The Improvement of Removal Rate in Countercurrent-Flow Frazier Scheme of Thermal Diffusion Columns with Optimal Plate Spacing

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Abstract

This work investigated the plate spacing effect on binary mixture separation removal rate in a countercurrent-flow Frazier scheme with N flat-plate thermal diffusion columns of the same size and with fixed total expense. The equations for estimating optimal plate spacing and the corresponding maximum removal rate are developed. Considerable improvement in performance is obtainable when thermal diffusion columns with optimal plate spacing are employed for operation. The fact that the countercurrent-flow operation is more effective than the cocurrent-flow operation in a Frazier scheme, is also confirmed.

Key Words: Thermal Diffusion, Mass Transfer, Frazier Scheme, Optimal Plate Spacing, Countercurrent Flow, Mathematical Modeling

1. Introduction

A concentration gradient is usually established if a temperature gradient is applied to a homogeneous solution. This phenomenon is called thermal diffusion or the Soret effect, which is an unusual process for separating a liquid or gas mixture. Mixtures that are difficult or impossible to separate using distillation, extraction or any other usual method, may be successfully separated by thermal diffusion. These separable mixtures include biological solutions and suspensions, aqueous solutions, polymer or organic solutions, isotopic solutions, liquid metal solutions and various gas mixtures [1].

It was the great achievement of Clusius and Dickel [2] to introduce the thermogravitational thermal diffusion column, in which convective currents could be utilized to produce a cascading effect analogous to the countercurrent multistage effect extraction, and thus obtain a relatively large separation. In addition to the desirable cascading effect, the convective currents in a C-D column also produce an undesirable remixing effect [3]. Therefore, it appears that proper control of the convection strength might effectively suppress this undesirable remixing effect while still preserving the desirable cascading effect, leading to improved separation. Based on this concept, some improved columns have been developed in the literature, such as the inclined column [3], wired column [4–6], and barrier column [7].

For practical applications, the thermal diffusion columns (C-D columns) are connected in series shown in Figure 1. This system is called the Frazier scheme [8,9]. The separation theory of thermal diffusion in C-D column was first presented by Furry et al. [10] and Jones and Furry [11], while that in the Frazier scheme was given by Rabinovich [12] and Sovorov and Rabinovich [13]. Many improved columns have been introduced using the Frazier scheme, such as the optimum plate-spacing column [14], optimum plate-aspect ratio column [15] and spiral wired column [6]. This study investigates the plate spacing effect on separation in a countercurrent-flow, instead of concurrent-flow [14], of the Frazier scheme, and the results will be discussed and compared.
2. Separation Theory

2.1 Separation Equation

Consider N flat-plate C-D columns of the same size with plate spacing (2\(\omega\)) and plate surface area (\(S = \text{length} L \times \text{width} B\)), connected in series, as shown in Figure 1. The delivery of supplies \(\sigma\) with feed concentration \(C_0\) is accomplished at the upper-end of the first column and at the lower end of the Nth (last) column. Products sampling at the same flow rate \(\sigma\) is carried out at the ends opposite to the supply entrances. The temperature gradient, \(\Delta T/2\omega\), applied between the surface of a flat-plate C-D column, as shown in Figure 2 for the \(i\)th column, has two effects: (1) a flux of one solution component relative to the other is brought about by thermal diffusion, and (2) natural convective currents are produced parallel to the plates owing to density differences. The combined results of these effects is to produce a concentration difference between the two ends of the column. Meanwhile, the concentration gradients produced by the combined effects of thermal diffusion and convection acts as the ordinary diffusion to oppose thermal diffusion and to limit the separation.

The separation equation in a countercurrent-flow Frazier scheme with N thermal-diffusion columns of the same size was derived in previous work [15]

\[
\Delta = C_{T,N} - C_{R,1}
\]  

\[
\left( \frac{1.5}{\Delta} \right)^2 + 12C_0(1 - C_0)\right)^{1/2} - \frac{1.5}{\Delta}
\]  

or

\[
\bar{\Lambda} = \frac{3\Delta}{12C_0(1 - C_0) - \Delta^2}
\]  

where

\[
\bar{\Lambda} = \frac{(LNH/2)}{(N + 1)K + \sigma L}
\]

Figure 1. Schematic diagram of the countercurrent-flow Frazier scheme.

