The Optimal Column Number for Deuterium Removal Rate from Water-Isotopes Mixture in the Frazier Scheme with the Total Sum of Column Heights Fixed

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Abstract

The effect of column number N, as well as the column height h of thermal diffusion columns, on the deuterium removal rate from water-isotope mixture in the countercurrent-flow Frazier scheme with total sum of column heights L (= Nh) fixed, has been investigated. The equations, which may be employed to predict the optimal numbers of column for the maximum performances, have been derived. Considerable improvement in deuterium removal rate is obtainable if the scheme is constructed with the optimal number of thermal diffusion columns, especially for large flow rate and smaller L.

Key Words: Thermal Diffusion, Frazier Scheme, Optimal Column Number, Recovery of Deuterium, Total Sum of Column Heights Fixed, Best Performance

1. Introduction

Thermal diffusion is a molecular phenomenon in which small concentration differences arise under the application of a temperature gradient. It can be used to separate gas or liquid mixtures, which are difficult or impossible to separation, such as isotopes and isomers. For separation of hydrogen isotopes, thermal diffusion process is more attractive because of the large ratio in molecular weights [1]. The experimental studies for separation of hydrogen isotopes were successfully carried out in cryogenic-wall thermal diffusion columns [2–4] and the correlation equations for prediction of deuterium (D) recovery were derived in previous work [5].

In industrial applications, thermal diffusion columns are connected in series such as that shown in Figure 1, called the Frazier scheme [6]. In this device, feeding method is in such a manner that the sampling streams do not pass through but move outside the columns. The theory of separation by thermal diffusion was first presented by Furry et al. [7] and Jones and Furry [8], while that in the Frazier scheme was given by Rabinovic [9] and Sovorov and Rabinovic [10]. A more detailed study of the mechanism of thermodiffusion separation in the thermogravitational thermal diffusion introduced by Clusius and Dickel [11,12], indicates that the convective currents produced by the temperature gradient in thermal diffusion columns actually have two conflict effects: the desirable cascading effect and the undesirable remixing effect. Therefore, the strength of convection flows must be properly reduced and controlled, leading to improved performance [13–16].

One of the flexible ways of properly controlling the convective strength in the concurrent-flow Frazier scheme is suitably adjusting the number of columns with the total sum of column heights fixed [17]. Instead of concurrent-flow operation, it is the purpose of present study to estimate the improvement in recovery of deuterium from water-isotope mixture by thermal diffusion in the countercurrent-flow Frazier scheme with optimal number of columns and with the total sum of column heights fixed.
2. Theory

The mass-fraction concentrations of \( \text{H}_2\text{O} (C_1), \text{HDO} (C_2) \) and \( \text{D}_2\text{O} (C_3) \) in water-isotope mixture may be considered to be locally equilibrium at every point in thermal diffusion columns [18], i.e.

\[
\text{H}_2\text{O} + \text{D}_2\text{O} \rightleftharpoons 2\text{HDO}
\] (1)

with the equilibrium constant defined as

\[
K_{eq} = \frac{[\text{HDO}]^2}{[\text{H}_2\text{O}][\text{D}_2\text{O}]} = \frac{(C_2/19)^2}{(C_1/18)(C_3/20)} \\
= 0.997 \frac{(C_2)}{(C_1)(C_3)} \\
\approx \frac{(C_2)}{(C_1)(C_3)}
\] (2)

2.1 Separation of \( \text{D}_2\text{O} \)

Figure 1 is the schematic diagram of a Frazier scheme with \( N \) flat-plate thermal diffusion columns operating under countercurrent flow, while Figure 2 illustrates the flows and fluxes prevailing in the \( i \)th thermogravitational thermal-diffusion column of the Frazier scheme. All flat-plate columns have the same dimensions of gap \( (2\alpha) \), height \( h \), and width \( B \). The temperature gradient applied to a water-isotope mixture has two effects: (1) a flux of heavy water, \( J_{\text{X-TD}} \), relative to the other is brought about by thermal diffusion toward cold surface, and (2) convective currents are produced parallel to the plates owing to density differences. The combined result of these two effects is to produce a concentration difference.
of heavy water between two ends of the column. Meanwhile, the concentration gradients in x and z directions produce the fluxes $J_{x-OD}$ and $J_{z-OD}$, respectively, due to ordinary diffusion, to counterbalance that resulting from thermal diffusion at steady state. The separation equation for heavy water in a countercurrent-flow Frazier scheme with N columns of same column height h and with the total sum of column heights L (= Nh), is [19]

