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ENRICHMENT OF HEAVY WATER BY THERMAL DIFFUSION IN COUNTERCURRENT-FLOW FRAZIER SCHEME INCLINED FOR IMPROVED PERFORMANCE

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ABSTRACT

The theory for enrichment of heavy water by thermal diffusion in a countercurrent-flow inclined Frazier scheme was developed and investigated. The equations for the optimal angles of inclination and the corresponding best performance were derived. Considerable improvement in performance is obtained when the Frazier scheme is operated at the optimal angle of inclination. Further improvement can be achieved if the operation is conducted in countercurrent flow instead of concurrent flow.

Key Words: Thermal diffusion; Heavy water; Inclined Frazier scheme; Countercurrent flow

INTRODUCTION

The convective currents in the thermogravitational thermal diffusion column developed by Clusius and Dickel have 2 conflicting effects (1,2): the desirable cascading effect and the undesirable remixing effect. The convection currents have a multistage effect that is necessary in securing high separation, and it is an essential feature of the Clusius-Dickel column. However, because the convection brings up the fluid at the bottom of the column, where it is rich in one component, to the top of the column, where it is rich in the other component, and vice versa, the components are remixed.

Powers and Wilke (3) demonstrated that a simple and flexible way of adjusting the convective strength is to tilt a flat-plate column with the hot plate on top so it reduces the effective gravitational force. The equations for the optimum angle of inclination and the best performance in the thermogravitational thermal diffusion column were later derived by Chueh and Yeh (4) and Yeh and Ward (5), and the theories were confirmed with their experimental results. They also showed that proper control of the convective strength obtained by tilting the column might effectively suppress the undesirable remixing effect while still preserving the desirable cascading effect, thereby leading to improved separation.

In industrial applications, the Clusius-Dickel columns are connected in series, as shown in Fig. 1, and called the Frazier scheme. Frazier and Rabinovich (6,7) gave a theory of this process on the basis of a simplified, modified representation while an analytical solution was given by Rabinovich and Suvorov (8,9). Later, a separation theory for the enrichment of heavy water in flat-plate thermal

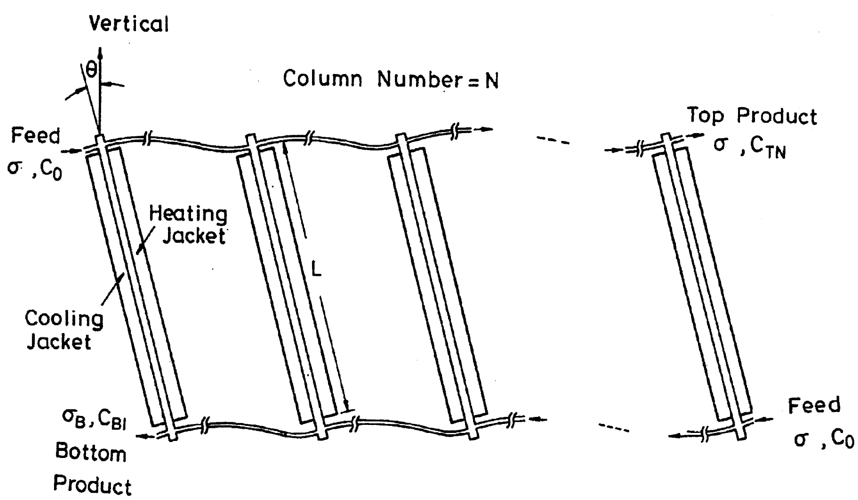

Figure 1. Schematic diagram of an inclined countercurrent-flow Frazier scheme.

diffusion columns of inclined concurrent-flow Frazier scheme was developed (10,11). The equations for the optimum angle of inclination and the corresponding best performance in the inclined countercurrent-flow Frazier scheme was derived in the present work, and the results are compared with those obtained in the concurrent-flow device.

