Mathematical Simulation Procedure of Multicomponent Separating Cascade

Application to Analysis of Hydrogen Isotope Separation System by Porous Membrane Method

Masahiro KINOSHITA and Yuji NARUSE
Division of Thermonuclear Fusion Research, Japan Atomic Energy Research Institute*

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A simulation procedure of multicomponent separating cascades is developed. This procedure is applicable and effective in such cases that the cut of mixture of each stage is prescribed and required to be independent of concentration, and the stage separation factors are given as input variables and very large. It can deal with cascades which have multiple feeds and sidestreams. The stage separation factors are allowed to be given as functions of compositions of down streams and up streams. The exact solutions are found out treating all the basic equations simultaneously by use of multi-dimensional Newton-Raphson method.

As an application of this procedure to actual cascade calculations, a computer simulation study is carried out for hydrogen isotope separation system by porous membrane method which consists of two cascades and two catalytic equilibrators. It is proved that the Newton-Raphson iterative calculation proceeds stably under the assumed input and output conditions, and the porous membrane method is worth while to investigate in further studies as one of promising methods for hydrogen isotope separation.

KEYWORDS: hydrogen isotopes, isotope separation, multicomponent separating cascade, separation factors, multi-dimensional Newton-Raphson method, porous membrane method, catalytic equilibrator, simulation

I. INTRODUCTION

In recent years, hydrogen isotope separation seems to be a major subject in isotope separation technology due to progress of thermonuclear fusion research and development. In design or analysis of hydrogen isotope separating cascades, multicomponents (i.e. the three isotopic elements or the six molecular species) must be treated, and the separation factors are usually very large.

The theories of multicomponent separating cascades were developed by de la Garza et al.(1)(2) and Yamamoto et al.(3) Since de la Garza’s basic equations are based on the assumption that the separating elements have small separation factors, they are not applicable to cases of large separation factors. Yamamoto’s analytical method can deal with processes of large separation factors. He derived explicit expressions in terms of heads and tails separation factors for the cascade variables such as interstage flow rates and concentration of each component. His method has the great advantage that these cascade variables can be calculated purely analytically. However, if the cut of mixture of each stage is pre-

* Tokai-mura, Ibaraki-ken 319-11.
scribed and required to be independent of concentration, the calculational procedure is not simple any longer and incorporation of successive approximation is necessary.

The purpose of the present study is to develop an alternative to Yamamoto's method in such cases that interstage flows are input variables and stage separation factors (=heads separation factors x tails separation factors) are given. The method finds out the exact solutions effectively by use of multi-dimensional Newton-Raphson method treating all the model equations simultaneously.

As an application of the method to actual cascade calculations, a computer simulation study is carried out for the hydrogen isotope separation system by porous membrane method which is assumed to be constructed in the main stream fuel circulation system for fusion reactors.

II. DEVELOPMENT OF MATHEMATICAL SIMULATION PROCEDURE

Our calculational procedure is based on a linearization of the model equations. This idea is similar to that reported by Naphtali & Sandholm(4) for multicomponent distillation calculations though the model equations are quite different from those of cascade calculations of our interest in the present study.

The model cascade for mathematical simulation is shown in Fig. 1. It is assumed that all the flows $L_j, L'_j$ and $L''_j$ are initially calculated and used as input variables. The model equations are derived from the requirements for conservation of material and for stage equilibrium. The component material balances are expressed by the following equations:

\[
L_jx_{i,j} = L'_jx'_{i,j} + L''_jx''_{i,j},
\]

\[
L_jx_{i+1} = L'_jx'_{i+1} + r_w L''_j x''_{i,1}
\]

\[
L_jx_{i,j} = L'_j x'_{i,j+1} + L''_j x''_{i,j-1} + F_j x_{i,j} - \sum_{j=2}^{N-1} U_j x_{i,j}
\]

\[
L_N x_{i,N} = L'_{N-1} x'_{i,N-1} + r_p L''_N x''_{i,N}
\]

\[
(i=1, \ldots, m; j=1, \ldots, N)
\]