$$\Delta_1 = C_{3,b} - C_{3,T}$$ (3)

$$\Delta_1 = \frac{2AhN(-H)}{(N+1)K + \sigma h}$$ (4)

where $(C_{3,b})$ and $(C_{3,T})$ denote the mass-fraction of heavy water at the bottom end of the first column and at the top end of the last (Nth) column, respectively, and the transport coefficients related with the fluid properties and temperature difference $(\Delta T)$ between the hot and cold plates are defined by

$$H = \frac{\alpha p \beta g(2\theta)^3 B(\Delta T)^2}{6!\mu T_n}$$ (5)

$$K = \frac{\rho B^2 g^2 (2\theta)^5 B(\Delta T)^2}{9!D_n}$$ (6)

while the product form of concentration is approximately considered as a constant A with the value taken at its feed concentration of heavy water $C_F$, as [18]

$$A = C_F[0.05263 - (0.05263 - 0.0135 K_{eq})C_F - 0.027(C_F K_{eq} [1 - (1 - 0.25 K_{eq})C_F]^{1/2})]$$ (7)

### 2.2 Separation of HDO

For $H_2O - HDO - D_2O$ system

$$C_1 + C_2 + C_3 = 1$$ (8)

Thus, the concentration of $HDO$ ($C_2$) can be solved from Eqs. (2) and (8) with the expression in term of the concentration of $D_2O$ ($C_3$) as

$$C_2 = \frac{K_{eq}}{2}C_3 + \left\{ C_1 K_{eq} [1 - (1 - \frac{K_{eq}}{4} C_3)]^{1/2} \right\}$$ (9)

Since the value of $K_{eq}$ is approximately 4 ($K_{eq} = 3.793$ at 30.5 °C and $K_{eq} = 3.80$ at 25 °C [18], Eq. (9) may be reduced to

$$C_2 = -(\frac{K_{eq} C_3}{2}) + K_{eq}^{1/2} C_3^{1/2}$$ (10)

Accordingly, the separation equation for $HDO$ in a countercurrent-flow Frazier scheme of N columns with same column height is

$$\Delta_2 = C_{2,b} - C_{2,T}$$ (11)

$$\Delta_2 = -(\frac{K_{eq}}{2})(C_{3,b} - C_{3,T}) + K_{eq}^{1/2} (C_{3,b}^{1/2} - C_{3,T}^{1/2})$$ (12)

Further, since $(C_{3,b} - C_{3,T})$ are very small and their values are very close, we have

$$C_{3,b}^{1/2} - C_{3,T}^{1/2} = \frac{(C_{3,b} - C_{3,T})}{(C_{3,b}^{1/2} + C_{3,T}^{1/2})} = \frac{(C_{3,b} - C_{3,T})}{(2C_3^{1/2})}$$ (13)

thus, Eq. (12) becomes

$$\Delta_2 = -(\frac{K_{eq}}{2})\Delta_3 + (K_{eq}^{1/2} / 2C_3^{1/2})\Delta_3$$ (14)

### 2.3 Degree of Deuterium Recovery

The degree of deuterium recovery $\Delta_D$ in a Frazier scheme may be defined from the molecular weights of $HDO$ and $D_2O$ as

$$\Delta_D = C_{D,b} - C_{D,T}$$ (15)

$$\Delta_D = \left[ \frac{2}{19} C_{2,b} + \frac{4}{20} C_{3,b} \right] - \left[ \frac{2}{19} C_{2,T} + \frac{4}{20} C_{3,T} \right]$$ (16)

Substitution of Eqs. (4) and (14) into Eq. (16) yieds

$$\Delta_D = \frac{2FhN(-H)}{(N+1)K + \sigma h}$$ (17)

where

$$F = A[0.2 - (K_{eq} / 19) + (K_{eq} / C_3^{1/2}) / 19]$$ (18)

### 2.4 Deuterium Removal Rate

The equation for estimating the output from given
degree of deuterium recovery, may be derived by rewriting Eq. (17) as

$$\sigma = \frac{2FN(-H)}{\Delta_D} - \frac{(N+1)K}{h}$$  \hspace{1cm} (19)$$

and the removal rate of deuterium D is

$$M = \sigma(C_{D,B} - C_F) + \sigma(C_F - C_{D,F}) = \sigma C_{D,B} - \sigma C_{D,F} = \sigma \Delta_D$$  \hspace{1cm} (20)$$

