THE INFLUENCE OF THE INCLINED ANGLE ON THE HEAVY WATER ENRICHMENT IN DOUBLE-PASS THERMAL-DIFFUSION COLUMNS WITH EXTERNAL RECYCLE

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ABSTRACT

The enhancement of the heavy water separation efficiency in double-flow thermal diffusion columns under inclined angle external refluxes at the both ends has been investigated analytically. The analytical solution is obtained by using the separation variable with an orthogonal expansion technique in terms of power series. The theoretical predictions of separation efficiencies are compared with those of Clusius-Dickel columns of the same working dimension. The recycle ratio, impermeable-sheet (or permeable-barrier) position and inclination angle are significant design parameters, resulting in substantial separation efficiency improvement, compared with that in single-flow operations (without inserting an impermeable sheet or permeable barrier).

KEYWORDS

Water isotopes; Orthogonal expansion techniques; Inclined Angle; Thermal diffusion.

INTRODUCTION

The transport phenomenon of thermal diffusion was studied from the kinetic theory of a gas mixture and was later demonstrated experimentally (Chapman and Dootson, 1917). In static systems were used in the study on the thermal-diffusion effect, in which the temperature gradient was established in such a behavior that mass convection was eliminated by the concentration gradient flux due to ordinary diffusion and resulting in no net bulk flow. These improved design include the inclined column (Yeh and Ward, 1971) wired column (Yeh and Ward, 1971) inclined moving-wall columns (Sullivan *et. al*., 1955a; Yeh and Tsai, 1972), rotary columns (Yeh and Tsai, 1981a; Yeh and Tsai, 1982), packed columns (Lorenz and Emery, 1959; Yeh and Chu, 1974) rotary wired columns (Yeh and Ho, 1975; Yeh and Tsai, 1981b), permeable barrier columns (Yeh *et. al*., 1986a & 1986b) and impermeable barrier columns (Tsai and Yeh, 1986c).

The introduction of recycle-effect concept in double-flow operations with external refluxes in parallel-plate mass exchangers enhances mass-transfer coefficient, resulting in improved device performance (Ho *et al*., 2001 & 2002). The purpose of this study is to consider various design parameters that influence the improved Clusius-Dickel column performance, such as inserting an impermeable sheet or a permeable barrier with inclination angle. The theoretical formulation of such conjugated Graetz problems were solved using the orthogonal expansion technique (Treacy and Rich, 1955b; Yeh *et. al*., 1986a; Ebadian and Zhang, 1989) with the orthogonality conditions to obtain the analytical results and to investigate the effects of various operating and design parameters on the device performance.

THEORETICAL FORMULATION

(1) Inclined Clusius-Dickel Column

The transport equation for heavy water separation enrichment in continuous Clusius-Dickel column has developed in our previous work (Ho *et al*., 2002), the separation equations for inclined column were obtained

$$\Delta_0 = C_b - C_t = (F_e + F_s)(1 - e^{-\sigma' L'/2}) / \sigma' \tag{1}$$

where transport constants and the dimensionless variables were defined as

$$H' = \frac{\alpha\beta\rho g \cos\theta (2\omega)^3 (\Delta T)^2}{6!\,\mu \overline{T}} \tag{2}$$

$$K' = \frac{\rho g^2 \cos^2\theta \beta^2 W^7 B (\Delta T)^2}{9!\,\mu^2 D} \tag{3}$$

and

$$\sigma' = \frac{\sigma}{(-H')}, \quad L' = \frac{L(-H')}{K'}. \tag{4}$$

The pseudo concentration products defined as

$$C\hat{C} = C\{0.05263 - (0.05263 - 0.0135K_{eq})C - 0.027\{[1 - (1 - \frac{K_{eq}}{4})C]CK_{eq}\}^{\frac{1}{2}}\} \tag{5}$$

in which

$$F_e (= C_e\hat{C}_e = \frac{1}{C_B - C_F}\int_{C_F}^{C_B} C_e\hat{C}_e dC_e) \tag{6}$$

$$F_s (= C_s\hat{C}_s = \frac{1}{C_F - C_T}\int_{C_T}^{C_F} C_s\hat{C}_s dC_s) \tag{7}$$

$$K_{eq} = \frac{C_2^2}{C_1 C_3} = \frac{[\mathrm{HDO}]^2}{[\mathrm{H_2O}][\mathrm{D_2O}]} \times \frac{19\times 19}{18\times 20} \tag{8}$$

(2) Inclined Double-Flow Clusius-Dickel Column With External Refluxes

A new design of continuous-type thermal diffusion columns inserts an impermeable sheet or a permeable barrier between the cold and hot plates, as shown in Figures 1 and 2. The channel thicknesses of two sections are W_A and W_B, respectively. And feed at the center of the column and output products from both ends.