We define the stage separation factors $\alpha_{i,j}$ with respect to an arbitrary key component —here, we choose the $n$-th component—in the form:

\[
\alpha_{i,j} = \frac{x'_{i,j}/x''_{i,j}}{(x'_{i,j}/x''_{i,j})},
\]

\[
(i \neq n; j=1, \ldots, N)
\]

It is obvious that the following equations must be satisfied:

\[
\sum_{i=1}^{m} x_{i,j} = \sum_{i=1}^{m} x'_{i,j} + \sum_{i=1}^{m} x''_{i,j} = 1,
\]

\[
(j=1, \ldots, N).
\]
For the purpose of calculating \( x_{i,j}, x'_{i,j} \) and \( x''_{i,j} \) from the input variables \( L_j, L'_j, L''_j, F_j, z_{i,j}, U_j \) and \( \alpha_{i,j} \), we choose the \( 3 mN \) variables \( x_{i,j}, x'_{i,j} \) and \( x''_{i,j} \) as the independent ones and define the \( 3 mN \) functions \( f_{i,j}, g_{i,j} \) and \( h_{i,j} \) by

\[
\begin{align*}
  f_{i,j} &= x_{i,j} - L_j x'_{i,j} / L_j - L''_j x''_{i,j} / L_j, \quad (i=1, \ldots, m; \quad j=1, \ldots, N), \\
  g_{i,j} &= x_{i,j} - L'_j z_{i,j} / L_j - r_w L''_j x''_{i,j} / L_j, \\
  h_{i,j} &= -F_j z_{i,j} / L_j + U_j x_{i,j} / L_j \\
  g_{i,N} &= x_{i,N} - L''_N x''_{i,N} / L_N - r_p L''_N x''_{i,N} / L_N,
\end{align*}
\]

\( (i=1, \ldots, m; \quad j=2, \ldots, N-1) \),

\[
\begin{align*}
  h_{i,j} &= -\alpha_{i,j} x_{i,j} x''_{i,j} - x'_{i,j} x''_{i,j}, \quad h_{i,N} &= \sum_{k=1}^{m} x_k' x''_{i,k} - \sum_{k=1}^{m} x_k'' / x_{i,k} \quad (i=m, j=1, \ldots, N). 
\end{align*}
\]

We define the \( 3 m \)-dimensional vectors \( \mathbf{x}_j \) and \( \mathbf{F}_j \) by

\[
\mathbf{x}_j = \begin{bmatrix} x_{1,j} \\ \vdots \\ x_{m,j} \\ x'_{1,j} \\ \vdots \\ x'_{m,j} \\ x''_{1,j} \\ \vdots \\ x''_{m,j} \end{bmatrix}, \quad \mathbf{F}_j = \begin{bmatrix} f_{1,j} \\ \vdots \\ f_{m,j} \\ g_{1,j} \\ \vdots \\ g_{m,j} \\ h_{1,j} \\ \vdots \\ h_{m,j} \end{bmatrix}, \quad (j=1, \ldots, N).
\]

In the above definition, \( \mathbf{x}_j \) is the vector of variables on the \( j \)-th stage and \( \mathbf{F}_j \) is the vector of functions on that stage. The simulation of the cascade is performed by solving the following \( 3 mN \)-dimensional nonlinear simultaneous equations:

\[
\begin{align*}
  \mathbf{F}_1(\mathbf{x}_1, \ldots, \mathbf{x}_N) &= 0, \\
  \mathbf{F}_N(\mathbf{x}_1, \ldots, \mathbf{x}_N) &= 0,
\end{align*}
\]

where \( \mathbf{x}'_j \) is the transposed vector of \( \mathbf{x}_j \). Modified Newton-Raphson method seems the most favorable one to find out the solutions of the equations. The \( 3 mN \)-dimensional square matrix, Jacobian \( \mathbf{G} \), is given by

\[
\mathbf{G} = \begin{bmatrix} \partial F_1 / \partial x_1 & \cdots & \partial F_1 / \partial x_N \\ \vdots & \ddots & \vdots \\ \partial F_N / \partial x_1 & \cdots & \partial F_N / \partial x_N \end{bmatrix},
\]

