\text{out}} + \frac{y_{3,j}^{\text{out}}}{2} \right) / \left(\frac{x_{1,j}^{\text{out}}}{2} + x_{2,j}^{\text{out}} + \frac{x_{3,j}^{\text{out}}}{2} \right), \\
 X_{H,j}^{\text{in,out}} &= \frac{x_{1,j}^{\text{in,out}}}{2} + x_{2,j}^{\text{in,out}} + \frac{x_{3,j}^{\text{in,out}}}{2}, & Y_{H,j}^{\text{in,out}} &= \frac{y_{1,j}^{\text{in,out}}}{2} + y_{2,j}^{\text{in,out}} + \frac{y_{3,j}^{\text{in,out}}}{2}, \\
 Z_{FH,j} &= \frac{Z_{F1,j}}{2} + Z_{F2,j} + \frac{Z_{F3,j}}{2}, & x_{i,j}^{\text{in}} &= x_{i,j-1}^{\text{out}}
 \end{aligned} \tag{8}$$

Similar equations are derived for atomic elements of D and T also.

We introduce the Murphree-type factor for the efficiency of the scrubbing trays also¹⁴:

$$\alpha = \frac{y_{i,j}^{\text{out}} - y_{i,j}^{\text{in}}}{y_{i,j}^{\text{out,eq}} - y_{i,j}^{\text{in}}}, \quad y_{i,j}^{\text{out,eq}} = \frac{(V + G)p_i^*(T_j)x_{i,j}^{\text{out}}}{VP_j} \tag{9}$$

For the liquid water phase, the three equilibrium reactions are described by



The equilibrium compositions can be obtained by

$$Q_1 x_{1,j}^{\text{out}^4} + Q_2 x_{1,j}^{\text{out}^3} + Q_3 x_{1,j}^{\text{out}^2} + Q_4 x_{1,j}^{\text{out}} + Q_5 = 0,$$

$$Q_1 = q_1^2 q_6 + q_1 q_4 q_8,$$

$$Q_2 = 2q_1 q_2 q_6 + q_2 q_4 q_8 + q_1 q_5 q_8 + q_1 q_4 q_7 + q_4^2 q_9,$$

$$Q_3 = 2q_1 q_3 q_6 + q_2^2 q_6 + q_1 q_5 q_7 + q_2 q_5 q_8 + q_2 q_4 q_7 + q_3 q_4 q_8 \\ + 2q_4 q_5 q_9 + q_4^2 q_{10},$$

$$Q_4 = 2q_2 q_3 q_6 + q_2 q_5 q_7 + q_3 q_5 q_8 + q_3 q_4 q_7 + q_5^2 q_9 + 2q_4 q_5 q_{10},$$

$$Q_5 = q_3^2 q_6 + q_3 q_5 q_7 + q_5^2 q_{10},$$

$$q_1 = \frac{2 - K_{46}}{2}, \quad q_2 = K_{46} (1 - X_{T,j}^{\text{out}}), \quad q_3 = -2K_{46} X_{D,j}^{\text{out}} X_{T,j}^{\text{out}}$$

$$q_4 = \frac{K_{46}}{2} - \sqrt{K_{46} K_{47} / K_{48}}, \quad q_5 = -K_{46} X_{H,j}^{\text{out}}, \quad q_6 = \frac{2 - K_{48}}{2},$$

$$q_7 = K_{48} (1 - X_{H,j}^{\text{out}}), \quad q_8 = \sqrt{K_{47} K_{48} / K_{46}} - \frac{K_{48}}{2}, \quad q_9 = K_{48} X_{T,j}^{\text{out}},$$

$$q_{10} = -2K_{48} X_{T,j}^{\text{out}} X_{D,j}^{\text{out}}, \quad x_{5,j}^{\text{out}} = (q_1 x_{1,j}^{\text{out}} + q_2 x_{1,j}^{\text{out}} + q_3) / (q_4 x_{1,j}^{\text{out}} + q_5),$$

$$x_{4,j}^{\text{out}} = X_{D,j}^{\text{out}} - \frac{x_{1,j}^{\text{out}}}{2} - \frac{x_{5,j}^{\text{out}}}{2}, \quad x_{6,j}^{\text{out}} = \frac{x_{5,j}^{\text{out}^2}}{K_{24} x_{4,j}^{\text{out}}},$$

$$x_{3,j}^{\text{out}} = 2 \left(X_{T,j}^{\text{out}} - x_{6,j}^{\text{out}} - \frac{x_{5,j}^{\text{out}}}{2} \right), \quad x_{2,j}^{\text{out}} = 1 - x_{1,j}^{\text{out}} - x_{3,j}^{\text{out}} - x_{4,j}^{\text{out}} - x_{5,j}^{\text{out}} - x_{6,j}^{\text{out}}$$