In obtaining the theoretical formulation, the following assumptions were made: (a) heat transfer between the space of the hot and cold plates by conduction only; (b) purely laminar flow of the fluid in the regions; (c) the influences of ordinary and thermal diffusions, end effects, and inertia terms on the velocity are neglected; (d) ordinary diffusion in the vertical direction and bulk flow in the horizontal direction are neglected; (e) constant fluxes because the thermal diffusion (i.e. $\alpha C\hat{C}/\tilde{T}$, $\alpha C_{Ae}\hat{C}_{Ae}/\tilde{T}$ and $\alpha C_{Be}\hat{C}_{Be}/\tilde{T}$ were regarded as some constant, say θ). After the following dimensionless variables for the inclined column with inserting an impermeable sheet or permeable barrier in the enriching section are introduced:

$$\eta_A = \frac{x_A}{W_A},\ \ \eta_B = \frac{x_B}{W_B},\ \ \varsigma = \frac{z}{L},\ \ \tilde{T} = \frac{2W_A{}^3T_1 + 2W_B{}^3T_2 - h_1(W_A{}^4 - W_B{}^2)}{2(W_A{}^3 + W_B{}^2)},$$

$$h_1 = \frac{\Delta T}{W_A + W_B + \dfrac{k_f\delta}{k_f\varepsilon + k(1-\varepsilon)}},\ \ h_2 = \frac{h_1 k_f}{k_f\varepsilon + k(1-\varepsilon)},\ \ \kappa = W_A/W,\ \ R = \frac{\sigma_R}{\sigma} \tag{9}$$

Since the space between the plates is small, the heat transfer is determined only by conduction. According, the temperature distribution in the fluid is linear and the temperature gradient in the various column regions is h_1. During the derivation of the h_1, we assumed that the thermal resistance in the barrier was composed of the resistance of the fluid and the barrier in parallel. Hence the temperature gradient of the fluid within the barrier is h_2.

The velocity distribution from the Navier-Stokes equation as follows

$$\rho\left(\frac{\partial V}{\partial t} + V_x\frac{\partial V}{\partial x} + V_y\frac{\partial V}{\partial y} + V_z\frac{\partial V}{\partial z}\right) = -\rho g\cos\theta - \frac{\partial P}{\partial z} + \mu\left(\frac{\partial^2 V}{\partial x^2} + \frac{\partial^2 V}{\partial y^2} + \frac{\partial^2 V}{\partial z^2}\right) \tag{10}$$

and the equation of continuity we can obtain

$$\frac{\partial V_x}{\partial x} + \frac{\partial V_y}{\partial y} + \frac{\partial V_z}{\partial z} = 0 \tag{11}$$

Substitution Eq. (11) into Eq. (10) we can obtain as follows

$$\mu\frac{\partial^2 V}{\partial x^2} = \rho g\cos\theta + \frac{\partial P}{\partial z} \tag{12}$$

where

$$P = P_n + P_f \tag{13}$$

The nature convection (P_n) is occurred by the weight of the fluid. From the mechanical energy balance

$$\rho = -\frac{\partial P_n}{g\partial z} \tag{14}$$

Because the ρ is the function of temperature, so that expression by the Tayler series as follows:

$$\rho(T) = \rho|_{\bar{T}} + \left.\frac{\partial\rho}{\partial z}\right|_{\bar{T}}(T - \bar{T}) = \bar{\rho} - \beta_T(T - \bar{T}) \tag{15}$$

and the temperature distribution can be obtained

$$T_A = T_1 + fx_A \tag{16}$$

Substitution Eqs. (13)~(16) into Eq. (12) can be written as follows:

$$\frac{d^2V_A}{d\eta_A^{\ 2}} = -\frac{\overline{\beta}_T g\cos\theta W_A^{\ 2}}{\mu}(T_1 - \overline{T} + fW_A\eta_A) + \frac{W_A^{\ 2}\Delta P_f}{2\mu L} \tag{17}$$

and the boundary conditions

$$\eta_A = 0 \qquad V_A = 0 \tag{18a}$$

$$\eta_A = 1 \qquad V_A = 0 \tag{18b}$$

Integration of the Eq. (17) with the use of B. C. (18) gives

$$V_A = -\frac{\overline{\beta}_T g\cos\theta W_A^{\ 2}}{\mu}[\frac{1}{6}fW_A(\eta_A^{\ 3} - \eta_A) + \frac{1}{2}(T_1 - \overline{T})(\eta_A^{\ 2} - \eta_A)] + \frac{W_A^{\ 2}\Delta P_f}{4\mu L}(\eta_A^{\ 2} - \eta_A) \tag{19}$$

The mass balance for the enriching section of *a* column as follows

$$-(1+R)\sigma_e = \int_0^W B\rho V_{Ae}dx \tag{20}$$

where

$$-\sigma_e = (-\sigma_{e,n}) + (-\sigma_{e,f}) \tag{21}$$

The mass balance for the nature convection is equal to zero, so that

$$\overline{T} = \frac{2W_A^{\ 3}T_1 + 2W_B^{\ 3}T_2 - f(W_B^{\ 4} - W_A^{\ 4})}{2(W_A^{\ 3} + W_B^{\ 3})} \tag{22}$$

Substitution Eqs. (22) and (19) into Eq. (20) we can obtain as follows:

$$\frac{\Delta P_f}{2L} = \frac{12\mu\sigma_e(1+R)}{(W_A^{\ 3} + W_B^{\ 3})\overline{\rho}B} \tag{23}$$

where

$$\sigma = \frac{W_A^{\ 3}}{W_A^{\ 3} + W_B^{\ 3}}\sigma_e \tag{24}$$

Combination of the Eqs. (19), (22) and (23) yields

$$V_{Ae} = -\frac{\overline{\beta}_T g\cos\theta f W_A^{\ 3}}{6\mu}(\eta_A^{\ 3} - \eta_A) + \frac{\overline{\beta}_T gW_A^{\ 2}[2W_B^{\ 3}\Delta T - f(W_B^{\ 4} - W_A^{\ 4})]}{4\mu(W_A^{\ 3} + W_B^{\ 3})}(\eta_A^{\ 2} - \eta_A)$$

$$+\frac{6\sigma}{\overline{\rho}BW_A}(1+R)(\eta_A - \eta_A^{\ 2}) \tag{25}$$

Similarly, for the enriching section of the *b* column

$$V_{Be} = \frac{\overline{\beta}_T g\cos\theta f W_B^{\ 3}}{6\mu}(\eta_B^{\ 3} - \eta_B) + \frac{\overline{\beta}_T gW_B^{\ 2}[2W_A^{\ 3}\Delta T - f(W_B^{\ 4} - W_A^{\ 4})]}{4\mu(W_A^{\ 3} + W_B^{\ 3})}(\eta_B^{\ 2} - \eta_B)$$

$$+\frac{6\sigma}{\overline{\rho}BW_B}R(\eta_B - \eta_B^{\ 2}) \tag{26}$$

Now, let

$$f_{1e}=\frac{\beta g\cos\phi h_1 W_A{}^3}{6\mu},\quad f_{2e}=\frac{\beta g\cos\phi W_A{}^2[2W_B{}^3\Delta T-h_1(W_A{}^4-W_B{}^4)]}{4\mu(W_A{}^3+W_B{}^3)},\quad f_{3e}=\frac{6W_A{}^2\sigma}{\rho B(W_A{}^3+W_B{}^3)},$$

$$g_{1e}=\frac{\beta g\cos\phi h_1 W_B{}^3}{6\mu},\quad g_{2e}=\frac{\beta g\cos\phi W_B{}^2[2W_A{}^3\Delta T+h_1(W_B{}^4-W_A{}^4)]}{4\mu(W_A{}^3+W_B{}^3)},\quad g_{3e}=\frac{6W_B{}^2\sigma}{\rho B(W_A{}^3+W_B{}^3)} \tag{27}$$