\( \begin{align*}
  \frac{\partial f_{i,j} / \partial x_{i,j}}{\partial F_1 / \partial x_1} & \cdots \frac{\partial f_{i,j} / \partial x_{m,j}}{\partial F_1 / \partial x_1}, \\
  \vdots & \ddots & \vdots \\
  \frac{\partial h_{i,j} / \partial x_{i,j}}{\partial F_N / \partial x_N} & \cdots \frac{\partial h_{i,j} / \partial x_{m,j}}{\partial F_N / \partial x_N}. 
\end{align*} \]

It should be noted that the functions for stage \( j \) involve only the variables on stage \( j-1 \), \( j \) and \( j+1 \). Therefore, it can be shown that Jacobian has the following specific structure:

\[
\mathbf{G} = \begin{bmatrix} \mathbf{B}_1 & \mathbf{C}_1 & \mathbf{0} & \cdots & \mathbf{0} \\ \mathbf{A}_2 & \mathbf{B}_2 & \mathbf{C}_2 & \cdots & \mathbf{0} \\ \vdots & \mathbf{A}_{N-1} & \mathbf{B}_{N-1} & \mathbf{C}_{N-1} & \mathbf{0} \\ \mathbf{0} & \cdots & \mathbf{A}_N & \mathbf{B}_N & \mathbf{C}_N \end{bmatrix},
\]

\( \begin{align*}
  \mathbf{B}_j &= \begin{bmatrix} \partial b_{1,j} / \partial x_{1,j} & \cdots & \partial b_{1,j} / \partial x_{m,j} \\ \vdots & \ddots & \vdots \\ \partial b_{m,j} / \partial x_{1,j} & \cdots & \partial b_{m,j} / \partial x_{m,j} \end{bmatrix}, \\
  \mathbf{C}_j &= \begin{bmatrix} \partial c_{1,j} / \partial x_{1,j} & \cdots & \partial c_{1,j} / \partial x_{m,j} \\ \vdots & \ddots & \vdots \\ \partial c_{m,j} / \partial x_{1,j} & \cdots & \partial c_{m,j} / \partial x_{m,j} \end{bmatrix}, \\
  \mathbf{A}_j &= \begin{bmatrix} \partial a_{1,j} / \partial x_{1,j} & \cdots & \partial a_{1,j} / \partial x_{m,j} \\ \vdots & \ddots & \vdots \\ \partial a_{m,j} / \partial x_{1,j} & \cdots & \partial a_{m,j} / \partial x_{m,j} \end{bmatrix}. 
\end{align*} \]
where the elements of the $3m$-dimensional square matrices $\mathbf{A}_j$, $\mathbf{B}_j$ and $\mathbf{C}_j$ are given in Appendix 1, and $\mathbf{0}$ denotes a $3m$-dimensional zero matrix.

The values of the independent variables are improved by the following equations at each iterative step of the Modified Newton-Raphson calculation:

\[
\begin{bmatrix}
\mathbf{d}x_1 \\
\vdots \\
\mathbf{d}x_N
\end{bmatrix} = - \begin{bmatrix}
F_1 \\
\vdots \\
F_N
\end{bmatrix},
\]

\[
x_j := x_j + \gamma \mathbf{d}x_j, \quad (j=1, \ldots, N),
\]

where $\gamma$ is either a moderator ($0 < \gamma \leq 1$) or an accelerator ($\gamma > 1$), and $a := b$ means the procedure that the value of $b$ is regarded as the new value of $a$.

After the values of the independent variables are improved by Eq. (12), they must be normalized so as to satisfy Eq. (4) by the following procedure:

\[
x_{i,j} := x_{i,j}/\sum_{i=1}^{m} x_{i,j}, \quad x'_{i,j} := x'_{i,j}/\sum_{i=1}^{m} x'_{i,j}, \quad x''_{i,j} := x''_{i,j}/\sum_{i=1}^{m} x''_{i,j},
\]

\[
(i=1, \ldots, m; \quad j=1, \ldots, N).
\]

There is no need to directly calculate the inverse matrix of Jacobian with the result that computational time is greatly saved: Eq. (12) can be solved effectively taking advantage of the specific feature that Jacobian has the tridiagonal matrix form if all the $3m$-dimensional square matrices are regarded as scalars\(^5\). The initial values of the independent variables are properly postulated (for instance, compositions of all the flows are postulated to be equal to the feed composition, for a cascade with a single feed) and the values continue to be improved until the following convergence criterion is satisfied:

\[
f := \sum_{j=1}^{N} \sum_{i=1}^{m} (|f_{i,j}| + |g_{i,j}| + |h_{i,j}|) < 3N \varepsilon, \quad 0 < \varepsilon \leq 1.
\]

In the present study, the order of magnitude of $\varepsilon$ is established to be $10^{-8}$, which seems small enough to find out the solutions.

