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AN ANALYTICAL STUDY ON THE ENRICHMENT OF HEAVY WATER IN THE CONTINUOUS THERMAL-DIFFUSION COLUMN WITH EXTERNAL REFLUXES

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ABSTRACT

A new device of inserting an impermeable sheet or a permeable barrier to divide the thermal-diffusion columns into two subchannels with external refluxes at the ends, resulting in substantial improvement of the separation efficiency of heavy water, has been developed and investigated using orthogonal expansion technique. The analytical results are represented graphically and compared with that in a Clusius–Dickel column of the same size with recycle. Considerable improvement on enrichment of heavy water is obtained by employing such devices with an impermeable sheet or a permeable barrier instead of using the Clusius–Dickel thermal-diffusion column. The effect of sheet or barrier location on the enhancement of the separation efficiency of heavy water has also been discussed.

Key Words: Continuous type; Orthogonal expansion techniques; Thermal diffusion; Water isotopes

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INTRODUCTION

Thermal diffusion is an unconventional process for separating liquid or gas mixtures that are difficult to separate by using traditional processes such as distillation or extraction. The well-known separation phenomenon of the thermogravitational thermal-diffusion column was first introduced by Clusius and Dickel[1,2] to make the purification technique used to concentrate highly desired valuable materials such as isotopes and rare gases, as well as to remove undesired ones technically and economically feasible. The practical application with the complete theory of the Clusius–Dickel column was first presented by Furry et al.[3,4] Recently, the enrichment of heavy water in the Clusius–Dickel column was investigated both theoretically and experimentally.[5–7]

The convective currents, producing a cascading effect analogous to the multi-stage effect of counter-current 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 devices for improving the performance in a relatively large separation are either a suppression of remixing effect or an enhancement of the cascading effect. Some improved columns have been proposed in the literature for the device of considerable improvement in separation efficiency, such as inclined column,[8,9] wired column,[10,11] inclined moving-wall columns,[12,13] rotary columns,[14–17] packed columns,[18,19] rotary wired columns,[20,21] permeable barrier columns,[22,23] and impermeable barrier columns.[24] The enrichments obtained from these improved columns are somewhat better than that from the conventional Clusius–Dickel column.

The introduction of a thin impermeable sheet or permeable barrier[25] in the working space of the column can also effectively increase both the magnitude and the rate of separation, resulting in two-channel thermal-diffusion columns, increases of both the magnitude and the rate of separation. The transport phenomena in such a new device belong to the category of conjugated Graetz problems with refluxes at both ends. The solutions to these theoretical formulations are obtained by using the method of separation of variables, where resulting eigenvalue problem is solved by an orthogonal expansion technique.[22,26–32] The aim of this work is to investigate the improvement in the separation efficiency of heavy water in such two-channel devices by inserting an impermeable sheet or a permeable barrier. The present study includes the effects of recycling on mass transfer with reflux ratio and sheet or barrier location as parameters. The results obtained in this work may also be used with other conjugated Greatz problems with counter-current flow and with reflux internally or externally at both ends.

THEORETICAL FORMULATIONS

The Clusius–Dickel Column

Consider a continuous flat-plate thermogravitational thermal-diffusion column filled with water isotopes, the permeable barrier is removed as shown in Fig. 1. The distance between the plates is W. Furry et al.[3] have presented an equation that gives the separation for the column where top and bottom products withdrawn at the same rate from both ends, with the feed introduced at the center of the column. Yeh and Yang[5] developed the transport equation for the enrichment of heavy water in batch-type Clusius–Dickel column. For the continuous operation, the transport equations may be modified from the previous results are given as follows:

$$\Delta_{\mathrm{e}} = C_{\mathrm{b}} - C_{\mathrm{f}} = F_{\mathrm{e}}[1 - \exp(-\sigma' L'/2)]/\sigma' \tag{1}$$

$$\Delta_{\mathrm{s}} = C_{\mathrm{f}} - C_{\mathrm{t}} = F_{\mathrm{s}}[1 - \exp(-\sigma' L'/2)]/\sigma' \tag{2}$$

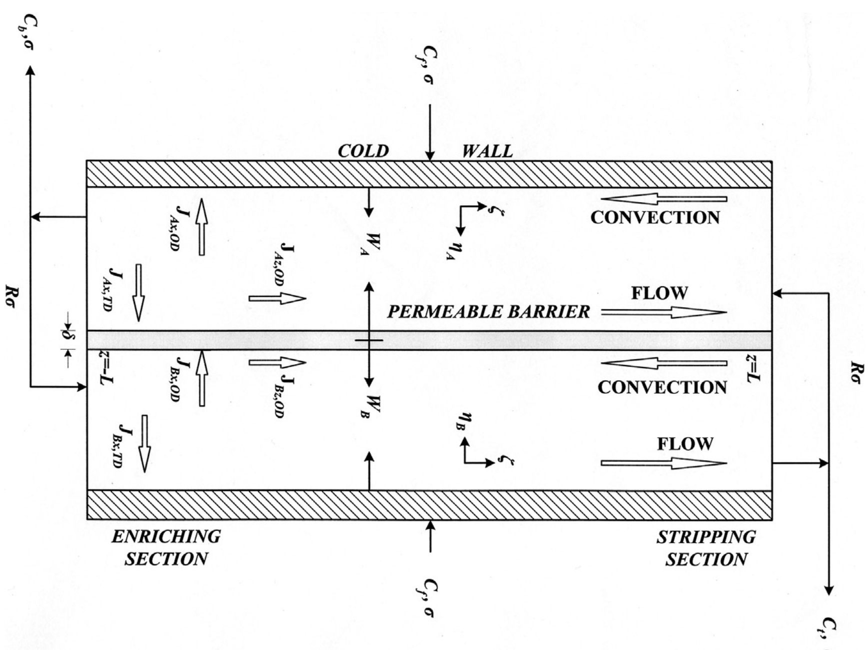

Figure 1. Schematic diagram of a continuous-type thermal-diffusion column with a vertical permeable barrier inserted.

Summing Eqs. (1) and (2) gives the degree of separation for the whole column

$$\Delta_0 = C_b - C_t = \Delta_e + \Delta_s = (F_e + F_s)[1 - \exp(-\sigma' L'/2)]/\sigma' \quad (3)$$

where the dimensionless variables are defined as

$$\sigma' = \frac{\sigma}{(-H)} \quad \text{and} \quad L' = \frac{L(-H)}{K} \quad (4)$$

The transport constants in the above equations are defined by

$$H = \frac{\alpha\beta\rho g (2\omega)^3 (\Delta T)^2}{6!\mu \bar{T}} < 0 \quad (5)$$

and

$$K = \frac{\rho g^2 \beta^2 W^7 B (\Delta T)^2}{9!\mu^2 D} + W\rho D B \quad (6)$$

while the pseudo-concentration products $C_e\hat{C}_e$ ($= F_e = \frac{1}{C_B - C_F}\int_{C_F}^{C_B} C_e\hat{C}_e \, dC_e$, an appropriate constant) and $C_s\hat{C}_s$ ($= F_s = \frac{1}{C_F - C_T}\int_{C_T}^{C_F} C_s\hat{C}_s \, dC_s$, another appropriate constant) defined in Eq. (7) for enriching and stripping sections, respectively, were considered as constant in the previous works,[6,7] are defined as

$$C\hat{C} = C\left\{0.05263 - (0.05263 - 0.0135K_{eq})C - 0.027\left\{\left[1 - \left(1 - \frac{K_{eq}}{4}\right)C\right]CK_{eq}\right\}^{1/2}\right\} \quad (7)$$

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

$$H_2O + D_2O \Leftrightarrow 2HDO \quad (8)$$

is

$$K_{eq} = \frac{C_2^2}{C_1 C_3} = \frac{[HDO]^2}{[H_2O][D_2O]} \times \frac{19 \times 19}{18 \times 20} \quad (9)$$

