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Theoretical and Experimental Studies on the Heavy Water Enrichment by Thermal Diffusion for Improved Performance Under Countercurrent-Flow Frazier Scheme and Flow-Rate Fraction Variations

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ABSTRACT

The separation equation for heavy water enrichment by thermal diffusion in countercurrent-flow Frazier schemes has been derived and the average value of concentration-product term was treated separately in the enriching and stripping sections. The effect of the flow-rate fraction variation of a countercurrent-flow device on separation efficiencies for a H_2O–HDO–D_2O system, has also been investigated theoretically and experimentally. A qualitative agreement is achieved between the analytical predictions

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and experimental results. The analytical predictions are represented graphically and are compared with that in a classical Frazier scheme of the concurrent-flow device. Considerable improvement in the device performance is obtained by using a countercurrent-flow Frazier scheme with flow-rate fraction variations, instead of using a concurrent-flow device.

Key Words: Thermal diffusion; Heavy water; Separation efficiency; Frazier scheme; Countercurrent flow.

INTRODUCTION

Thermal diffusion is a powerful technique for concentrating desired highly valuable substances such as isotopes and rare gases, which are not possibly separated by conventional separation techniques such as extraction and distillation. In thermal-diffusion columns, a temperature gradient for establishing a net transport of one component in the radial direction (opposite direction for the other component) and density differences resulting in convective flow either upward or downward in the vicinity of the hot and cold walls. The utility of the thermogravitational column, in which a cascading effect, due to natural convection, multiplies the thermal-diffusion effect, and has an advantage in small-scale operations to purify materials because of a resultant concentration gradient. The important application of thermal-diffusion column for practical isotope separation was first developed by Clusius and Dickel;[1,2] subsequently, a complete theory presentation for a Clusius–Dickel column was derived by Furry et al.[3,4] The isotope mixtures of uranium at Oak Ridge during World War II[5] and heavy water by the US government under the Manhattan District Program[6] were successfully separated by thermal-diffusion column. The practical applications of thermal-diffusion columns are commonly used to purify tritium in production and to recover hydrogen isotopes in the fusion nuclear fuel cycle.[7–10] Recently, several theoretical and experimental approaches have been presented[11–13] for the enrichment of heavy water in the Clusius–Dickel column.

In addition, there are two conflicting effects to be confronted in the significant factor of convective flow in thermal-diffusion columns: the desirable cascading effect and an undesirable remixing effect. Consequently, many significant papers are devoted entirely or largely to enhance the former effect and to suppress the latter effect within the operating conditions in thermal diffusion. With proper adjustment in designing improved columns, the separation efficiency improvements on thermal-diffusion columns with inclination,[14] rotation,[15] moving wall,[16] packing,[17] and winding a wire helix[18] were accomplished.

The connection of vertical thermodiffusion columns with the same column length resulted in a tremendous increase in the separation obtainable in a single operation, the so-called Frazier-scheme thermal-diffusion columns, as shown in Fig. 1, was given by Frazier[19] and Grasseli and Frazier.[20] Rabinovich[21] and Sovorov and Rabinovich[22] have developed and presented a general formalism to the Frazier-scheme thermal-diffusion column for binary mixtures. The separation theory of concurrent-flow thermal-diffusion columns on modified Frazier scheme for the enrichment of heavy water with column length varied at a constant ratio has been developed,[13] and the equations for the best column number, best column-length ratio, as well as for the maximum separation efficiency under flow-rate fraction variations, also have been derived. Considerable device performance is obtained when a modified concurrent-flow Frazier scheme is operated at the best corresponding column-length ratio. In contrast to concurrent-flow thermal-diffusion columns, the change in the concentration difference for each column of the countercurrent flow is nowhere as large as it is for the inlet column of the concurrent flow. The mass transfer of countercurrent flow in the present study could be analogized from that of heat exchangers, hence, the separation efficiency improvement under the same working dimensions is greater for the countercurrent flow than for the concurrent-flow arrangement. It is the purpose of the present work to develop and to investigate theoretically and experimentally the separation theory of thermal diffusion on the Frazier scheme with flow-rate fraction variations under countercurrent-flow operations, instead of operating under concurrent flow.

THE THERMAL-DIFFUSION COLUMN IN THE FRAZIER SCHEME

Rabinovich[21] and Sovorov and Rabinovich[22] developed the theoretical analysis to the Frazier-scheme thermal-diffusion columns with the same column length, while a modified device with column length varied at a constant ratio was given by Ho and Chen.[13] Figure 2 illustrates the Frazier-scheme concentric-tube thermal-diffusion column, having a column length L and transverse-flow streams of opposite directions under flow-rate fraction variations. The mass flow rates $r\sigma$ and $(1-r)\sigma$ with feed concentration C_F are accomplished at the upper and lower ends, respectively, with the product end and the supply entrance on the opposite sides.

The transport equations of D_2O for the ith column of countercurrent-flow Frazier scheme were modified from the previous results[3] for the top and the bottom of the column, respectively

$$-\tau_i = -HC\hat{C} + K\left.\frac{dC_i}{dz}\right|_{z=L} = r\sigma \cdot (C_{\mathrm{T},i-1} - C_{\mathrm{T},i}) \tag{1}$$

$$-\tau_i = -HC\hat{C} + K\left.\frac{dC_i}{dz}\right|_{z=0} = (1-r)\sigma \cdot (C_{\mathrm{B},i} - C_{\mathrm{B},i+1}) \tag{2}$$