In this simulation procedure, the cut of mixture of each stage is independent of concentration. It should be also noted that the stage separation factors are allowed to be functions of $x'_{i,j}$ and $x''_{i,j}$.

### III. Simulation Study of Hydrogen Isotope Separation System by Porous Membrane Method

The design of fusion reactors and its associated systems is still in a very early stage. For this reason, the input conditions and the output requirements for hydrogen isotope separation system in the fuel circulation loop have not been determined yet. In the present study, the flow rates and the compositions of the input streams and the separation requirements are assumed as shown in Fig. 2. They are the same as those assumed by Naito et al.\(^6\)(\(^7\)) in their analysis of the cascade system using Pd-Ag alloy membrane. However, the separation requirements are much more relaxed than those assumed for the cryogenic distillation system in our previous work\(^8\) or other works\(^9\)(\(^10\)).

The system configuration developed in the present study is shown in Fig. 3. The system is composed of two cascades (Cascade 1 and 2) and two catalytic equilibrators which promote the isotopic exchange reaction $\text{HT} + \text{D}_2 \rightleftharpoons \text{HD} + \text{DT}$. The equilibrators are necessary because HT and D\(_2\) cannot be separated by porous membrane diffusers alone on account of equality of their molecular weights. Each cascade has a sidestream which
An Essentially Tritium-free Stream of $H_2$ (, HD ) for Waste Disposal to the Atmosphere

External Feed
30.654 g·mol/h
$H_2 : 0.617 \times 10^{-4}$
$HD : 0.928 \times 10^{-2}$
$HT : 0.431 \times 10^{-2}$
$D_2 : 0.430$
$DT : 0.459$
$T_2 : 0.117$

Hydrogen Isotope Separation System

A High-purity Stream of $D_2$ for Plasma Heating as a Neutral Beam
(atom fraction of $D > 0.99$)

A Basically Protium-free Stream of D-T Mixture for Refueling
(atom fraction of $H < 10^{-4}$)

Fig. 2 Input conditions and separation requirements for hydrogen isotope separation system in fuel circulation loop for fusion reactors

is recycled to the feed stream after being passed through the equilibrator. In the present simulation, the reflux ratio defined by

$$R = \frac{L_j}{P}, \quad (j = 1, \ldots, N-1) \quad (15)$$

is given as one of the input data, and $r_P$ and $r_W$ are postulated to be zero.

Calculations are made for the two cascades following the procedure described in the previous chapter and the successive iteration method (Appendix 2).

In order to find out design specifications which meet the separation requirements, brief parametric surveys are separately made for each cascade. Parameters to be studied are the number of total stages, reflux ratio, feed location, sidestream flow rate, sidestream location and top product flow rate. In the present study, several cases are assumed and the design specifications are determined from the calculation result for each case. The flow rate of the top product is properly established considering the requirements for the cascade and the flow rate of the sidestream is assumed to be equal to that of the external feed stream. The stage separation factors are assumed to have ideal values given by

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Fig. 3 System configuration of cascade system by porous membrane method

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$\alpha_{i,j} = \sqrt{\frac{M_i}{M_j}}$, \quad (i=1, \cdots, m; \ i \neq n). \quad (16)$

These brief parametric surveys appear to be satisfactory for our purpose. The calculational conditions (assumed cases) and the results are summarized in Tables 1 and 2. Parametric surveys for Cascade 2 are made after design specifications of Cascade 1 are determined.