(11)

2.3 Basic equations for the electrolysis cell

We introduce two factors to consider the separation efficiency at the electrolysis cell.

$$\begin{aligned}\eta_D &= \left\{ (1 - Z_{D,N}) / Z_{D,N} \right\} / \left\{ (1 - Y_{D,N}^{\text{in}}) / Y_{D,N}^{\text{in}} \right\}, \\ \eta_T &= \left\{ (1 - Z_{T,N}) / Z_{T,N} \right\} / \left\{ (1 - Y_{T,N}^{\text{in}}) / Y_{T,N}^{\text{in}} \right\}, \\ Z_{D,N} &= z_{4,N} + \frac{z_{1,N}}{2} + \frac{z_{5,N}}{2}, \quad Z_{T,N} = z_{6,N} + \frac{z_{3,N}}{2} + \frac{z_{5,N}}{2}\end{aligned}\quad (12)$$

From the mass balances of atomic elements of D and T around the electrolysis cell,

$$\begin{aligned}L_N X_{D,N}^{\text{out}} &= (G + W) Z_{D,N} + v Y_{D,N}^{\text{in}}, \\ L_N X_{T,N}^{\text{out}} &= (G + W) Z_{T,N} + v Y_{T,N}^{\text{in}}\end{aligned}\quad (13)$$

Substituting Eqs. (12) into (13), we obtain

$$\begin{aligned}Y_{D,N}^{\text{in}} &= \left(\sqrt{o_1^2 - 4o_2o_3} - o_1 \right) / 2o_2, \\ Y_{T,N}^{\text{in}} &= \left(\sqrt{o_4^2 - 4o_5o_6} - o_4 \right) / 2o_5, \\ o_1 &= v\eta_D + G + W - L_N X_{D,N}^{\text{out}} (1 - \eta_D), \\ o_2 &= v(1 - \eta_D), \quad o_3 = L_N X_{D,N}^{\text{out}} \eta_D, \\ o_4 &= v\eta_T + G + W - L_N X_{T,N}^{\text{out}} (1 - \eta_T), \\ o_5 &= v(1 - \eta_T), \quad o_6 = L_N X_{T,N}^{\text{out}} \eta_T\end{aligned}$$

In the liquid phase, the equilibrium reactions of (10) must be considered:

$$\begin{aligned}
 y_{1,N}^{in} / (y_{2,N}^{in} y_{4,N}^{in}) &= K_{22} \left\{ (p_1^*(T_e))^2 / p_2^*(T_e) p_4^*(T_e) \right\}, \\
 y_{3,N}^{in} / (y_{2,N}^{in} y_{6,N}^{in}) &= K_{23} \left\{ (p_3^*(T_e))^2 / p_2^*(T_e) p_6^*(T_e) \right\}, \\
 y_{5,N}^{in} / (y_{6,N}^{in} y_{4,N}^{in}) &= K_{22} \left\{ (p_5^*(T_e))^2 / p_6^*(T_e) p_4^*(T_e) \right\}
 \end{aligned} \tag{15}$$

If the atom fractions of D and T in the water stream to the electrolysis cell are given, $y_{i,N}^{in}$'s and $z_{i,n}$'s can be calculated from Eqs. (14), (15) and (11).

2.4 Bubble point calculation

The temperatures of sieve trays, T_j 's, can be calculated by the Newton method:

$$\begin{aligned}
 f(T_j) &= \sum_{i=1}^m y_{i,j}^{out,eq} - 1 = 0, \\
 T_j^{k+1} &= T_j^k + \Delta T_j^k, \\
 \Delta T_j^k &= -f(T_j^k) / \frac{df(T_j^k)}{dT_j^k}
 \end{aligned} \tag{16}$$

2.5 Physical properties

We need the vapor pressure of water and equilibrium constants of the reactions. The vapor pressure of the six isotopic species of water can be calculated by the equations reported by Van Hook¹⁶.

$$p_i^*(T_j) = 1.013 \times 10^5 \exp\left[\phi_{1,i}/T_j^2 + \phi_{2,i}/T_j + \phi_{3,i}/(T_j + \phi_{4,i}) + \phi_{5,i}\right] \quad (17)$$

We describe the equilibrium constants as a function of temperature from the values at several temperatures listed in Ref. 17.

$$\ln(K) = \lambda_1/T + \lambda_2 \quad (18)$$

The parameters of Eqs. (17) and (18) are presented in Tables 1 and 2, respectively.