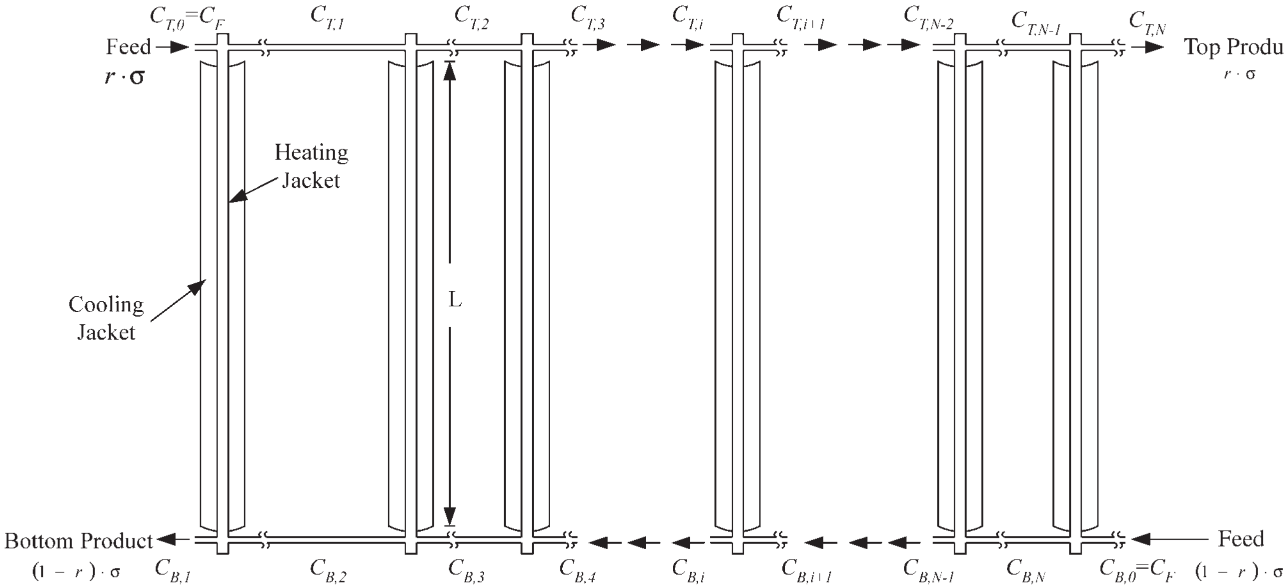

Figure 1. Schematic diagram of the countercurrent-flow Frazier-scheme thermal-diffusion columns with flow-rate fraction variations.

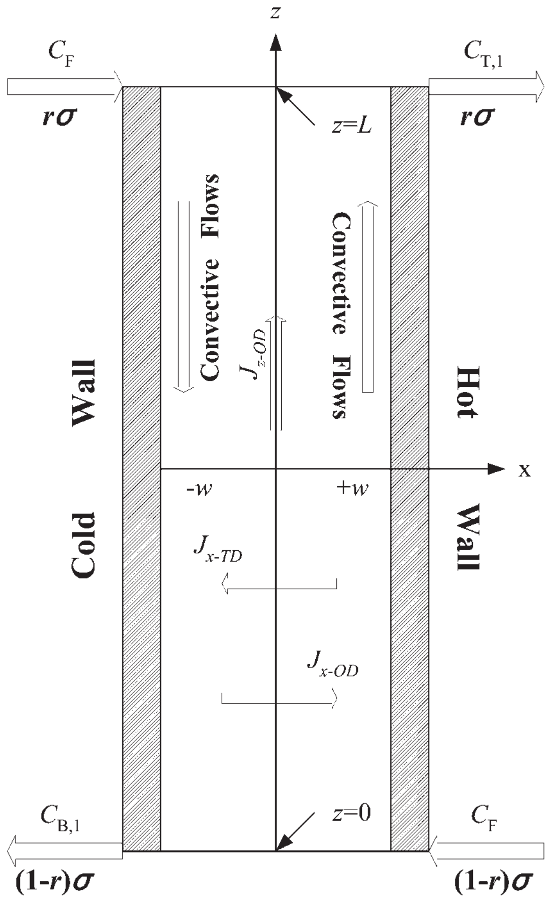

Figure 2. The one-column Frazier-scheme thermal-diffusion column.

and also making material balances around the *i*th column yields

$$r\sigma(C_{T,i-1} - C_{T,i}) = (1 - r)\sigma \cdot (C_{B,i} - C_{B,i+1}) \tag{3}$$

The pseudoconcentration products, $C\hat{C}$ is defined as follows:

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

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

$$H_2O + D_2O \Longleftrightarrow 2HDO \tag{5}$$

is

$$K_{eq} = \frac{[HDO]^2}{[H_2O] \cdot [D_2O]} \times \frac{19 \times 19}{18 \times 20} \tag{6}$$

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, at temperature $T = 25°C$ and 30.5°C, respectively.[23]

The appropriate value of $C\hat{C} = A$ (= constant) in Eqs. (1) and (2) maybe assumed to obtain the simultaneous solution of this set of nonlinear equations. A rather reasonable approximate for $C\hat{C} = A$ in a Frazier-scheme thermal-diffusion column is determined by using the least-squares method in previous works.[12] Accordingly, minimizing the following integration

$$R = \int_{C_T}^{C_B} (C\hat{C} - A)^2 dC \tag{7}$$

i.e.,

$$\frac{dR}{dA} = \int_{C_T}^{C_B} -2(C\hat{C} - A) dC = 0 \tag{8}$$

one obtains

$$A = \frac{1}{C_B - C_T} \int_{C_T}^{C_B} C\hat{C} dC \tag{9}$$

The appropriate value of A, thus obtained in Eq. (9), is exactly the average value of $C\hat{C}$ in the concentration range. Also, the transport constants in above equations are defined by

$$H = \frac{\alpha \beta_T \rho g (2\omega)^3 B (\Delta T)^2}{6! \mu \bar{T}} < 0 \quad \text{for } \alpha < 0 \tag{10}$$

and

$$K = \frac{\rho g^2 \beta_T^2 (2\omega)^7 B (\Delta T)^2}{9! \mu^2 D} + 2\omega \rho D B \tag{11}$$

Substitution of Eqs. (1) and (2) into Eq. (3) gives

$$-HC\hat{C} + K\frac{dC_i}{dz}\bigg|_{z=L} = -HC\hat{C} + K\frac{dC_i}{dz}\bigg|_{z=0} \tag{12}$$

or

$$\frac{dC_i}{dz}\bigg|_{z=L} = \frac{dC_i}{dz}\bigg|_{z=0} = \frac{dC_i}{dz} = \text{constant} \tag{13}$$

Equation (13) indicates that the concentration gradient in the z direction is constant through the entire column.

Countercurrent-Flow Operations

One-Column Operations

For one-column operations, $i = 1$, $C_{\mathrm{T},0} = C_\mathrm{F}$, and $C_{\mathrm{B},2} = C_\mathrm{F}$. Thus, the degree of separation is obtained from Eqs. (1) to (3)

$$C_{\mathrm{T},1} - C_{\mathrm{B},1} = \frac{HL}{K}\left[A + \frac{r\sigma}{H}(C_\mathrm{F} - C_{\mathrm{T},1})\right] \tag{14}$$

$$C_{\mathrm{T},1} - C_{\mathrm{B},1} = \frac{HL}{K}\left[A + \frac{(1-r)\sigma}{H}(C_{\mathrm{B},1} - C_\mathrm{F})\right] \tag{15}$$