So that, the velocity distribution for the enriching section can be written as

$$V_{Ae}(\eta_A)=-f_{1e}(\eta_A{}^3-\eta_A)+f_{2e}(\eta_A{}^2-\eta_A)+(1+R)f_{3e}(\eta_A-\eta_A{}^2) \tag{28}$$

$$V_{Be}(\eta_B)=g_{1e}(\eta_B{}^3-\eta_B)+g_{2e}(\eta_B{}^2-\eta_B)+Rg_{3e}(\eta_B-\eta_B{}^2) \tag{29}$$

For the stripping section, all equations were still valid expect the subscript “*e*” replaced by “*s*”, and (1+*R*) in equation (28) and *R* in equation (29) replaced by (*1+R*) and *R*, respectively.

(2-1) An Impermeable Sheet Inserted

For the double-flow continuous-type thermal diffusion column, the impermeable sheet is neglected thermal resistance and inserted between the plates, as shown in figure 1. The equations of mass transfer for each slit in the enriching section in dimensionless form may be obtained as

$$\frac{\partial^2 C_{Ae}}{\partial \eta_A{}^2}=(\frac{W_A{}^2 V_{Ae}}{LD})\frac{\partial C_{Ae}}{\partial \zeta} \tag{30}$$

$$\frac{\partial^2 C_{Be}}{\partial \eta_B{}^2}=(\frac{W_B{}^2 V_{Be}}{LD})\frac{\partial C_{Be}}{\partial \zeta} \tag{31}$$

The thermal diffusion term has been eliminated in both Eqs. (30) and (31) by the assumption (d). The associated boundary conditions were:

$$\frac{-\partial C_{Ae}}{\partial \eta_A}+\alpha\theta h_1 W_A=0 \qquad at \quad \eta_A=0 \tag{32}$$

$$\frac{\partial C_{Be}}{\partial \eta_B}+\alpha\theta h_1 W_B=0 \qquad at \quad \eta_B=0 \tag{33}$$

$$-\frac{\partial C_{Ae}}{\partial \eta_A}+\alpha\theta h_1 W_A=0 \qquad at \quad \eta_A=1 \tag{34}$$

$$\frac{\partial C_{Be}}{\partial \eta_B}+\alpha\theta h_1 W_B=0 \qquad at \quad \eta_B=1 \tag{35}$$

$$C_{Ae}=C_{Be}=C_b \qquad at \quad \xi=-1 \tag{36}$$

where α was the thermal diffusion constant, equations (32) and (33) were the hot and cold plates. Eqs. (34) and (35) were the conditions of the impermeable plate. Since it was imperative to having a mixing zone at the ends, we have imposed equation (36).

(2.2) A Permeable Barrier Inserted

Similarly, a permeable barrier is inserted, as shown in figure 2, for the double-flow device. The equations of mass transfer for each slit in the enriching section may also be obtained in the same dimensionless form as in Eqs. (30) and (31), with the boundary conditions of Eqs. (34) and (35) being changed as follows:

$$\frac{-\partial C_{Ae}}{\partial \eta_A}+\alpha\theta h_1 W_A=\frac{W_A}{W_B}[\frac{\partial C_{Be}}{\partial \eta_B}+\alpha\theta h_1 W_B] \qquad at \quad \eta_A=\eta_B=1 \tag{37}$$

$$-\frac{\partial C_{Ae}}{\partial \eta_A}+\alpha\theta h_1 W_A=\frac{W_A\varepsilon}{\delta}[\alpha\theta h_2\delta-\frac{C_{Be}}{D}+\frac{C_{Ae}}{D}] \quad at \quad \eta_A=\eta_B=1 \tag{38}$$

