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## **The Study of Continuous Thermal-Diffusion Columns on Modified Frazier-Scheme for the Enrichment of Heavy Water with Column Length Varied at a Constant Ratio**

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### **ABSTRACT**

A study of heavy water in continuous thermal-diffusion columns on modified Frazier-scheme, resulting in substantial improvement of the separation efficiency of heavy water, was developed and investigated with column length varied at a constant ratio and with flow-rate fraction variations. The analytical results are represented graphically and compared with that in a classical Frazier-scheme of the same column length. Considerable improvement in the enrichment of heavy water was obtained by employing such a modified Frazier-scheme, with column length varied at a constant ratio and with flow-rate fraction variations instead of using a classical Frazier-scheme thermal diffusion column with

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the same column length. The effect of the ratio of column length and the column number as well as the flow-rate fraction variation on the enhancement of the separation efficiency of heavy water is discussed.

*Key Words:* Thermal diffusion; Frazier-scheme; Column length variation; Water isotopes.

## INTRODUCTION

The general term, thermal-diffusion column, denotes a method that includes a temperature gradient for giving rise to one component accumulated near the hot wall and the other component accumulated near the cold wall, leading to concentration gradient and purifying materials. The concept was discovered and first introduced by Clusius and Dickel,<sup>[1,2]</sup> and later, a complete theory presentation for a Clusius–Dickel column was developed by Furry et al.<sup>[3,4]</sup> It is a powerful purification technique for separating the mixtures, which are difficult to separate by means of conventional methods such as extraction and distillation. The first application of thermal diffusion was to separate the isotope mixtures of uranium at Oak Ridge during World War II. This method is exceptionally attractive for the separation of hydrogen isotopes, which may serve as an indirect nuclear fuel for fusion reactors, due to the large ratios of molecular weights. Recently, the enrichment of heavy water in the Clusius–Dickel column was investigated both theoretically and experimentally.<sup>[5–7]</sup>

A series of connected vertical thermodiffusion columns of the same column length can be used in a single operation, as in Frazier-scheme thermal-diffusion columns.<sup>[8,9]</sup> The advantages of the Frazier-scheme are that operating time can be saved and the separation efficiency is tremendously increased. The desirable cascading effect and an undesirable remixing effect are the two conflicting effects to be confronted in the significant factor of convective current of thermal-diffusion separation columns. Further, if a suppression of the remixing effect and an enhancement of the cascading effect are suitably adjusted in designing improved columns, the maximum degree of separation efficiency can be accomplished. The governing equation for modeling thermal-diffusion separation according to the Frazier-scheme for binary mixture, was presented previously.<sup>[10,11]</sup> Consequently, many investigators studied the separation efficiency of thermal-diffusion columns. It depends on the column with inclination,<sup>[12,13]</sup> rotation,<sup>[14]</sup> moving wall,<sup>[15]</sup> packing,<sup>[16]</sup> and winding a wire helix.<sup>[17]</sup>

There are two purposes in this work: first, to develop and to investigate the separation theory in a modified Frazier-scheme thermal-diffusion column with column length varied at a constant ratio and with flow-rate fraction variations; second, to study theoretically the effects of column lengths and flow-rate fractions, which are varied in each column, on the performance of modified Frazier-scheme thermal-diffusion columns. The best column number, the best ratio of column length, and the maximum degree of separation efficiency is also determined as a function of dimensionless mass-flow rate.

## THEORETICAL FORMULATIONS

### Modified Frazier-Scheme Thermal-Diffusion Column

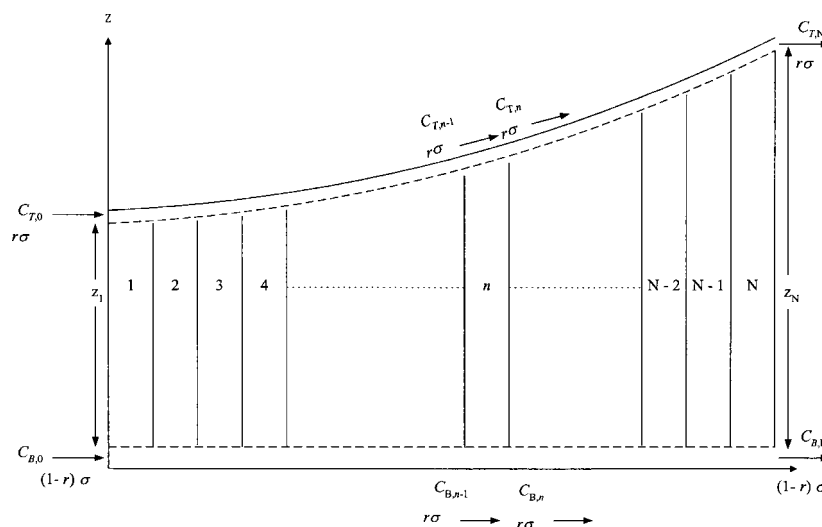
A theoretical analysis of a Frazier-scheme thermal-diffusion column of the same column length was introduced and developed by Rabinovich.<sup>[10,11]</sup> A modified Frazier-scheme, concentric-tube, thermal-diffusion column having summing column length  $L$ , perimeter length  $B$ , and annular spacing  $2w$ , which is to carry out forward transverse sampling streams of the same direction and with column length varied at a constant ratio is shown in Fig. 1. The mass-flow rates  $r\sigma$  and  $(1-r)\sigma$  with concentration  $C_0$  are accomplished at the upper and lower ends, respectively, with the product end and the supply entrance on the opposite sides.