Figure 2. Flows and fluxes in \(i\)th thermal diffusion column.
in which
\[ H = \frac{\alpha \beta g (2\omega)^3 B (\Delta T)^2}{6! \mu T_w} \] (5)
\[ K = \frac{\rho B^2 g^2 (2\omega)^3 B (\Delta T)^2}{9! D \mu^2} \] (6)

Noted that \( \Delta \) denotes the approximate degree of separation when \( C_0 \) is within the range, \( 0.3 < C_0 < 0.7 \). In this case, \( C_0 (1 - C_0) \equiv 1/4 \), while \( \Delta = \Delta/\sqrt[3]{1 - (\Delta^2/3)} \approx \Delta \) since \( \Delta << 1 \).

### 2.2 Separation Equation for Fixed Expense

The plate spacing \( 2\omega \) in a thermal diffusion column is generally so small that changing \( 2\omega \) will not cause any additional fixed charge. The expenditure of making a separation by thermal diffusion essentially includes two parts: a fixed charge and an operating expense. The fixed charge is roughly proportional to the equipment cost, say mainly the material cost of a thermal diffusion column (BL). The operating expense is chiefly the heat transfer rate obtainable from the expression, \( KBL (\Delta T/2\omega) \). Based on these terms, we account for the plate-spacing change influence on the degree of separation and removal rate considering the fixed total expense.

Accordingly, with the consideration of approximately fixed total expense \( (\Delta T/2\omega = \text{constant}) \), Eqs. (5) and (6) may be rewritten as
\[ a = \frac{\alpha \beta g (\Delta T/2\omega)^3 B}{6! \mu T_w} = H/(2\omega)^3 = \text{constant} \] (7)
\[ b = \frac{\rho B^2 g^2 (\Delta T/2\omega)^3 B}{9! D \mu^2} = K/(2\omega)^3 = \text{constant} \] (8)

and Eq. (4) becomes
\[ \Delta = \frac{(LN/2)(2\omega)^4}{(N + 1)b(2\omega)^4} + \sigma L \] (9)

### 2.3 Maximum Removal Rate

The component 1 removal rate in the countercurrent-flow Frazier scheme may be defined as
\[ M = \sigma(C_{x,n} - C_0) + \sigma(C_0 - C_{r,1}) = \sigma \Delta \] (10)

The optimum plate spacing \( (2\omega)^* \) for maximum removal rate \( M_{\text{max}} \) with specified column length \( L \), column width \( B \), column number \( N \) and flow rate \( \sigma \), is obtained by partially differentiating Eq. (10) with respect to \( 2\omega \) and setting \( \partial M/\partial (2\omega) = 0 \), or \( \partial \Delta/\partial (2\omega) = 0 \). After differentiation and simplification, we have
\[ \frac{\partial \Delta}{\partial (2\omega)} = 0 \] (11)

This result indicates that \( (2\omega)^* \) does not depend on the feed concentration \( C_0 \). Equation (11) can also be obtained from Eq. (3) once \( \partial \Delta/\partial (2\omega) = 0 \). Substitution of Eq. (4) into Eq. (11) results in
\[ (2\omega)^* = \left[ \frac{5\sigma L}{4b(N + 1)} \right]^{1/6} \] (12)

### 2.4 The Best Performance for Concurrent-Flow Operation

The plate spacing effect on the removal rate for a binary mixture in a concurrent-flow Frazier scheme with \( N \) flat-plate thermal diffusion-column columns of the same size and with total fixed expense, was investigated in a previous work [14]. Considerable improvement in performance is obtainable when thermal diffusion columns with optimal plate-spacing are employed for operation. The equation for estimating the optimal plate spacing for maximum removal rate, are
\[ M_{\text{max}} = \sigma \Delta_{\text{max}} \] (14)

where
\[ \Delta_{\text{max}} = \left[ \left( \frac{1.5}{\Delta_{\text{max}}} \right)^{1/2} + 12C_0(1 - C_0) \right]^{1/2} - \frac{1.5}{\Delta_{\text{max}}} \] (15)