Multiplying Eq. (14) by $(1 - r)$ and Eq. (15) by r, and add the results to get

$$(C_{\mathrm{T},1} - C_{\mathrm{B},1}) = \frac{HL}{K}\left\{A + \frac{\sigma r(1-r)}{H}[C_\mathrm{F} - C_{\mathrm{T},1} + C_{\mathrm{B},1} - C_\mathrm{F}]\right\} \tag{16}$$

$$\frac{\sigma Lr(1-r) - K}{K}S_1 = \frac{-HLA}{K} \tag{17}$$

where

$$S_1 = C_{\mathrm{B},1} - C_{\mathrm{T},1} = \Delta_1$$

Multicolumn Operations

For two-column operations, $i = 2$, $C_{\mathrm{T},0} = C_\mathrm{F}$, and $C_{\mathrm{B},3} = C_\mathrm{F}$. Thus, the degree of separation is obtained from Eqs. (1) to (3) for the first and second columns separately

$$C_{\mathrm{T},1} - C_{\mathrm{B},1} = \frac{HL}{K}\left[A + \frac{r\sigma}{H}(C_\mathrm{F} - C_{\mathrm{T},1})\right] \tag{18}$$

$$C_{T,1} - C_{B,1} = \frac{HL}{K}\left[A + \frac{(1-r)\sigma}{H}(C_{B,1} - C_{B,2})\right] \tag{19}$$

$$C_{T,2} - C_{B,2} = \frac{HL}{K}\left[A + \frac{r\sigma}{H}(C_{T,1} - C_{T,2})\right] \tag{20}$$

$$C_{T,2} - C_{B,2} = \frac{HL}{K}\left[A + \frac{(1-r)\sigma}{H}(C_{B,2} - C_F)\right] \tag{21}$$

Multiplying Eqs. (18) and (20) by $(1 - r)$ and Eqs. (19) and (21) by r, and add the results to get

$$C_{T,1} - C_{B,1} = \frac{HL}{K}\left\{A + \frac{\sigma r(1-r)}{H}(C_F - C_{T,1} + C_{B,1} - C_{B,2})\right\} \tag{22}$$

and

$$C_{T,2} - C_{B,2} = \frac{HL}{K}\left\{A + \frac{\sigma r(1-r)}{H}(C_{T,1} - C_{T,2} + C_{B,2} - C_F)\right\} \tag{23}$$

Combination of Eqs. (22) and (23) gives

$$-\Delta_1 - \Delta_2 = \frac{HL}{K}\left\{2A + \frac{\sigma r(1-r)}{H}S_2\right\} \tag{24}$$

or

$$S_2 = -\frac{H}{\sigma r(1-r)}\left[\frac{HL}{K}(\Delta_1 + \Delta_2) + 2A\right] \tag{25}$$

in which $\Delta_1 = C_{B,1} - C_{T,1}$, $\Delta_2 = C_{B,2} - C_{T,2}$, and $S_2 = C_{B,1} - C_{T,2}$.

For further-column operations, the degree of separation is readily obtained if we perform the calculations of Eqs. (18)–(25) repetitively. The resultant expressions of multicolumn operations for heavy-water enhancement in countercurrent-flow thermal-diffusion column are as follows:

$$-\sum_{i=1}^{N}\Delta_i = \frac{HL}{K}\left\{NA + \frac{\sigma r(1-r)}{H}S_N\right\} \tag{26}$$

or

$$S_N = -\frac{H}{\sigma r(1-r)}\left[\frac{HL}{K}\sum_{i=1}^{N}\Delta_i + NA\right] \tag{27}$$

in which $\Delta_N = C_{B,N} - C_{T,N}$ and $S_N = C_{B,1} - C_{T,N}$.

Concurrent-Flow Operations

The calculation procedure for a concurrent-flow Frazier-scheme thermal-diffusion column of the same working dimension is rather simpler than that for a countercurrent-flow device. The degree of separation for concurrent-flow Frazier-scheme thermal-diffusion column[13] is

$$S_N^0 = -\frac{HLA}{K}\left[1 - \left(\frac{L\sigma r(1-r)}{L\sigma r(1-r)+K}\right)^N\right] \tag{28}$$

SEPARATION DEGREE IMPROVEMENT

The improvement of the continuous Frazier-scheme thermal-diffusion columns by operating under the countercurrent-flow pattern is best illustrated by calculating the percentage increase in separation efficiency based on the concurrent-flow device as

$$I_r(\%) = \frac{S_N(\text{countercurrent}) - S_N^0(\text{concurrent}, r = 0.5)}{S_N^0(\text{concurrent}, r = 0.5)} \times 100\% \tag{29}$$

Similarly, the flow-rate fraction improvement may be defined as

$$I_c(\%) = \frac{S_N(\text{countercurrent}) - S_N(\text{countercurrent}, r = 0.5)}{S_N(\text{countercurrent}, r = 0.5)} \times 100\% \tag{30}$$

As an illustration, some equipment parameters and physical properties of the mixture[23] to calculate the separation degree improvement as a numerical example for the separation of heavy water is given as follows:

$$H = -1.473 \times 10^{-4}\,\text{g sec} = -0.53\,\text{g hr}^{-1}$$

$$K = 1.549 \times 10^{-3}\,\text{g cm sec}^{-1} = 5.5763\,\text{g cm hr}^{-1}$$

$$K_{\text{eq}} = 3.793, \quad L = 122\,\text{cm}, \quad B = 10\,\text{cm},$$

$$\sigma = 2.1\,\text{g hr}^{-1}, \quad 2\omega = 0.04\,\text{cm}$$

$$\Delta T = 35\,\text{K}, \quad \bar{T} = 30.5\,\text{K}, \quad C_{\text{F}} = 0.381, \quad \frac{\Delta T}{2\omega} = 875\,\text{K cm}^{-1}$$

with these values, some results are presented in Tables 1 and 2 for the countercurrent-flow Frazier-scheme thermal-diffusion column.

Table 1. The separation degree improvement with the flow-rate fraction variation and column number as parameters; $C_F = 0.381$, $\sigma = 2.1\,\mathrm{g\,hr^{-1}}$.

	I_r (%)				
r	$N = 1$	$N = 2$	$N = 3$	$N = 4$	$N = 5$
0.1	143.16	128.39	115.49	104.16	94.15
0.2	49.52	46.44	43.70	41.26	39.07
0.3	17.26	16.46	15.81	15.30	14.90
0.4	3.82	3.72	3.71	3.81	3.98
0.5	0	0.08	0.25	0.50	0.84
0.6	3.82	3.76	3.80	3.93	4.15
0.7	17.26	16.57	16.03	15.62	15.33
0.8	49.52	46.69	44.23	42.04	40.11
0.9	143.16	129.35	117.36	106.87	97.63

MAXIMUM SEPARATION WITH BEST COLUMN NUMBER

The best column number N^* for maximum overall separation degree $S_{N,\max}$ with r and σ as parameters in a countercurrent-flow Frazier-scheme thermal-diffusion column with a fixed sum of the column length (as an illustration, the column length $L = 5000/N$ cm for each column) are calculated by using the method of two variable univariant search.[24] The calculation procedure is to find N^* first and then $S_{N,\max}$ is obtained in Eq. (27). Similarly, for a concurrent-flow Frazier-scheme thermal-diffusion column, the best column number $N_0{}^*$ and the maximum degree of separation $S^0_{N,\max}$ have been calculated and presented in Tables 3 and 4.