2.5. Separation Equations with Total Sum of Column Heights Fixed

Consider a countercurrent-flow Frazier scheme of N thermal diffusion columns with same column height h. If the total sum of column heights of a Frazier scheme L is fixed, then

$$L = Nh = \text{constant}$$  \hspace{1cm} (21)$$

and Eqs. (17) and (19) may be rewritten as

$$\Delta_D = \frac{2FL(-H)}{(N+1)K + (\sigma L / N)}$$  \hspace{1cm} (22)$$

$$\sigma = \frac{2FN(-H)}{\Delta_D} - \frac{N(N+1)K}{L}$$  \hspace{1cm} (23)$$

3. Optimal Column Numbers for Maximum Performances

3.1 Optimal Column Numbers for Maximum $\Delta_D$ and Maximum $\sigma$

The optimal column number $N_{\Delta,\text{opt}}$ for maximum recovery of deuterium with flow rate $\sigma$ and total sum of column heights L specified, is obtained by partially differentiating Eq. (22) with respect to $N$ and setting $\partial \sigma / \partial N = 0$. The result is

$$N_{\Delta,\text{opt}} = \frac{AL(-H)}{\Delta_D K} - \frac{1}{2}$$  \hspace{1cm} (25)$$

3.2 Most Optimal Column Numbers and Maximum Performances

Mathematically, the optimal column numbers, $N_{\Delta,\text{opt}}$ and $N_{\sigma,\text{opt}}$, calculated from Eqs. (24) and (25) are generally not the integers, while the practical column numbers should be the positive integers. Therefore, the most optimal column numbers, $N_{\Delta}^*$ and $N_{\sigma}^*$, must be the positive integers which are nearest to, and smaller than the optimal column numbers, i.e. $0 \leq N_{\Delta,\text{opt}} - N_{\Delta}^* \leq 1$ and $0 \leq N_{\sigma,\text{opt}} - N_{\sigma}^* \leq 1$. Accordingly, the practical maximum degree of deuterium recovery and maximum production rate should be calculated from Eqs. (22) and (23) with $N$ replaced by $N_{\Delta}^*$ ($= N_{\Delta,\text{opt}}$) and $N_{\sigma}^*$ ($= N_{\sigma,\text{opt}}$), respectively, i.e.,

$$\Delta_D = \frac{2FL(-H)}{(N_{\Delta}^*+1)K + (\sigma L / N_{\Delta}^*)}$$  \hspace{1cm} (26)$$

$$\sigma_{\max} = \frac{2FN_{\Delta}^*(-H)}{\Delta_D} - \frac{N_{\Delta}^*(N_{\Delta}^*+1)K}{L}$$  \hspace{1cm} (27)$$

Finally, there are two maximum removal rates of deuterium based on $\Delta_D, \max$ and $\sigma_{\max}$, respectively, as

$$M_{\Delta} = \Delta_D, \max \sigma \bigg|_{N_{\Delta}^*}$$  \hspace{1cm} (28)$$

$$M_{\sigma} = \sigma_{\max} \Delta_D \bigg|_{N_{\sigma}^*}$$  \hspace{1cm} (29)$$

If $N_{\Delta}^* = N_{\sigma}^*$ or $N_{\Delta,\text{opt}} = N_{\sigma,\text{opt}}$, then

$$M_{\Delta} = M_{\sigma} = \Delta_D, \max \sigma_{\max}$$  \hspace{1cm} (30)$$

Provided that, from Eqs. (24) and (25)

$$\frac{\sqrt{\sigma L}}{K} = \frac{FL(-H)}{\Delta_D K} - \frac{1}{2}$$  \hspace{1cm} (32)$$
4. The Improvement in Performances with Most Optimal Column Numbers

The improvement in the deuterium removal rate by operating at the most optimal column number \( N^* \) is best illustrated by calculating the percentage increase in recovery based on that \( \Delta D_{N^*, \text{max}} \) obtained in the Frazier scheme of \( N \) (specified) columns with the same total sum of column heights \( L \), i.e.,

\[
I_\Delta = \frac{M_\Delta - M}{M} = \frac{\Delta D_{\text{max}} - \Delta D_{N, \text{max}}}{\Delta D_{N, \text{max}}} \tag{33}
\]

Similarly, the improvement in removal rate may be defined as

\[
I_\sigma = \frac{M_\sigma - M}{M} = \frac{\sigma_{\text{max}} - \sigma}{\sigma} \tag{34}
\]

where \( \sigma \) denotes the output obtained in the device of \( N \) columns.