### Table 1 Calculational conditions and results for Cascade 1

<table>
<thead>
<tr>
<th>Case</th>
<th>$P$ (g-mol/h)</th>
<th>$R$</th>
<th>$N$</th>
<th>$N_F$</th>
<th>$N_{SC}$</th>
<th>$y_{1, H}$ (%)</th>
<th>$c_{N, T}$ (Ci/h)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.2864</td>
<td>800</td>
<td>80</td>
<td>32</td>
<td>16</td>
<td>0.233</td>
<td>3.15</td>
</tr>
<tr>
<td>2</td>
<td>0.2864</td>
<td>800</td>
<td>100</td>
<td>40</td>
<td>20</td>
<td>0.216</td>
<td>0.432</td>
</tr>
<tr>
<td>3</td>
<td>0.2864</td>
<td>800</td>
<td>120</td>
<td>48</td>
<td>24</td>
<td>0.213</td>
<td>0.0628</td>
</tr>
<tr>
<td>4</td>
<td>0.2864</td>
<td>500</td>
<td>100</td>
<td>40</td>
<td>20</td>
<td>0.246</td>
<td>4.52</td>
</tr>
<tr>
<td>5</td>
<td>0.2864</td>
<td>500</td>
<td>100</td>
<td>30</td>
<td>15</td>
<td>0.249</td>
<td>4.55</td>
</tr>
<tr>
<td>6</td>
<td>0.2864</td>
<td>500</td>
<td>100</td>
<td>50</td>
<td>30</td>
<td>0.254</td>
<td>7.30</td>
</tr>
</tbody>
</table>

(1) Reflux ratio $R$ is defined by the following equation: $R = L_j' / P$ (j=1, ..., N-1).

(2) The conditions of Case 3 were chosen for Cascade 1.

$\begin{align*}
&c_{N, T}: \text{Tritium flow rate in top product} \\
&N: \text{Number of total stages} \\
&N_F: \text{Feed location (feed stage number)} \\
&N_{SC}: \text{Sidestream location} \\
&P: \text{Flow rate of top product} \\
&R: \text{Reflux ratio} \\
&y_{1, H}: \text{Atom percentage of protium in bottom product}
\end{align*}$

### Table 2 Calculational conditions and results for Cascade 2

<table>
<thead>
<tr>
<th>Case</th>
<th>$P$ (g-mol/h)</th>
<th>$R$</th>
<th>$N$</th>
<th>$N_F$</th>
<th>$N_{SC}$</th>
<th>$y_{1, H}$ (%)</th>
<th>$y_{N, D}$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>13.25</td>
<td>20</td>
<td>120</td>
<td>48</td>
<td>24</td>
<td>0.00809</td>
<td>96.7</td>
</tr>
<tr>
<td>2</td>
<td>13.25</td>
<td>25</td>
<td>120</td>
<td>48</td>
<td>24</td>
<td>0.00673</td>
<td>99.1</td>
</tr>
</tbody>
</table>

(1) The conditions of Case 2 were chosen for Cascade 2.

$y_{N, D}: \text{Atom percentage of deuterium in top product}$

In the Newton-Raphson iterative calculations, the initial estimates of the independent variables are matters of vital importance. In cases of small reflux ratio, compositions of all the flows are postulated to be equal to the feed composition. In cases of relatively large reflux ratio, the final values of the independent variables in cases of smaller reflux ratio are used as the initial ones. These proper estimates result in stable progress of the iterative calculations or great decrease in the iteration numbers. Under the assumed conditions in the present study, all the calculations proceed considerably stably.

The specifications of the three output streams produced by the cascade system are