EXPERIMENTAL STUDIES

The experimental apparatus used in this study is almost the same as that in Yeh and Ward's work,[18] except that two same dimensions were connected in series. Two continuous-type concentric-tube thermal-diffusion columns to separate heavy water from water–isotope mixtures were constructed of stainless steel with column length $L = 122$ cm and annular space $2w = 0.04$ cm, as shown in Fig. 3. Hot and cold waters with high enough rates and controlled temperatures, circulating countercurrently through the inner tube and jacket, respectively, were supplied from two thermostat tanks to heat and cool

Table 2. Separation degree improvements by operating countercurrent-flow devices with the flow-rate fraction variation and number of columns as parameters; $C_F = 0.381$.

	I_c (%)														
	$N = 1$			$N = 2$			$N = 3$			$N = 4$			$N = 5$		
	σ (g hr^{-1})														
r	0.85	2.1	3.55	0.85	2.1	3.55	0.85	2.1	3.55	0.85	2.1	3.55	0.85	2.1	3.55
0.1	111.29	143.16	155.53	86.80	127.55	144.75	41.28	92.02	118.09	5.05	53.25	84.84	−20.39	12.89	42.98
0.2	42.10	49.52	52.06	35.83	46.15	49.88	21.86	37.71	44.14	6.19	26.62	36.03	−11.62	10.88	23.49
0.3	15.16	17.26	17.95	13.27	16.31	17.34	8.81	13.87	15.74	3.13	10.45	13.38	−5.20	5.07	9.50
0.4	3.40	3.82	3.95	2.98	3.61	3.82	1.97	3.09	3.49	0.61	2.34	2.99	−1.67	1.11	2.14
0.5	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
0.6	3.40	3.82	3.95	3.16	3.69	3.87	2.49	3.32	3.62	1.62	2.79	3.25	0.32	1.94	2.62
0.7	15.16	17.26	17.95	13.72	16.51	17.46	10.05	14.43	16.08	5.42	11.54	14.05	−1.17	7.04	10.68
0.8	42.10	49.52	52.06	36.86	46.64	50.18	24.45	39.03	44.98	10.39	29.04	37.61	−5.86	14.79	26.12
0.9	111.29	143.16	155.53	89.85	129.28	145.89	47.17	96.14	121.02	11.77	59.41	89.72	−14.24	20.02	49.57

Table 3. Comparison of the degree of separation obtained in the concurrent- and countercurrent-flow operations with the mass flow rate as a parameter; $C_F = 0.381$, $r = 0.50$.

	Concurrent flow		Countercurrent flow	
$\sigma\,(\mathrm{g\,hr^{-1}})$	N_0^*	$S_{N,\max}^0$	N^*	$S_{N,\max}$
0.1	5	0.45849	7	0.50157
0.2	8	0.33015	9	0.36192
0.4	11	0.23645	13	0.25994
0.6	13	0.19414	16	0.21368
0.8	15	0.16865	19	0.18584
1.0	17	0.15117	21	0.16667
2.0	24	0.10746	30	0.11867
4.0	34	0.07627	42	0.08431
6.0	41	0.06239	52	0.06899
8.0	48	0.05408	60	0.05982
10.0	53	0.04841	67	0.05355

the tube surfaces within ±0.5°C at all times. Four copper-constantan thermocouples located on the surface of each plate were used to monitor the mean surface temperatures of hot and cold surfaces, which were 47°C and 14°C, respectively. Hence, the temperature difference between the hot and cold plates (33°C) and mean temperature of the mixture (30.5°C) were used during the calculation procedure. Four holes were drilled at the top and bottom of the hot and cold plates, respectively, to serve as feed and product ports. The needle valve was installed in each product stream to control and to measure the flow rates. Deaerated feed, consisting of various feed concentration C_F and feed rate σ at 30.5°C, was regulated due to gravitational force with flow-rate fraction variations by a constant-head tank into the top of the first column and the bottom of the last column at each time. The two product streams, the bottom of the first column and the top of the last column, were attuned by needle valves, withdrawn continuously through cooling coils and were measured by rotameters with the flow-rate fraction r and $(1 - r)$, respectively, before being collected as product samples. The product samples were analyzed at 25 ± 0.05°C by an automatic density meter (model DA-210, Kyoto Electronics, Tokyo, Japan). The precision of $\pm 1.0 \times 10^{-4}\,\mathrm{g\,cm^{-3}}$ as well as $\pm 5.0 \times 10^{-4}$ D_2O mass fraction was obtainable in this instrument. The theoretical predictions and experimental results are compared and represented in Fig. 4 for one- and two-column operations with $C_F = 0.381$, and $r = 0.5$ and 0.7. It is found from this figure that the theoretical predictions confirm pretty well with experimental results.

Table 4. Comparison of the best column number obtained in both concurrent- and countercurrent-flow operations with the mass flow rate as a parameter; $C_F = 0.381$, $r = 0.50$.

σ (g hr^{-1})	$S^0_{N,\max}$ (or $S_{N,\max}$)	Concurrent flow N^*_0	Countercurrent flow N^*
0.1	0.45849	5	4
0.2	0.33015	8	6
0.4	0.23645	11	8
0.6	0.19414	13	10
0.8	0.16865	15	12
1.0	0.15117	17	13
2.0	0.10746	24	18
4.0	0.07627	34	26
6.0	0.06239	41	32
8.0	0.05408	48	37
10.0	0.04841	53	42

The accuracy of the experimental results maybe calculated by using the definition

$$E = \frac{1}{M}\sum_{i=1}^{M}\frac{|S_{N,i} - \hat{S}_{N,i}|}{S_{N,i}} \tag{31}$$

where $S_{N,i}$ denotes the theoretical prediction of S_N, while S_N and M are the experimental data of S_N and the number of experimental measurements, respectively. The error analysis of the four measurements for each experimental run in Fig. 4 is calculated by Eq. (31). As an illustration, the accuracy of the experimental results are 9.0×10^{-2} and 8.0×10^{-2} for $r = 0.5$ and 0.7 of two-column operations, respectively.