The optimum plate spacing \( (2\omega)^* \) for maximum removal rate \( M_{\text{max}} \) with specified column length \( L \), column width \( B \), column number \( N \) and flow rate \( \sigma \), is obtained by partially differentiating Eq. (10) with respect to \( (2\omega) \) and setting \( \partial M/\partial (2\omega) = 0 \), or \( \partial \Delta/\partial (2\omega) = 0 \). After differentiation and simplification, we have

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\[ (2\omega)^* = \left[ \frac{5\sigma L}{4b(N + 1)} \right]^{1/6} \] (12)

substituting of Eq. (12) into Eq. (9) yields
\[ \Delta_{\text{max}} = \frac{2Na(2\omega)^{3/2}}{9\sigma} = \left( \frac{2Na}{9\sigma} \right)^{1/3} \left[ \frac{5\sigma L}{4b(N + 1)} \right]^{1/6} \] (13)

and the maximum removal rate is
\[ M_{\text{max}} = \sigma \Delta_{\text{max}} \] (14)

where
\[ \Delta_{\text{max}} = \left[ \left( \frac{1.5}{\Delta_{\text{max}}} \right)^{1/2} + 12C_0(1 - C_0) \right]^{1/2} - \frac{1.5}{\Delta_{\text{max}}} \] (15)
\[ M_{\text{max}} = \sigma \Delta \text{max} \]
\[ = \sigma \left( \frac{1.5}{\Delta \text{max}} \right)^2 + 12C_0(1-C_0) \left[ \frac{1.5}{\Delta \text{max}} \right]^{1/2} \]  
(17)
\[ \Delta \text{max} = \frac{ab}{4b} \left[ \frac{\sigma L}{2b} (U-1) \right]^{4/9} (1-U^{-N}) \]  
(18)
where
\[ a = H/(2\omega)^{3} = 1.43 \times 10^{4} \text{ g/min/cm}^{5} \]
(19)
(20)
(21)
(22)
In which the parameter values \( U \) is determined from the following equation:
\[ 4U^{N-1} - (9N+4)U + 9N = 0 \]
(18)

3. Results and Discussion

The improvement in performance resulting from operating at the optimum plate-spacing with total fixed expense, may be illustrated numerically using the experimented data from Chueh and Yeh’s work [16] for n-heptane enrichment from a benzene-and n-heptance system:
\[ T = 69 \text{ °F} = 38.3 \text{ °C}; (2\omega) = 0.09 \text{ cm}; L = 185 \text{ cm}; B = 10 \text{ cm}; H = 0.845 \text{ g/min}; K = 419 \text{ g/cm/min}. \]

If the total expense is kept unchanged, i.e. \( \Delta T/(2\omega) = 38.3/(0.09) = 4.25 \times 10^{2} \text{ °C/cm} \) (fixed value), then from Eqs. (7) and (8)
\[ a = H/(2\omega)^{3} = 1.43 \times 10^{4} \text{ g/min/cm}^{5} \]
(21)
\[ b = K/(2\omega)^{3} = 1.0815 \times 10^{2} \text{ g/min/cm}^{8} \]  
(22)

From these values, the optimal plate spacing \( (2\omega)^{*} \) and the corresponding maximum separation \( \Delta \text{max} \), as well as the maximum removal rate \( M_{\text{max}} \), are calculated from the appropriate equation. Further, the difference in plate temperature \( (\Delta T)^{*} \) needed to maintain fixed expense can be estimated by
\[ (\Delta T)^{*} = 425.5(2\omega)^{*}, \text{ °C} \]  
(23)

The results are listed in Table 1.