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 $T = 25$ and 30.5°C.[33] A graphical representation of $C\hat{C}$ vs. C with $K_{eq} = 3.80$ at 25°C is plotted in Fig. 2. During the derivation of the above equations, all the physical properties, defined in the Nomenclature, are evaluated at the reference temperature

$$\bar{T} = (T_1 + T_2)/2 \quad (10)$$

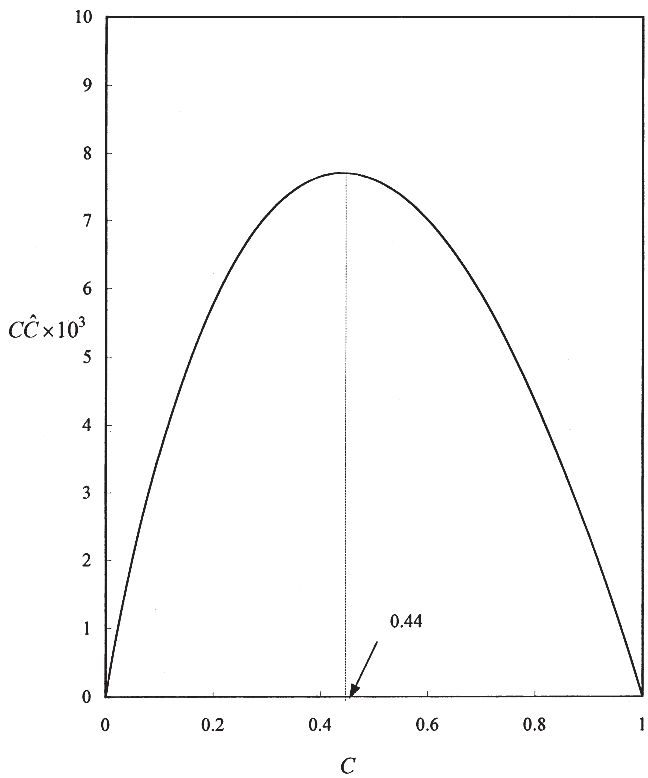

Figure 2. The function $C\hat{C}$ vs. C.

An Improved Column

An impermeable sheet or a permeable barrier with negligible thickness δ is inserted in parallel into a continuous flat-plate thermal-diffusion column with thickness ($W = W_A + W_B$) to divide the open column into two channels, channel A (left channel) and channel B (right channel), with thicknesses W_A and W_B, respectively. Feeds are introduced from center of the column and the products are withdrawn from both ends continuously with a variable flow rate ratio. In the present study, the whole column is separately composed of enriching and stripping sections and each section is divided into channels by the impermeable sheet or permeable barrier.

Temperature Distributions

The heat transfer by conduction only was assumed since the space between the plates is small. Accordingly, the temperature distribution in the fluid is linear and the temperature gradient of the fluid in the regions is

$$h_1 = \frac{\Delta T}{W_A + W_B + \frac{k_f \delta}{k_f \varepsilon + k(1-\varepsilon)}} \tag{11}$$

where k_f is the thermal conductivity of the fluid, and k, δ, and ε are the thermal conductivity, thickness, and permeability of the barrier, respectively. During the derivation of Eq. (11), we assumed that the thermal resistance in the barrier was composed of the resistances of the fluid and the barrier in parallel. Hence, the temperature gradient of the fluid within the barrier can be expressed as follows:

$$h_2 = \frac{h_1 k_f}{k_f \varepsilon + k(1-\varepsilon)} \tag{12}$$

The Governing Equations for the Degree of Separation

The following assumptions are made in the present analysis:

1. Fully developed laminar flow of the fluid in the regions and neglecting the influences of ordinary and thermal diffusions, end effects, and inertia terms on the velocity.
2. Neglect ordinary diffusion in the vertical direction and bulk flow in the horizontal direction.
3. Concentration changes or the separation is small for the entire column, the fluxes due to thermal diffusion, $\alpha C\hat{C}/\tilde{T}$, $\alpha C_{Ae}\hat{C}_{Ae}/\tilde{T}$, and $\alpha C_{Be}\hat{C}_{Be}/\tilde{T}$ may be regarded as constant.
4. Within the barrier, no bulk flow exists and the change of concentration is small. Thus, a linear relationship of concentration is expressed at $\eta_A = \eta_B = 1$. Moreover, due to the internal reflux at the end, the boundary condition is $C_{Ae} = C_{Be} = C_t$ at $\zeta = 1$.

The velocity distributions may be written as

$$V_{Ae}(\eta_A) = -f_{1e}\left(\eta_A - \eta_A^3\right) + f_{2e}\left(\eta_A^2 - \eta_A\right) + f_{3e}(1+R)\left(\eta_A - \eta_A^2\right) \tag{13}$$

$$V_{Be}(\eta_B) = g_{1e}\left(\eta_B - \eta_B^3\right) - g_{2e}\left(\eta_B^2 - \eta_B\right) + g_{3e}R\left(\eta_B - \eta_B^2\right) \tag{14}$$

in which

$$f_{1\mathrm{e}} = \frac{\beta g h_1 W_\mathrm{A}^3}{6\mu}, \quad f_{2\mathrm{e}} = \frac{\beta g W_\mathrm{A}^2 \left[2W_\mathrm{B}^3 \Delta T - h_1\left(W_\mathrm{A}^4 - W_\mathrm{B}^4\right)\right]}{4\mu\left(W_\mathrm{A}^3 + W_\mathrm{B}^3\right)},$$

$$f_{3\mathrm{e}} = \frac{6W_\mathrm{A}^2\sigma}{\rho B\left(W_\mathrm{A}^3 + W_\mathrm{B}^3\right)}, \quad g_{1\mathrm{e}} = \frac{\beta g h_1 W_\mathrm{B}^3}{6\mu},$$

$$g_{2\mathrm{e}} = \frac{\beta g W_\mathrm{B}^2 \left[2W_\mathrm{A}^3 \Delta T + h_1\left(W_\mathrm{B}^4 - W_\mathrm{A}^4\right)\right]}{4\mu\left(W_\mathrm{A}^3 + W_\mathrm{B}^3\right)},$$

$$g_{3\mathrm{e}} = \frac{6W_\mathrm{B}^2\sigma}{\rho B\left(W_\mathrm{A}^3 + W_\mathrm{B}^3\right)},$$

$$\tilde{T} = \frac{2W_\mathrm{A}^3 T_1 + 2W_\mathrm{B}^3 T_2 - h_1\left(W_\mathrm{A}^4 - W_\mathrm{B}^2\right)}{2\left(W_\mathrm{A}^3 + W_\mathrm{B}^2\right)}, \quad \eta_\mathrm{A} = \frac{x_\mathrm{A}}{W_\mathrm{A}},$$

$$\eta_\mathrm{B} = \frac{x_\mathrm{B}}{W_\mathrm{B}}, \quad \zeta = \frac{z}{L}, \quad \kappa = W_\mathrm{A}/W \tag{15}$$

A Permeable Barrier Inserted

Consider a flat-plate thermal-diffusion column with a permeable vertical barrier inserted between the plates, as shown in Fig. 1. The barrier has a negligible thickness comparable with the distance between the plates. The whole column is composed of two sections and each section is divided into two regions by the barrier. Fully developed fluids in both regions are a counter-current operation with internal reflux at the ends.

Equations of mass balances in dimensionless form may be obtained:[22]

$$\frac{\partial^2 C_\mathrm{Ae}}{\partial \eta_\mathrm{A}^2} = \left(\frac{W_\mathrm{A}^2 V_\mathrm{Ae}}{LD}\right)\frac{\partial C_\mathrm{Ae}}{\partial \zeta} \tag{16}$$

$$\frac{\partial^2 C_\mathrm{Be}}{\partial \eta_\mathrm{B}^2} = \left(\frac{W_\mathrm{B}^2 V_\mathrm{Be}}{LD}\right)\frac{\partial C_\mathrm{Be}}{\partial \zeta} \tag{17}$$

