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The effect of column number N, as well as the column height h of thermal diffusion columns, on the degree of separation in the Frazier scheme with the total sum of the column height L (=Nh) fixed, has been investigated. The equations, which may be employed to predict the optimum column number N^* for the maximum separation $\Delta_{N,\max}$, have been derived. Considerable performance in separation is obtainable if the column number, as well as the column height, in a Frazier scheme with the total sum of the column height fixed is properly assigned for a certain flow-rate operation.

Keywords Frazier scheme; optimum column number; thermal diffusion; total sum of column height fixed

INTRODUCTION

Thermal diffusion occurs when a temperature gradient in a mixture of two gases or liquids gives rise to a concentration gradient with one component concentrated near the hot wall and the other component concentrated near the cold wall. This process can be used to separate mixtures that are difficult to separate by conventional methods such as distillation and extraction. It was the great achievement of Clusius and Dickel (1,2) to introduce the thermogravitational thermal diffusion column, in which convective currents were created to produce a cascading effect, analogous to the multistage effect of countercurrent extraction, giving a relatively large separation.

For practical applications, thermal diffusion columns are connected in series such as that shown in Fig. 1, called the Frazier Scheme (3,4). The feeding method of the Frazier Scheme is different from that of a conventional column. In a conventional thermal diffusion column, the feed is introduced at the middle and the products are withdraw from the top and bottom while, in the Frazier Scheme, the sampling streams do not pass through but move outside the columns, as shown in the figure.

The separation theory of thermal diffusion in conventional columns was first presented by Furry et al. (5,6), while that in the Frazier Scheme was given by Rabinovich and Sovorov (7,8). Many improved columns have been introduced for the best performances in the Frazier Scheme, such as inclined columns (9), optimum plate-spacing columns (10), and optimum plate aspect-ratio columns (11). It is the purpose of this work to investigate the effect of the column number, as well as the column height, on the performance in flat-pate thermal diffusion columns of the Frazier scheme with the total sum of the column height fixed. The optimal column numbers for maximum separation will be determined with various mass flow rates.

THEORY

Separation in the Frazier Scheme of N Flat-Plate Columns

The scheme proposed by Frazier, to connect N flat-plate thermal diffusion columns of the same size with forward transverse sampling streams, is shown in Fig. 1. The delivery of the supply σ with feed concentration C_F of a binary mixture is accomplished at the upper and lower ends in a flat-plate thermal diffusion column with plate spacing ($2w$) and height h, where both streams have the same direction. Sampling of the product is carried out at the ends opposite to the supply entrance.

The temperature gradient applied between the surfaces of a flat-plate thermal diffusion column for separation of a binary mixture has two effects:

1. a flux of component 1 relative to the other is brought about by thermal diffusion toward the hot surface, and
2. natural convective currents are produced, parallel to plate surfaces and up the hot surface.

The combined result of these two effects is to produce a concentration difference of component 1 between the two

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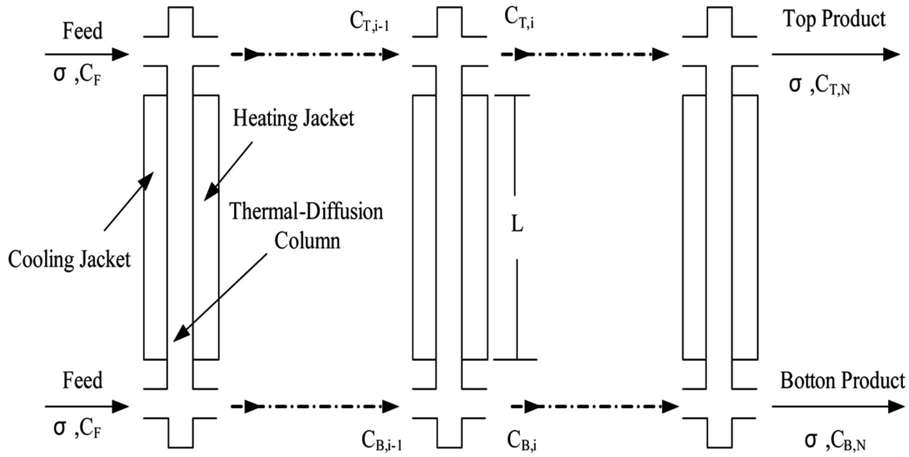