Furry, Jones, and Onsager<sup>[3]</sup> give a well-developed theory to present the separation equation for a thermal-diffusion column in continuous operations. If the perimeter length, the annular spacing, the flow-rate ratio, and the temperature difference in each column are all the same, except the column length, for continuous operation, the transport equations for the  $n$ th column may be modified from the previous results as follows:

$$-\tau_n = -HC\hat{C} + K \frac{dC_n}{dz} \Big|_{z=z_n} = r\sigma(C_{T,n-1} - C_{T,n}) \quad (1)$$

for the stripping section, and

$$-\tau_n = -HC\hat{C} + K \frac{dC_n}{dz} \Big|_{z=0} = (1-r)\sigma(C_{B,n} - C_{B,n-1}) \quad (2)$$



**Figure 1.** The modified Frazier-scheme thermal-diffusion columns with column length varied at a constant ratio and with the flow-rate fraction variation.

for the enriching section. The pseudo concentration product,  $C\hat{C}$  is defined as

$$C\hat{C} = C \left\{ 0.05263 - (0.05263 - 0.0135K_{eq})C - 0.027 \left\{ \left[ 1 - \left( 1 - \frac{K_{eq}}{4} \right) C \right] CK_{eq} \right\}^{\frac{1}{2}} \right\} \quad (3)$$

in which the equilibrium constant  $K_{eq}$  for the following equilibrium relation



is

$$K_{eq} = \frac{[\text{HDO}]^2}{[\text{H}_2\text{O}][\text{D}_2\text{O}]} \times \frac{19 \times 19}{18 \times 20} \quad (5)$$

$K_{eq}$  does not vary sensitively within the operating temperature range.

For instance, the values of the equilibrium constant are  $K_{eq} = 3.80$  and  $3.793$ , respectively, at  $T = 25^\circ\text{C}$  and  $30.5^\circ\text{C}$ .<sup>[18]</sup>

The simultaneous solution of this set of equations is complicated and inconvenient to analyze due to the nonlinear forms of eqs. (1) and (2). In the present study, a rather reasonable approximate for  $C\hat{C} = A$  in a Frazier-scheme thermal-diffusion column, the appropriate value of  $C\hat{C} = A$ , may be determined using the least squares method as in the previous works.<sup>[7]</sup> The calculation of this constant was conducted in the whole column from  $C_T$  to  $C_B$ . Accordingly, minimizing the following integration

$$E = \int_{C_T}^{C_B} (C\hat{C} - A)^2 dC \quad (6)$$

that is,

$$\frac{dE}{dA} = \int_{C_T}^{C_B} -2(C\hat{C} - A) dC = 0 \quad (7)$$

one obtains

$$A = \frac{1}{C_B - C_T} \int_{C_T}^{C_B} C\hat{C} dC \quad (8)$$

It is noted from eq. (8) that the appropriate value of  $A$ , thus obtained, is exactly the average value of  $C\hat{C}$  in the concentration range. Also, the transport constants in the above equations are defined by

$$H = \frac{\alpha\beta_T\rho g(2\omega)^3 B(\Delta T)^2}{6!\mu\bar{T}} < 0 \quad \text{for } \alpha < 0 \quad (9)$$

and

$$K = \frac{\rho g^2 \beta_T^2 (2\omega)^7 B(\Delta T)^2}{9!\mu^2 D} + 2\omega\rho DB \quad (10)$$

Equations (3) and (4) were obtained according to Jones and Furry theory with the physical properties considered as constants.

Making material balance around the  $n$ th column yields

$$r\sigma(C_{T,n-1} - C_{T,n}) = (1 - r)\sigma(C_{B,n} - C_{B,n-1}) \quad (11)$$

Substitution of eqs. (1) and (2) into eq. (11) gives

$$\left. \frac{dC_n}{dz} \right|_{z=z_n} = \left. \frac{dC_n}{dz} \right|_{z=0} \quad (12)$$

Thus,  $(dC_n/dz)$  may be assumed to be constant through the entire column. Integrating eqs. (1) and (2) from  $z = 0$  to  $z = z_n$  for the  $n$ th column, we have

$$C_{T,n} - C_{B,n} = \frac{AHz_n}{K} + \frac{r\sigma z_n}{K}(C_{T,n-1} - C_{T,n}) \quad (13)$$

$$C_{T,n} - C_{B,n} = \frac{AHz_n}{K} + \frac{(1-r)\sigma z_n}{K}(C_{B,n} - C_{B,n-1}) \quad (14)$$

Combination of eqs. (13) and (14) with setting  $\Delta_n = C_{B,n} - C_{T,n}$ , one obtains

$$\Delta_n - \frac{\frac{z_n\sigma(1-r)}{2K}}{1 + \frac{z_n\sigma(1-r)}{2K} + \frac{1-2r}{2r}} \cdot \Delta_{n-1} + \frac{\frac{Hz_nA}{K} \cdot \left(1 + \frac{1-2r}{2r}\right)}{1 + \frac{z_n\sigma(1-r)}{2K} + \frac{1-2r}{2r}} = 0 \quad (15)$$

Equation (15) may be rewritten into a first-order difference equation of  $\Delta_n$  as follows:

$$\Delta_n - \rho\Delta_{n-1} = \phi \quad (16)$$

where

$$\rho = \frac{\frac{z_n\sigma(1-r)}{2K}}{1 + \frac{z_n\sigma(1-r)}{2K} + \frac{1-2r}{2r}} = \frac{Z_n \cdot r(1-r)}{Z_n \cdot r(1-r) + u} \quad (17)$$

$$\begin{aligned} \phi &= \frac{\frac{-Hz_nA}{K} \cdot \left(1 + \frac{1-2r}{2r}\right)}{1 + \frac{z_n\sigma(1-r)}{2K} + \frac{1-2r}{2r}} = \frac{-Hz_nA}{K}(1-\rho) \\ &= -Au \frac{Z_n}{Z_n \cdot r(1-r) + u} \end{aligned} \quad (18)$$

in which

$$Z_n = \frac{Hz_n}{K} \quad (19)$$

$$u = \frac{H}{\sigma} \quad (20)$$

# Continuous Thermal-Diffusion Columns

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The solution of eq. (16) for the degree of separation obtained in modified Frazier-scheme thermal-diffusion columns is calculated as follows:

$$\begin{aligned}\Delta_N &= C_{B,N} - C_{T,N} \\ &= -Au \cdot \left\{ \frac{Z_N}{Z_N \cdot r(1-r) + u} + \frac{Z_N Z_{N-1} [r(1-r)]}{[Z_N \cdot r(1-r) + u][Z_{N-1} \cdot r(1-r) + u]} \right. \\ &\quad \left. + \dots + \frac{Z_N Z_{N-1} \dots Z_1 [r(1-r)]^{N-1}}{[Z_N \cdot r(1-r) + u][Z_{N-1} \cdot r(1-r) + u] \dots [Z_1 \cdot r(1-r) + u]} \right\}\end{aligned}\quad (21)$$