The boundary conditions are

$$\frac{-\partial C_\mathrm{Ae}}{\partial \eta_\mathrm{A}} + \frac{\alpha C_\mathrm{Ae}\hat{C}_\mathrm{Ae} h_1 W_\mathrm{A}}{D\tilde{T}} = 0 \quad \text{at } \eta_\mathrm{A} = 0 \tag{18}$$

$$\frac{\partial C_{\mathrm{Be}}}{\partial \eta_{\mathrm{B}}} + \frac{\alpha C_{\mathrm{Be}} \hat{C}_{\mathrm{Be}} h_1 W_{\mathrm{B}}}{D\tilde{T}} = 0 \quad \text{at } \eta_{\mathrm{B}} = 0 \tag{19}$$

$$-\frac{\partial C_{\mathrm{Ae}}}{\partial \eta_{\mathrm{A}}} + \frac{\alpha C_{\mathrm{Ae}} \hat{C}_{\mathrm{Ae}} h_1 W_{\mathrm{A}}}{D\tilde{T}} = \frac{W_{\mathrm{A}}}{W_{\mathrm{B}}} \left[\frac{\partial C_{\mathrm{Be}}}{\partial \eta_{\mathrm{B}}} + \frac{\alpha C_{\mathrm{Be}} \hat{C}_{\mathrm{Be}} h_1 W_{\mathrm{B}}}{D\tilde{T}} \right] \quad \text{at } \eta_{\mathrm{A}} = \eta_{\mathrm{B}} = 1 \tag{20}$$

$$-\frac{\partial C_{\mathrm{Ae}}}{\partial \eta_{\mathrm{A}}} + \frac{\alpha C_{\mathrm{Ae}} \hat{C}_{\mathrm{Ae}} h_1 W_{\mathrm{A}}}{D\tilde{T}} = \frac{W_{\mathrm{A}} \varepsilon}{D\delta} \left[\frac{\alpha C \hat{C} h_2 \delta}{\tilde{T}} - C_{\mathrm{Be}} + C_{\mathrm{Ae}} \right] \quad \text{at } \eta_{\mathrm{A}} = \eta_{\mathrm{B}} = 1 \tag{21}$$

$$C_{\mathrm{Ae}} = C_{\mathrm{Be}} = C_{\mathrm{t}} \quad \text{at } \zeta = 1 \tag{22}$$

where α is the thermal-diffusion constant. During the derivation of the above equations, all the physical properties were assumed constant and could be evaluated at the reference temperature. Likewise, the velocity distributions in the stripping section were obtained with the mass flow rate σ and the subscript e being replaced by $-\sigma$ and s, respectively, in Eqs. (13)–(22).

By following similar calculation procedures performed in our previous works,[22] with the eigenvalues $(\lambda_{\mathrm{e},1}, \lambda_{\mathrm{e},2}, \ldots, \lambda_{\mathrm{e},m}, \ldots)$ calculated from the following equations:

$$\frac{S_{\mathrm{Ae},m}}{S_{\mathrm{Be},m}} = \frac{W_{\mathrm{A}} \varepsilon F_{\mathrm{Be},m}(1)}{W_{\mathrm{A}} \varepsilon F_{\mathrm{Ae},m}(1) + \delta F'_{\mathrm{Ae},m}(1)} = -\frac{W_{\mathrm{A}}}{W_{\mathrm{B}}} \frac{F'_{\mathrm{Ae},m}(1)}{F'_{\mathrm{Be},m}(1)} \tag{23}$$

Likewise, the eigenvalues $(\lambda_{\mathrm{s},1}, \lambda_{\mathrm{s},2}, \ldots, \lambda_{\mathrm{s},m}, \ldots)$ calculated from Eq. (24) are as follows:

$$\frac{S_{\mathrm{As},m}}{S_{\mathrm{Bs},m}} = \frac{W_{\mathrm{A}} \varepsilon F_{\mathrm{Bs},m}(1)}{W_{\mathrm{A}} \varepsilon F_{\mathrm{As},m}(1) + \delta F'_{\mathrm{As},m}(1)} = -\frac{W_{\mathrm{A}}}{W_{\mathrm{B}}} \frac{F'_{\mathrm{As},m}(1)}{F'_{\mathrm{Bs},m}(1)} \tag{24}$$

The degree of separation for whole column was obtained by solving Eqs. (16) and (17) analytically with the use of Eqs. (18)–(22). The expressions of $F_{\mathrm{Ae},m}$ and $F_{\mathrm{Be},m}$ (or $F_{\mathrm{As},m}$ and $F_{\mathrm{Bs},m}$) are shown in Appendix A. The results of both enriching and stripping sections are in terms of the eigenvalues ($\lambda_{\mathrm{e},m}$ and $\lambda_{\mathrm{s},m}$), expansion coefficients ($S_{\mathrm{Ae},m}$, $S_{\mathrm{Be},m}$, $S_{\mathrm{As},m}$, and $S_{\mathrm{Bs},m}$), location of permeable barrier (κ) and eigenfunctions ($F_{\mathrm{Ae},m}$, $F_{\mathrm{Be},m}$, $F_{\mathrm{As},m}$, and $F_{\mathrm{Bs},m}$) as follows:

$$\begin{aligned}\Delta &= C_{\mathrm{t}} - C_{\mathrm{b}} \\ &= \left(\frac{LD\sum_{m=0}^{\infty}\left[\frac{S_{\mathrm{Ae},m}F'_{\mathrm{Ae},m}(1)}{\lambda_{\mathrm{e},m}}\right][1-\exp(-\lambda_{\mathrm{e},m})]}{(\kappa W)^2\int_0^1 V_{\mathrm{Ae}}\,\mathrm{d}\eta_{\mathrm{A}}} + \frac{\int_0^1 V_{\mathrm{Ae}}C_{\mathrm{Ae}}(\eta_{\mathrm{A}},0)\,\mathrm{d}\eta_{\mathrm{A}}}{\int_0^1 V_{\mathrm{Ae}}\,\mathrm{d}\eta_{\mathrm{A}}} \right) \\ &\quad - \left(\frac{\int_0^1 V_{\mathrm{As},m}C_{\mathrm{As}}(\eta_{\mathrm{A}},0)\,\mathrm{d}\eta_{\mathrm{A}}}{\int_0^1 V_{\mathrm{As}}\,\mathrm{d}\eta_{\mathrm{A}}} - \frac{LD\sum_{m=0}^{\infty}\left[\frac{S_{\mathrm{As},m}F'_{\mathrm{As},m}(1)}{\lambda_{\mathrm{s},m}}\right][\exp(\lambda_{\mathrm{s},m})-1]}{(\kappa W)^2\int_0^1 V_{\mathrm{As}}\,\mathrm{d}\eta_{\mathrm{A}}} \right)\end{aligned} \tag{25}$$

or

$$\begin{aligned}\Delta &= C_{\mathrm{t}} - C_{\mathrm{b}} \\ &= \left(\frac{LD\sum_{m=0}^{\infty}\left[\frac{S_{\mathrm{Be},m}F'_{\mathrm{Be},m}(1)}{\lambda_{\mathrm{e},m}}\right][1-\exp(-\lambda_{\mathrm{e},m})]}{(1-\kappa)^2 W^2\int_0^1 V_{\mathrm{Be}}\,\mathrm{d}\eta_{\mathrm{B}}} + \frac{\int_0^1 V_{\mathrm{Be}}C_{\mathrm{Be}}(\eta_{\mathrm{B}},0)\,\mathrm{d}\eta_{\mathrm{B}}}{\int_0^1 V_{\mathrm{Be}}\,d\eta_{\mathrm{B}}} \right) \\ &\quad - \left(\frac{\int_0^1 V_{\mathrm{Bs},m}C_{\mathrm{Bs}}(\eta_{\mathrm{B}},0)\,\mathrm{d}\eta_{\mathrm{B}}}{\int_0^1 V_{\mathrm{Bs}}\,\mathrm{d}\eta_{\mathrm{B}}} - \frac{LD\sum_{m=0}^{\infty}\left[\frac{S_{\mathrm{Bs},m}F'_{\mathrm{Bs},m}(1)}{\lambda_{\mathrm{s},m}}\right][\exp(\lambda_{\mathrm{s},m})-1]}{(1-\kappa)^2 W^2\int_0^1 V_{\mathrm{Bs}}\,\mathrm{d}\eta_{\mathrm{B}}} \right)\end{aligned} \tag{26}$$

Figure 3 is the graphical representation of the degree of separation (Δ) for the whole column.