4.1 Numerical Example

The experimental data obtained in previous work [18] for separation of \( \text{H}_2\text{O}-\text{HDO}-\text{D}_2\text{O} \) system in a thermal diffusion column will be employed for predicting the improvement in performances. They are:

\[
\begin{align*}
\Delta T &= 47 \, ^\circ\text{C} - 14 \, ^\circ\text{C} = 33 \, ^\circ\text{C}; \\
K_{\text{eq}} &= 3.793 \text{ at } T_m = 30.5 \, ^\circ\text{C} (= 303.5 \, \text{K}); \\
B &= 10.12 \, \text{cm}; \\
(2\alpha) &= 0.04 \, \text{cm}; \\
H &= -0.53 \, \text{g/h}; \\
K &= 5.58 \, \text{g cm/h}; \\
F &\times 10^2 = 0.1165 \, \textnormal{(C}_3\text{F} = 0.1), \\
0.1329 \, \text{(C}_3\text{F} = 0.3), \\
0.1106 \, \text{(C}_3\text{F} = 0.5), \\
0.0726 \, \text{(C}_3\text{F} = 0.7), \quad 0.0257 \, \text{(C}_3\text{F} = 0.9).
\end{align*}
\]

With the use of these numerical values, the performances in the Frazier schemes with the total sum of column heights: \( L = 25 \, \text{m}, 50 \, \text{m}, \) and \( 75 \, \text{m} \), were calculated from the appropriate equations, and the results are given in Tables 1–3, for the maximum recovery, maximum output and maximum removal rate, respectively.

4.2 Results and Discussion

The comparison of deuterium recoveries obtained in the Frazier schemes of \( N^* \) columns and those obtained in the Frazier scheme of \( N \) (specified) columns, with given values of flow rate \( \sigma \) and same total sum of column heights \( L \), are given in Table 1. It is shown in this table that the most optimal column number \( N^* \) for the best deuterium recovery increases as the flow rate \( \sigma \), or the total sum of column heights \( L \), increases. The reason for this result is that increasing \( \sigma \) will decrease the contact time for the transversely flowing streams mixed with the fluids at the top and bottom ends of each thermal diffusion column. In this case the transport path, as well as the transport time, for fluids flowing through a thermal diffusion column should be properly shortened, while still somewhat preserving the effective separation section in the column, leading to increased the most optimal column numbers for the total sum of column heights fixed.

<table>
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<th>( \sigma ) (g/h)</th>
<th>( \Delta D/F )</th>
<th>( N^* )</th>
<th>( h^*_a ) (m)</th>
<th>( \Delta D_{\text{max}}/F )</th>
<th>( I_\Delta ) (%)</th>
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$N'_\lambda = N = 20$ and $(\Delta D_{\text{max}}/F) = (\Delta D/F) = 23.17$; also when $L = 75$ m and $\sigma = 0.6696$ g/h, $N'_\lambda = N = 30$ and $(\Delta D_{\text{max}}/F) = (\Delta D/F) = 23.36$. Therefore, the experimental system in the previous work [18] carried out at these operating conditions will be exactly the cases where the column height $h$ and column number $N$ are optimal and the separations are maximum. Consequently, the improvement in deuterium recovery $I_\lambda$ increases as the most optimal column number $N'_\lambda$ goes far from the specified column number $N$, and the increment is more obvious for smaller total sum of column heights.

Table 2 shows the comparison of production rates obtained in the Frazier scheme of $N'_\lambda$ columns and those obtained in the Frazier scheme of $N$ (specified) columns, with given values of $L$ and $\Delta D$. It is shown in this table that a larger value of the most optimal column number $N'_\lambda$ for the best production rate $\sigma_{\text{max}}$ is needed for a lower value of $\Delta D$, as well as for a larger value of $L$. The reason for this fact is the same as that described for the results in Table 1 because that decreasing the specified value of $\Delta D$ will lead to increased the flow rate. Further, when $L = 25$ m and $\Delta D/F = 22.58$, $N'_\lambda = N = 10$ and $\sigma_{\text{max}} = \sigma = 0.2232$ g/h; again when $L = 50$ m and $\Delta D/F = 23.17$, $N'_\lambda = N = 20$ and $\sigma_{\text{max}} = \sigma = 0.4464$ g/h; also when $L = 75$ m and $\Delta D/F = 23.36$, $N'_\lambda = N = 30$ and $\sigma_{\text{max}} = \sigma = 0.6696$ g/h. Therefore, the experimental system in previous work [18] performing at these operating conditions will be exactly the