2.6 Simulation procedure

The basic equations derived in preceding sections can be solved by the Newton-Raphson method. The simulation procedure is as follows.

- (i) Assume the atom fractions of D and T in liquid water streams leaving the sieve trays, $X_{D,j}^{\text{out}}$'s and $X_{T,j}^{\text{out}}$'s. A practical method is that all the atom fractions of D and T are equal to the average values of all the feed streams:

$$\begin{aligned}
 X_{D,j}^{\text{out}} S &= \sum_{j=1}^N \left(Z_{F4,j} + \frac{Z_{F1,j}}{2} + \frac{Z_{F5,j}}{2} \right) / \sum_{j=1}^N O_j, \\
 X_{T,j}^{\text{out}} S &= \sum_{j=1}^N \left(Z_{F6,j} + \frac{Z_{F5,j}}{2} + \frac{Z_{F3,j}}{2} \right) / \sum_{j=1}^N O_j
 \end{aligned}
 \tag{19}$$

(ii) Assume the temperatures of the sieve trays. From Eq. (11), $x_{i,j}^{\text{out}}$'s are calculated.

(iii) By the bubble point calculation, Eq. (16), T_j 's and $y_{i,j}^{\text{out,cq}}$'s become all known. If the temperatures assumed in step (ii) are not equal to the values calculated by the bubble point calculation, return to step (ii).

(iv) Since $x_{i,N}^{\text{out}}$'s are known, $y_{i,N}^{\text{in}}$'s and $z_{i,N}$'s can be calculated from Eqs. (11), (14) and (15).

(v) By accounting for the scrubbing efficiencies for the N'th tray, $y_{i,N}^{\text{out}}$'s are calculated from Eq. (9).

(vi) Then, the catalyst beds are treated. From $y_{i,N}^{\text{out}}$'s and $z_{i,N}$'s, $y_{i,N-1}^{\text{in}}$ and $z_{i,N-1}$'s can be calculated with the efficiency of the catalyst bed by Eqs. (5) and (6).

(vii) Steps (v) and (vi) are sequentially carried out from $j=N-1$ to 1. Thus, $y_{i,j}^{\text{out}}$'s, $y_{i,j}^{\text{in}}$'s, and $z_{i,j}$'s for $j=1$ to N become known.

(viii) Now we can evaluate all the values of A_j 's, B_j 's, C_j 's, and E_j 's in Eq. (8). The new values of $X_{H,j}^{\text{out}}$'s, $X_{D,j}^{\text{out}}$'s, and $X_{T,j}^{\text{out}}$'s are obtained by solving the tridiagonal equations of Eq. (8) three times. Normalize the new atom fractions of H, D and T.

(ix) Denote the value of $X_{D,j}^{\text{out}}$'s and $X_{T,j}^{\text{out}}$'s assumed at step (i) by $(X_{D,j}^{\text{out}})_0$ and $(X_{T,j}^{\text{out}})_0$. The values of $X_{D,j}^{\text{out}}$'s and $X_{T,j}^{\text{out}}$'s are not equal to $(X_{D,j}^{\text{out}})_0$ and $(X_{T,j}^{\text{out}})_0$.

We define the functions of S_j 's by

$$\begin{aligned}
 S_j &= (X_{D,j}^{\text{out}})_0 - X_{D,j}^{\text{out}} = 0 \quad (j=1, N), \\
 S_{j+N} &= (X_{T,j}^{\text{out}})_0 - X_{T,j}^{\text{out}} = 0 \quad (j=1, N)
 \end{aligned}
 \tag{20}$$

The simulation can be understood to find out the solution of Eq. (21). This is carried out by the 2N-variable Newton-Raphson method.

$$\begin{aligned}
 \mathbf{X}^{k+1} &= \mathbf{X}^k + \Delta \mathbf{X}^k, \\
 \Delta \mathbf{X}^k &= -\mathbf{J}_S^{-1} \mathbf{S},
 \end{aligned}$$

$$\mathbf{X} = \begin{pmatrix} X_{D,1}^{\text{out}} \\ \cdot \\ X_{D,N}^{\text{out}} \\ X_{T,1}^{\text{out}} \\ \cdot \\ X_{T,N}^{\text{out}} \end{pmatrix}, \quad \mathbf{S} = \begin{pmatrix} S_1 \\ \cdot \\ S_N \\ S_{N+1} \\ \cdot \\ S_{2N} \end{pmatrix},$$