FIG. 1. Schematic diagram of cocurrent-flow Frazier scheme.

ends of the column. Figure 2 illustrates the flows and fluxes prevailing in the ith thermal diffusion column with transverse sampling streams. From the combined result of the above two effects, the transport equation for component 1 in a steady continuous-flow ith column of the Frazier scheme was given (5,6) as

$$\tau_i = HC(1-C) - K\frac{dC_i}{dZ} \tag{1}$$

where the transport coefficients, H and K, are defined as

$$H = \frac{\alpha\rho\beta_{\overline{T}}g(2w)^3B(\Delta T)^2}{6!\mu\overline{T}} \tag{2}$$

$$K = \frac{\rho\beta_{\overline{T}}^2g^2(2w)^7B(\Delta T)^2}{9!D\mu^2} \tag{3}$$

In obtaining Eq. (1), the product form of concentration $C(1-C)$ was assumed to be a constant, i.e.,

$$A = C(1-C) \tag{4}$$

The first term on the right-hand side of Eq. (1) represents the effectiveness of separation by thermal diffusion while the second term denotes the countereffect of remixing due to convection in the ith column.

By integrating Eq. (1) through the ith column with the boundary conditions:

$C_i\ (z=0) = C_{B,i}$ and $C_i(z=h) = C_{T,i}$, one obtains the steady (τ_i = constant) degree of separation in the ith column as

$$\Delta_i = C_{T,i} - C_{B,i} \tag{5}$$

$$= [AH - (\tau_i/H)](Hh/K) \tag{6}$$

Making material balances for the top and the bottom of the column, one has, respectively

$$\tau_i = \sigma(C_{T,i} - C_{T,i-1}) \tag{7}$$

$$= \sigma(C_{B,i-1} - C_{B,i})Z \tag{8}$$

or

$$\tau_i = \sigma(\Delta_i - \Delta_{i-1})/2 \tag{9}$$

Substitution of Eq. (9) into Eq. (6) yields

$$\Delta_i - [(2K/\sigma h)+1]^{-1}\Delta_{i-1} - (2H/\sigma)[(2K/\sigma h)+1]^{-1}A = 0 \tag{10}$$

Eq. (10) is the first-order difference equation of Δ_i whose solution, subject to the condition:

$$i = 0, \quad C_{0,T} = C_{0,B} = C_F, \quad \Delta_0 = C_{0,T} - C_{0,B} = 0 \tag{11}$$

is

$$\Delta_N = C_{N,T} - C_{N,B} \tag{12}$$

$$= (AHh/K)\left\{1 - \left[\frac{1}{(2K/\sigma h)+1}\right]^N\right\} \tag{13}$$

in which Δ_N denotes the degree of separation (concentration difference) of component 1 in a whole Frazier scheme of N flat-plate thermal diffusion column.

Furry et al. (5) suggested that the constant A defined by Eq. (4) may be taken as 0.25 for the equifraction solution ($0.3 < C < 0.7$), and the equation of separation, Eq. (13), reduces to

$$\overline{\Delta}_N = (Hh/4K)\left\{1 - \left[\frac{1}{(2K/\sigma h)+1}\right]^N\right\} \tag{14}$$