### Table 3 Specifications of output streams from cascade system

<table>
<thead>
<tr>
<th>Flow rate (g-mol/h)</th>
<th>Stream ①</th>
<th>Stream ②</th>
<th>Stream ③</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.2864</td>
<td>13.25</td>
<td>17.1176</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Composition</th>
<th>Stream ①</th>
<th>Stream ②</th>
<th>Stream ③</th>
</tr>
</thead>
<tbody>
<tr>
<td>$H_2$:</td>
<td>$0.180 \times 10^{-1}$</td>
<td>$0.141 \times 10^{-4}$</td>
<td>$0.825 \times 10^{-11}$</td>
</tr>
<tr>
<td>$HD$:</td>
<td>0.981</td>
<td>0.764 $\times 10^{-2}$</td>
<td>0.895 $\times 10^{-6}$</td>
</tr>
<tr>
<td>$HT$:</td>
<td>$0.734 \times 10^{-3}$</td>
<td>$0.190 \times 10^{-2}$</td>
<td>$0.134 \times 10^{-3}$</td>
</tr>
<tr>
<td>$D_2$:</td>
<td>$0.964 \times 10^{-3}$</td>
<td>0.984</td>
<td>0.709 $\times 10^{-1}$</td>
</tr>
<tr>
<td>$DT$:</td>
<td>$0.140 \times 10^{-4}$</td>
<td>$0.640 \times 10^{-2}$</td>
<td>0.649</td>
</tr>
<tr>
<td>$T_2$:</td>
<td>$0.107 \times 10^{-6}$</td>
<td>0.423 $\times 10^{-5}$</td>
<td>0.280</td>
</tr>
</tbody>
</table>
shown in Table 3. According to Table 3, the separation requirements described before are certainly met.

The number of total stages of the cascade system by porous membrane method is larger than that by Pd-Ag alloy membrane method because of the smaller separation factors. The values of \(L_jN\) \((j=1, \ldots, N-1)\) for these two methods are as follows:

Pd-Ag alloy membrane \(\left(L_jN\right)_{\text{total}}=(L_jN)_{\text{Cascade 1}}+(L_jN)_{\text{Cascade 2}}=59,000\ \text{g-atom/h}\)

Porous membrane \(\left(L_jN\right)_{\text{total}}=134,000\ \text{g-atom/h}\)

The value of \(\left(L_jN\right)_{\text{total}}\) of porous membrane method is approximately two times larger than that of Pd-Ag alloy membrane method. It should be noted, however, that the permeation coefficient is much larger for porous membrane. Hence, the above comparison never leads to the conclusion that the dimensions of the diffusers are larger for porous membrane. On the contrary, the membrane area of porous membrane is expected to be smaller. This indicates that the porous membrane method is worth while to investigate in further studies as one of promising methods for hydrogen isotope separation.

IV. CONCLUSIONS

1) A mathematical simulation procedure is developed for multicomponent separating cascades. It is applicable and effective in such cases that the cut of mixture of each stage is prescribed and required to be independent of concentration, and the interstage flows and the stage separation factors are given as input variables.

2) The procedure is applied to analysis of separation characteristics of hydrogen isotope separation system by porous membrane method. The multi-dimensional Newton-Raphson iterative calculations proceed stably under the assumed conditions. It is proved that the porous membrane method is worth while to investigate in further studies as a possible method for hydrogen isotope separation in the fuel circulation system for fusion reactors.

[NOMENCLATURE]

\[
F_j: \text{Flow rate of feed stream supplied to} \ j\text{-th stage (g-mol/h)}
\]

\[
L_j: \text{Flow rate of stream entering} \ j\text{-th stage (g-mol/h)}
\]

\[
L'_j: \text{Flow rate of stream leaving} \ j\text{-th stage upward (g-mol/h)}
\]

\[
L''_j: \text{Flow rate of stream leaving} \ j\text{-th stage downward (g-mol/h)}
\]

\[
m: \text{Number of components}
\]

\[
M: \text{Molecular weight (g/g-mol)}
\]

\[
N: \text{Number of total stages}
\]

\[
r_p: \text{Reflux ratio from top product}
\]

\[
r_w: \text{Reflux ratio from bottom product}
\]

\[
U_j: \text{Flow rate of sidestream on} \ j\text{-th stage (g-mol/h)}
\]

\[
x_{i,j}: \text{Mole fraction of} \ i\text{-th component in stream entering} \ j\text{-th stage}
\]

\[
z_{i,j}: \text{Mole fraction of} \ i\text{-th component in stream leaving} \ j\text{-th stage upward}
\]

\[
z''_{i,j}: \text{Mole fraction of} \ i\text{-th component in stream leaving} \ j\text{-th stage downward}
\]

\[
z_{i,j}: \text{Mole fraction of} \ i\text{-th component in feed stream supplied to} \ j\text{-th stage}
\]

For instance, \(x\), \(F\) and \(G\) denote a scalar, a vector and a matrix, respectively.