SEPARATION EFFICIENCY ENHANCEMENT

The improvement of separation, I and I_m, for an impermeable sheet and permeable barrier inserted, respectively, are defined relative to on the vertical Clusius-Dickel column of the same working dimensions as follows:

$$I=\frac{\Delta-\Delta_0}{\Delta_0} \tag{39}$$

and

$$I_m=\frac{\Delta_m-\Delta_0}{\Delta_0} \tag{40}$$

CALCULATION METHOD FOR DEGREE OF SEPARATION EFFICIENCIES

The analytical solution to this problem can be obtained by using the separation of variable and power series expansion techniques. Separation of variables results in the following form

$$C_{Ae}(\eta_A,\zeta)=\theta h_1 W_A\eta_A+\sum_{m=0}^{\infty}S_{Ae,m}F_{Ae,m}(\eta_A)G_{e,m}(\zeta) \tag{41}$$

$$C_{Be}(\eta_B,\zeta)=-\theta h_1 W_B\eta_B+\sum_{\mathrm{m}=0}^{\infty}S_{Be,m}F_{Be,m}(\eta_B)G_{e,m}(\zeta)+\theta(\Delta T) \tag{42}$$

Applied to Eq. (30) and (31) leads to

$$G_{e,m}(\xi)=\exp[\lambda_{em}(1+\xi)] \tag{43}$$

For the stripping section Eqs. (30)-(36) were still valid except the symbol "*e*" replaced by "*s*", and the Eqs. (36) and (43) are replaced by Eqs. (44) and (45), respectively.

$$C_{As}=C_{Bs}=C_t, \quad \text{at } \zeta=1 \tag{44}$$

$$G_{e,m}(\xi)=\exp[-\lambda_{sm}(1-\xi)] \tag{45}$$

Without loss of generality, we may assume the eigenfunctions $F_{Ae,m}(\eta_A)$ and $F_{Be,m}(\eta_B)$ to be polynomials and express in the following forms:

$$F_{Ae,m}(\eta_A)=\sum_{n=0}^{\infty}d_{mn}\eta_A^{\ n}, \quad d_{m0}=1 \ \text{(selected)}, \quad d_{m1}=0 \tag{46}$$

$$F_{Be,m}(\eta_B)=\sum_{n=0}^{\infty}e_{mn}\eta_B^{\ n}, \quad e_{m0}=1 \ \text{(selected)}, \quad e_{m1}=0 \tag{47}$$

All the coefficients d_{mn} and e_{mn} may be calculated by governing equation and boundary conditions of the enrichment and stripping sections in terms of eigenvalue, $\lambda_{Ae,m}$, $\lambda_{Be,m}$, $\lambda_{As,m}$, and $\lambda_{Bs,m}$.

Combining the average concentration in the enriching and stripping section yields the degree of separation for a whole column in terms of the eigenvalues, expansion coefficients, impermeable sheet position, recycle ratio and angle of inclination. The degree of separation for double-pass thermal diffusion column with an impermeable sheet is

$$\Delta = C_b - C_t = \left(\frac{LD\sum_{m=0}^{\infty}[\frac{S_{Ae,m}F'_{Ae,m}(1)}{\lambda_{Ae,m}}][\exp(\lambda_{Ae,m})-1]}{(\kappa W)^2\int_0^1 V_{Ae}d\eta_A} + \frac{\int_0^1 V_{Ae}C_{Ae}(\eta_A,0)d\eta_A}{\int_0^1 V_{Ae}d\eta_A} \right)$$

$$- \left(\frac{\int_0^1 V_{As,m}C_{As}(\eta_A,0)d\eta_A}{\int_0^1 V_{As}d\eta_A} - \frac{LD\sum_{m=0}^{\infty}[\frac{S_{As,m}F'_{As,m}(1)}{\lambda_{As,m}}][1-\exp(-\lambda_{As,m})]}{(\kappa W)^2\int_0^1 V_{As}d\eta_A} \right) \quad (48)$$

The calculation procedure is similar to that in the previous section, except the eigenvalues for the two subchannel. The degree of separation for double-pass thermal diffusion column with a permeable barrier is

$$\Delta_m = C_b - C_t = \left(\frac{LD\sum_{m=0}^{\infty}[\frac{S_{Ae,m}F'_{Ae,m}(1)}{\lambda_{e,m}}][\exp(\lambda_{e,m})-1]}{(\kappa W)^2\int_0^1 V_{Ae}d\eta_A} + \frac{\int_0^1 V_{Ae}C_{Ae}(\eta_A,0)d\eta_A}{\int_0^1 V_{Ae}d\eta_A} \right)$$

$$- \left(\frac{\int_0^1 V_{As,m}C_{As}(\eta_A,0)d\eta_A}{\int_0^1 V_{As}d\eta_A} - \frac{LD\sum_{m=0}^{\infty}[\frac{S_{As,m}F'_{As,m}(1)}{\lambda_{s,m}}][1-\exp(-\lambda_{s,m})]}{(\kappa W)^2\int_0^1 V_{As}d\eta_A} \right) \quad (49)$$