ENRICHMENT OF HEAVY WATER IN AN INCLINED COUNTERCURRENT-FLOW FRAZIER SCHEME

The scheme proposed by Frazier to connect several flat-plate thermogravitational tharmal diffusion columns with countercurrent-flow sampling streams is shown in Fig. 1. All flat-plate columns have the same dimensions of gap (2*w*), length *L*, and width *B*. Figure 2 illustrates the flows and fluxes prevailing in the

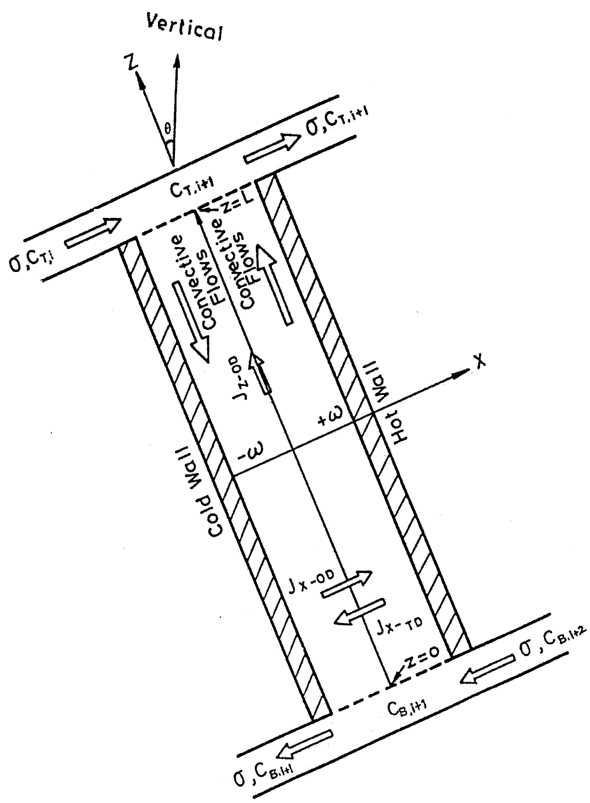

Figure 2. Flows and fluxes prevailing in the $(i + 1)$th thermal diffusion column of the Frazier scheme.

$(i+1)$th column. The transport equation for D_2O in a vertical Clusius-Dickel column was derived (12). For the inclined column, the transport equation is modified by replacing g with $g \cos\theta$. Accordingly, the steady transport of D_2O in the $(i+1)$th column of the inclined Frazier scheme is (10, 11)

$$-\tau_{i+1} = -PH\cos\theta + K\cos^2\theta\left(\frac{dC_{i+1}}{dz}\right) \tag{1}$$

$$= \sigma\,(C_{T,i} - C_{T,i+1}) \tag{2}$$

$$= \sigma(C_{B,i+1} - C_{B,i+2}) \tag{3}$$

where

$$H = \frac{\alpha\rho\beta g\,(2w)^3\,B(\Delta T)^2}{6!\mu T_m} \tag{4}$$

$$K = \frac{\rho\beta^2\,g^2(2w)^7\,B(\Delta T)^2}{9!D\mu^2} \tag{5}$$

$$P = C_o[0.05263 - (0.05263 - 0.0135K_{eq})C_o - 0.027\{C_oK_{eq}[1 - (1 - 0.25K_{eq})C_o]\}^{1/2}] \tag{6}$$

The product form of concentration, P, in Eq. (1) was assumed to be a constant. For enrichment of heavy water from the H_2O—HDO—D_2O system, α (thermal diffusion constant) < 0, and thus, $H < 0$ and $\tau_{i+1} < 0$.

By integrating Eq. (2) through the (i + 1)th column from $z = 0$ ($C_{i+1} = C_{B,i+1}$) to $z = L$ ($C_{i+1} = C_{T,i+1}$), the following equation is obtained:

$$C_{B,i+1} = C_{T,i+1} - \left\{\frac{[PH\cos\theta + \sigma\,(C_{T,i} - C_{T,i+1})]L}{(K\cos^2\theta)}\right\} \tag{7}$$

By replacing i with $(i+1)$, Eq. (7) becomes

$$C_{B,i+2} = C_{T,i+2} - \left\{\frac{[PH\cos\theta + \sigma(C_{T,i+1} - C_{T,i+2})]L}{(K\cos^2\theta)}\right\} \tag{8}$$

Substitution of Eqs. (7) and (8) into Eq. (3) to eliminate $C_{B,i+1}$ and $C_{B,i+2}$ yields

$$C_{T,i+2} - 2C_{T,i+1} + C_{T,i} = 0 \tag{9}$$

Equation (9) is a second-order difference equation in which the general solution is

$$C_{T,i} = A_1 + A_2 i, \quad 0 \le i \le N+1 \tag{10}$$

in which A_1 and A_2 are arbitrary constants to be determined. Substitution of Eq. (10) into Eq. (7) with i in Eq. (7) replaced by $(i-1)$ results in

$$C_{B,i} = A_1 - \left(\frac{PHL}{K\cos\theta}\right) + A_2\left[\left(\frac{\sigma L}{K\cos^2\theta}\right) + i\right] \tag{11}$$