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<th>$\Delta D/F$</th>
<th>$\sigma$ (g/h)</th>
<th>$N'_\lambda$</th>
<th>$h'_\lambda$ (m)</th>
<th>$\sigma_{\text{max}}$ (g/h)</th>
<th>$I_\lambda$ (%)</th>
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(b) 37.28 0.1 12 4.17 0.167 66.99 31.70 0.2 14 3.57 0.234 17.00 24.40 0.4 18 2.78 0.400 0 16.71 0.8 28 1.79 0.870 8.75 10.25 1.6 45 1.11 2.344 46.50 5.78 3.2 85 0.59 7.430 132.19 3.09 6.4 153 0.33 26.190 309.22 10.25 1.6 33 2.27 0.807 0.88 13.87 1.6 50 1.50 1.924 20.25 8.17 3.2 86 0.87 5.591 74.72 4.48 6.4 158 0.47 18.693 192.08

(c) 40.16 0.1 18 4.17 0.221 122.10 35.65 0.2 19 3.95 0.282 41.00 29.12 0.4 24 3.13 0.427 6.75 (23.36) (0.6696) (30) (2.50) (0.6696) (0) 21.31 0.8 33 2.27 0.807 0.88 13.87 1.6 50 1.50 1.924 20.25 8.17 3.2 86 0.87 5.591 74.72 4.48 6.4 158 0.47 18.693 192.08

Table 2. Comparison of production rates ($\sigma$ and $\sigma_{\text{max}}$) obtained in the Frazier schemes with $N$ and $N'_\lambda$ columns, respectively, with $h = L/N = 2.5$ m: (a) $L = 25$ m and $N = 10$; (b) $L = 50$ m and $N = 50$; (c) $L = 75$ m and $N = 30$

Table 3(a). Optimal column numbers ($N'_\lambda$ and $N'_\alpha$) and the corresponding maximum removal rates of deuterium ($M'_\lambda$ and $M'_\alpha$) with $L = 25$ m: $F \times 10^2 = 0.1165$ ($C_{3,F} = 0.1$), 0.1329 ($C_{3,F} = 0.3$), 0.1106 ($C_{3,F} = 0.5$), 0.0726 ($C_{3,F} = 0.7$), 0.0257 ($C_{3,F} = 0.9$)

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<th>$N'<em>\lambda$ or $N'</em>\alpha$</th>
<th>$\sigma$ or $\sigma_{\text{max}}$</th>
<th>$(\Delta D_{\text{max}}/F)$ or $(\Delta D/F)$</th>
<th>$(M'<em>\lambda/F)$ or $(M'</em>\alpha/F)$</th>
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<td>9(9)</td>
<td>0.2</td>
<td>23.80</td>
<td>4.760</td>
<td>0.555</td>
</tr>
<tr>
<td>10(10)</td>
<td>0.2</td>
<td>22.58</td>
<td>4.516</td>
<td>0.526</td>
</tr>
<tr>
<td>13</td>
<td>0.4</td>
<td>17.09</td>
<td>6.836</td>
<td>0.796</td>
</tr>
<tr>
<td>16</td>
<td>0.586</td>
<td>14.22</td>
<td>8.333</td>
<td>0.971</td>
</tr>
<tr>
<td>18</td>
<td>0.8</td>
<td>12.20</td>
<td>9.760</td>
<td>1.137</td>
</tr>
<tr>
<td>22</td>
<td>1.17</td>
<td>10.14</td>
<td>11.864</td>
<td>1.382</td>
</tr>
<tr>
<td>26</td>
<td>1.6</td>
<td>8.70</td>
<td>13.920</td>
<td>1.622</td>
</tr>
<tr>
<td>37</td>
<td>3.2</td>
<td>6.19</td>
<td>19.808</td>
<td>2.308</td>
</tr>
<tr>
<td>40</td>
<td>3.726</td>
<td>5.74</td>
<td>21.387</td>
<td>2.492</td>
</tr>
<tr>
<td>77</td>
<td>13.095</td>
<td>3.08</td>
<td>40.333</td>
<td>4.699</td>
</tr>
</tbody>
</table>
cases where the column height $h$ and column number $N$ are the optimal and the production rate is maximum. Consequently, the improvement in the production rate of deuterium $I_o$ increases when $N^*_N - N^*$ increases.