RESULTS AND CONCLUSIONS

Separation efficiency of double-flow thermal-diffusion columns with inserting an impermeable sheet or a permeable barrier has been investigated and solved analytically by using the orthogonal expansion technique with eigenfunction expanding in terms of an extended power series. The improvement of separation for an impermeable sheet and permeable barrier inserted, respectively, are defined relative to the vertical Clusius-Dickel column of the same working dimension is best illustrated by calculating Eqs. (39), (40) and the results are shown in figure 3. It is seen from the table that the improvement of separation, I and I_m, increases with inclination angle and the position of the sheet and membrane moving away 0.5. Furthermore, the higher separation efficiency can be obtained, as indicated in figure 3, with the sheet or membrane position, κ, moving away from 0.5 and the inclination angle, ϕ, about 40°~50°. The best separation efficiency was obtained with the design parameters $R = 1.0$, $\kappa = 0.75$ and $\phi = 40^\circ \sim 50^\circ$.

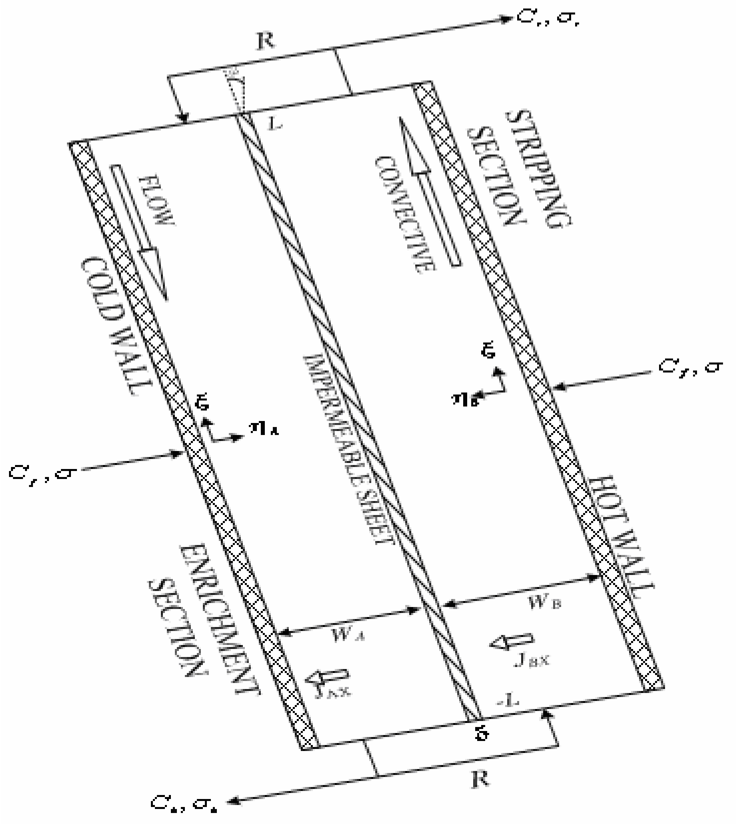

Fig. 1　Schematic diagram of a double-flow inclined thermal-diffusion column with a vertical impermeable sheet inserted.