Applying 2 inlet conditions of feed at the top and bottom, respectively,

$$i = o,\ C_{T,i} = C_0 \tag{12}$$

$$i = N + 1,\ C_{B,i} = C_0 \tag{13}$$

one obtains

$$A_1 = C_0 \text{ and } A_2 = \frac{PHL/(K\cos\theta)}{N + 1 + [\sigma L/(K\cos^2\theta)]} \qquad (14), (15)$$

In addition, the equation for the degree of separation Δ is readily obtained from Eqs. (10) and (11) through the use of Eqs. (14) and (15):

$$\Delta = C_{B,1} - C_{T,N}$$

$$= \frac{2(-H)\,PLN\cos\theta}{(N+1)\,K\cos^2\theta + \sigma L} \tag{16}$$

MAXIMUM SEPARATION WITH FIXED COLUMN LENGTH

The optimum angle of inclination θ_Δ for a maximum separation Δ_{max} is obtained by partially differentiating Eq. (16) with respect to θ and setting $\partial\Delta/\partial\theta = 0$. The result is

$$\theta_\Delta = \cos^{-1}\left[\frac{\sigma L}{K(N+1)}\right]^{1/2} \tag{17}$$

Substitution of Eq. (17) into Eq. (16) yields the maximum separation as

$$\Delta_{max} = \left[\frac{P^2 H^2 L}{\sigma K}\right]^{1/2}\left[\frac{N}{(N+1)^{1/2}}\right] \tag{18}$$

The existence of θ_Δ is determined from Eq. (17) by setting $\cos\theta_\Delta < 1$:

$$\sigma < \left(\frac{K}{L}\right)(N+1) \tag{19}$$

MAXIMUM OUTPUT WITH FIXED COLUMN LENGTH

The equation of output can be derived from Eq. (16) as

$$\sigma = 2\left(\frac{-HPN}{\Delta}\right)\cos\theta - \frac{(N+1)\,K\cos^2\theta}{L} \tag{20}$$

The optimum angle of inclination θ_σ required to obtain the maximum production rate σ_{max} for a given scheme (that is, to attain a specified degree of separation Δ) can be obtained by partially differentiating Eq. (20) with respect to θ and setting

$\partial\sigma/\partial\theta = 0$. The results for θ_σ and σ_{max} are

$$\theta_\sigma = \cos^{-1}\left[\left(\frac{-HLP}{K\Delta}\right)\left(\frac{N}{N+1}\right)\right] \tag{21}$$

$$\sigma_{max} = \left(\frac{H^2 LP^2}{K\Delta^2}\right)\left(\frac{N^2}{N+1}\right) \tag{22}$$

and the restriction on the existence of θ_σ is

$$\left(\frac{-HLP}{K\Delta}\right)\left(\frac{N}{N+1}\right) < 1 \tag{23}$$

MINIMUM COLUMN LENGTH

Equation (16) can be rewritten to obtain the expression for column length as

$$L = \frac{(N+1)\,K\cos^2\theta}{[2(-HNP/\Delta)\cos\theta - \sigma]} \tag{24}$$

To find the minimum column length L_{min} required to accomplish a specified degree of separation Δ and production rate σ, L from Eq. (24) can be minimized with respect to θ. The results are

$$\theta_L = \cos^{-1}\left[\frac{\sigma\Delta}{(-HNP)}\right] \tag{25}$$

$$L_{min} = \left(\frac{\sigma\Delta^2 K}{H^2P^2}\right)\left(\frac{N+1}{N^2}\right) \tag{26}$$

Note that the restriction on the existence of θ_L is from Eq. (25)

$$\sigma < \left(\frac{-HNP}{\Delta}\right) \tag{27}$$

IMPROVEMENT IN PERFORMANCE

The improvement in performance by operating at optimum inclination angles is best illustrated by the calculated percentages of increased performance based on the vertical scheme:

$$I_\Delta = \frac{\Delta_{max} - \Delta_0}{\Delta_0} = \frac{(\Delta_{max}/P) - (\Delta_0/P)}{\Delta_0/P} \tag{28}$$

$$I_\sigma = \frac{\sigma_{max} - \sigma_0}{\sigma_0} \tag{29}$$

$$I_{\Delta/L} = \frac{(\Delta/L_{min}) - (\Delta/L_0)}{(\Delta/L_0)} = \left(\frac{L_0}{L_{min}}\right) - 1 \tag{30}$$

where Δ_0, σ_0, and L_0 denote, respectively, the degree of separation, production rate, and column length in the vertical columns, which are readily calculated from Eqs. (16), (20), and (24) when $\theta = 0$.