RESULTS AND DISCUSSIONS

The Degree of Separation in Countercurrent-Flow Frazier-Scheme Thermal-Diffusion Columns

The present study for improving the performance in Frazier-scheme thermal-diffusion columns under flow-rate fraction variations countercurrent-flow operations is to suppress the remixing effect and to enhance the

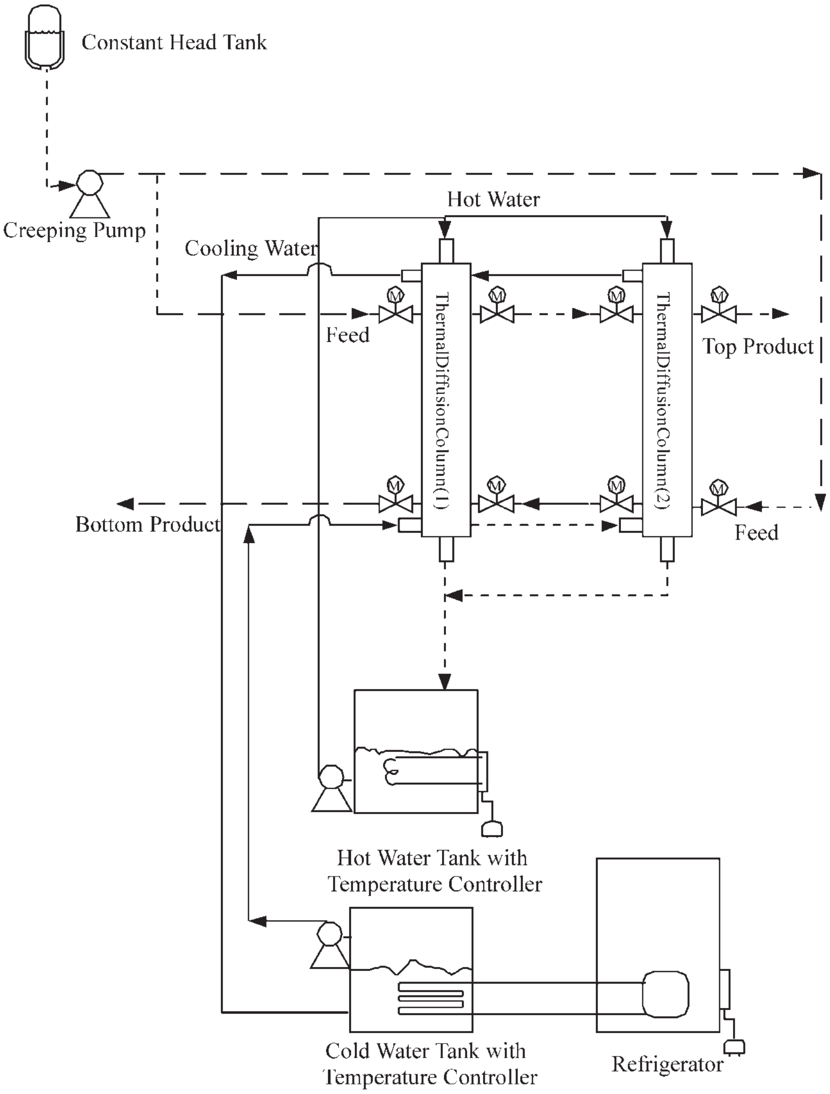

Figure 3. Flow diagram of the experimental system.

cascading effect. Some experimental results and graphical representations calculated from Eqs. (27) and (28) are shown in Figs. 4–8. Figures 5 and 6 show, respectively, the overall degree of separation vs. flow-rate fraction with feed concentration as a parameter for one- and two-column operations.

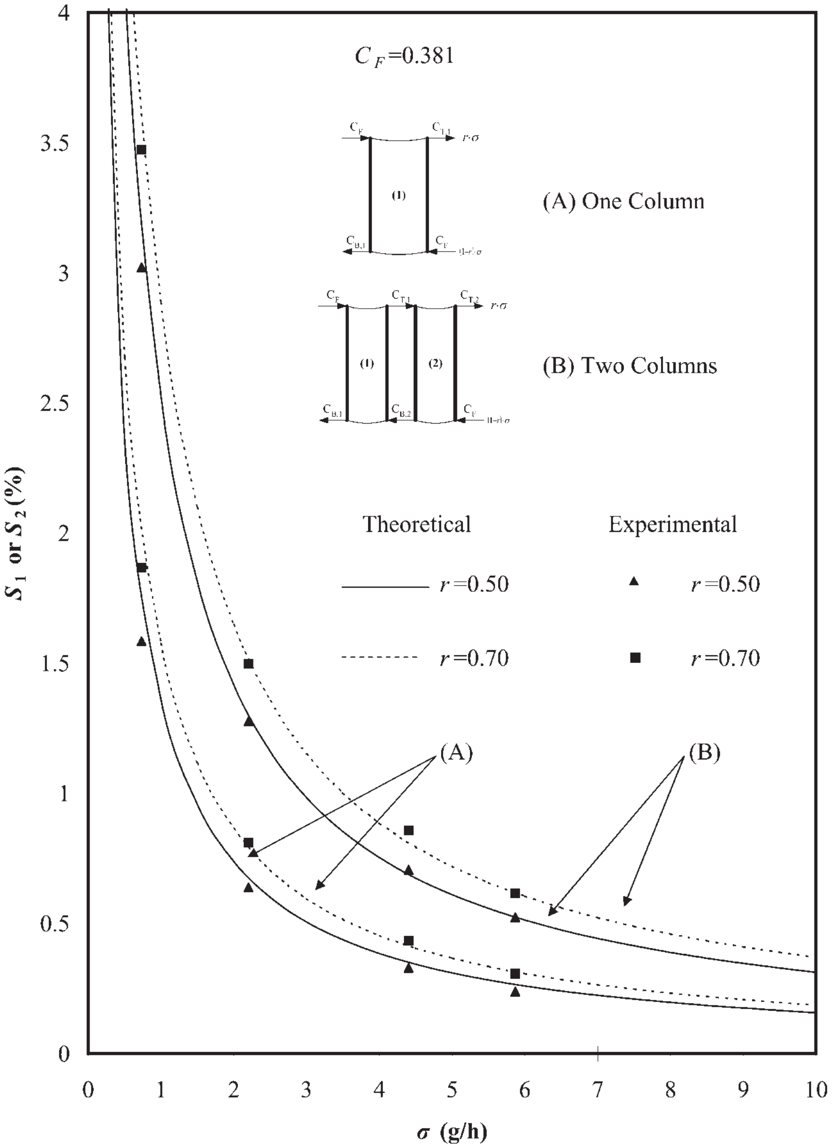

Figure 4. Comparisons of the overall degree of separation obtained from theoretical predictions and experimental results.