An Impermeable Sheet Inserted

Similarly, an impermeable sheet is inserted as shown in Fig. 1. Feeds are introduced at the center of the column and the products are withdrawn from both ends for each section. The value of inserting the impermeable sheet, which as a metal sheet, is to produce two open columns side-by-side, the two having equal heat fluxes and a common temperature at their junction. Since the two sides, sections A and B, are separated everywhere by a sheet with negligible thermal resistance, each of the two columns will operate independently, with no mass exchange, except for the shared temperature gradient. At steady state, the compositions in each section will therefore depend on the feed composition. The equations of mass balances in dimensionless form are the same as in Eqs. (16) and (17), except that the boundary conditions in Eqs. (20) and (21) are replaced by

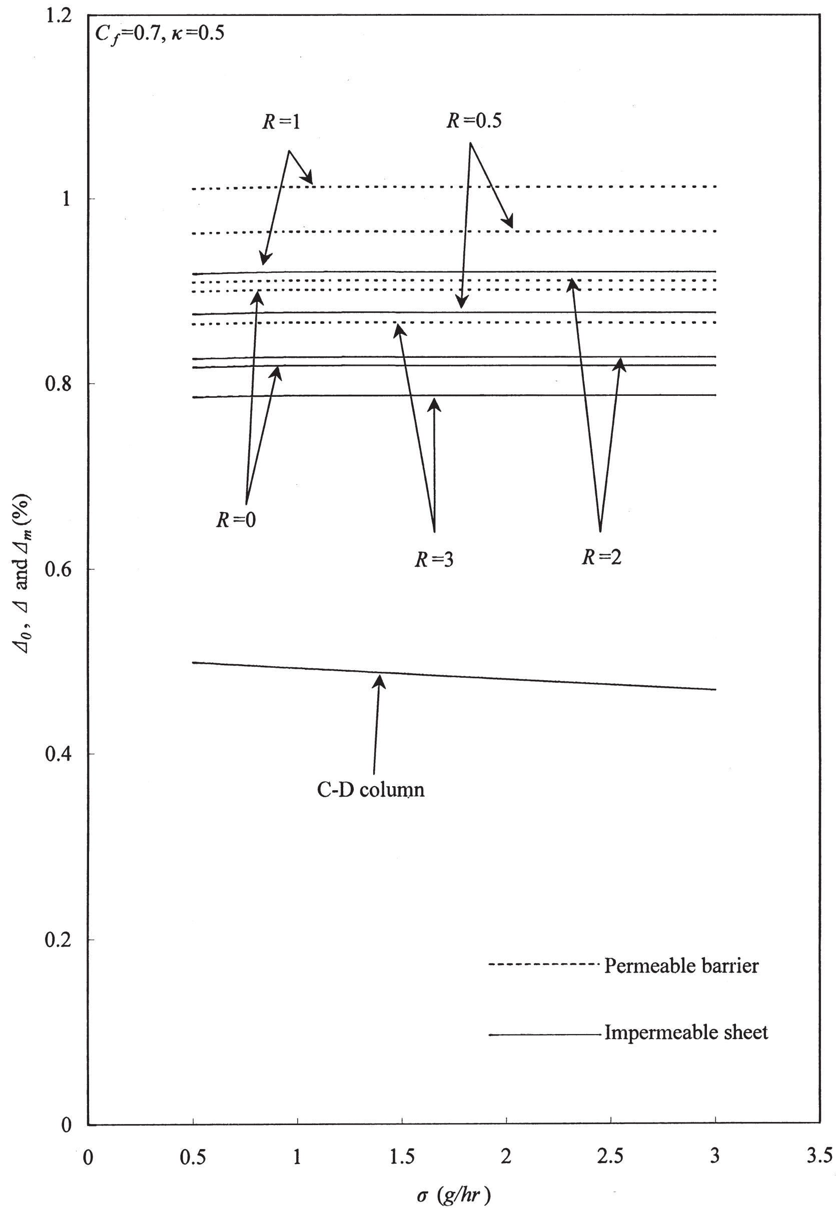

Figure 3. The degree of separation of three devices vs. flow rate with reflux ratio as a parameter; $C_\mathrm{f} = 0.7$, $\kappa = 1/2$.

Eqs. (27) and (28) are given as follows:

$$-\frac{\partial C_{\mathrm{Ae}}}{\partial \eta_{\mathrm{A}}}+\frac{\alpha C_{\mathrm{Ae}}\hat{C}_{\mathrm{Ae}}h_1 W_{\mathrm{A}}}{\tilde{T}}=0 \quad \text{at } \eta_{\mathrm{A}}=1 \tag{27}$$

$$\frac{\partial C_{\mathrm{Be}}}{\partial \eta_{\mathrm{B}}}+\frac{\alpha C_{\mathrm{Be}}\hat{C}_{\mathrm{Be}}h_1 W_{\mathrm{B}}}{\tilde{T}}=0 \quad \text{at } \eta_{\mathrm{B}}=1 \tag{28}$$

The mathematical treatment is similar to that in the previous section, except the eigenvalues calculated for each subchannel individually. The degree of separation (Δ_m) for whole column with an impermeable sheet inserted was also obtained in terms of the eigenvalues ($\lambda_{\mathrm{Ae},m}$, $\lambda_{\mathrm{As},m}$, $\lambda_{\mathrm{Be},m}$, and $\lambda_{\mathrm{Bs},m}$), expansion coefficients ($S_{\mathrm{Ae},m}$, $S_{\mathrm{Be},m}$, $S_{\mathrm{As},m}$, and $S_{\mathrm{Bs},m}$), location of permeable barrier (κ) and eigenfunctions ($F_{\mathrm{Ae},m}$, $F_{\mathrm{Be},m}$, $F_{\mathrm{As},m}$, and $F_{\mathrm{Bs},m}$) are given as follows:

$$\begin{aligned}\Delta_m &= C_{\mathrm{t}}-C_{\mathrm{b}}\\ &=\left(\frac{LD\sum_{m=0}^{\infty}\left[\frac{S_{\mathrm{Ae},m}F'_{\mathrm{Ae},m}(1)}{\lambda_{\mathrm{Ae},m}}\right][1-\exp(-\lambda_{\mathrm{Ae},m})]}{(\kappa W)^2\int_0^1 V_{\mathrm{Ae}}\,\mathrm{d}\eta_{\mathrm{A}}}+\frac{\int_0^1 V_{\mathrm{Ae}}C_{\mathrm{Ae}}(\eta_{\mathrm{A}},0)\,\mathrm{d}\eta_{\mathrm{A}}}{\int_0^1 V_{\mathrm{Ae}}\,\mathrm{d}\eta_{\mathrm{A}}}\right)\\ &\quad-\left(\frac{\int_0^1 V_{\mathrm{As},m}C_{\mathrm{As}}(\eta_{\mathrm{A}},0)\,\mathrm{d}\eta_{\mathrm{A}}}{\int_0^1 V_{\mathrm{As}}\,\mathrm{d}\eta_{\mathrm{A}}}-\frac{LD\sum_{m=0}^{\infty}\left[\frac{S_{\mathrm{As},m}F'_{\mathrm{As},m}(1)}{\lambda_{\mathrm{As},m}}\right][\exp(\lambda_{\mathrm{As},m})-1]}{(\kappa W)^2\int_0^1 V_{\mathrm{As}}\,\mathrm{d}\eta_{\mathrm{A}}}\right)\end{aligned} \tag{29}$$

or

$$\begin{aligned}\Delta_m &= C_{\mathrm{t}}-C_{\mathrm{b}}\\ &=\left(\frac{LD\sum_{m=0}^{\infty}\left[\frac{S_{\mathrm{Be},m}F'_{\mathrm{Be},m}(1)}{\lambda_{\mathrm{Be},m}}\right][1-\exp(-\lambda_{\mathrm{Be},m})]}{(1-\kappa)^2W^2\int_0^1 V_{\mathrm{Be}}\,\mathrm{d}\eta_{\mathrm{B}}}+\frac{\int_0^1 V_{\mathrm{Be}}C_{\mathrm{Be}}(\eta_{\mathrm{B}},0)\,\mathrm{d}\eta_{\mathrm{B}}}{\int_0^1 V_{\mathrm{Be}}\,\mathrm{d}\eta_{\mathrm{B}}}\right)\\ &\quad-\left(\frac{\int_0^1 V_{\mathrm{Bs},m}C_{\mathrm{Bs}}(\eta_{\mathrm{B}},0)\,\mathrm{d}\eta_{\mathrm{B}}}{\int_0^1 V_{\mathrm{Bs}}\,\mathrm{d}\eta_{\mathrm{B}}}-\frac{LD\sum_{m=0}^{\infty}\left[\frac{S_{\mathrm{Bs},m}F'_{\mathrm{Bs},m}(1)}{\lambda_{\mathrm{As},m}}\right][\exp(\lambda_{\mathrm{As},m})-1]}{(1-\kappa)^2W^2\int_0^1 V_{\mathrm{Bs}}\,\mathrm{d}\eta_{\mathrm{B}}}\right)\end{aligned} \tag{30}$$

Figure 3 is the graphical representation of the degree of separation (Δ_m) for the whole column.