$$\mathbf{J}_S = \begin{pmatrix} \partial S_1 / \partial (X_{D,1}^{\text{out}})_0 & \cdot & \cdot & \partial S_{2N} / \partial (X_{D,1}^{\text{out}})_0 \\ \cdot & \cdot & \cdot & \cdot \\ \partial S_1 / \partial (X_{D,N}^{\text{out}})_0 & \cdot & \cdot & \partial S_{2N} / \partial (X_{D,N}^{\text{out}})_0 \\ \partial S_1 / \partial (X_{T,1}^{\text{out}})_0 & \cdot & \cdot & \partial S_{2N} / \partial (X_{T,1}^{\text{out}})_0 \\ \cdot & \cdot & \cdot & \cdot \\ \partial S_1 / \partial (X_{T,N}^{\text{out}})_0 & \cdot & \cdot & \partial S_{2N} / \partial (X_{T,N}^{\text{out}})_0 \end{pmatrix} \quad (21)$$

The partial derivatives are numerically calculated:

$$\partial S_j / (X_{D,k}^{\text{out}})_0 = \left(S_j |_{(X_{D,k}^{\text{out}})_0} - S_j |_{(X_{D,k}^{\text{out}})_0 - \Delta (X_{D,k}^{\text{out}})_0} \right) / \Delta (X_{D,k}^{\text{out}})_0 \quad (22)$$

(x) Return to step (ii). If S_j 's are equal to zero, the variables in steps (i) through (x) satisfy all the basic equations derived. The simulation problem is considered solved once

$$\sum_{j=1}^{2N} |S_j| / 2N \leq \varepsilon, \quad \varepsilon \approx 10^{-10} \quad (23)$$

is satisfied. The simulation procedure is illustrated in Fig. 3.

3. Reduction of computation time

The order of Jacobian matrix is only twice the number of sieve trays in the present simulation code. However, if the number of sieve trays is large, long computation time is expected for evaluation of the Jacobian matrix and its inversion. To overcome this problem, we introduce Broyden's method¹⁸. The evaluation of the Jacobian matrix and its inversion are needed only once in his method. In the present section, his method is applied to the solution of Eq. (20).

The Jacobian matrix J_s is given in Eq. (21). Expressing $-J_s^{-1}$ by H , the simulation procedure can be summarized as follows:

- (i) Obtain an initial estimate X^0 .
- (ii) Calculate J_s and invert it. Thus H^0 is obtained. Also calculate S^0 .
- (iii) Compute $\Delta X^0 = H^0 S^0$.
- (iv) Calculate $X^1 = X^0 + \tau \Delta X^0$. In the case of $\varphi^1 \leq \varphi^0$, τ is set at unity. In the case of $\varphi^1 \geq \varphi^0$, τ is calculated from

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$$\tau = \left\{ (1 + 6\theta)^{1/2} - 1 \right\} / 3\theta, \quad \theta = \varphi^1 / \varphi^0,$$

$$\varphi^1 = \sum_{j=1}^{2N} \left(S_j |_{X^0 + \Delta X^0} \right)^2, \quad \varphi^0 = \sum_{j=1}^{2N} \left(S_j |_{X^0} \right)^2 \quad (24)$$

Thus, X^1 and S^1 are calculated.

(v) If the convergence has not been achieved yet, calculate H^1 from

$$H^1 = H^0 - \left\{ H^0 (S^1 - S^0) + \tau \Delta X^0 \right\} (\Delta X^0)^T H^0 / \left\{ (\Delta X^0)^T H^0 (S^1 - S^0) \right\},$$

$$(X)^T = (X_1, \dots, X_{2N}) \quad (25)$$

(vi) The values of X^1, S^1, H^1 are substituted for X^0, S^0, H^0 , and repeat from step (iii).

The Newton-Raphson method is replaced by this method.

The Newton-Raphson method sometimes fails to get convergence. If a set of appropriate initial values is given, we can get the convergence even for this case. The initial values of the present simulation code are atom fractions of D and T on sieve trays. A possible method is that either top or bottom product composition is assumed¹³. The other product composition can be calculated from the overall mass balance of the column. Then, the atom fractions of D and T on sieve trays are linearly set from top and bottom atom fractions. However, it is not easy task to assume an appropriate product composition. We propose the following procedure for setting the initial values for the case where it is difficult to get the convergence:

- (i) The initial values are calculated from Eq. (19).
- (ii) Solve Eqs. (5), (6), (9), (11), (14), and (15). Evaluate all the values in Eq. (8).
- (iii) Solve the tridiagonal equations of Eq. (7) three times. Normalize the new atom fractions of H, D and T. These values are used for the initial values of the Newton-Raphson iterations.