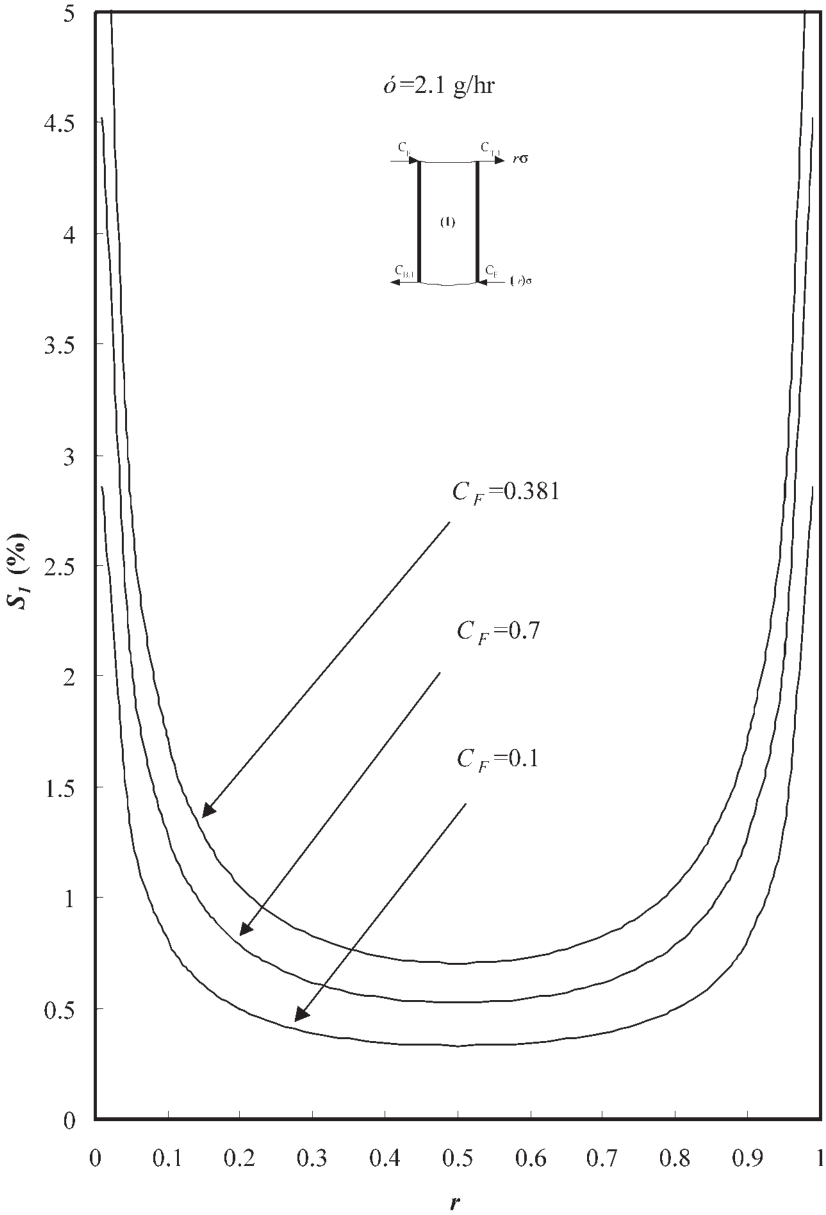

Figure 5. Effect of flow-rate fraction variation on the overall degree of separation with feed concentration as a parameter for one-column operations; $\sigma = 2.1\,\mathrm{g\,hr^{-1}}$.

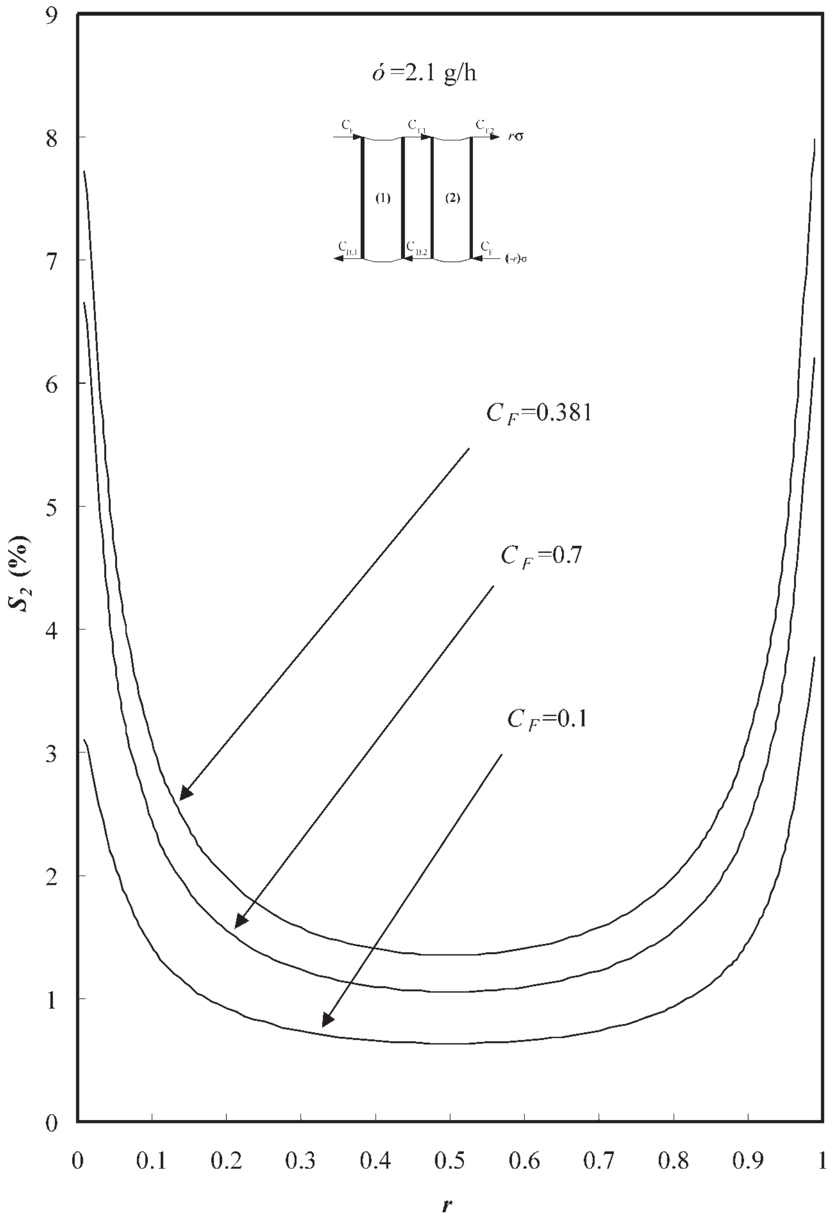

Figure 6. Effect of flow-rate fraction variation on the overall degree of separation with feed concentration as a parameter for two-column operations; $\sigma = 2.1\,\mathrm{g\,hr^{-1}}$.