For the purpose of illustration, let us employ some numerical values obtained in a previous work (12) for the enrichment of heavy water from the H_2O—HDO—D_2O system:

$$H = -1.473 \times 10^{-4}\ \text{(g/s)} = 0.53\ (g\text{/h})$$
$$K = -1.549 \times 10^{-3}\ (\text{g}\cdot\text{cm/s}) = 5.576\ (\text{g}\cdot\text{cm/h})$$

where L (or L_0) = 177 cm; B = 10 cm; ΔT = 30.5°C; K_{eq} = 3.793; $P \times 10^3$ = 3.619 (C_0 = 0.1); 7.088 (C_0 = 0.3); 7.610 (C_0 = 0.5); 5.910 (C_0 = 0.7); and 3.528 (C_0 = 0.9). From these values, the optimum angles of inclination were calculated from the corresponding equations, and consequently the improvements in performance were obtained by substituting the appropriate values of performance into Eqs. (28–30). The results are presented in Tables 1–3.

Table 1. Comparison of Separation Obtained in Inclined (Δ_{max} and $\dot{\Delta}_{max}$) and Vertical (Δ_0 and $\dot{\Delta}_0$) Columns as Well as in Countercurrent-Flow (Δ_{max} and Δ_0) and Concurrent-Flow ($\dot{\Delta}_{max}$ and $\dot{\Delta}_0$) Columns with L = 177 cm and Various σ

		Concurrent Flow				Countercurrent Flow						
	σ or $\dot{\sigma}$ (g/h)	$\dot{\Delta}_0$/P	$\dot{\theta}_\Delta$ (deg)	$\dot{\Delta}_{max}$/P (%)	$\dot{I}_\Delta$ (%)	Δ_0/P	θ_Δ (deg)	Δ_{max}/P (%)	I_Δ (%)	$\frac{\Delta_0 - \dot{\Delta}_0}{\dot{\Delta}_0}$ (%)	$\frac{\Delta_{max} - \dot{\Delta}_{max}}{\dot{\Delta}_{max}}$ (%)	$\frac{\Delta_0 - \dot{\Delta}_{max}}{\dot{\Delta}_{max}}$ (%)
(a)	0.5	15.26	45.4	16.79	10.00	18.26	29.62	18.44	0.98	19.66	9.83	8.88
	1.0	11.87	6.8	11.88	0.08	12.77	—	—	—	7.58	—	7.49
	1.5	9.44	—	—	—	9.81	—	—	—	3.92	—	—
(b)	0.5	16.69	60.1	23.91	43.24	23.68	51.53	26.40	11.47	41.19	10.41	−0.96
	1.0	15.37	45.2	16.90	9.95	18.51	28.37	18.66	0.82	20.43	10.41	9.53
	1.5	13.59	30.4	13.81	1.59	15.20	—	—	—	11.85	—	10.07
(c)	0.5	16.83	69.5	33.97	101.85	27.80	63.73	37.56	35.09	65.18	10.57	−18.16
	1.0	16.70	60.3	24.01	43.79	23.89	51.25	26.56	11.17	43.05	10.62	−0.50
	1.5	16.21	52.7	19.62	21.03	20.94	39.94	21.68	3.55	29.18	9.56	6.73

(a) N = 20, $\sigma < 0.66$ g/h for θ_Δ, $\dot{\sigma} < 1.01$ g/h for $\dot{\theta}_\Delta$; (b) N = 40, $\sigma < 1.29$ g/h for θ_Δ, $\dot{\sigma} < 2.02$ g/h for $\dot{\theta}_\Delta$; (c) N = 80, $\dot{\sigma} < 2.55$ g/h for θ_Δ, $\dot{\sigma} < 9.72$ g/h for $\dot{\theta}_\Delta$.