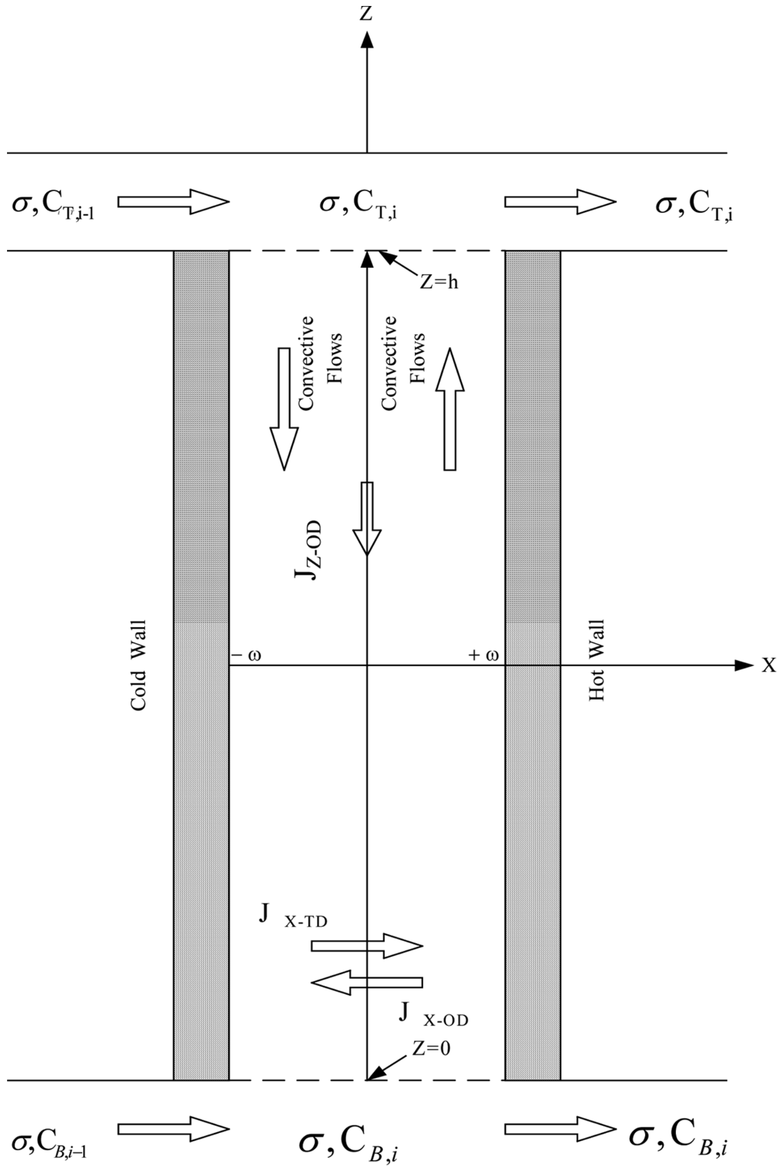

FIG. 2. Flows and fluxes in ith column.

For the whole range of concentration ($0 < C < 1$), the appropriate choice of constant A was determined by the method of least squares,

$$\text{minimum} \quad E = \int_{C_{N,B}}^{C_{N,T}} [C(1-C) - A]^2 dC$$

and the result is (12,13)

$$A = C_F(1 - C_F) - \Delta_N^2/12 \tag{15}$$

The explicit form of separation equation for whole range of concentration then obtained by substituting Eq. (15) into Eq. (13) with the use of Eq. (14)

$$\Delta_N = [C_F(1 - C_F) - \Delta_N^2/12](4\overline{\Delta}_N) \tag{16}$$

After rearrangement, one has

$$\Delta_N = \left[\left(\frac{1.5}{\overline{\Delta}_N}\right)^2 + 12C_F(1 - C_F)\right]^{1/2} - \left(\frac{1.5}{\overline{\Delta}_N}\right) \tag{17}$$

or

$$\overline{\Delta}_N = \frac{\Delta_N}{12C_F(1 - C_F) - \Delta_N^2} \tag{18}$$

Optimum Column Number for Best Performance

For a Frazier scheme of N thermal diffusion columns with the same column height h and the total sum column height L fixed

$$h = L/N \tag{19}$$

Substitution of Eq. (19) into Eq. (14) results in

$$\begin{aligned}\overline{\Delta}_N &= (HL/4KN)\left\{1 - \left[\frac{1}{(2KN/\sigma L)+1}\right]^N\right\} \\ &= (u/4)\left\{\frac{\xi}{N}\left[1 - \frac{1}{(N/\xi)+1}\right]^N\right\}\end{aligned} \tag{20}$$