The optimal column numbers ($N^*_N$ and $N^*_o$) and the corresponding maximum removal rates ($M_A$ and $M_o$) of deuterium calculated by Eqs. (28) and (29), respectively, are combined from Tables 1 and 2, and the results are listed in Table 3. It is observed from Table 3 that the removal rates increase when the flow rate (or the production rate), or the total sum of column heights $L$, increases, and that $M_A = M_o$ as $N^*_N = N^*_o$. This fact has been already verified by Eq. (31). Considerable improvements in deuterium removal rates, $I_o$ and $I_o$, are achieved if the Frazier schemes of fixed total sum of column heights are constructed with the optimal numbers.

### Table 3(b). Optimal column numbers ($N^*_N$ and $N^*_o$) and the corresponding maximum removal rates of deuterium ($M_A$ and $M_o$) with $L = 50 \text{m}$: $F \times 10^2 = 0.1165 (C_{3,F} = 0.1), 0.1329 (C_{3,F} = 0.3), 0.1106 (C_{3,F} = 0.5), 0.0726 (C_{3,F} = 0.7), 0.0257 (C_{3,F} = 0.9)$

<table>
<thead>
<tr>
<th>$N^<em>_N$ or $N^</em>_o$</th>
<th>$\sigma$ or $\sigma_{max}$</th>
<th>$(\Delta_{D,max}/F)$ or $(\Delta_{D}/F)$</th>
<th>$(M_A/F)$ or $(M_o/F)$</th>
<th>$M_A \times 10^2$ and $M_o \times 10^2 \text{ (g/h)}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$C_{3,F} = 0.1 \text{ C}<em>{3,F} = 0.3 \text{ C}</em>{3,F} = 0.5 \text{ C}<em>{3,F} = 0.7 \text{ C}</em>{3,F} = 0.9$</td>
</tr>
<tr>
<td>9</td>
<td>0.1</td>
<td>47.60</td>
<td>4.760</td>
<td>0.555 0.633 0.526 0.346 0.122</td>
</tr>
<tr>
<td>12</td>
<td>0.167</td>
<td>37.28</td>
<td>6.226</td>
<td>0.725 0.827 0.689 0.452 0.160</td>
</tr>
<tr>
<td>13</td>
<td>0.2</td>
<td>34.18</td>
<td>6.836</td>
<td>0.796 0.909 0.756 0.496 0.176</td>
</tr>
<tr>
<td>18(18)</td>
<td>0.4</td>
<td>24.41</td>
<td>9.764</td>
<td>1.138 1.298 1.080 0.709 0.251</td>
</tr>
<tr>
<td>20(20)</td>
<td>0.4464</td>
<td>23.17</td>
<td>10.343</td>
<td>1.205 1.375 1.144 0.751 0.266</td>
</tr>
<tr>
<td>26</td>
<td>0.8</td>
<td>17.74</td>
<td>14.192</td>
<td>1.653 1.886 1.570 1.030 0.365</td>
</tr>
<tr>
<td>28</td>
<td>0.87</td>
<td>16.71</td>
<td>14.538</td>
<td>1.694 1.932 1.608 1.055 0.374</td>
</tr>
<tr>
<td>37</td>
<td>1.6</td>
<td>12.38</td>
<td>19.808</td>
<td>2.308 2.632 2.191 1.438 0.509</td>
</tr>
<tr>
<td>45</td>
<td>2.344</td>
<td>10.25</td>
<td>24.026</td>
<td>2.799 3.193 2.657 1.744 0.617</td>
</tr>
<tr>
<td>53</td>
<td>3.2</td>
<td>8.79</td>
<td>28.128</td>
<td>3.277 3.738 3.111 2.042 0.723</td>
</tr>
<tr>
<td>75</td>
<td>6.4</td>
<td>6.23</td>
<td>39.872</td>
<td>4.645 5.299 4.410 2.895 1.025</td>
</tr>
<tr>
<td>85</td>
<td>7.43</td>
<td>5.78</td>
<td>42.945</td>
<td>5.003 5.707 4.750 3.118 1.104</td>
</tr>
</tbody>
</table>