Table 2. Comparison of Production Rates (σ_{max} and σ_0) Obtained in Inclined and Vertical Columns with L = 177 cm and Various Δ/P or Δ_0/P

	$\frac{\Delta}{P}$ or $\frac{\Delta_0}{P}$ (%)	σ_0 (g/h)	θ_0 (degree)	σ_{max} (g/h)	I_σ (%)
(a)	18.26	0.5	28.16	0.510	2.00
	12.27	1.0	—	—	—
	9.81	1.5	—	—	—
(b)	23.68	0.5	46.10	0.621	24.20
	18.51	1.0	27.48	1.017	1.70
	15.20	1.5	—	—	—
(c)	27.80	0.5	53.27	0.912	82.40
	23.89	1.0	45.90	1.236	23.60
	20.94	1.5	37.45	1.608	7.20

(a) N = 20, (Δ/P) >16.03; (b) N = 40, (Δ/P) >16.42 (c) N = 80, (Δ/P) >16.62.

RESULTS AND DISCUSSION

The comparison of separations, Δ_{max} and Δ_0, obtained at the optimum angle of inclination θ_Δ and at the vertical position ($\theta = 0$) in countercurrent-flow devices under various flow rates is presented in Table 1. This table shows that the

Table 3. Comparison of Column Lengths (L_{min} and L_0 = 177 cm) Required in Inclined and Vertical Columns with Various σ (or σ_0) and Δ (or Δ_0)

	$\frac{\Delta}{P}$ or $\frac{\Delta_0}{P}$ (%)	σ or σ_0 (g/h)	θ_0 (degree)	L_{min} (g/h)	$I_{\Delta/L}$ (%)
(a)	18.26	0.5	30.59	173.57	1.98
	12.27	1.0	—	—	—
	9.81	1.5	—	—	—
(b)	23.68	0.5	56.06	142.48	24.23
	18.51	1.0	29.22	174.11	1.66
	15.20	1.5	—	—	—
(c)	27.80	0.5	70.87	96.99	82.49
	23.89	1.0	55.71	143.25	32.75
	20.94	1.5	42.21	165.08	7.22

(a) N = 20, σ (Δ/P) < 10.61 g/h; (b) N = 40, σ(Δ/P) < 21.21 g/h; (c) N = 80, σ(Δ/P) < 42.42 g/h.

optimum angle of inclination for maximum separation increases when the flow rate decreases or as the column number increases. An improvement in separation I_Δ is obtained, especially for low flow-rate operation, or for the scheme of large column number.

The comparison of production rates, σ_{max} and σ_0, obtained at the best corresponding angle of inclination θ_σ and at the vertical position ($\theta = 0$) under various degrees of separation is given in Table 2. The optimum angle of inclination for maximum production rate increases when the specified degree of separation or the column number increases. An improvement in production rate I_σ is obtained, especially for a higher degree of separation, for the scheme having a large column number.

The minimum column length L_{min} and the corresponding angle of inclination θ_L under various flow rates and specified degree of separation are shown in Table 3. The optimum angle of inclination for minimum column length increases when the specified degree of separation or column number increases or when the specified flow rate decreases. The shorter column length, or the improvement in the degree of separation per unit column length $I_{\Delta/L}$, is obtained at lower flow rate, larger column number, or higher degree of separation.

Table 1 also gives the calculated results of separation in concurrent-flow schemes to use for comparison with those obtained in countercurrent devices. The equations for calculating the optimum angle of inclination $\dot{\theta}_\Delta$ and the corresponding maximum separation $\dot{\Delta}_{max}$ in concurrent-flow devices were already derived in a previous work (11). They are

$$\frac{\dot{\Delta}_0}{P} = \frac{(-H)L}{K}\left\{\left[1 - \left(\frac{2K}{\sigma L}\right)\cos^2\theta + 1\right]^{-N}\right\} \tag{31}$$

$$\dot{\theta}_\Delta = \cos^{-1}\left[\frac{\sigma L(1 - W)}{2KW}\right]^{1/2} \tag{32}$$

$$\frac{\dot{\Delta}_{max}}{P} = \left[\frac{2H^2LW}{\sigma K\,(1 - W)}\right]^{1/2}(1 - W^N) \tag{33}$$

in which the value of W is determined from the following equation:

$$2NW^{N+1} - (2N + 1)W^N + 1 = 0 \tag{34}$$

and the restriction on the existence of $\dot{\theta}_\Delta$ is

$$\sigma < \frac{2KW}{L(1 - W)} \tag{35}$$

Table 1 shows that in a vertical column ($\theta = 0$), the performance (Δ_0) of countercurrent flow is much better than the (Δ_0) of concurrent flow, especially for lower flow-rate operations, or for the scheme with a larger column number. In an inclined scheme the countercurrent-flow operation still overcomes the concurrent-flow operation: $\Delta_{max} - \dot{\Delta}_{max}$ is much smaller than $\Delta_0 - \dot{\Delta}_0$. In other words, the improvement of separation $\dot{I}_\Delta$ created by tilting the scheme in the concurrent-flow device, is much larger than I_Δ in the countercurrent-flow device. Also, at certain flow rates and column numbers (e.g., $N = 40$, $\sigma = 1.5g$ / h), the performance in a vertical device operating under countercurrent-flow ($\Delta_0 / P = 15.20$) is better than the $\dot{\Delta}_{max}/P = 13.81$ obtained in an inclined scheme but operating under concurrent flow.

CONCLUSIONS

The undesirable remixing effect in a countercurrent-flow Frazier scheme for enrichment of heavy water can be effectively reduced and controlled by tilting the columns, resulting in substantial improvement in performance. Furthermore, a Frazier scheme operating with countercurrent flow is better than that with concurrent flow. The equations of the optimum angle of inclination for the best performance (maximum separation, maximum output, and minimum column length) were derived. The regions in which inclination improves performance were also delineated.

A numerical example for enrichment of heavy water from the H_2O—HDO—D_2O system, in which considerable improvement in performance is obtained by operating at the optimum angle of inclination, is shown in Tables 1–3. Performance can be further improved by operating the system with countercurrent flow, instead of concurrent flow, as demonstrated by the data in Table 1.

SYMBOLS

B	column width (cm)
C	fractional mass concentration of heavy water in H_2O—HDO—D_2O system
C_0	C in feed streams
C_i	C in ith column
$C_{T,i}$, $C_{B,i}$	C in product streams exiting from ith column, for top and bottom ends, respectively
D	ordinary diffusion coefficient, cm^2/s
g	gravitational acceleration (cm/s^2)
H	system constant, defined by Eq. (4) (g/s)
I_Δ, I_σ, $I_{\sigma/L}$	improvement of performance with countercurrent flow, defined by Eqs. (28–30)
$\dot{I}_\Delta$	I_Δ with concurrent flow
J_{x-OD}, J_{x-TD}	mass flux of heavy water in x direction due to ordinary diffusion; thermal diffusion ($g/cm^2 \cdot s$)

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J_{Z-OD}	mass flux of heavy water in z direction due to ordinary diffusion ($g/cm^2 \cdot s$)
K	system constant, defined by Eq. (5) ($g \cdot cm/s$)
K_{eq}	equilibrium constant of H_2O—HDO—D_2O system
L	column length (cm)
L_{min}	minimum value of L obtained at the optimal angle of inclination θ_L (cm)
N	total column number
P	product form of concentration defined by Eq. (6)
T_m	mean absolute temperature (K)
ΔT	difference in temperature of hot and cold plates (K)
x	axis in temperature-gradient direction (cm)
z	axis in transport direction (cm)

Greek letters

α	thermal diffusion constant for H_2O—HDO—D_2O system (<0)
β	$(-\partial\rho / \partial T)$ evaluated at T_m under constant pressure ($g/cm^3 \cdot K$)
Δ	$C_{B,1} - C_{T,N}$, degree of separation in a countercurrent-flow Frazier Scheme
$\dot{\Delta}$	$C_{T,N} - C_{B,N}$, degree of separation in a concurrent-flow Frazier scheme
Δ_0, $\dot{\Delta}_0$	Δ, $\dot{\Delta}$ obtained in a vertical Frazier scheme
Δ_{max}, $\dot{\Delta}_{max}$	maximum value of Δ; $\dot{\Delta}$ obtained at the optimal angle of inclination θ_Δ
θ	column angle of inclination (degree)
θ_Δ, θ_σ, θ_L	the optimal angle of inclination for Δ_{max}, for σ_{max}, for L_{min} (degree)
μ	absolute viscosity ($g/cm \cdot s$)
ρ	mass density evaluated at T_m (g/cm^3)
σ	mass flow rate (g/s)
σ_{max}	maximum value of σ obtained at the optimal angle of inclination θ_σ (g/s)
θ_σ	σ obtained in a vertical Frazier scheme (g/s)
τ_i	transport of heavy water in z direction in ith column (< 0) (g/s)
w	half of plate spacing (g/s)

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