The sum of the column length,  $L$ , of  $N$  columns in the Frazier-scheme is given and the ratio,  $R$ , of every two neighboring columns is kept constant, that is,

$$\frac{z_2}{z_1} = \frac{z_3}{z_2} = \dots = \frac{z_N}{z_{N-1}} = R \quad (22)$$

$$L = z_1 + z_2 + \dots + z_N = z_1(1 + R + R^2 + \dots + R^{N-1}) \quad (23)$$

Then

$$Z_n = \frac{R^{n-1}(R-1)\zeta u}{R^N - 1} \quad (24)$$

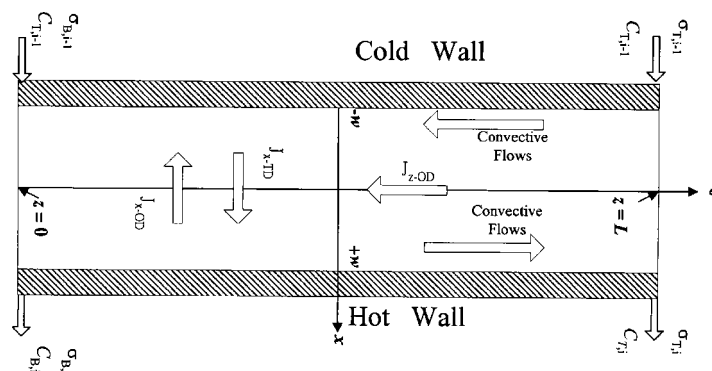
in which

$$\zeta = \frac{HL}{Ku} = \frac{\sigma L}{K} \quad (25)$$

Substituting eq. (24) into eq. (21), the degree of separation obtained in a modified Frazier-scheme thermal-diffusion column may be expressed as follows:

$$\begin{aligned}\Delta_N &= -Au \cdot \left\{ \left[ r(1-r) + \frac{R^{N-1}}{R^{N-1}(R-1)\zeta} \right]^{-1} + \right. \\ &\quad \left[ r(1-r) + \frac{R^{N-1}}{R^{N-1}(R-1)\zeta} \right]^{-1} \cdot \left[ r(1-r) + \frac{R^{N-1}}{R^{N-2}(R-1)\zeta} \right]^{-1} \cdot [r(1-r)] \\ &\quad + \dots + \\ &\quad \left[ r(1-r) + \frac{R^{N-1}}{R^{N-1}(R-1)\zeta} \right]^{-1} \cdot \left[ r(1-r) + \frac{R^{N-1}}{R^{N-2}(R-1)\zeta} \right]^{-1} \cdot \dots \\ &\quad \left. \left[ r(1-r) + \frac{R^{N-1}}{(R-1)\zeta} \right]^{-1} \cdot [r(1-r)]^{N-1} \right\}\end{aligned}\quad (26)$$





**Figure 2.** A single column of the classical Frazier-scheme thermal-diffusion column.

or

$$\Delta_N = -Au \left\{ \frac{1 + \sum_{\beta=0}^{N-2} \left\{ [r(1-r)]^{-(\beta+1)} \cdot \prod_{\alpha=0}^{\beta} \left[ 1 + \frac{R^N - 1}{R^{\alpha}(R-1)\xi} \right] \right\}}{[r(1-r)]^{-(N-1)} \cdot \prod_{n=0}^{N-1} \left[ 1 + \frac{R^N - 1}{R^n(R-1)\xi} \right]} \right\} \\ = -Au \{ f(N, R, \xi, r) \} \quad (27)$$

Equation (27) reduces to the case that the same length of each column,  $R = 1$ , in the classical Frazier-scheme, as shown in Fig. 2.

$$\begin{aligned}\Delta_N^0 &= C_{B,N} - C_{T,N} = \frac{-AHL}{KN} \cdot \left\{ 1 - \left[ \frac{\frac{HL}{NK}}{H\left(\frac{1}{\sigma_T} + \frac{1}{\sigma_B}\right) + \frac{HL}{NK}} \right]^N \right\} \\ &= \frac{-AHL}{NK} \cdot \left\{ 1 - \left[ \frac{\frac{HL}{NK}}{H\left(\frac{1}{r\sigma} + \frac{1}{(1-r)\sigma}\right) + \frac{HL}{NK}} \right]^N \right\} \\ &= -Au \cdot \frac{\zeta}{N_0} \cdot \left\{ 1 - \left[ \frac{\zeta}{\zeta + \frac{N_0}{r(1-r)}} \right]^N \right\} = -Au \{f(N_0, \zeta, r)\} \quad (28)\end{aligned}$$

**Best Column Number and Best Length Ratio**

The best column number  $N^*$  and best ratio of column length  $R^*$  for maximum degree of separation  $\Delta_{n,\max}$  with  $r$  and  $\zeta$  as parameters in a modified Frazier-scheme thermal-diffusion column are calculated by using the method of two variable univariant search.<sup>[19]</sup> The calculation procedure is to find  $N^*$  and  $R^*$  first and then  $\Delta_{n,\max}$  is obtained in eq. (27). The results show that  $N^*$ ,  $R^*$ , and  $\Delta_{n,\max}$  are almost proportional to  $\zeta$  for a given flow-rate fraction and thus may be expressed in straight lines. As an illustration,  $r = 0.5$

$$N^* = 0.7766\zeta^{0.5} \quad (29)$$

$$R^* = 3.110\zeta^{-0.5} + 1 \quad (30)$$

$$\frac{\Delta_{N,\max}}{-Au} = 1.226\zeta^{0.5} \quad (31)$$

Similarly, for a classical Frazier-scheme thermal-diffusion column with  $R = 1$  and  $r = 0.5$ , the best column number  $N_0^*$  and the maximum degree of separation  $\Delta_{n,\max}^0$  have been calculated and are presented in Figs. 6 and 7.