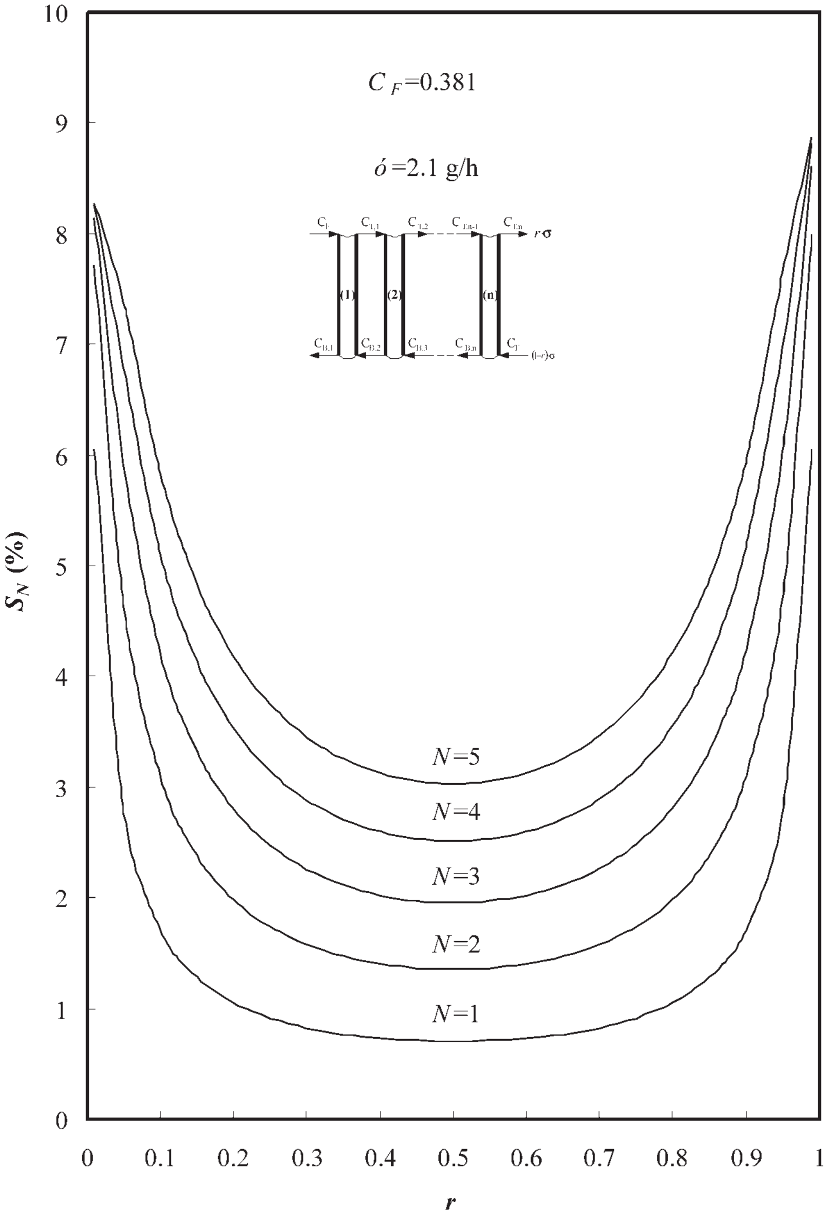

Figure 7. Effect of flow-rate fraction variations on the overall degree of separation with number of columns as a parameter; $C_F = 0.381$, $\sigma = 2.1\,\mathrm{g\,hr^{-1}}$.

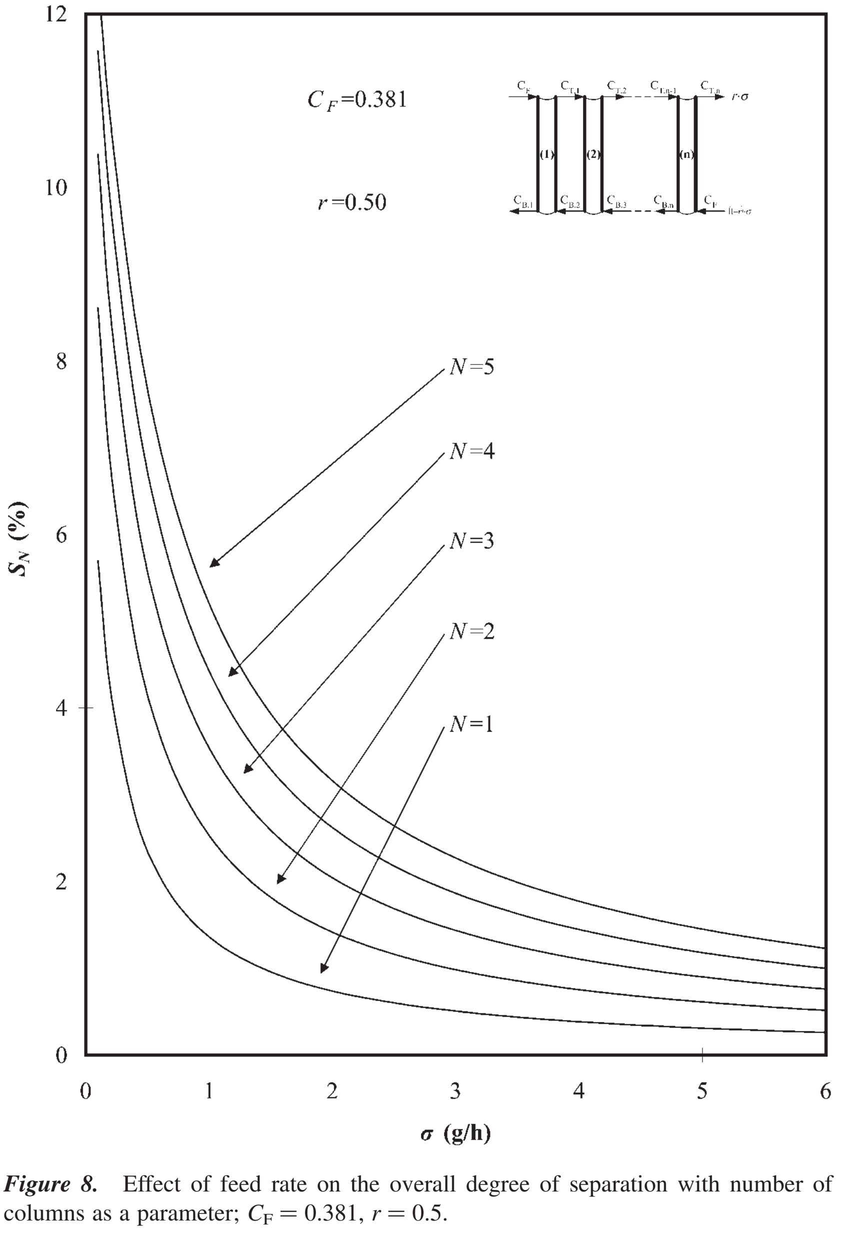

Figure 8. Effect of feed rate on the overall degree of separation with number of columns as a parameter; $C_F = 0.381$, $r = 0.5$.