4. Numerical examples

A computer program was written in FORTRAN statements. Computations were performed in the double precision. The system used was FACOM M-780 at the Japan Atomic Energy Research Institute. For all the cases tested, no difficulty for the convergence was observed. No technique for the setting the initial values was required. An example case is presented in Table 3 and 4. The convergence is obtained by only three iterations. The composition of the water supplied to the column is equal to that of the waste water of ITER¹. The tritium concentration can be reduced to $\sim 1/400$ by the column having only 35 sieve trays. If we get the same separation performance by the water distillation column, about 200-300 stages are required¹. The chemical exchange column is thus attractive process.

Broyden's method was also tried to reduce the computation time. As shown in Table 4, the method can reduce the computation time to half of that for the Newton-Raphson method without increasing the number of iterations. Kinoshita reported that it was required to evaluate the Jacobian matrix only once¹⁵. He obtained the convergence by the Jacobian matrix which were calculated for the initial values for his code of H-D system. Even Broyden's method was not necessary for his code. However, for the present code of H-D-T system, we could not get the convergence by the Jacobian matrix for the initial values; in spite of the setting method of initial values. The application of Broyden's method is thus quite effective for the present simulation code.

5. Conclusion

A simulation code for the multistage chemical exchange column has been developed. Some significant features of this code are summarized here.

- (1) The code can deal with all the twelve molecular species of hydrogen gas and water. The equilibrium of atomic elements of H, D and T in the liquid phase is also considered in the code. The separation factor between the isotopic species depends on the temperature and composition. This dependency is fully taken into account.
- (2) The Murphree-type factors are introduced in the code to evaluate the efficiencies for both of the sieve trays and catalyst beds.
- (3) The main calculation loop is based on the Newton-Raphson iteration. The order of the Jacobian matrix is only twice the number of the sieve trays.
- (4) The great stability and high converging speed were verified. It was also verified that Broyden's method was effective for this code: The computation time was reduced to $\sim 1/2$ without increasing the number of the iterations.

Acknowledgment

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Nomenclature

- D : Flow rate of bottom product (mol/h)
- G : Flow rate of hydrogen gas (mol/h)
- J : Jacobian matrix (-)
- K : Equilibrium constant of reaction (-)
- L : Flow rate of liquid water (mol/h)
- N : Total number of sieve trays (-)
- N_o : Feed tray number (-)
- O : Flow rate of feed stream (mol/h)
- P : Total pressure (Pa)
- p^* : Vapor pressure of pure component for water (Pa)
- R : Reflux ratio = $(V+G-D)/D$ (-)
- T : Temperature (K)
- T_{av} : Temperature of hot-water bath (operating temperature) (K)
- T_c : Temperature at electrolysis cell (K)
- U : Flow rate of side stream of water (mol/h)
- V : Flow rate of water vapor stream (mol/h)
- v : Flow rate of water vapor stream from electrolysis cell (mol/h)
- X : Atomic fraction of element in liquid water (-)
- x : mole fraction of component in liquid water (-)
- Y : Atomic fraction of element in water vapor (-)
- y : Mole fraction of component in water vapor (-)
- Z : Atomic fraction of element in hydrogen gas (-)
- z : Mole fraction of component in hydrogen gas (-)
- Z_F : Atomic fraction of element in feed water (-)
- z_F : Mole fraction of component in feed water (-)
- α : Murphree-type scrubbing efficiency (-)

γ : Murphree-type catalytic efficiency (-)

Subscripts

i : Component or element index

1 : HD, HDO; 2 : H₂, H₂O; 3 : HT, HTO; 4 : D₂, D₂O; 5 : DT, DTO; 6 : T₂, T₂O

H : Hydrogen; D : Deuterium; T : Tritium

j : Tray index

Superscripts

eq : Equilibrium composition

k : Iteration number

in : Entering

out : Leaving

For Instance,

$x_{1,j}^{\text{out}}$: Mole fraction of HDO in liquid water stream leaving j -th tray

$Y_{D,j}^{\text{in}}$: Atomic fraction of deuterium in water vapor stream entering j -th tray

$z_{6,j}$: Mole fraction of T₂ in hydrogen gas leaving j -th catalyst bed and going through j -th tray

In reaction equations, l means liquid phase, g means hydrogen gas phase, and v means vapor phase.