3.1 Numerical Example

The improvement in removal rate by operating at the optimal plate spacing is best illustrated by calculating the percentage increase in removal rate based on that ob-

<table>
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<tr>
<th>( \sigma ) (g/min)</th>
<th>( \Delta ) (%)</th>
<th>( (2\omega)^{*} \times 10^{-2} )</th>
<th>( (\Delta T)^{*} \times 10^{-2} )</th>
<th>( \Delta \text{max} ) (%)</th>
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<th>Co = 0.3 or 0.7</th>
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tained at \((2\alpha) = 0.09 \text{ cm}\), i.e.

\[
I = \frac{M_{\text{max}} - M}{M} = \frac{\Delta_{\text{max}} - \Delta}{\Delta}
\]  

(24)

Table 1 shows that the optimal plate spacing \((2\alpha)\) for maximum removal rate \(M_{\text{max}}\) increases when the flow rate increases, or as the column number decreases. Considerable improvement in removal rate \(I\) based on the removal rate obtained at \((2\alpha) = 0.09 \text{ cm}\), is achieved, especially for the case that the optimal plate spacing goes far 0.09 cm, as shown in Table 1 and Figure 3. The flow rates, where \((2\alpha) = (2\alpha)^*\), increase with the column number, as indicated by points a, b and c in Figure 3. For \((2\alpha)^* < 0.09 \text{ cm}\), \(I\) increases as flow rate \(\sigma\) decreases. In the case that \(N = 40\), \(\sigma = 4 \text{ g/min}\) and \(C_0 = 0.3\) (or 0.7), one has \(I = 111.5\%\).

### 3.3 Comparison of Separations between Two Flow-Pattern Operations

The concurrent-flow and countercurrent-flow operations removal rate comparisons are presented in Table 2. This table shows that the removal rates obtained by the countercurrent-flow operation were better than those obtained by concurrent-flow operation at \((2\alpha) = 0.09 \text{ cm}\) or at \((2\alpha)^*\) and that \((M - \dot{M}) / M_{\text{max}}\) also increases when the flow rate \(\sigma\) decreases, or as number of columns \(N\) increases. \((M_{\text{max}} - \dot{M}_{\text{max}}) / \dot{M}_{\text{max}}\) also increases with \(N\) but nearly

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**Table 2.** Comparison of removal rates of n-heptane obtained at \((2\alpha)\) and at \((2\alpha) = 0.09 \text{ cm}\) for \(C_0 = 0.5\): (a) \(N = 10\); (b) \(N = 20\); (c) \(N = 40\)

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<th>(\sigma) (g/min)</th>
<th>((2\alpha)^* \times 10^2) (cm)</th>
<th>((\Delta T)^*) (°C)</th>
<th>(\Delta_{\text{max}}) (%)</th>
<th>(\dot{M}_{\text{max}}) (g/min)</th>
<th>(\dot{M}) (g/min)</th>
<th>((\dot{M}<em>{\text{max}} - \dot{M}) / \dot{M}) (%), (M</em>{\text{max}} - \dot{M}_{\text{max}}) (%)</th>
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does not depend on $\sigma$. Furthermore, in some cases, even when $M > M_{\text{max}}$; the countercurrent flow effect on separation operating at $(2\omega) = 0.09$ cm may overcome the concurrent flow effect even operating at optimal plate spacing.

### 4. Conclusion

The study showed that for the thermal diffusion column separation, proper convection strength control by suitably adjusting the plate spacing may effectively suppress the undesirable remixing effect while still preserving the desirable cascading effect. This results in substantial performance improvement. A Frazier scheme operating with feed countercurrent flow is better than a system operating with concurrent flow.

The optimal plate spacing equation for maximum removal rate for separating a binary mixture using thermal diffusion in a countercurrent-flow Frazier scheme with fixed total expense, was derived. A numerical example using the experimental data obtained in a previous work [16] was given. Considerable improvement in performance was obtained by operating at the optimal plate spacing as shown in Table 1. The performance can be further improved in a countercurrent-flow scheme, as shown in Table 2. In some cases, the countercurrent flow effect on performance operating at $(2\omega) = 0.09$ cm even overcomes the concurrent flow effect operating at optimal plate spacing. In other word, $(M - M_{\text{max}})/M_{\text{max}} > 0$ as shown in Table 2.