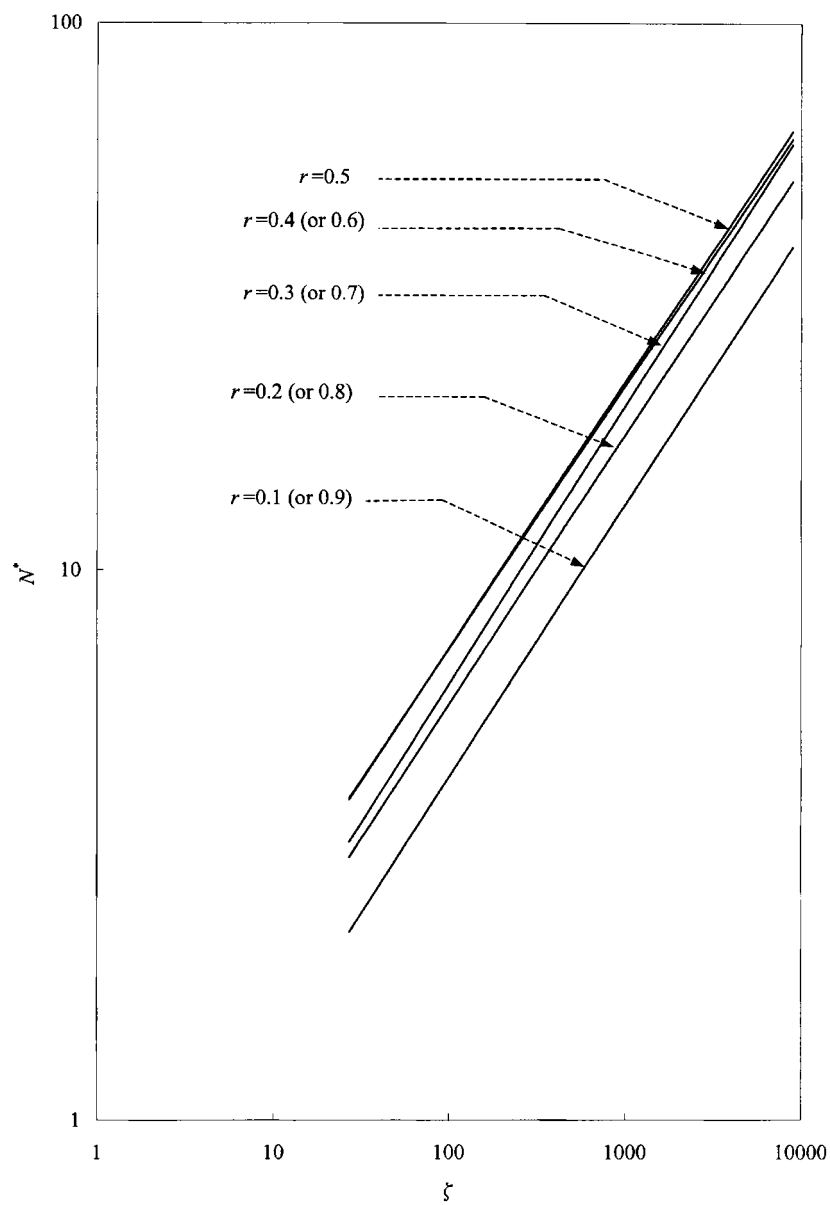
$$N_0^* = 0.6683\zeta^{0.5} \quad (32)$$

$$\frac{\Delta_{N,\max}^0}{-Au} = 1.161\zeta^{0.5} \quad (33)$$

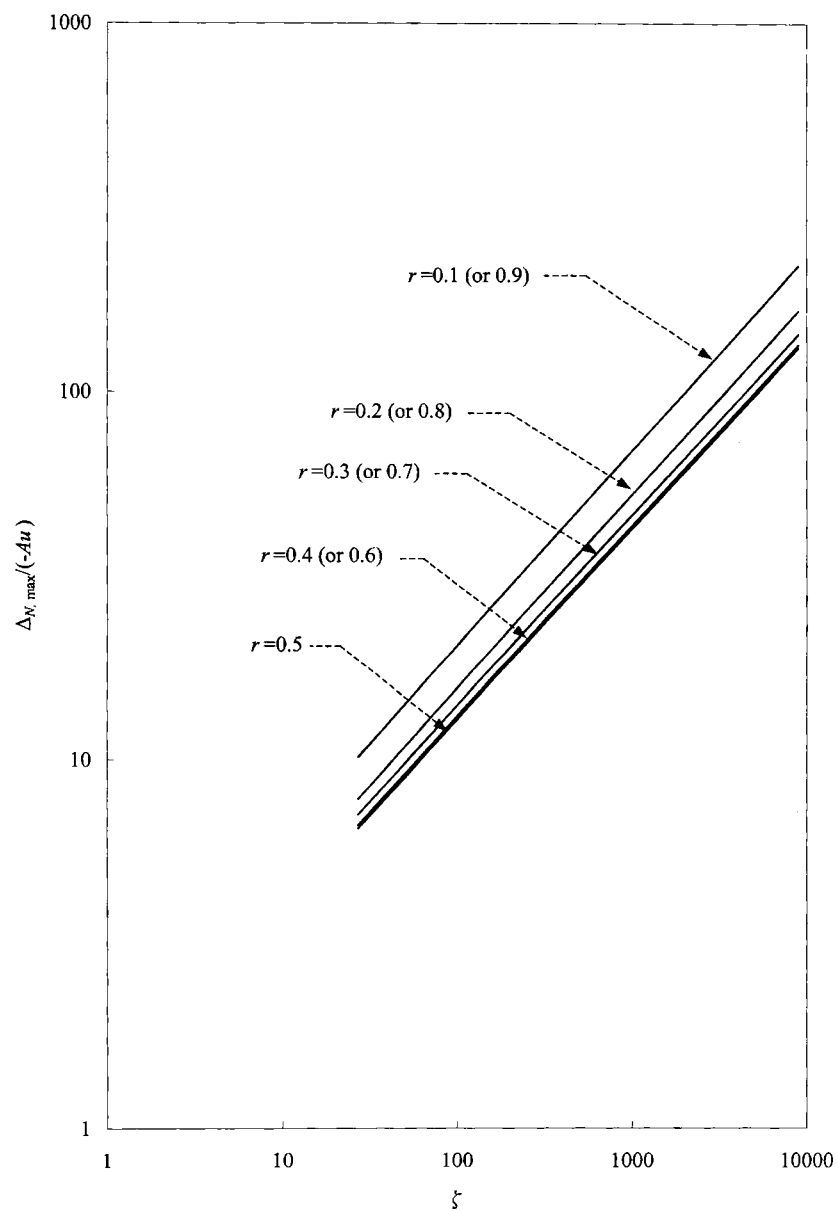
The relationship of the best column number,  $N^*$  and  $N_0^*$ , between both Frazier-scheme thermal-diffusion columns with and without modification can be obtained from eqs. (29) and (32)