Table 1 Parameters of Equation (17)

	ϕ_1	ϕ_2	ϕ_3	ϕ_4	ϕ_5
H ₂	0.0	0.0	-3578.5	-54.15	11.20
HD	-26398.8	89.6065	-3578.5	-54.15	11.124198
HT	-37813.2	136.751	-3578.5	-54.15	11.075904
D ₂	-49314.9	164.266	-3578.5	-54.15	11.059951
DT	-59313.4	204.941	-3578.5	-54.15	11.017314
T ₂	-68702.3	244.687	-3578.5	-54.15	10.975612

Table 2 Parameters of Equation (18)

	λ_1	λ_2
K ₁	-77.5	1.441
K ₂	-176.0	1.528
K ₃	-20.2	1.406
K ₄	472.0	-0.297
K ₅	673.0	-0.434
K ₆	923.0	-0.667
K ₇	1072.0	-0.7354
K ₈	1189.0	-0.669
K ₄₆	-13.0	1.419
K ₄₇	-71.7	1.469
K ₄₈	-35.6	1.397

Table 3 Input specifications for an example case

N	35	
N _O	30	
O	1111 mol/h	
D	1000 mol/h	
B	111 mol/h	
P	101.3 kPa	
Pressure Drop per Tray	66 Pa	
R	1.0	
T _{av}	333.15 K	
Feed	H ₂ O:0.9996	HDO:4.0x10 ⁻⁴
Composition	HTO:6.0x10 ⁻⁸	D ₂ O:4.0x10 ⁻⁸
	DTO:1.2x10 ⁻¹¹	T ₂ O: 1.0x10 ⁻¹⁵

$$R = (V + G - D)/D$$

The values of α and γ are unity.

Table 4 Calculated results for an example case

Top Product	H ₂ O:0.99995	HDO:5.3x10 ⁻⁵
Composition	HTO:1.6x10 ⁻¹⁰	D ₂ O:8.2x10 ⁻¹⁰
	DTO:5.3x10 ⁻¹⁵	T ₂ O:9.3x10 ⁻²¹
Bottom Product	H ₂ :0.99647	HD:3.5x10 ⁻³
Composition	HT:6.0x10 ⁻⁷	D ₂ :3.8x10 ⁻⁶
	DT:1.4x10 ⁻⁹	T ₂ :1.4x10 ⁻¹³
Iteration Number	3	
	3*	
Computation Time	2.18	
	1.09*	
v	51.66 mol/h	
V	398.1 mol/h	
G	1601.9 mol/h	

* Broyden's method is used.