### Table 3(c). Optimal column numbers ($N^*_N$ and $N^*_o$) and the corresponding maximum removal rates of deuterium ($M_A$ and $M_o$) with $L = 75 \text{m}$: $F \times 10^2 = 0.1165 (C_{3,F} = 0.1), 0.1329 (C_{3,F} = 0.3), 0.1106 (C_{3,F} = 0.5), 0.0726 (C_{3,F} = 0.7), 0.0257 (C_{3,F} = 0.9)$

<table>
<thead>
<tr>
<th>$N^<em>_N$ or $N^</em>_o$</th>
<th>$\sigma$ or $\sigma_{max}$</th>
<th>$(\Delta_{D,max}/F)$ or $(\Delta_{D}/F)$</th>
<th>$(M_A/F)$ or $(M_o/F)$</th>
<th>$M_A \times 10^2$ and $M_o \times 10^2 \text{ (g/h)}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$C_{3,F} = 0.1 \text{ C}<em>{3,F} = 0.3 \text{ C}</em>{3,F} = 0.5 \text{ C}<em>{3,F} = 0.7 \text{ C}</em>{3,F} = 0.9$</td>
</tr>
<tr>
<td>11</td>
<td>0.1</td>
<td>58.83</td>
<td>5.883</td>
<td>0.685 0.782 0.651 0.427 0.151</td>
</tr>
<tr>
<td>16</td>
<td>0.2</td>
<td>42.15</td>
<td>8.430</td>
<td>0.982 1.120 0.932 0.612 0.217</td>
</tr>
<tr>
<td>18</td>
<td>0.221</td>
<td>40.16</td>
<td>8.875</td>
<td>1.034 1.180 0.982 0.644 0.228</td>
</tr>
<tr>
<td>19</td>
<td>0.282</td>
<td>35.65</td>
<td>10.053</td>
<td>1.171 1.336 1.112 0.730 0.258</td>
</tr>
<tr>
<td>23</td>
<td>0.4</td>
<td>30.07</td>
<td>12.028</td>
<td>1.401 1.599 1.330 0.873 0.309</td>
</tr>
<tr>
<td>24</td>
<td>0.427</td>
<td>29.12</td>
<td>12.434</td>
<td>1.449 1.653 1.375 0.903 0.320</td>
</tr>
<tr>
<td>30(30)</td>
<td>0.6696</td>
<td>23.36</td>
<td>15.642</td>
<td>1.822 2.079 1.730 1.136 0.402</td>
</tr>
<tr>
<td>32</td>
<td>0.8</td>
<td>21.98</td>
<td>17.584</td>
<td>2.049 2.337 1.945 1.277 0.452</td>
</tr>
<tr>
<td>33</td>
<td>0.807</td>
<td>21.31</td>
<td>17.197</td>
<td>2.003 2.286 1.902 1.249 0.442</td>
</tr>
<tr>
<td>46</td>
<td>1.6</td>
<td>15.20</td>
<td>24.320</td>
<td>2.833 3.232 2.690 1.766 0.625</td>
</tr>
<tr>
<td>50</td>
<td>1.924</td>
<td>13.87</td>
<td>26.686</td>
<td>3.109 3.547 2.951 1.937 0.686</td>
</tr>
<tr>
<td>65</td>
<td>3.2</td>
<td>10.78</td>
<td>34.496</td>
<td>4.019 4.585 3.815 2.504 0.887</td>
</tr>
<tr>
<td>86</td>
<td>5.591</td>
<td>8.17</td>
<td>45.678</td>
<td>5.322 6.071 5.052 3.316 1.174</td>
</tr>
<tr>
<td>92</td>
<td>6.4</td>
<td>7.21</td>
<td>46.144</td>
<td>5.376 6.133 5.104 3.350 1.186</td>
</tr>
</tbody>
</table>
of column. For instance, when \( L = 75 \text{ m} \) and \( \frac{\Delta_T}{F} = 4.48 \), one has \( N'_{\text{opt}} = 158 \), \( h'_{\text{opt}} = 0.47 \text{ m} \) and \( I = 192.08\% \) based on that obtained in the device with \( N = 30 \) and \( h = 2.5 \text{ m} \), as shown in Table 2.