$$\Delta_{N,\max} = 1.056\Delta_{N,\max}^0 \quad (34)$$

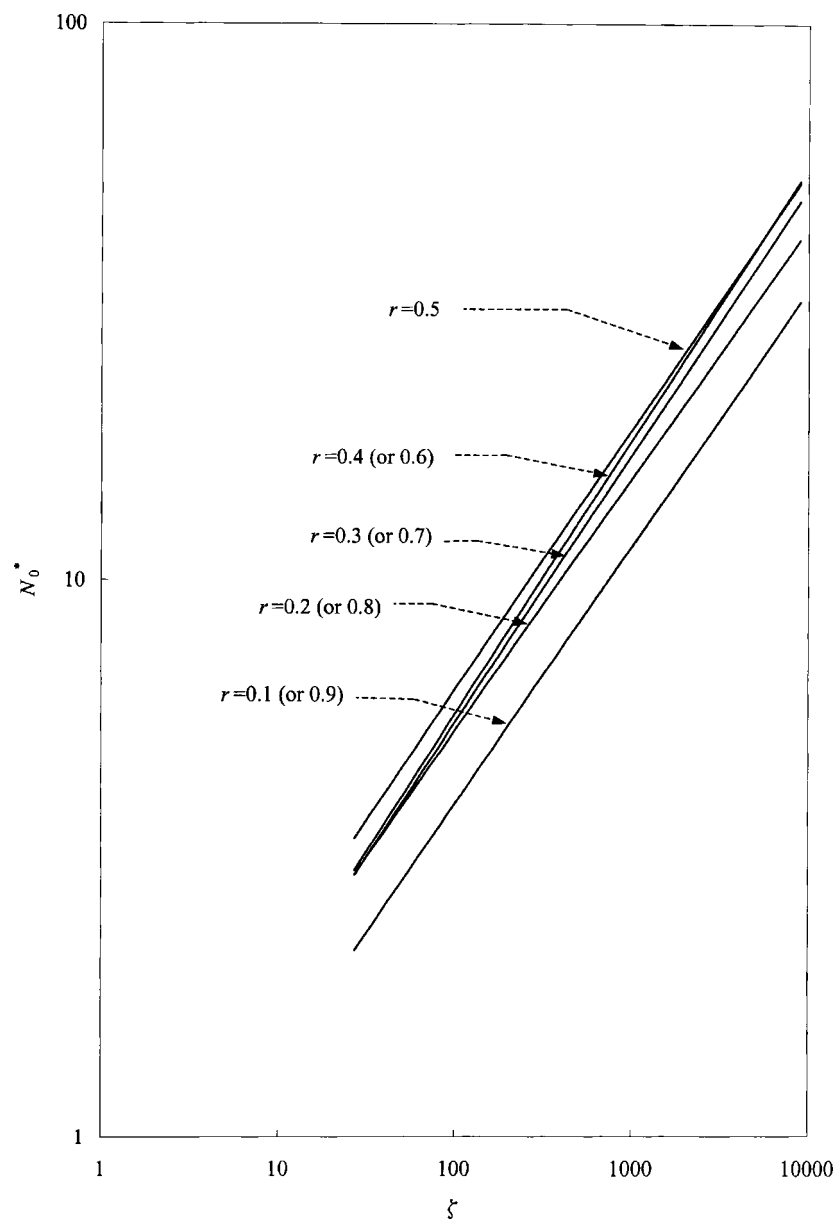
The results in the columns of constant and varied and Tables 1 and 2 column length are presented in Figs. 3 through 7.



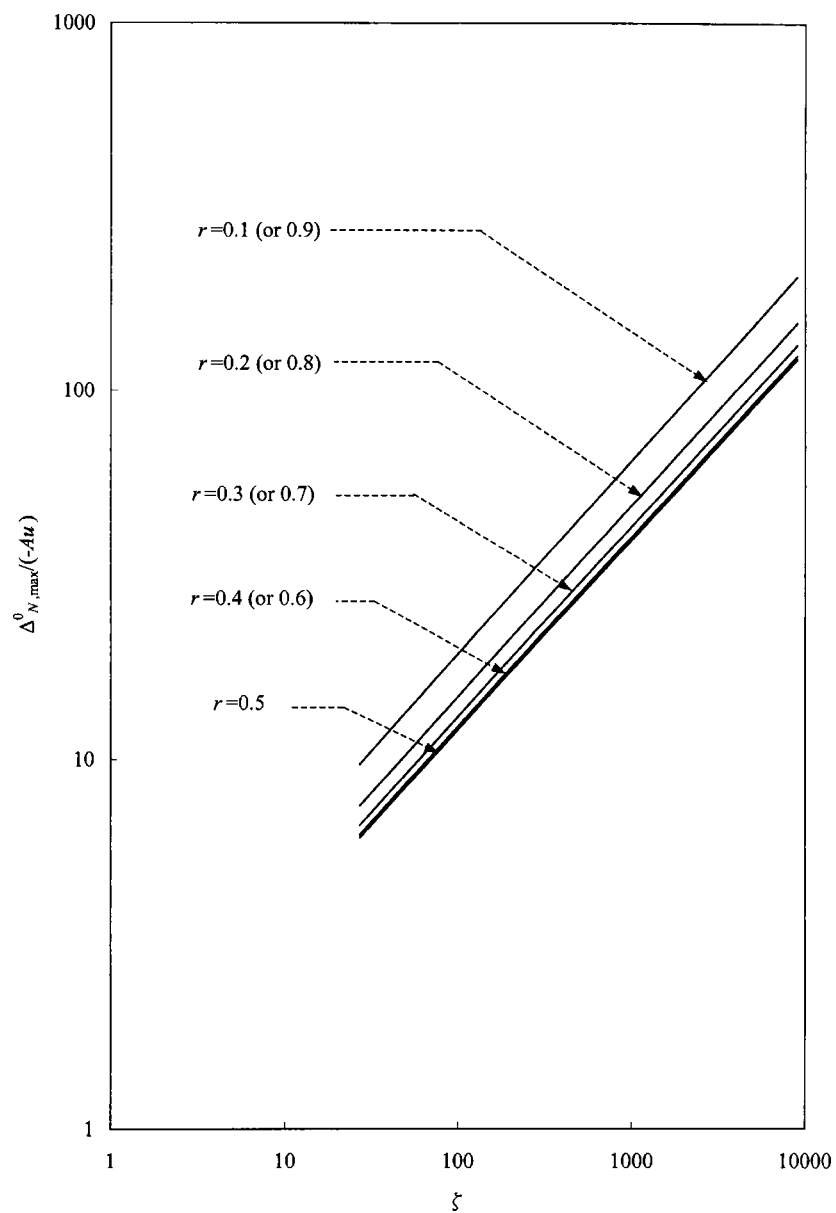
**Figure 3.** The best column number in modified Frazier-scheme thermal-diffusion columns.