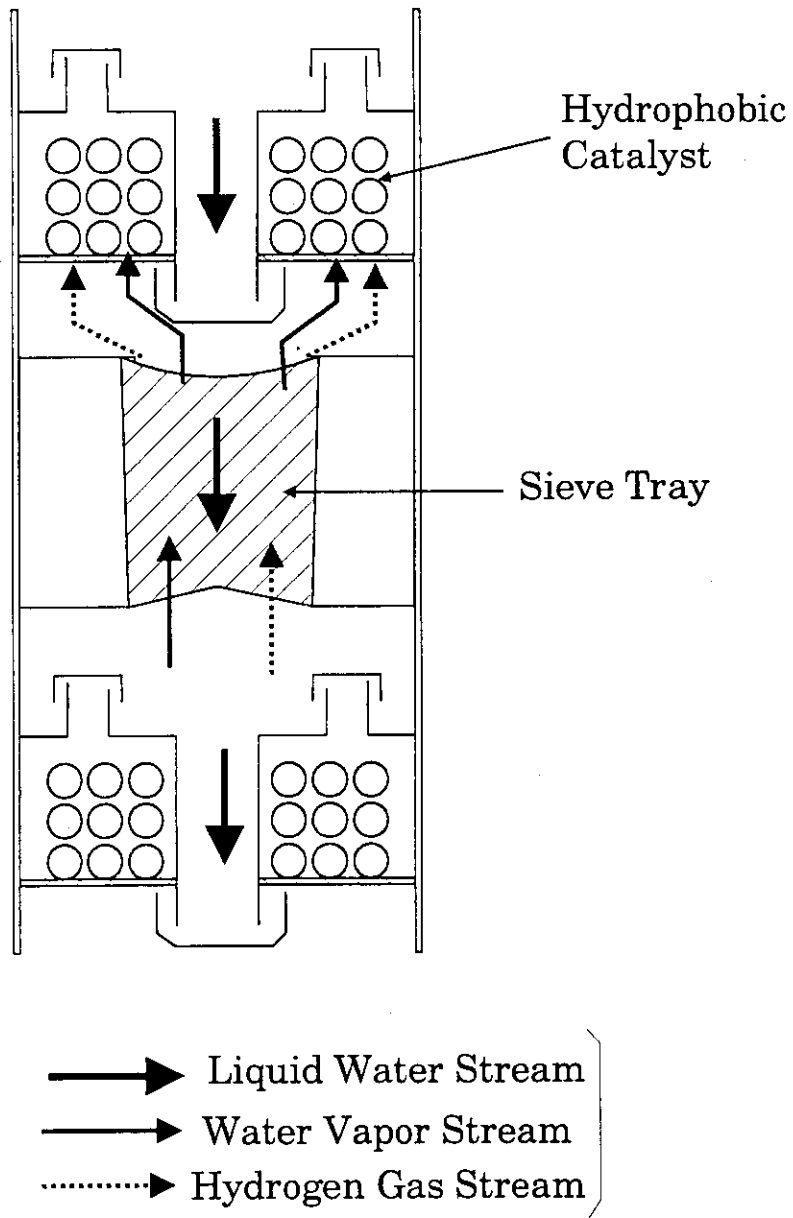


Figure 1 Structure of multistage water/hydrogen exchange column.

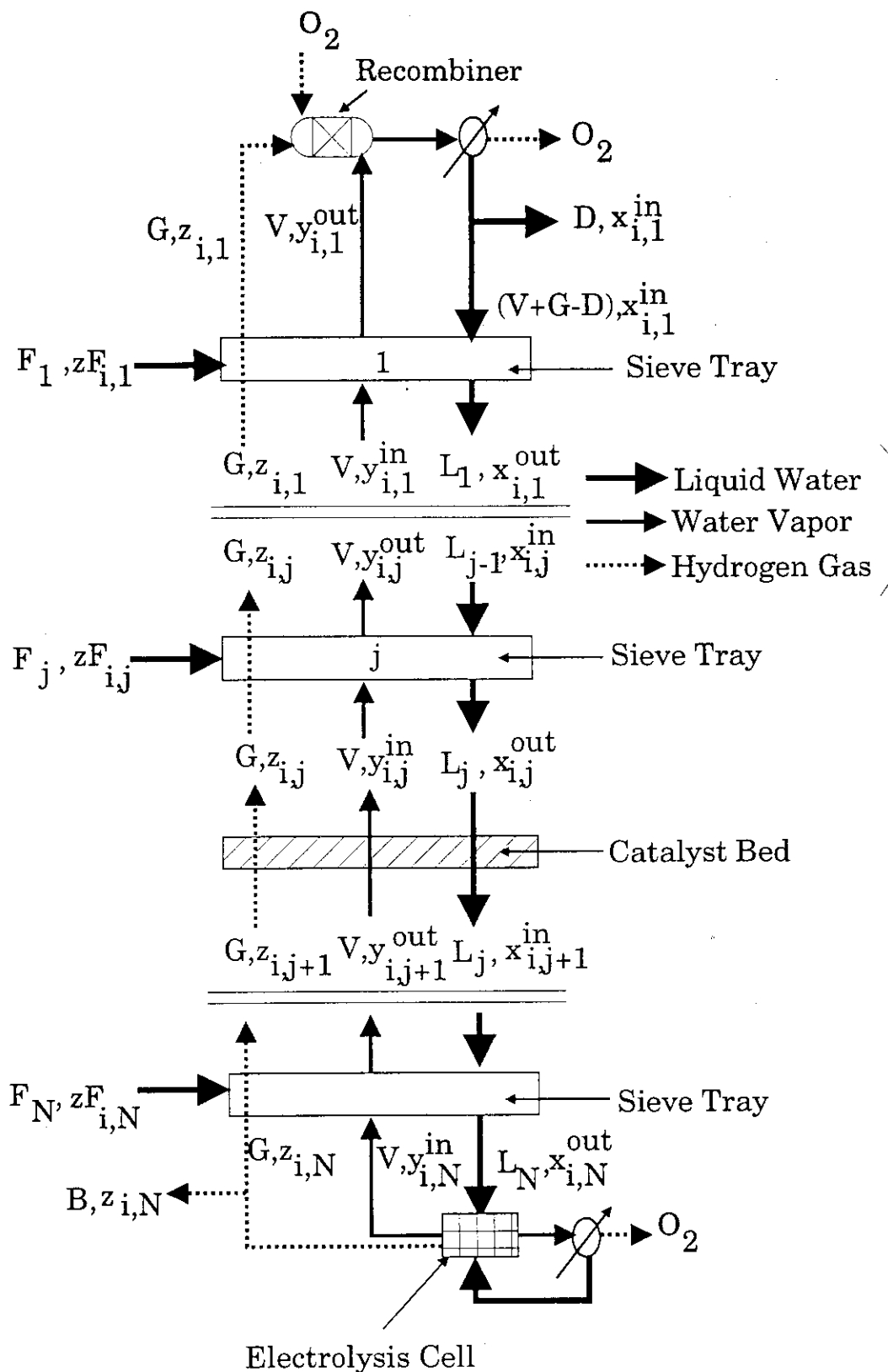


Figure 2 Model column for simulation on digital computers.