IMPROVEMENT OF SEPARATION

The improvement of separation, I and I_m, for such devices by inserting a permeable barrier and by inserting an impermeable sheet, respectively, at the optimal condition are best illustrated by calculating the percentage increase in the degree of separation, based on the degree of separation of the open column as

$$I = \frac{\Delta - \Delta_0}{\Delta_0} \tag{31}$$

and

$$I_m = \frac{\Delta_m - \Delta_0}{\Delta_0} \tag{32}$$

A numerical example for the separation of water isotopes is given as follows. Some equipment parameters and physical properties of the mixture were found[33] as:

$$B = 10.12\,\text{cm}, \quad g = 980\,\text{cm/sec}^2, \quad W_A + W_B = 0.08\,\text{cm},$$

$$\Delta T = 30.5\text{K}, \quad \tilde{T} = 303.25\text{K}, \quad \alpha = -0.0184, \quad L = 144\,\text{cm},$$

$$\mu = 1.0\,\text{g/cm sec}, \quad K_{eq} = 3.8, \quad \delta = 0.02\,\text{cm},$$

$$\beta = 5 \times 10^{-3}\,\text{g/cm}^3\,K, \quad \varepsilon = 0.378, \quad \rho = 1.0\,\text{g/cm}^3,$$

$$D = 3.9 \times 10^{-5}\,\text{cm}^2/\text{sec}, \quad k_f = 0.00132\,\text{cal/cm sec K},$$

$$k = 3.8 \times 10^{-2}\,\text{cal/cm sec K}$$

Some results are presented in Tables 1 and 2 for the device by inserting a permeable barrier and by inserting an impermeable sheet, respectively.

RESULTS AND DISCUSSION

The Degree of Separation in the Devices with External Refluxes

Tables 3 and 4 show some calculation results of the first two eigenvalues and their associated expansion coefficients, as well as the degree of separation with $\kappa = 1/2$, $\sigma = 1.0\,\text{g/hr}$ and $C_f = 0.1$, for the devices by inserting a permeable barrier and by inserting an impermeable sheet, respectively. It was observed that due to the rapid convergence, only the first negative eigenvalue is necessary to be considered during the calculation of the degree of separation. Figure 3 shows, respectively, the degree of separation Δ and Δ_m vs. mass flow rate for $C_f = 0.7$ with the reflux ratio as a parameter for $\kappa = 1/2$ while Figs. 4 and 5 show the degree of separation Δ and Δ_m vs. the ratio of channel thickness κ for $C_f = 0.7$ and $C_f = 0.1$ with the reflux ratio as a parameter for $\sigma = 1.0\,\text{g/hr}$. It

Table 1. The Improvement of the Degree of Separation by Inserting a Permeable Barrier with Reflux Ratio and Barrier Position as Parameters; $\sigma = 1.0\,g/hr$

	I (%)													
	$C_f = 0.7$							$C_f = 0.1$						
R	$\kappa = 1/8$	$\kappa = 1/4$	$\kappa = 3/8$	$\kappa = 1/2$	$\kappa = 5/8$	$\kappa = 3/4$	$\kappa = 7/8$	$\kappa = 1/8$	$\kappa = 1/4$	$\kappa = 3/8$	$\kappa = 1/2$	$\kappa = 5/8$	$\kappa = 3/4$	$\kappa = 7/8$
0	63.59	137.96	113.00	66.25	199.41	248.95	138.65	63.59	137.95	113.62	66.25	199.39	248.93	138.64
0.5	75.04	154.61	127.91	77.89	220.37	273.38	155.35	47.23	114.15	92.26	49.62	169.45	214.03	114.77
1.0	83.79	167.34	139.30	86.78	236.39	292.05	168.12	32.51	92.74	73.03	34.66	142.51	182.63	93.30
2.0	65.41	140.61	115.37	68.10	202.75	252.84	141.31	25.88	83.10	64.38	27.93	130.38	168.50	83.63
3.0	57.14	128.58	104.61	59.70	187.62	235.20	129.24	19.59	73.95	56.16	21.53	118.86	155.07	74.45

Table 2. The Improvement of the Degree of Separation by Inserting an Impermeable Sheet with Reflux Ratio and Sheet Position as Parameters; $\sigma = 1.0\,g/hr$

	I_m (%)													
	$C_f = 0.7$							$C_f = 0.1$						
R	$\kappa = 1/8$	$\kappa = 1/4$	$\kappa = 3/8$	$\kappa = 1/2$	$\kappa = 5/8$	$\kappa = 3/4$	$\kappa = 7/8$	$\kappa = 1/8$	$\kappa = 1/4$	$\kappa = 3/8$	$\kappa = 1/2$	$\kappa = 5/8$	$\kappa = 3/4$	$\kappa = 7/8$
0	78.31	152.23	82.49	82.88	214.39	262.91	152.97	78.31	152.22	130.71	214.36	217.35	262.88	152.96
0.5	90.79	169.89	95.27	95.68	236.39	288.31	170.67	60.48	127.00	107.64	182.93	153.88	226.59	127.66
1.0	100.33	183.38	105.03	105.46	253.21	307.73	184.21	44.43	104.30	86.87	154.63	115.80	193.93	104.89
2.0	80.30	155.05	84.53	84.91	217.89	266.96	155.79	37.21	94.09	77.53	141.90	94.22	179.24	94.65
3.0	71.29	142.29	75.30	75.67	202.00	248.61	143.00	30.35	84.38	68.65	129.81	84.51	165.28	84.92

Table 3. Eigenvalues and Expansion Coefficients as Well as the Degree of Separation with External Refluxes of the Device by Inserting a Permeable Barrier for $\kappa = 1/2$ and $C_f = 0.1$

R	m	λ_e	λ_s	$S_{Ae,m}$	$S_{Be,m}$	$S_{As,m}$	$S_{Bs,m}$	$\Delta(\lambda_{e,1}, \lambda_{s,1})$ (%)	$\Delta(\lambda_{e,1}, \lambda_{e,2}, \lambda_{s,1}\lambda_{s,2})$ (%)
0.5	1	−3.59	3.59	-3.27×10^{-3}	8.92×10^{-3}	-8.68×10^{-3}	3.38×10^{-3}	0.4931	0.4931
	2	−28.32	28.32	4.58×10^{-7}	1.11×10^{-7}	-1.08×10^{-7}	-4.68×10^{-7}		
1.0	1	−2.03	2.03	-4.12×10^{-3}	7.56×10^{-3}	-7.48×10^{-3}	4.06×10^{-3}	0.4438	0.4441
	2	−21.09	21.09	3.98×10^{-7}	9.59×10^{-8}	-9.41×10^{-8}	-3.97×10^{-7}		
2.0	1	−1.13	1.13	-4.43×10^{-3}	6.48×10^{-3}	-6.32×10^{-3}	4.37×10^{-3}	0.4216	0.4226
	2	−13.93	13.93	2.91×10^{-7}	8.62×10^{-8}	-8.52×10^{-8}	-2.88×10^{-7}		
3.0	1	−0.79	0.79	-4.68×10^{-3}	6.12×10^{-3}	-6.01×10^{-3}	4.68×10^{-3}	0.4005	0.4006
	2	−10.37	10.37	2.26×10^{-7}	8.20×10^{-8}	-7.92×10^{-8}	-2.28×10^{-7}		

Table 4. Eigenvalues and Expansion Coefficients as Well as the Degree of Separation with External Refluxes of the Device by Inserting an Impermeable Sheet for $\kappa = 1/2$ and $C_f = 0.1$