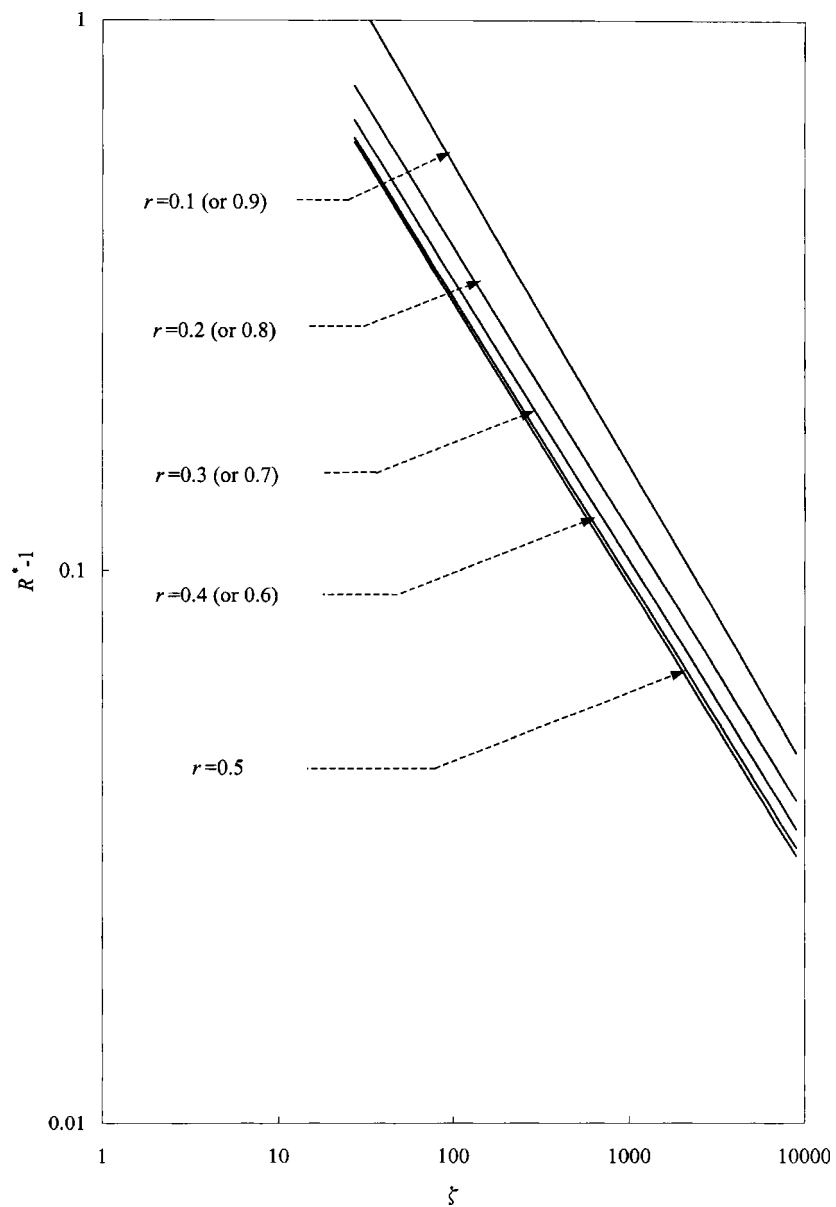
**Figure 4.** The maximum degree of separation in modified Frazier-scheme thermal-diffusion columns.



**Figure 5.** The best column number in classical Frazier-scheme thermal-diffusion columns.



**Figure 6.** The maximum degree of separation in classical Frazier-scheme thermal-diffusion columns.



**Figure 7.** The best column length ratio in modified Frazier-scheme thermal-diffusion columns.

**Table 1.** Comparison of separation obtained in the columns of constant and varied column length with flow-rate fraction as a parameter ( $\sigma = 0.1$  g/hr).

$r$	$R = 1$		$R \neq 1$		
	$N_0^*$	$\Delta_{N,\max}^0$	$N^*$	$R^*$	$\Delta_{N,\max}$
0.1	3	0.5045	4	1.56	0.5268
0.2	4	0.3867	5	1.41	0.4059
0.3	5	0.3419	5	1.35	0.3642
0.4	5	0.3203	6	1.33	0.3381
0.5	5	0.3135	6	1.32	0.3302
0.6	5	0.3203	6	1.33	0.3381
0.7	5	0.3419	5	1.35	0.3642
0.8	4	0.3867	5	1.41	0.4059
0.9	3	0.5045	4	1.56	0.5268

### IMPROVEMENT IN THE DEGREE OF SEPARATION EFFICIENCY

The improvement of a Frazier-scheme thermal-diffusion column by operating with modification is best illustrated by calculating the percentage increase in separation efficiency based on the a classical Frazier-scheme thermal-diffusion column ( $r = 0.5$ ) without column length variation in each

**Table 2.** Comparison of separation obtained in the columns of constant and varied column length with flow-rate fraction as a parameter ( $\sigma = 1.0$  g/hr).