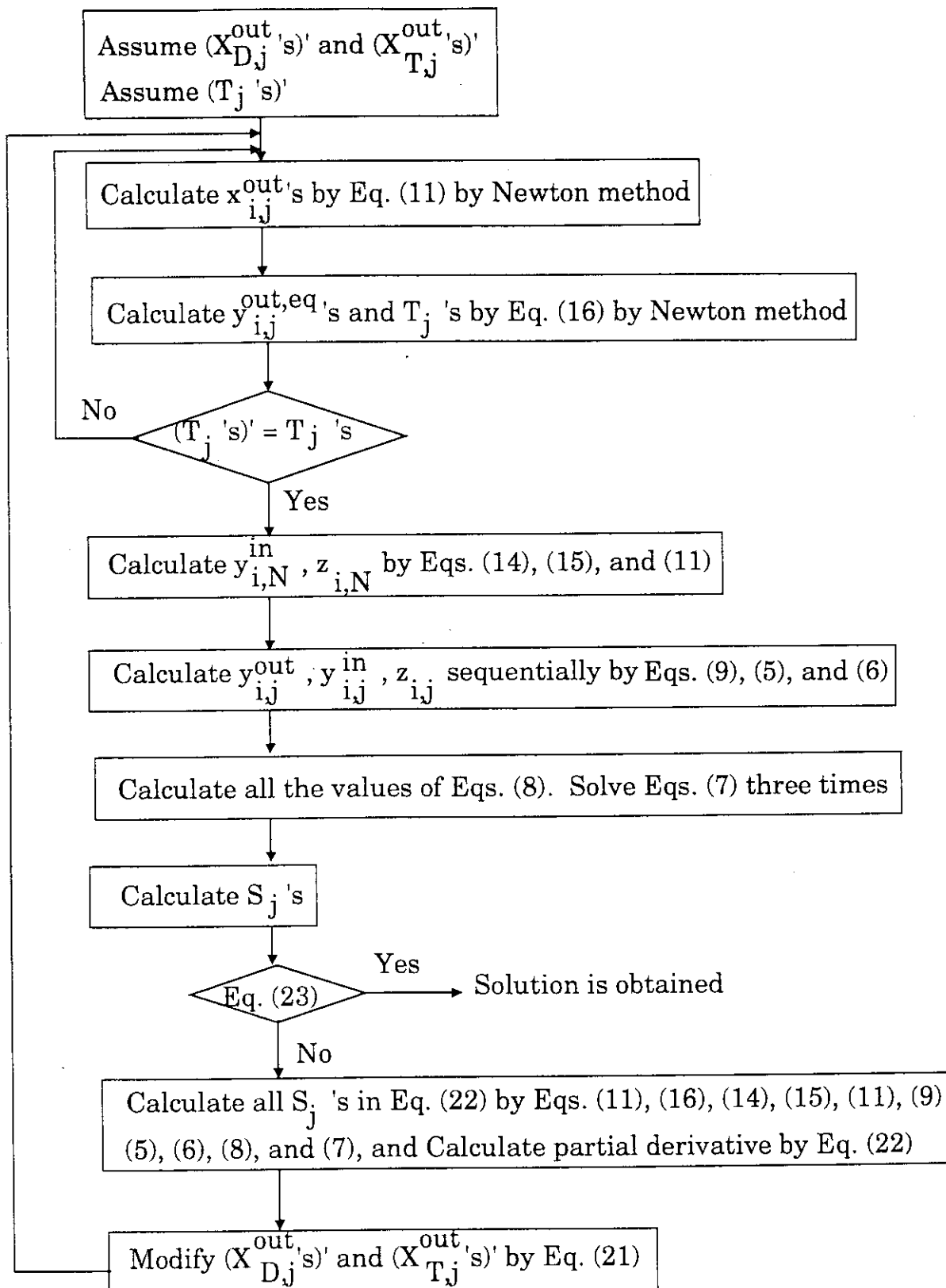


Figure 3 Flow chart of simulation procedure.