R	m	$\lambda_{Ae,m}$	$\lambda_{Be,m}$	$S_{Ae,m}$	$S_{Be,m}$	$\lambda_{As,m}$	$\lambda_{Bs,m}$	$S_{As,m}$	$S_{Bs,m}$	$\Delta_m(\lambda_{Ae,1},\lambda_{As,1},\lambda_{Be,1},\lambda_{Bs,1})$ (%)	$\Delta_m(\lambda_{Ae,1},\lambda_{As,1},\lambda_{Ae,2},\lambda_{As,2},\lambda_{Be,1},\lambda_{Bs,1},\lambda_{Be,2},\lambda_{Bs,2})$ (%)
0.5	1	−25.94	−77.19	8.28×10^{-6}	2.11×10^{-6}	25.94	77.19	2.11×10^{-6}	8.28×10^{-6}	0.4482	0.4482
	2	−53.01	−162.32	5.17×10^{-11}	1.32×10^{-10}	53.01	162.32	1.32×10^{-10}	5.17×10^{-11}		
1.0	1	−12.97	−19.30	-7.16×10^{-6}	-5.51×10^{-6}	12.97	19.30	-5.51×10^{-6}	-7.16×10^{-6}	0.4034	0.4035
	2	−39.88	−81.21	-4.43×10^{-11}	-3.42×10^{-10}	39.88	81.21	-3.42×10^{-10}	-4.43×10^{-11}		
2.0	1	−12.97	−19.30	8.28×10^{-11}	9.83×10^{-6}	12.97	19.30	9.83×10^{-6}	8.28×10^{-6}	0.3833	0.3834
	2	−26.61	−40.62	5.15×10^{-11}	6.12×10^{-10}	26.61	40.62	6.12×10^{-10}	5.15×10^{-11}		
3.0	1	−9.73	−12.87	-0.95×10^{-6}	-5.52×10^{-6}	9.73	12.87	-5.52×10^{-6}	-0.95×10^{-6}	0.3641	0.3641
	2	−19.87	−27.07	-6.18×10^{-11}	-3.42×10^{-10}	19.87	27.07	-3.42×10^{-10}	-6.18×10^{-11}		

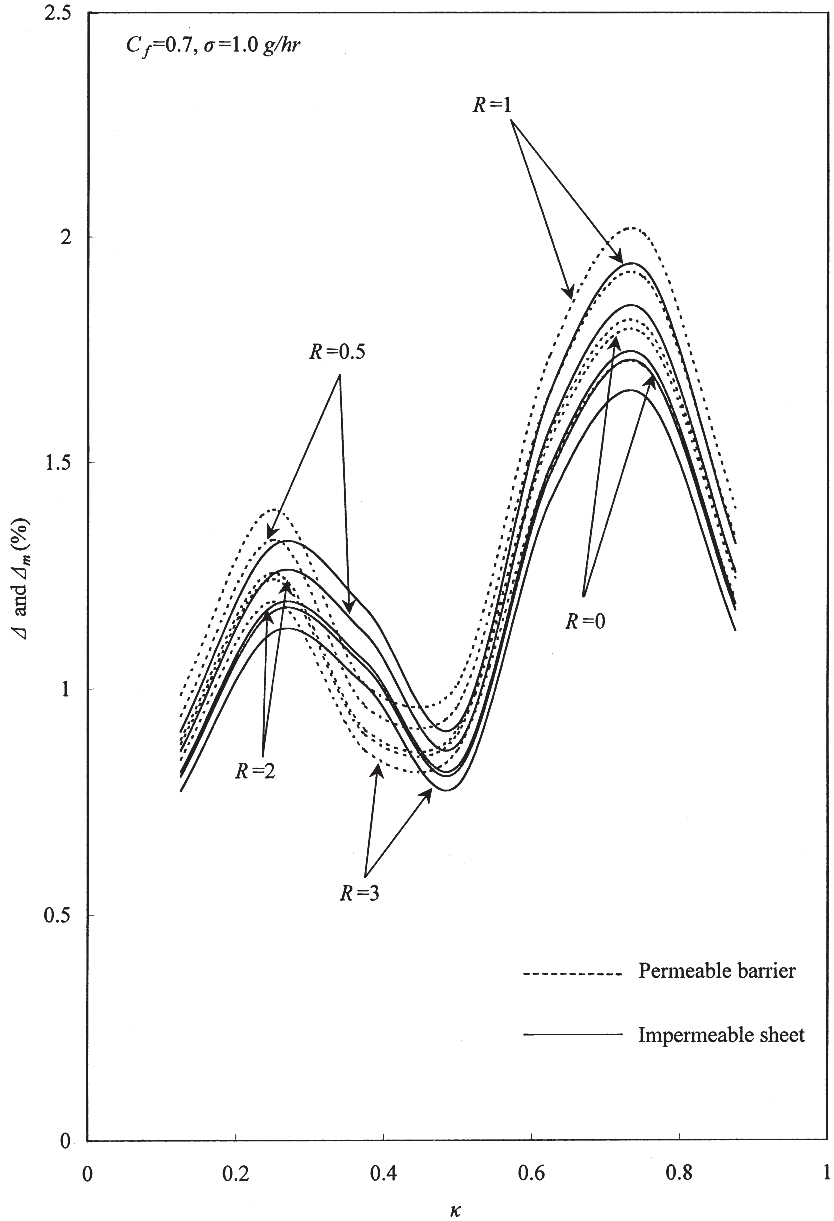

Figure 4. Effect of barrier position or sheet position on the degree of separation with reflux ratio as a parameter; $C_f = 0.7$, $\sigma = 1.0\,g/hr$.

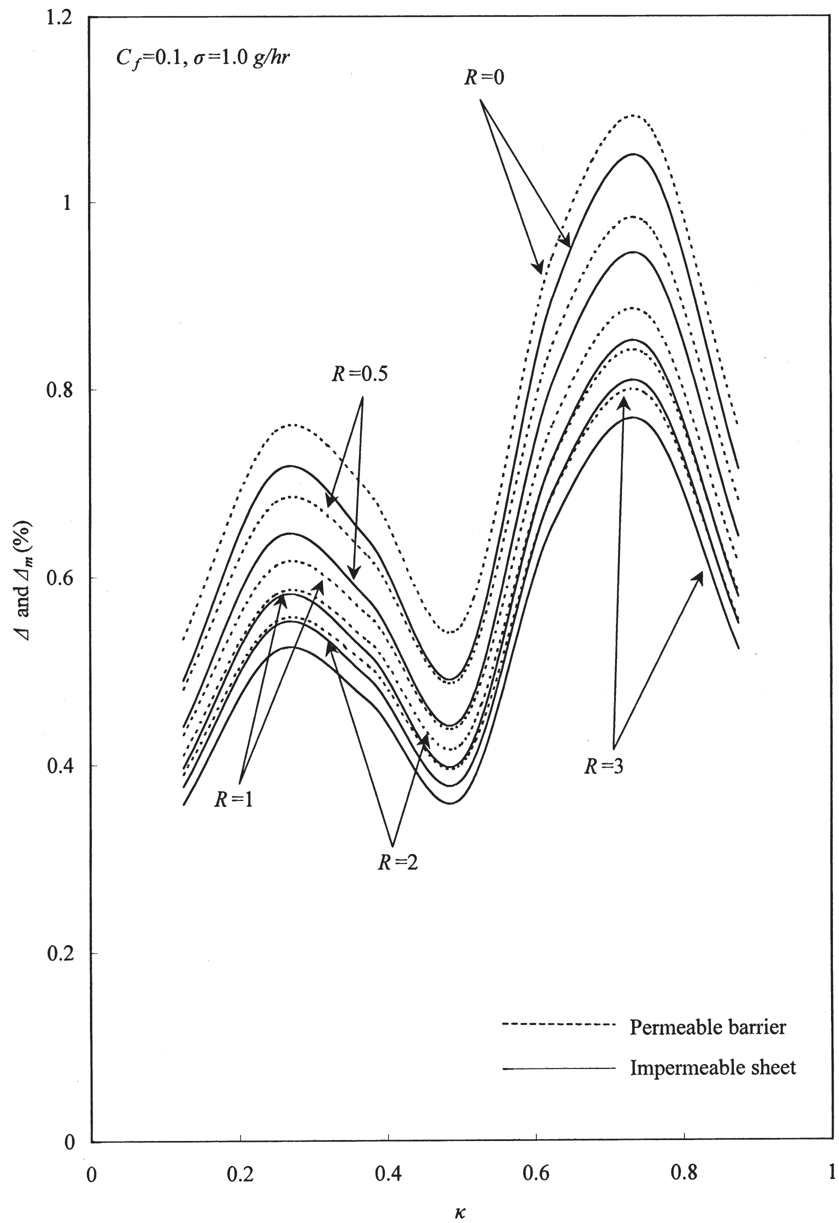

Figure 5. Effect of barrier position or sheet position on the degree of separation with reflux ratio as a parameter; $C_f = 0.1$, $\sigma = 1.0\,g/hr$.

is shown in Figs. 3–5 that the maximum degree of separation for both devices by inserting a permeable barrier and an impermeable sheet is obtained at $R = 1.0$ for $C_\mathrm{f} = 0.7$ while the degree of separation for both devices decreases with reflux ratio for $C_\mathrm{f} = 0.1$ and with κ going away from two peaks, say $\kappa = 1/4$ and $\kappa = 3/4$. Although the recycle effect has positive influences on the degree of separation for the device by inserting a permeable barrier or by inserting an impermeable sheet, the remixing effect by increasing the reflux ratio cannot compensate for the decrease of the concentration product, $C\hat{C}$, as shown in Fig. 2, and hence the degree of separation decreases with increasing reflux ratio for concentration being less than $C_\mathrm{f} \approx 0.44$ or as the feed concentration is larger than $C_\mathrm{f} \approx 0.44$ for large reflux ratio, say $R > 1$ for $C_\mathrm{f} = 0.7$, the improvement of the degree of separation decreases in the devices with external refluxes. It was also found in Fig. 5 that for a fixed reflux ratio, the degree of separation decreases slightly with increasing feed flow rate for both devices with external refluxes.