ACKNOWLEDGMENT

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REFERENCES

1. Elements of Matrices $\mathbf{A}_j$, $\mathbf{B}_j$ and $\mathbf{C}_j$

<table>
<thead>
<tr>
<th>$\mathbf{A}_j$ (j=2, ..., N)</th>
<th>$\mathbf{B}_j$ (j=1, ..., N)</th>
<th>$\mathbf{C}_j$ (j=1, ..., N-1)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\frac{\partial f_{i,j}}{\partial x_{k,j-1}} = 0$</td>
<td>$\frac{\partial g_{j,i}}{\partial x_{k,j-1}} = 0$</td>
<td>$\frac{\partial h_{i,j}}{\partial x_{k,j-1}} = 0$</td>
</tr>
<tr>
<td>$\frac{\partial f_{i,j}}{\partial x_{k,j-1}} = 0$</td>
<td>$\frac{\partial g_{j,i}}{\partial x_{k,j-1}} = 0$</td>
<td>$\frac{\partial h_{i,j}}{\partial x_{k,j-1}} = 0$</td>
</tr>
<tr>
<td>$\frac{\partial f_{i,j}}{\partial x''_{k,j-1}} = 0$</td>
<td>$\frac{\partial g_{j,i}}{\partial x''_{k,j-1}} = 0$</td>
<td>$\frac{\partial h_{i,j}}{\partial x''_{k,j-1}} = 0$</td>
</tr>
<tr>
<td>$\frac{\partial f_{i,j}}{\partial x''_{k,j-1}} = 0$</td>
<td>$\frac{\partial g_{j,i}}{\partial x''_{k,j-1}} = 0$</td>
<td>$\frac{\partial h_{i,j}}{\partial x''_{k,j-1}} = 0$</td>
</tr>
<tr>
<td>$\frac{\partial f_{i,j}}{\partial x''_{k,j-1}} = 0$</td>
<td>$\frac{\partial g_{j,i}}{\partial x''_{k,j-1}} = 0$</td>
<td>$\frac{\partial h_{i,j}}{\partial x''_{k,j-1}} = 0$</td>
</tr>
</tbody>
</table>

APPENDIX
In these equations, $\delta_{i,j}$ denotes the Kronecker's delta defined by

$$\delta_{i,j} = \begin{cases} 1 & (i=j) \\ 0 & (i \neq j) \end{cases}.$$ 

2. Calculational Procedure for Cascade with Feedback Stream

(1) Assume the atom fraction of each isotopic element in the sidestream $z_H$, $z_D$ and $z_T$.

(2) Calculate the mole fraction of each molecular in the stream recycled to the feed stream. It is assumed that the equilibrium composition established by the following isotopic exchange reactions at 25°C is obtained at the exit of the equilibrator:

$$\begin{align*}
H_2 + D_2 & \rightleftharpoons 2HD \quad (k_1 = 3.25^{(11)}) \\
H_2 + T_2 & \rightleftharpoons 2HT \quad (k_2 = 2.57) \\
D_2 + T_2 & \rightleftharpoons 2DT \quad (k_3 = 3.82).
\end{align*}$$

(3) Calculate the composition of the feed supplied to the cascade, and perform the cascade calculation following the procedure developed in the present study.

(4) Calculate the atom fraction of each element in the sidestream $z'_H$, $z'_D$ and $z'_T$, and calculate the residual function defined by

$$J = \frac{|z_H - z'_H| / z'_H + |z_D - z'_D| / z'_D + |z_T - z'_T| / z'_T}{3}.$$ 

(5) Examine whether $J$ exceeds $\varepsilon$ or not ($0 < \varepsilon \ll 1$).

- If $J < \varepsilon$, the calculation is terminated and the result is printed out.
- If $J > \varepsilon$, return to step (2) after substituting the values of $z'_H$, $z'_D$ and $z'_T$ into $z_H$, $z_D$ and $z_T$, respectively.

It should be noted that $\varepsilon$ must be considerably small ($\varepsilon < 1.0 \times 10^{-5}$) to find out accurate solutions.