where

$$u = 2H/\sigma \tag{21}$$

$$\xi = \frac{\sigma L}{2K} \tag{22}$$

The optimum column number for the maximum degree of separation with the flow rate σ and the total sum of the column height L specified, is obtained by partially differentiating Eq. (17) with respect to N and setting $\partial\Delta_N/\partial N = 0$. It is found that this also leads to $\partial\overline{\Delta}_N/\partial N = 0$. In other words, the optimum column number is independent on the solution concentration C_F. However, the optimal column number is not easy to be determined from the cumbersome equation, $\partial\overline{\Delta}_N/\partial N = 0$. Therefore, the best column number N^* for the maximum degree of separation, $\Delta_{N,\max}$, as well as $\overline{\Delta}_{N,\max}$, is obtained by employing the method of variable univariant search (14) for Eq. (20) with ξ as parameter, as illustrated in Fig. 3. Once N^* is known, $\overline{\Delta}_{N,\max}$ and $\Delta_{N,\max}$ are calculated from Eqs. (20) and (17), respectively. The results for N^* under various values of ξ were thus obtained and some of them are presented graphically in Fig. 4. It is found from this figure that the result is nearly a straight line, and thus the following expression is reached

$$N^* = 1.126\xi^{0.5} \tag{23}$$

Note that N^* is independent on the feed concentration C_F, and that the value of N^* calculated from Eq. (23) must be taken as a positive integer for the practical application. Accordingly, $\overline{\Delta}_{N,\max}$ can be calculated from Eq. (20) with the calculated values of N replaced by the best integer value, N^*, and finally, the equation for the maximum degree of separation is obtained by modifying Eq. (17) as

$$\Delta_{N,\max} = \left[\left(\frac{1.5}{\overline{\Delta}_{N,\max}}\right)^2 + 12C_F(1-C_F)\right] - \frac{1.5}{\overline{\Delta}_{N,\max}} \tag{24}$$

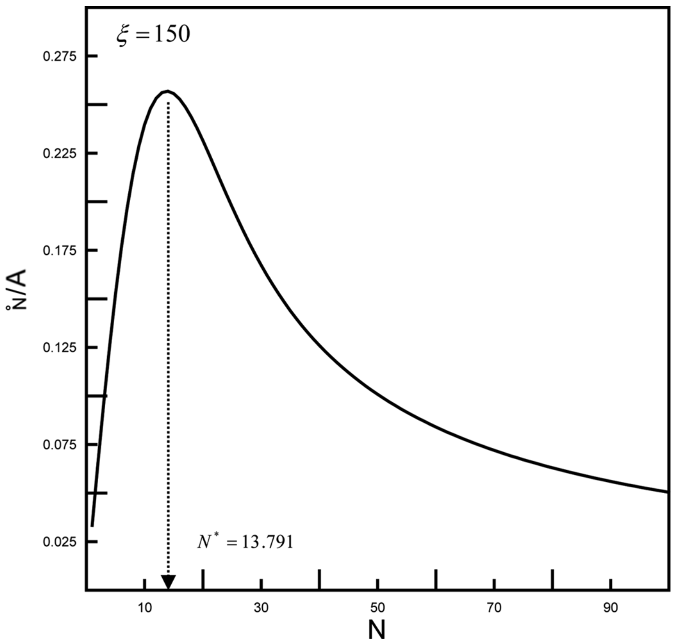

FIG. 3. Determination of the optimal column number.

IMPROVEMENT IN SEPARATION

The improvement in separation by employing the Frazier scheme of the optimum column number is best illustrated by calculating the percentage increase in the maximum separation based on the Frazier scheme of

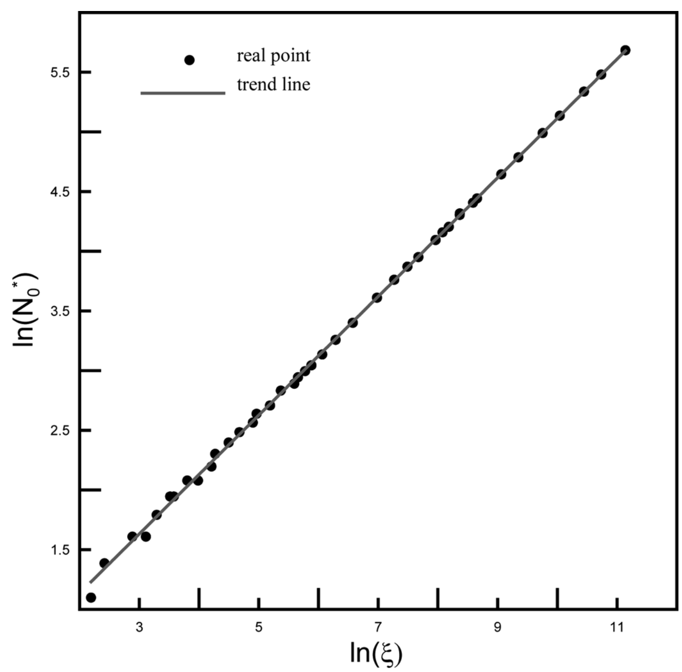

FIG. 4. Correlation equation of N^* vs. ξ.