$r$	$R = 1$		$R \neq 1$		
	$N_0^*$	$\Delta_{N,\max}^0$	$N^*$	$R^*$	$\Delta_{N,\max}$
0.1	10	0.1702	11	1.17	0.1818
0.2	14	0.1285	14	1.12	0.1388
0.3	15	0.1125	16	1.11	0.1201
0.4	17	0.1054	17	1.10	0.1137
0.5	17	0.1034	17	1.10	0.1111
0.6	17	0.1054	17	1.10	0.1137
0.7	15	0.1125	16	1.11	0.1201
0.8	14	0.1285	14	1.12	0.1388
0.9	10	0.1702	11	1.17	0.1818



column as

$$I_L = \frac{\Delta_{N,\max} - \Delta_{N,\max}^0(r = 0.5)}{\Delta_{N,\max}^0(r = 0.5)} \times 100\% \quad (35)$$

A numerical example for the separation of heavy water is given as follows. Some equipment parameters and physical properties of the mixture were found<sup>[18]</sup> as:

$$H = -1.473 \times 10^{-4} \text{ g/s} = -0.53 \text{ g/hr}$$

$$K = 1.549 \times 10^{-3} \text{ g}\cdot\text{cm/s} = 5.576 \text{ g}\cdot\text{cm/hr}$$

$$K_{eq} = 3.793$$

$$L = 5000 \text{ cm}$$

$$B = 10 \text{ cm}$$

$$2w = 0.04 \text{ cm}$$

$$\Delta T = 35^\circ\text{C}$$

$$\bar{T} = 30.5^\circ\text{C}$$

$$C_0 = 0.381$$

**Table 3.** Improvement in the degree of separation efficiency with flow-rate fraction as a parameter based on the a classical Frazier-scheme thermal-diffusion column ( $\frac{\Delta_{N,\max}^0}{-Au\xi^{0.5}} = 1.161$ ).

$r$	$\frac{\Delta_{N,\max}}{-Au\xi^{0.5}}$	$I_L$ (%)
0.1	1.772	52.63
0.2	1.385	19.33
0.3	1.293	11.33
0.4	1.208	4.04
0.5	1.184	1.95
0.6	1.208	4.04
0.7	1.293	11.33
0.8	1.385	19.33
0.9	1.772	52.63



Some results are presented in Table 3 for the modified Frazier-scheme thermal-diffusion column.

## RESULTS AND DISCUSSION

### Degree of Separation in Modified Frazier-Scheme Thermal-Diffusion Columns

Figures 3 and 5 show, respectively, the best column number  $N^*$  and  $N_0^*$  while Figs. 4 and 6 show, respectively, the maximum degree of separation  $\Delta_{n,\max}$  and  $\Delta_{n,\max}^0$  vs  $\zeta$  for  $C_0 = 0.381$  with flow-rate fraction as a parameter. It is found in Figs. 3 through 6 and Tables 1 and 2 that the best column number and maximum degree of separation for both devices in modified and classical Frazier-scheme thermal-diffusion columns decrease with increasing mass-flow rate and as the flow-rate fraction moves away from  $r = 0.5$ . The convective currents, producing a cascading effect analogous to the multistage effect of countercurrent extraction, actually create two conflict effects: the desirable cascading effect and the undesirable effect of remixing with the diffusion along the column axis and across the column. Thus, the studies for improving the performance in Frazier-scheme thermal-diffusion columns are either considering flow-rate fraction and column length variations. Figure 7 and Tables 1 and 2 show the best ratio of column length in modified Frazier-scheme thermal-diffusion columns with flow-rate fraction as a parameter. The best ratio of column length increases as flow-rate fraction moves away from  $r = 0.5$  but decreases with mass-flow rate. It is concluded that the devices with flow-rate fraction and column length ratio variation can enhance the degree of separation efficiency in a Frazier-scheme.

### Improvement in the Degree of Separation in Modified Frazier-Scheme Thermal-Diffusion Columns

Figures 4 and 6 show that the degree of separation efficiency of modified Frazier-scheme thermal-diffusion columns is much larger than that of classical Frazier-scheme thermal-diffusion columns. The improvements of the degree of separation,  $I_L$ , in the modified Frazier-scheme thermal-diffusion columns are shown in Table 3 with flow-rate fraction as a parameter based on a classical Frazier-scheme thermal-diffusion column with the same column length and the same flow rate at both ends. It is noted that the improvements of



the degree of separation,  $I_L$ , increase when flow-rate fraction moves away from 1/2.

## CONCLUSION

The theoretical study of the separation efficiency for the enrichment of heavy water in modified Frazier-scheme thermal-diffusion columns with column length varied at a constant ratio were investigated in the present study. The equations of the best column number, the best column length ratio, and the maximum degree of separation in modified Frazier-scheme thermal-diffusion columns were calculated from eqs. (29) through (31) by using the given transport coefficients and equilibrium constant. Some graphic representations calculated from eqs. (29) through (32) are given in Figs. 3, 4, and 7 as well as in Tables 1 and 2. Considerable improvement of the degree of separation in modified Frazier-scheme thermal-diffusion columns is obtainable. This is the value of the present study in designing Frazier-scheme thermal-diffusion columns with modification on the column length. The effects of the ratio of column length and flow-rate fraction on the degree of separation for the modified Frazier-scheme thermal-diffusion columns are illustrated in Tables 1 and 2. The feed rate is also the parameter of the analytical calculations, as shown Tables 1 and 2.