The improvement of the degree of separation is defined by Eqs. (31) and (32), so the higher improvement of performance is really obtained by employing the device with a permeable barrier inserted, instead of using an impermeable sheet. It is also seen from Fig. 5 that the difference $(\Delta - \Delta_m)$ of the degree of separation decreases with reflux ratio. Since the position has much influence on the separation, the first eigenvalues for the enriching and stripping sections at various barrier and sheet positions are calculated and presented in Figs. 6 and 7 with R as a parameter for the devices by inserting a permeable barrier and by inserting an impermeable sheet, respectively.

Improvement in the Degree of Separation in An Improved Column

Figures 3–5 show that the degree of separation of the device by inserting a permeable barrier is larger than that of the device by inserting an impermeable sheet. It is found in Figs. 3–5 that all Δ and Δ_m increase as κ goes away from 1/2, especially for $\kappa > 1/2$ while Δ and Δ_m increase with decreasing reflux ratio for $C_\mathrm{f} = 0.1$ or decrease as the reflux ratio goes away from $R = 1$ for $C_\mathrm{f} = 0.7$, especially for $R > 1$. The improvements of the degree of separation, I and I_m, in the devices with external refluxes are shown in Tables 1 and 2 with the reflux ratio and the ratio of thickness κ as parameters for $C_\mathrm{f} = 0.1$ and $C_\mathrm{f} = 0.7$. It is noted that the improvements of the degree of separation, I and I_m, increase when κ goes away from 1/2, especially for $\kappa > 1/2$.

CONCLUSION

The theoretical study of the separation efficiency for the enrichment of heavy water in continuous thermal-diffusion columns has been investigated in the present

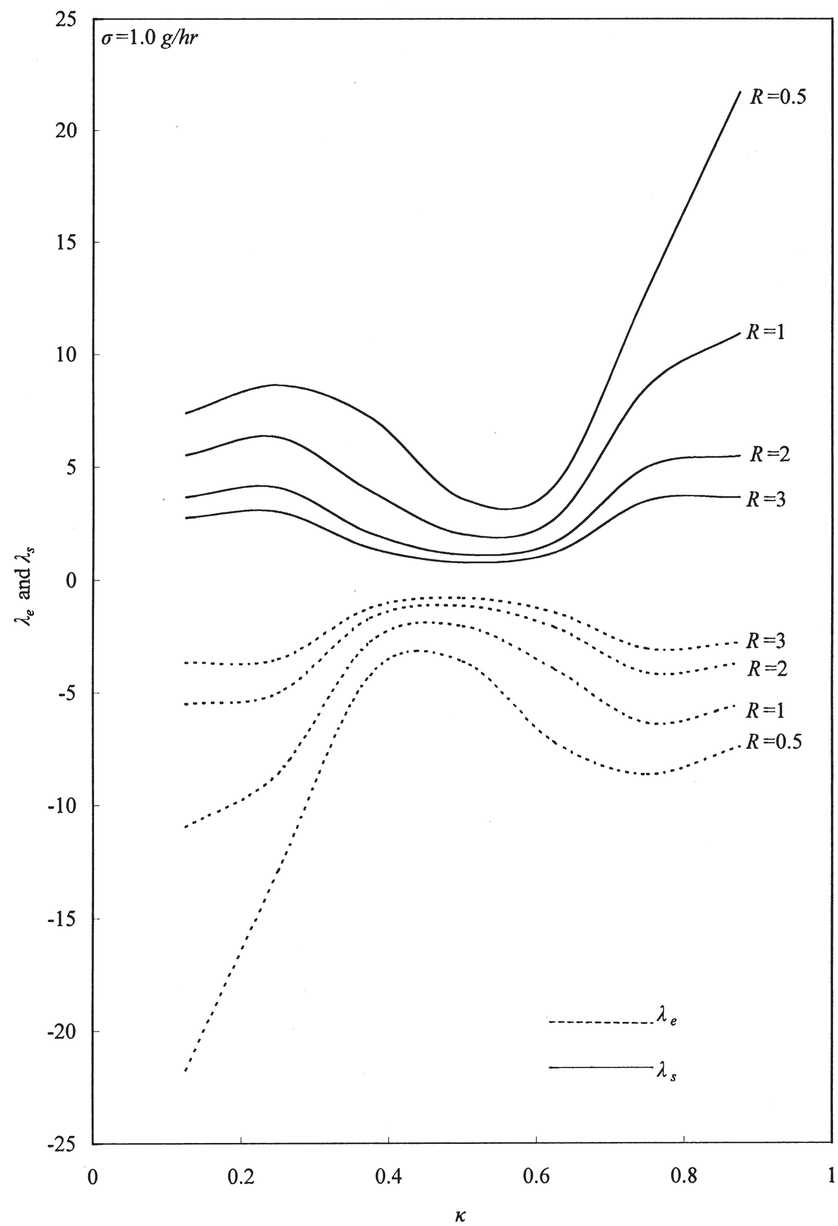

Figure 6. First eigenvalues at various barrier positions with reflux ratio as a parameter for the device with a permeable barrier inserted; $\sigma = 1.0\,\mathrm{g/hr}$.

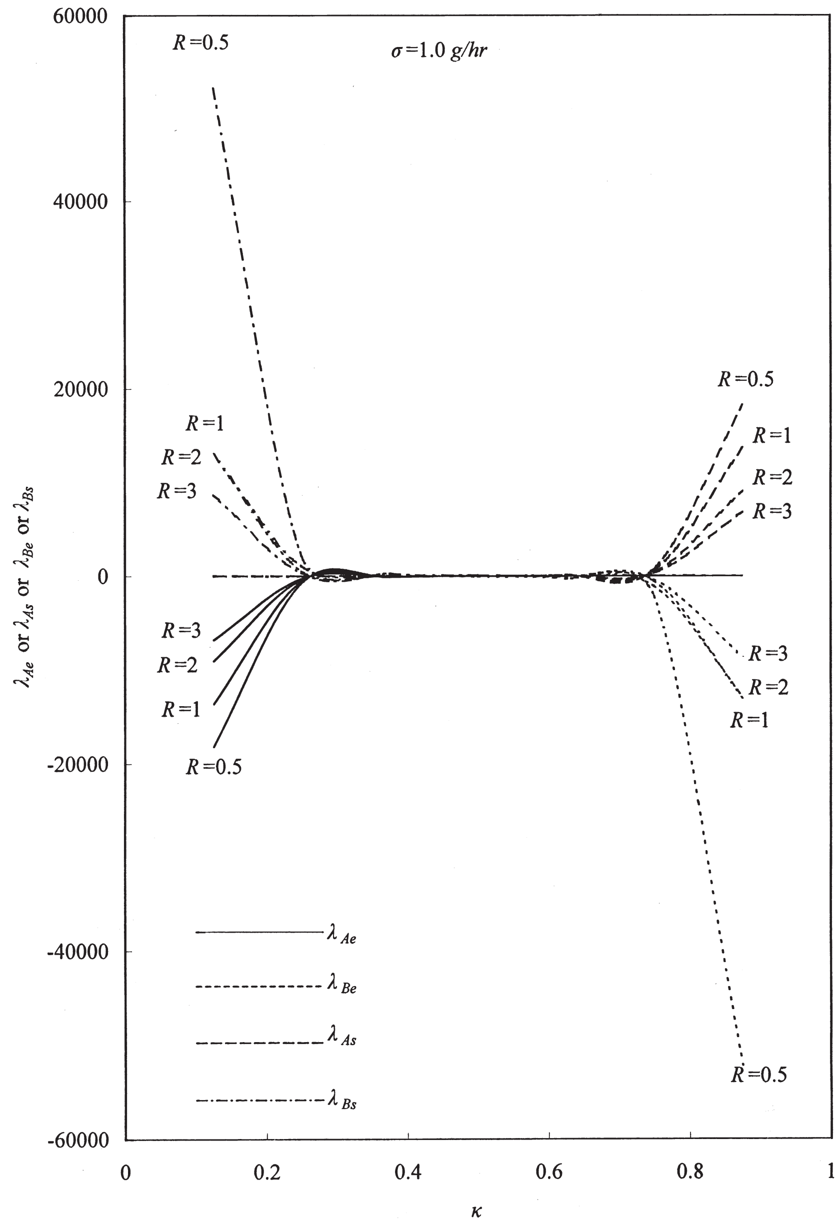

Figure 7. First eigenvalues at various sheet positions with reflux ratio as a parameter for the device with an impermeable barrier inserted; $\sigma = 1.0\,\mathrm{g/hr}$.