5. Conclusion

The equations which may be employed to predict the optimal column numbers, \( N_\text{A,opt} \) and \( N_\text{B,opt} \), for the maximum performances of deuterium recovery from water isotope mixture in the Frazier scheme of thermal diffusion columns with total sum of column height fixed, have been derived. They are Eqs. (24) and (25) for the maximum degree of recovery and the maximum production rate, respectively. As mentioned in the previous section, the most optimal column numbers, \( N'_{\text{A}} \) and \( N'_{\text{B}} \), should be the positive integers, which are nearest to, and smaller than, \( N_\text{A,opt} \) and \( N_\text{B,opt} \), respectively. Accordingly, the best degree of deuterium recovery \( \Delta_{\text{D,max}} \) and production rate \( \sigma_{\text{max}} \) may be calculated from Eqs. (22) and (23), respectively, by setting \( N = N'_{\text{A}} \) and \( N = N'_{\text{B}} \). The improvements in performances were illustrated numerically by employing the experimental data obtained in the previous work, and the result are presented in Tables 1–3.

It is seen in these tables that considerable improvements in performances for recovery of deuterium from water-isotope mixture by thermal diffusion in the Frazier schemes can be achieved if a Frazier schemes is constructed with the most optimal numbers of thermal diffusion columns with column height \( h'_{\text{opt}} = L / N'_{\text{A}} \) or \( h'_{\text{opt}} = L / N'_{\text{B}} \). We may also observe that the flow rate of feed affects very significantly the most optimal column number, as well as the optimal column height and should be limited for practical applications. As shown in Table 2 for small \( L \), the optimal column height may decrease impractically when the flow rate \( \sigma \) (or production rate) continuously increases due to the decrease in \( \Delta_{\text{D}} \). For instance, when \( L = 25 \text{ m} \), \( \frac{\Delta_{\text{D}}}{F} = 1.6 \), and \( \sigma = 6.4 \text{ g/h} \), one has the impractical optimal column height as \( h'_{\text{opt}} = 0.17 \text{ m} \) too small. Therefore, the optimal design of column number in a Frazier scheme should be limited without low flow rate or high degree of recovery specified.

Actually, the manufacture and assembly costs will increase with the column number in a Frazier scheme. In practical applications, therefore, the increase in manufacture assembly costs due to the increase of column number in a Frazier scheme should be also taken into consideration.

### Nomenclature

- \( B \): column width, (cm)
- \( C_j \): fractional mass concentration of j component or atom in H$_2$O-HDO-D$_2$O binary system, \( j = 1 \) (H$_2$O), 2 (HDO), 3 (D$_2$O), D (deuterium atom)
- \( C_F \): C in feed streams
- \( C_{T,T}, C_{B} \): C in product streams exiting from the top end of last column, the bottom end of first column, respectively
- \( C_{j,T}, C_{j,B} \): C for component j
- \( D \): ordinary diffusion coefficient (cm$^2$/s)
- \( g \): gravitational acceleration (cm/s$^2$)
- \( H \): system constant defined by Eq. (5) (g/s)
- \( h \): column height (cm)
- \( I_{\text{A}}, I_{\sigma} \): improvements of deuterium removal rate, defined by Eqs. (33) and (34), respectively
- \( J_{X,OD}, J_{Z,OD} \): mass flux of heavy water in x-direction, in z-direction, due to ordinary diffusion (g/cm$^2$ s)
- \( J_{X,TD} \): mass flux of heavy water in x-direction due to thermal diffusion (g/cm$^2$ s)
- \( K \): system constant defined by Eq. (6) (g cm/s)
- \( L \): total sum of column heights, \( Nh \), for \( N \) columns in a Frazier scheme (cm)
- \( M \): deuterium removal rate (g/h)
- \( N \): column number in a Frazier scheme
- \( T_m \): mean absolute temperature (K)
- \( \Delta T \): difference in temperature of hot and cold plates (K)
- \( x \): axis in temperature-gradient direction (cm)
- \( z \): axis in transport direction (cm)

### Greek Letters

- \( \alpha \): thermal-diffusion constant
- \( \beta \): \(-(1/\rho)(\partial \rho/\partial T)\) evaluated at \( T_m \) under constant pressure (1/K)
- \( \Delta \): degree of separation, \( C_0-C_T \) in the counter-current-flow Frazier scheme
- \( \mu \): absolute viscosity, (g/cm s)
ρ mass density evaluated at Tm (g/cm³)
σ mass flow rate, or production rate (g/s)
ω half of plate spacing (cm)

Superscripts
* most optimal value

Subscripts
B bottom end of the column
j j component
N number of columns
T top end of the column
opt optimal
Δ degree of recovery
ρ production rate
max maximum value

References


[18] Yeh, H. M. and Yang, S. C., “The Enrichment of


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