## NOMENCLATURE

$A$	$C\hat{C}$ , defined in eq. (6) (—)
$B$	column width (cm)
$C$	fraction concentration of $D_2O$ in $H_2O-HDO-D_2O$ system (—)
$C_{B,n}$	$C$ in the bottom product stream of $n$ th column (—)
$C_{T,n}$	$C$ in the top product stream of $n$ th column (—)
$D$	ordinary diffusion coefficient ( $cm^2/s$ )
$C\hat{C}$	pseudo product form of concentration for $D_2O$ defined by eq. (3) (—)
$g$	gravitational acceleration ( $cm/s^2$ )
$H$	transport coefficient defined by eq. (9) ( $g/s$ )
$I_L$	improvement of the degree of separation defined by eq. (35) (—)
$J_{x-OD}$	mass flux of component $D_2O$ in the $x$ direction owing to ordinary diffusion ( $g/cm^2 \cdot s$ )



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$J_{x-TD}$	mass flux of component D <sub>2</sub> O in the $x$ direction owing to thermal diffusion (g/cm <sup>2</sup> ·s)
$J_{z-OD}$	mass flux of component D <sub>2</sub> O in the $z$ direction owing to ordinary diffusion (g/cm <sup>2</sup> ·s)
$K$	transport coefficient defined by eq. (10) (g/s·cm)
$K_{eq}$	mass-fraction equilibrium constant of H <sub>2</sub> O–HDO–D <sub>2</sub> O system (—)
$L$	total length of all columns (cm)
$N$	column number in modified Frazier-scheme (—)
$N^*$	the best column number in modified Frazier-scheme (—)
$N_0$	column number in classical Frazier-scheme (—)
$N_0^*$	the best column number in classical Frazier-scheme (—)
$r$	flow-rate fraction (—)
$R$	column length ratio (—)
$R^*$	the best column length ratio (—)
$T$	absolute temperature (K)
$\bar{T}$	arithmetic mean value of $T$ of hot wall and cold wall (K)
$u$	$\frac{H}{\sigma}$
$x$	coordinate in the horizontal direction (cm)
$z$	coordinate in the vertical direction (cm)
$z_n$	length of the $n$ th column (cm)
$Z_n$	$\frac{Hz_n}{K}$ (—)

## Greek Symbols

$\alpha$	thermal diffusion constant for D <sub>2</sub> O in H <sub>2</sub> O–HDO–D <sub>2</sub> O system (—)
$\beta$	$\left(\frac{\partial \rho}{\partial T}\right)$ evaluated at reference temperature (g/cm <sup>3</sup> ·K)
$\Delta_n$	degree of separation in modified Frazier-scheme, $C_{B,n} - C_{T,n}$ (—)
$\Delta_n^0$	degree of separation in classical Frazier-scheme, $C_{B,n} - C_{T,n}$ (—)
$\Delta_{N,\max}$	maximum degree of separation in modified Frazier-scheme (—)
$\Delta_{N,\max}^0$	maximum degree of separation in classical Frazier-scheme (—)
$\Delta T$	difference in temperature of hot and cold walls (K)
$\zeta$	$\frac{HL}{Ku} = \frac{\sigma L}{K}$ (—)



$\mu$	viscosity of fluid (g/cm·s)
$\rho$	density of fluid (g/cm <sup>3</sup> )
$\sigma$	mass-flow rate of top or bottom product (g/s)
$\tau_n$	transport of component D <sub>2</sub> O along $z$ direction in $n$ th column (g/s)
$w$	one half of the plate spacing of columns (cm)

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### REFERENCES

1. Clusius, K.; Dickel, G. Neues Verfahren zur Gasentmischung und Isotopentrennung. *Naturwissenschaften* **1938**, *26*, 546–552.
2. Clusius, K.; Dickel, G. Grundlagen eines neuen Verfahrens zur Gasentmischung und Isotopentrennung durch Thermodiffusion. *Z. Phys. Chem.* **1939**, *B44*, 397–450.
3. Furry, W.H.; Jones, R.C.; Onsager, L. On the theory of isotope separation by thermal diffusion. *Phys. Rev.* **1939**, *55*, 1083–1095.
4. Jones, R.C.; Furry, W.H. The separation of isotopes by thermal diffusion. *Rev. Mod. Phys.* **1946**, *18*, 151–224.
5. Yeh, H.M.; Ho, C.D.; Yen, Y.L. Further study on the enrichment of heavy water in continuous-type thermal-diffusion columns. *Sep. Sci. Technol.* **2002**, *37*, 1–20.
6. Yeh, H.M. Enrichment of heavy water by thermal diffusion. *Chem. Eng. Commun.* **1998**, *167*, 167–179.
7. Ho, C.D.; Yeh, H.M.; Guo, J.J. An analytical study on the enrichment of heavy water in the continuous thermal diffusion column with external refluxes. *Sep. Sci. Technol.* **2002**, *37*, 1–20.
8. Frazier, D. Analysis of transverse-flow thermal diffusion. *Ind. Chem. Eng. Proc. Des. Dev.* **1962**, *1*, 237–240.
9. Grasselli, R.; Frazier, D. A comparative study of continuous liquid thermal diffusion systems. *Ind. Chem. Eng. Proc. Des. Dev.* **1962**, *1*, 240–243.
10. Rabinovich, G.D. Theory of thermodiffusion separation according to the Frazier scheme. *Inzh. Fiz. Zh.* **1976**, *31*, 514–552.



## Continuous Thermal-Diffusion Columns

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11. Suvorov, A.V.; Rabinovich, G.D. Theory of a thermal diffusion apparatus with transverse flows. *Inzh. Fiz. Zh.* **1981**, *41*, 231–238.
12. Power, J.E.; Wilke, C.R. Separation of liquid by thermal diffusion column. *AIChE J.* **1957**, *3*, 213–222.
13. Chueh, P.L.; Yeh, H.M. Thermal diffusion in a flat-plate column inclined for improved performance. *AIChE J.* **1967**, *13*, 37–41.
14. Sullivan, L.J.; Ruppel, T.C.; Willingham, C.B. Rotary and packed thermal diffusion fractionating columns for liquids. *Ind. Eng. Chem.* **1955**, *47*, 208–212.
15. Ramser, J.H. Theory of thermal diffusion under linear fluid shear. *Ind. Eng. Chem.* **1957**, *49*, 155–158.
16. Sullivan, L.J.; Ruppel, T.C.; Willingham, C.B. Packed thermal diffusion column. *Ind. Eng. Chem.* **1957**, *49*, 110–113.
17. Yeh, H.M.; Ward, H.C. The improvement in separation of concentric tube thermal diffusion columns. *Chem. Eng. Sci.* **1971**, *26*, 937–947.
18. Standen, A. *Encyclopedia of Chemical Technology*, 3rd Ed.; Wiley: New York, 1978; Vol. 7, 549.
19. Poloujadoff, M.; Christaki, E.; Bergmann, C. Univariant search: an opportunity to identify and solve conflict problems in optimization. *IEEE Trans. Energy Conversion* **1994**, *9*, 659–663.

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