study. The methods for improving the performance in the degree of separation are either with external refluxes by inserting a permeable barrier or by inserting an impermeable sheet with external refluxes. The theoretical values of the degree of separation for various ratios of thickness and reflux ratio were calculated from Eqs. (26) and (30) [or Eqs. (27) and (31)] for the devices by inserting a permeable barrier and by inserting an impermeable sheet, respectively, by using the given transport coefficients and equilibrium constant. Some graphical representations calculated from Eqs. (26) and (30) [or Eqs. (27) and (31)] are given in Figs. 3–5 as well as in Tables 1 and 2. Considerable improvement of the degree of separation in the devices by inserting either a permeable barrier or an impermeable sheet is obtained; this is the value of the present study in designing continuous-type thermal-diffusion columns with external refluxes. The most important assumption in this work is the pseudo-product form of the concentration [Eq. (7)] was considered as constant in accordance with the small degree of separation for the enrichment of water isotopes in a thermal-diffusion column as well as in other separation devices. The effects of reflux ratio and ratio of thickness on the degree of separation for the devices by inserting a permeable barrier and by inserting an impermeable sheet are illustrated in Tables 1 and 2, respectively. The feed concentration and feed rate are also the parameters of the analytical calculations, as shown in Fig. 3 and Tables 1 and 2.

The design of recycle devices with a permeable barrier or an impermeable sheet inserted will produce two conflicting effects on the convective flow: the desirable cascading effect and the undesirable remixing effect, and hence the analytical method could easily predict and adjust some operating parameters on the degree of separation. The suitable adjustment of the ratio of channel thickness by inserting a permeable barrier or an impermeable sheet can effectively restrain the undesirable remixing effect and conserve the desirable cascading effect, and hence thereby lead to improved separation. This is the value of the present study. It is concluded that suitable adjustment of the ratio of thickness, κ, can effectively enhance the degree of separation for water isotopes in a continuous-type thermal-diffusion column under recycle-effect devices by inserting a permeable barrier and by inserting an impermeable sheet with κ going away from 1/2, especially for $\kappa > 1/2$.

APPENDIX A

$$F_{\mathrm{Ae},m}(\eta_{\mathrm{A}}) = \sum_{n=0}^{\infty} d_{mn}\eta_{\mathrm{A}}^{n}, \quad d_{m0} = 1\,(\text{selected}), \;\; d_{m1} = 0 \tag{A1}$$

$$F_{\mathrm{Be},m}(\eta_{\mathrm{B}}) = \sum_{n=0}^{\infty} e_{mn}\eta_{\mathrm{B}}^{n}, \quad e_{m0} = 1\,(\text{selected}), \;\; e_{m1} = 0 \tag{A2}$$

in which

$$d_{m2} = 0, \quad d_{m3} = \frac{\lambda_e W_A^2}{6LD} a_1, \quad d_{m4} = \frac{\lambda_e W_{A>}^2}{12LD} a_2,$$

$$d_{m5} = \frac{\lambda_e W_A^2}{20LD} a_3,$$

$$d_{m(n+2)} = \frac{\lambda_e W_A^2}{(n+2)(n+1)LD} [a_1 d_{m(n-1)} + a_2 d_{m(n-2)} + a_3 d_{m(n-3)}], \quad n \geq 4 \tag{A3}$$

$$e_{m2} = 0, \quad e_{m3} = \frac{\lambda_e W_B^2}{6LD} b_1, \quad e_{m4} = \frac{\lambda_e W_B^2}{12LD} b_2,$$

$$e_{m5} = \frac{\lambda_e W_B^2}{20LD} b_3,$$

$$e_{m(n+2)} = \frac{\lambda_e W_B^2}{(n+2)(n+1)LD} [b_1 e_{m(n-1)} + b_2 e_{m(n-2)} + b_3 e_{m(n-3)}], \quad n \geq 4 \tag{A4}$$

where $a_1 = -f_{1,e} - f_{2,e} + (1+R)f_{3,e}$, $a_2 = f_{2,e} - (1+R)f_{3,e}$, $a_3 = f_{1,e}$, $b_1 = g_{1,e} + g_{2,e} + Rg_{3,e}$, $b_2 = -g_{2,e} - Rg_{3,e}$, and $b_3 = -g_{1,e}$. Likewise, the eigenfunctions $F_{As,m}$ and $F_{Bs,m}$ in the stripping section were obtained with the subscript e being replaced by s in Eqs. (A1)–(A4).

NOMENCLATURE

B	column width (cm)
C	fraction concentration of D_2O in H_2O–HDO–D_2O system (–)
D	ordinary diffusion coefficient (cm^2/sec)
d_{mn}	coefficient in the eigenfuction F_m for region A (–)
e_{mn}	coefficient in the eigenfuction F_m for region B (–)
F_m	eigenfuction associated with eigenvalue λ_m (–)

$C\hat{C}$	pseudo-product form of concentration for D_2O defined by Eq. (7) (–)
F_e, F_s	appropriate values of $C\hat{C}$ in enriching section, in stripping section (–)
f_1, f_2, f_3	constants defined in the velocity distribution of region A (–)
G_m	function defined during the use of orthogonal expansion method (–)
g	gravitational acceleration (cm/sec^2)
g_1, g_2, g_3	constants defined in the velocity distribution of region B (–)
H	transport coefficient defined by Eq. (5) (g/sec)
I, I_m	improvement of the degree of separation defined by Eqs. (31) and (32) (–)
J_x	mass flux of component 1 in the x direction (g/cm^2 sec)
K	transport coefficient defined by Eq. (6) (g/sec cm)
K_{eq}	mass-fraction equilibrium constant of H_2O–HDO–D_2O system (–)
k, k_f	thermal conductivity of the barrier and the fluid, respectively (cal/cm sec K)
L	one-half of column length (cm)
L'	dimensionless coordinate defined by Eq. (4) (–)
q_m	ratio of expansion coefficients associated with eigenvalue λ_m (–)
R	reflux ratio at both ends of the column (–)
S_m	expansion coefficient associated with eigenvalue λ_m (–)
$\bar{T}$	reference temperature evaluated by Eq. (10) (K)
$\tilde{T}$	reference temperature evaluated by Eq. (15) (K)
T_1, T_2	temperatures of the cold and hot plates, respectively (K)
ΔT	difference in temperature of hot and cold surfaces (K)
V	velocity distribution of fluid in the vertical direction (cm/sec)
W	thickness of the region (cm)
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 (–)
β	$\partial\rho/\partial T$ evaluated at reference temperature (g/cm^3 K)
Δ	degree of separation, $C_B - C_T$ (–)
Δ_e, Δ_s	$C_B - C_F$, $C_F - C_T$ (–)
δ	thickness of the barrier (cm)
ε	permeability of the barrier (–)
ζ	dimensionless coordinate in the vertical direction, defined by Eq. (15) (–)

η	dimensionless coordinate in the horizontal direction, defined by Eq. (15) (–)
θ	constant defined by $\alpha C(1 - C)/\tilde{T}$
λ_m	eigenvalue (–)
μ	viscosity of fluid (g/cm sec)
ρ	density of fluid (g/cm^3)
σ	mass flow rate of top or bottom product (g/hr)
σ'	dimensionless mass flow rate defined by Eq. (4) (–)

Subscripts

A	in the channel A
B	in the channel B
b	at end of the enriching section
e	in the enriching section
f	of feed stream
t	C at end of stripping section
s	in the stripping section

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