certain column number N

$$I = \frac{\Delta_{N,\max} - \Delta_N}{\Delta_N} = \frac{\Delta_{N,\max}}{\Delta_N} - 1 \tag{25}$$

Further, from Eq. (18)

$$\frac{\overline{\Delta}_{N,\max}}{\overline{\Delta}_N} = \left(\frac{\Delta_{N,\max}}{\Delta_N}\right) \frac{12C_F(1-C_F) - \Delta_N^2}{12C_F(1-C_F) - \Delta_{N,\max}^2} \tag{26}$$

Since in the thermal diffusion column, both Δ_N^2 and $\Delta_{N,\max}^2$ are much less than $12C_F\,(1-C_F)$, Eq. (26) may be approximated to

$$\frac{\overline{\Delta}_{N,\max}}{\overline{\Delta}_N} \approx \frac{\Delta_{N,msx}}{\Delta_N} \tag{27}$$

Accordingly, Eq. (25) may be simplified to

$$I \approx \frac{\overline{\Delta}_{N,\max}}{\overline{\Delta}_N} - 1 \tag{28}$$

For the purpose of illustration, let us employ the equipment parameters for the separation of the benzene and n-heptane (component 1) mixture given in the previous work (15) as follows: $B = 10\,\text{cm}$, $2w = 0.09\,\text{cm}$, $\Delta T = 38.3^\circ C$, $\overline{T} = 328\,\text{K}$. The transport coefficients thus obtained from the experimental data are: $H = 0.845\,\text{g/min}$ and $K = 419\,\text{g}\cdot\text{cm/min}$. Here we assign the total sums of column height in the N thermal diffusion columns as $L = Nh = 25\,\text{m}, 50\,\text{m}$, and $75\,\text{m}$. Substitution of the preceding values into the appropriate equations, the optimum column number and the corresponding degrees of separation, as well as the improvement in separation, are calculated. The results are presented in Tables 1–3.

DISCUSSION AND CONCLUSION

The equations which may be employed to predict the optimum column number N^* for maximum degrees of separation, $\overline{\Delta}_{N,\max}$ and $\Delta_{N,\max}$, for the separation of a binary mixture in the Frazier Scheme of thermal diffusion columns with the total sum of column height ($L = \text{Nh}$) fixed, have been derived. They are Eqs. (23) and (24) for N^* and $\Delta_{N,\max}$, respectively. As mentioned in Section 2.2., the practical value of the optimum column number N^* calculated from Eq. (23) must be taken as a positive integer and according, the actual value of $\Delta_{N,\max}$ should be calculated from Eqs. (20) and (24) with the calculated value of N replaced by the practical value of integer N^*.

The improvement in separation was illustrated numerically by employing the experimental data of benzene and n-heptane mixture obtained in previous work (15), and the results are presented in Tables 1–3 for $L = 25\,\text{m}$ ($N = 10$), $50\,\text{m}$ ($N = 20$) and $75\,\text{m}$ ($N = 30$), respectively, with $h = 2.5\,\text{m}$. It has been shown in these tables that considerable performance in the separation of a binary mixture by thermal diffusion in the Frazier scheme can be achieved if a Frazier scheme is constructed by N^* thermal diffusion columns with the total sum of column height fixed. However, for the particular cases that when $\sigma = 24$, 59.538 and 89.307 g/min for $L = 25, 50$, and $75\,\text{m}$, respectively, $N = N^* = 10, 20$, and 30 as well as $h = h^*$. As mentioned earlier in Section 2.2, the optimum column number is independent on the feed concentration C_F, while the improvement in separation I is nearly independent on C_F, as indicated by Eq. (28). Further, a larger value of