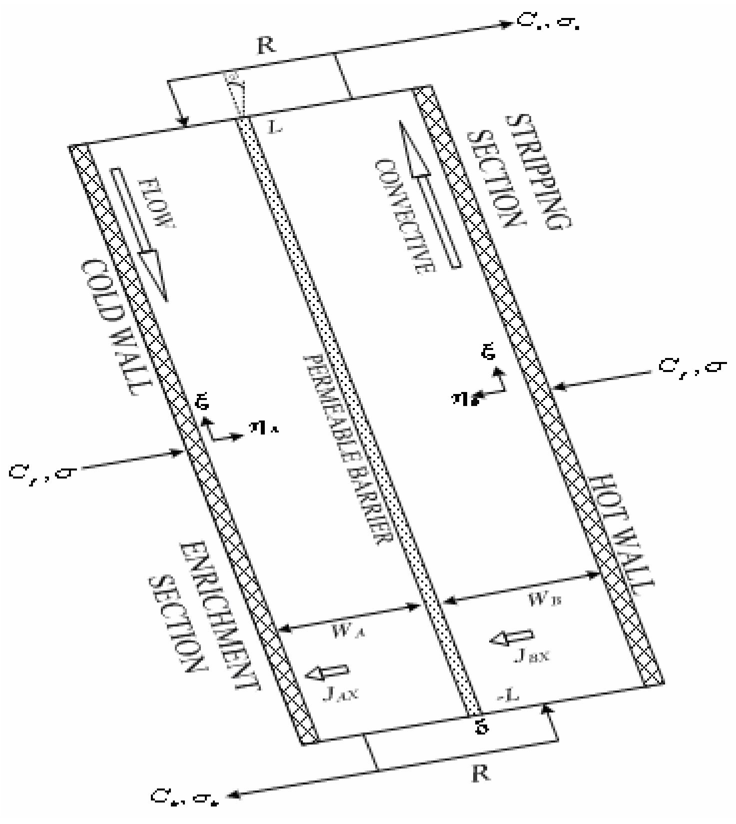

Fig. 2　Schematic diagram of a double-flow inclined thermal-diffusion column with a vertical permeable barrier inserted.

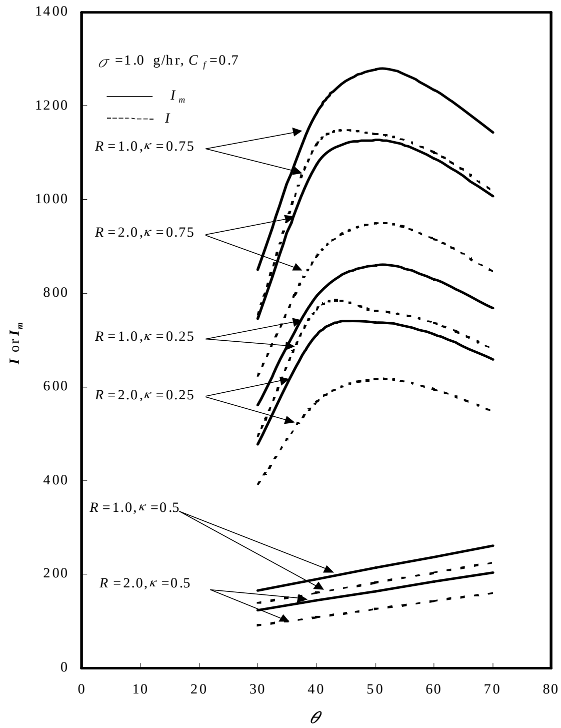

Fig. 3 The separation degree improvement for the device with an impermeable sheet and a permeable barrier with the angles of inclination, channel thickness ratio as parameters, with $C_F = 0.7$ $\sigma = 1.0 g/hr$ and $R = 1$ and 2

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NOMENCLATURE

B	column width (cm)
C	fraction concentration of D_2O in H_2O-HDO-D_2O system (-)
D	ordinary diffusion coefficient ($\mathrm{cm^2/s}$)
g	gravitational acceleration ($\mathrm{cm/s^2}$)
k , k_f	thermal conductivity of the barrier and the fluid, respectively ($\mathrm{cal/cm \cdot s \cdot K}$)
L	one-half of column length (cm)
R	reflux ratio at both ends of the column (-)
T_1, T_2	temperatures of the cold and hot plates, respectively (K)
W	thickness of the region (cm)
x	coordinate in the horizontal direction (cm)
z	coordinate in the vertical direction (cm)
δ	thickness of the barrier (cm)
ζ	dimensionless coordinate in the vertical direction, defined by Eq. (9) (-)
η	dimensionless coordinate in the horizontal direction, defined by Eq. (9) (-)
θ	angle of inclination of column plate from the vertical (^{o})
λ_m	eigen-value (-)
μ	viscosity of fluid ($\mathrm{g/cm \cdot s}$)
ρ	density of fluid ($\mathrm{g/cm^3}$)
σ	mass flow rate of top or bottom product (g / hr)
κ	the position of an impermeable sheet or a permeable barrier