It was mentioned before that $(2\omega)^*$ increases as the flow rate increases or as the column number decreases. Although the plate spacing in a thermal diffusion column is generally so small that changing $(2\omega)^*$ will not cause any additional fixed charge, however, increasing $(2\omega)^*$ will lead to an increase in $(\Delta T)^*$, the corresponding temperature difference between the two plate surfaces, in order to maintain the operating cost (i.e. $\Delta T/2\omega$) constant. Therefore, some additional cost may be needed to maintain the higher temperature difference between the two plate surfaces.

### Nomenclature

- $a$: a constant defined by Equation (7), g/s cm$^5$
- $b$: a constant defined by Eq. (8), g/s cm$^8$
- $C$: fractional mass concentration of component 1
- $C_O$: C in feed streams
- $C_{T,j}, C_{B,i}$: C in product streams exiting from $i$th column, for top and bottom ends, respectively
- $C_{T,N}, C_{B,1}$: $C_{T,j}, C_{B,i}$ from $N$th column and first column, respectively
- $D$: ordinary diffusion coefficient, cm$^2$/s
- $g$: gravitational acceleration, cm/s$^2$
- $H$: transport coefficient defined by Eq. (5), g/s
- $J_{x,OD}$: mass flux of component 1 in x-direction due to ordinary diffusion, g/s cm$^2$
- $J_{x,TD}$: mass flux of component 1 in x-direction due to thermal diffusion, g/s cm$^2$
- $J_{z,OD}$: mass flux of component 1 in z-direction due to ordinary diffusion, g/s cm$^2$
- $K$: transport coefficient defined by Eq. (6), g cm/s
- $L$: column length, cm
- $M, M_{\text{max}}$: removal rate in countercurrent- and concurrent-flow operations, respectively g/s
- $M_{\text{max}}, M_{\text{max}}$: Maximum value of $M, M_{\text{max}}$ g/s
- $N$: column number of a Frazier scheme
- $T_m$: mean absolute temperature, K
- $\Delta T$: difference in temperature of hot and cold plates, K
- $(\Delta T)^*$: $\Delta T$ for maximum removal rate, K
- $U$: value determined by Eq. 20
- $z$: axis of transport direction, cm

**Greek Symbols**

- $\alpha$: thermal diffusion constant
- $\beta$: $-(\partial \rho/\partial T)$ evaluated at $T_m$ under constant pressure, g/cm$^3$ K
- $\Delta, \dot{\Delta}$: $C_{T,N}, C_{B,1}$ in countercurrent- and concurrent-flow operations, respectively
- $\bar{\Delta}, \dot{\bar{\Delta}}$: $\Delta, \dot{\Delta}$ when $0.3 < C_O < 0.7$
- $\Delta_{\text{max}}, \dot{\Delta}_{\text{max}}$: maximum value of $\Delta, \dot{\Delta}$
- $\mu$: absolute viscosity, g/cm s
- $\rho$: mass density evaluated at $T_m$, g/cm$^3$
- $\sigma$: mass flow rate, g/s
- $2\omega$: plate spacing, distance between hot and cold plates, cm
- $(2\omega)^*$: optimum $(2\omega)$ for maximum separation, cm
References


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Tamkang University was founded in 1950 and Tamkang Journal was published since 1962. Starting from 1998, Tamkang Journal was divided into two, which are Tamkang Journal of Science and Engineering and Tamkang Journal of Humanity and Social Science. Tamkang Journal of Science and Engineering became an international journal since 2000 and four issues are published each year. Herein we are very delightful to announce that Tamkang Journal of Science and Engineering has been published under the new title Journal of Applied Science and Engineering (JASE) since 2012. This peer-reviewed journal with a new-look cover will continue to be published quarterly in print and online.

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