TABLE 1
Comparison of the degrees of separation ($\overline{\Delta}_{N=10}$ and $\overline{\Delta}_{N,\max}$) obtained in the Frazier Scheme of same column height (L/N) with $L = 25\,\text{m}$, $N = 10$ ($h = 2.5\,\text{m}$) and $N = N^*$

					$\Delta_{N,\max}$ (%)			
σ (g/min)	$\overline{\Delta}_{10}$ (%)	N^*	h^* (m)	$\overline{\Delta}_{N,\max}$ (%)	$C_F = 0.1$ or 0.9	$C_F = 0.3$ or 0.7	$C_F = 0.5$	I (%)
3	12.60	3	8.3333	24.36	8.71	20.14	23.90	48.29
6	12.46	5	5.0000	17.85	6.40	14.87	17.67	30.23
12	11.53	7	3.5714	12.85	4.62	10.74	12.78	10.25
(24)	(9.19)	(10)	(2.5000)	(9.19)	(3.31)	(7.71)	(9.17)	(0)
36	7.43	12	2.0833	7.56	2.72	6.34	7.54	1.68
48	6.19	14	1.7857	6.56	2.36	5.51	6.55	5.74
60	5.29	15	1.6667	5.89	2.12	4.94	5.88	10.26
120	3.04	21	1.1905	4.19	1.51	3.52	4.19	27.54
240	1.63	30	0.8333	2.98	1.07	2.50	2.97	45.13
360	1.12	37	0.6757	2.43	0.88	2.04	2.43	54.16
480	0.85	43	0.5814	2.11	0.76	1.77	2.11	59.84
600	0.68	48	0.5208	1.89	0.68	1.59	1.89	63.84

TABLE 2
Comparison of the degrees of separation ($\overline{\Delta}_{N=20}$ and $\overline{\Delta}_{N,\max}$) obtained in the Frazier Scheme of same column height (L/N) with $L = 50$ m, $N = 20$ ($h = 2.5$ m) and $N = N^*$

					$\Delta_{N,\max}$(%)			
σ (g/min)	$\overline{\Delta}_{20}$ (%)	N^*	h^* (m)	$\overline{\Delta}_{N,\max}$ (%)	$C_F = 0.1$ or 0.9	$C_F = 0.3$ or 0.7	$C_F = 0.5$	I (%)
3	12.60	5	10.0000	35.71	12.66	28.99	34.31	64.70
6	12.60	7	7.1429	25.70	9.18	21.20	25.15	50.96
12	12.51	10	5.0000	18.39	6.59	15.30	18.19	31.95
24	11.68	13	3.8462	13.13	4.72	10.97	13.05	11.01
36	10.48	17	2.9412	10.77	3.87	9.01	10.73	2.66
48	9.34	19	2.6316	9.35	3.36	7.84	9.33	0.17
(52.876)	(8.92)	(20)	(2.5000)	(8.92)	(3.21)	(7.47)	(8.89)	(0)
60	8.35	21	2.3810	8.38	3.01	7.02	8.36	0.28
120	5.34	30	1.6667	5.95	2.14	4.99	5.94	10.26
240	3.05	43	1.1628	4.22	1.52	3.54	4.22	27.64
360	2.13	52	0.9615	3.45	1.24	2.90	3.45	38.19
480	1.64	60	0.8333	2.99	1.08	2.51	2.99	45.24
600	1.33	67	0.7463	2.68	0.96	2.25	2.68	50.34

the optimum column number for maximum separation is needed for a higher flow rate σ, as well as for a larger value of total sum of column height L.

Actually, increasing the column height h of a single ($N = 1$) thermal diffusion column will increase the effective separation section of the device, leading to increased the degree of separation Δ_1. However, the increment of Δ_1 by further increasing h, has a limiting value, especially for a larger value of the flow rate σ. Taking $h = L(N = 1) = 10$ m, m, 25 m, and 50 m in a single-column device for instance, one has from Eq. (20) that as $\sigma = 600$ g/min, $\overline{\Delta}_1 =$ 0.002813, 0.002815, and 0.002816, respectively. Therefore, the column height of a thermal diffusion column should be properly controlled, and a Frazier scheme constructed with thermal diffusion columns of the optimum column number N^*, as well as with the optimal column height, $h = L/N^*$, is preferably employed to achieve the best performance.