It is found that in Figs. 5 and 6 and Table 3 the degree of separation for the countercurrent-flow Frazier-scheme thermal-diffusion columns decrease with increasing mass flow rate and as the flow-rate fraction goes away from $r = 0.5$. Figures 7 and 8 show the overall degree of separation vs. flow-rate fractions and feed concentration with the number of columns as a parameter up to five-column operations. The overall degree of separation increases with number of columns and as flow-rate fraction goes away from $r = 0.5$ but decreases with mass flow rate. These results would suggest that the devices with flow-rate fraction variations and countercurrent-flow operations could enhance the separation efficiency in Frazier-scheme thermal-diffusion columns.

Separation Degree Improvement in Countercurrent-Flow Operations

The separation degree improvements, I_r and I_c, after N column operations were calculated from Eqs. (29) and (30). Table 1 shows that the separation degree improvement, I_r, of countercurrent-flow Frazier-scheme thermal-diffusion columns is much larger than that of concurrent-flow Frazier-scheme thermal-diffusion columns, while Table 2 shows the influence of the flow-rate fraction on the separation degree improvement, I_c, of countercurrent-flow devices. The separation degree improvements, I_r, in the countercurrent-flow Frazier-scheme thermal-diffusion columns with flow-rate fraction as a parameter based on a concurrent-flow Frazier-scheme thermal-diffusion column with the same working dimensions and the same mass flow rate at both ends. The minimum degree of separation is operated at $r = 0.5$ under concurrent-flow devices, as concluded from our previous work.[13] Therefore, the separation degree improvements, I_r and I_c, were defined by Eqs. (29) and (30), respectively, and calculated with respect to the operation at $r = 0.5$ to show the percentage increase of separation efficiency, and, thus, the zero value is obtained at $r = 0.5$. It seems apparent that the separation degree improvements increase when flow-rate fraction goes away from $1/2$. The minus signs in Table 2 indicate that when $N = 5$, no improvement in separation efficiency can be achieved as $\sigma < 0.85\,\mathrm{g\,hr^{-1}}$, and, in this case, the countercurrent-flow device with $r = 0.5$ is preferred to be used rather than operating the device with $r \neq 0.5$.

The best column number N^* and N_0^* with the maximum separation degree for both operations in concurrent- and countercurrent-flow Frazier-scheme thermal-diffusion columns are calculated and presented in Tables 3 and 4 with mass flow rate as a parameter for $C_{\mathrm{F}} = 0.381$ and $r = 0.5$. Table 3 shows the achievable maximum separation degree of countercurrent-flow

Frazier-scheme thermal-diffusion columns is larger than that of concurrent-flow operations under the same feed rate, while the comparison of the best column number between these two flow-type operations under the same maximum separation degree is shown in Table 4. The column number of countercurrent-flow operations is smaller than that in concurrent-flow operations to obtain the same maximum separation degree, as confirmed from Tables 3 and 4, and, hence, the less operating cost is needed.

CONCLUSIONS

The theoretical and experimental study of the separation efficiency for heavy-water enrichment in countercurrent-flow Frazier-scheme thermal-diffusion columns has been investigated with the column number as a parameter. It is shown in Figs. 5–8 that the degree of separation decreases almost with the mass flow rate, but increases with the column number and as flow-rate fraction goes away from $1/2$. The separation degree improvement by operating at the best column number is illustrated for both concurrent- and countercurrent-flow Frazier-scheme thermal-diffusion columns, as shown in Tables 3 and 4. The countercurrent-flow Frazier-scheme thermal-diffusion columns can enhance the separation degree of heavy water compared with that in concurrent-flow Frazier-scheme thermal-diffusion columns under the same working dimensions and operating parameters. Considerable improvement in separation degree is obtainable by using such a countercurrent-flow Frazier-scheme thermal-diffusion device, instead of using the concurrent-flow one. Also, there exists the separation efficiency enhancement by suitable adjusting the flow-rate fractions of two product streams; this is the value of present study in designing countercurrent-flow Frazier-scheme thermal-diffusion columns under the smaller column number.

NOMENCLATURE

A	$C\hat{C}$, defined in Eq. (9)
B	column width (cm)
C	fraction mass concentration of heavy water in H_2O–HDO–D_2O system
$C_{B,i}$	C in the bottom product stream of ith column
C_F	C in the feed stream
$C_{T,i}$	C in the top product stream of ith column
D	ordinary diffusion coefficient ($cm^2\,sec^{-1}$)
$C\hat{C}$	pseudoproduct form of concentration for D_2O defined by Eq. (4)

g	gravitational acceleration ($\mathrm{cm\,sec^{-2}}$)
H	transport coefficient defined by Eq. (10) ($\mathrm{g\,sec^{-1}}$)
I_r	improvement of the degree of separation defined by Eq. (29)
I_c	improvement of the degree of separation defined by Eq. (30)
$J_{x-\mathrm{OD}}$	mass flux of heavy water in the x-direction due to ordinary diffusion ($\mathrm{g\,cm^{-2}\,sec^{-1}}$)
$J_{x-\mathrm{TD}}$	mass flux of heavy water in the x-direction due to thermal diffusion ($\mathrm{g\,cm^{-2}\,sec^{-1}}$)
$J_{z-\mathrm{OD}}$	mass flux of heavy water in the z-direction due to ordinary diffusion ($\mathrm{g\,cm^{-2}\,sec^{-1}}$)
K	transport coefficient defined by Eq. (11) ($\mathrm{g\,sec^{-1}\,cm^{-1}}$)
K_{eq}	mass-fraction equilibrium constant of $H_2O{-}HDO{-}D_2O$ system
M	the number of experimental measurements
N	total column number
r	flow-rate fraction
S_N	the degree of separation
$S_{N,i}$	theoretical prediction of S_N
S_N	experimental data of S_N
T	mean absolute temperature (K)
$\bar{T}$	arithmetic mean value of T of hot wall and cold wall (K)
x	coordinate in the horizontal direction (cm)
z	coordinate in the vertical direction (cm)

Greek Symbols

α	thermal-diffusion constant for D_2O in $H_2O{-}HDO{-}D_2O$ system, <0
β_r	$(-\partial\rho/\partial T)$ evaluated at reference temperature ($\mathrm{g\,cm^{-3}\,K^{-1}}$)
Δ_N	degree of separation in a countercurrent-flow Frazier scheme, $C_{\mathrm{B},1} - C_{\mathrm{T,N}}$
ΔT	difference in temperature of hot and cold plates (K)
μ	absolute viscosity of fluid ($\mathrm{g\,cm^{-1}\,sec^{-1}}$)
ρ	mass density of fluid ($\mathrm{g\,cm^{-3}}$)
σ	mass flow rate of top or bottom product ($\mathrm{g\,sec^{-1}}$)
τ_i	transport of heavy water along z-direction in ith column, <0 ($\mathrm{g\,sec^{-1}}$)
ω	one half of the plate spacing of columns (cm)

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