TABLE 3
Comparison of the degrees of separation ($\overline{\Delta}_{N=30}$ and $\overline{\Delta}_{N,\max}$) obtained in the Frazier Scheme of same column height (L/N) with $L = 75$ m, $N = 30$ ($h = 2.5$ m) and $N = N^*$

					$\Delta_{N,\max}$(%)			
σ (g/min)	$\overline{\Delta}_{30}$ (%)	N^*	h^* (m)	$\overline{\Delta}_{N,\max}$ (%)	$C_F = 0.1$ or 0.9	$C_F = 0.3$ or 0.7	$C_F = 0.5$	I (%)
3	12.60	6	12.5000	44.23	15.57	35.32	41.67	71.50
6	12.60	8	9.3750	31.70	11.28	25.92	30.71	60.24
12	12.60	12	6.2500	22.67	8.11	18.78	22.30	44.44
24	12.35	17	4.4118	16.15	5.80	13.47	16.01	23.50
36	11.73	20	3.7500	13.24	4.76	11.07	13.16	11.37
48	10.94	23	3.2609	11.49	4.13	9.61	11.44	4.75
60	10.14	26	2.8846	10.29	3.70	8.62	10.25	1.49
(79.314)	(8.96)	(30)	(2.5000)	(8.96)	(3.22)	(7.51)	(8.94)	(0)
120	7.09	37	2.0270	7.30	2.63	6.12	7.29	2.91
240	4.29	52	1.4423	5.18	1.86	4.34	5.17	17.10
360	3.06	64	1.1719	4.23	1.52	3.55	4.23	27.68
480	2.37	74	1.0135	3.67	1.32	3.08	3.66	35.22
600	1.94	82	0.9146	3.28	1.18	2.75	3.28	40.86

NOMENCLATURE

A	$=C(1-C)$
B	=column width (cm)
C	=fractional mass concentration of component 1 in a binary mixture
$C_{B,i}$	$=C$ in the bottom product stream of ith column
C_F	$=C$ in the feed streams
$C_{T,i}$	$=C$ in the top product stream of ith column
D	=ordinary diffusion coefficient (cm^2/s)
g	=gravitational acceleration (cm/s^2)
H	=system constant defined by Eq. (2) (g/s)
h	=column height (cm)
I	=improvement in separation defined by Eqs. (25) and (28)
$J_{x\text{-}OD}$	=mass flux of component 1 in x direction owing to ordinary diffusion (g/m^2s)
$J_{x\text{-}TD}$	=mass flux of component 1 in x direction owing to thermal diffusion (g/m^2s)
$J_{z\text{-}OD}$	=mass flux of component 1 in z direction owing to ordinary diffusion (g/m^2s)
K	=system constant defined by Eq. (3) (gcm/s)
L	=total sum of column height, Nh (cm)
N	=column number
N^*	=the optimum column number
$\overline{T}$	=mean absolute temperature (K)
ΔT	=difference in temperature between hot and cold plates (K)
u	$=2H/\sigma$
z	=axis parallel to the surfaces in flat-plate column (cm)

Greek Letters

α	=thermal diffusion constant
$\beta_{\overline{T}}$	$=-(1/\rho)(\partial\rho/\partial T)$ evaluated at $\overline{T}$ $(1/K)$
Δ	=degree of separation obtained in the Frazier Scheme
Δ_i	$=\Delta$ obtained in ith column, $C_{T,i}-C_{B,i}$
Δ_N	$=\Delta$ obtained in Nth column, $C_{T,N}-C_{B,N}$
$\overline{\Delta_N}$	$=\Delta_N$ obtained for equifraction solution
$\Delta_{N,\max}$	=maximum value of Δ_N obtained in the Frazier Scheme with optimum column number
ρ	=mass density evaluated at $\overline{T}$ (g/cm^3)
μ	=absolute viscosity (g/cm s)
σ	=mass flow rate (g/s)
w	=half of the plate spacing (cm)
τ_i	=transport of component 1 along z-direction in ith column (g/s)
ξ	$=\sigma L/2K$

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