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## Thermal Diffusion in an Ideal Column

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### Abstract

An ideal thermal diffusion column in which the separation efficiency is 100% is defined. The separation theory of such a column has also been derived. In the illustration given, the inclined moving-wall column acts as an ideal column because its efficiency reaches 98%. However, due to other operating and design conditions, there may be some better column still to be developed. The best thermal diffusion column for certain separation conditions is related to the highest separation efficiency based on the ideal column definition.

### INTRODUCTION

The thermogravitational thermal diffusion column (C-D column), introduced by Clusius and Dickel (2), has been employed as a device for the separation of liquid or gas mixtures which are difficult or impossible to separate by ordinary methods. This column was used to separate uranium isotopes at Oak Ridge in World War II, and a commercial plant to produce lubricating oil with a high viscosity index has also been designed. This method is more effective for the separation of hydrogen isotopes because of the large ratio in molecular weights (15).

A more detailed study of the mechanism of separation in the Clusius-Dickel column indicates that the convective currents actually have two conflicting effects: the desirable cascading effect and the undesirable remixing effect. The convective currents have a multistage effect which is necessary to secure high separation, and that effect is an essential feature of the Clusius-Dickel column. However, since convection brings down the

fluid at the top of the column, where it is rich in one component, to the bottom of the column, where it is rich in the other component, and vice versa, there is a remixing of the two components. It therefore appears that proper control of the convective strength might effectively suppress the undesirable remixing effect while still preserving the desirable cascading effect, and thereby lead to improved separation.

Accordingly, it has been demonstrated that separation can be improved in inclined columns (1, 5), wired columns (9, 16), inclined moving-wall columns (6, 13), rotary columns (7, 10), packed columns (4, 11), rotated wired columns (12), and barrier columns (8, 14). When the velocity profiles as well as the convective strengths are properly adjusted, the enrichments obtainable from these improved columns are much better than that from a C-D column.

Just as the ideal gas and the ideal stage do not exist in reality, we here define an ideal thermal diffusion column in which the convective strength as well as the velocity profile is most properly adjusted and the separation obtainable is the largest. Then, separations obtained from improved columns which have been developed may be compared with those obtainable from the ideal column.

### SEPARATION THEORY IN THERMAL DIFFUSION COLUMNS

Consider a flat-plate thermogravitational thermal diffusion column. Figure 1 illustrates the flows and fluxes in such a column. Since the space between the surfaces of the column is small, we assume that the convective flow produced by the density gradient is laminar and that the temperature distribution is determined by conduction in the  $x$ -direction only. We also assume that the convective velocity is in the  $z$ -direction only, that both end effects can be neglected, and that the mass fluxes due to thermal and ordinary diffusion are too small to affect the velocity and temperature profiles.

The horizontal mass flux of Component 1 of a binary mixture is related to the velocity by the differential mass balance equation

$$\frac{\partial J_x}{\partial \eta} + \rho \omega v_z \frac{\partial C}{\partial z} = 0 \quad (1)$$

Under the assumptions that diffusion in the  $z$ -direction is negligible and that bulk flow in the  $x$ -direction is negligible, the expression for  $J_x$  in terms of the two contributions (ordinary and thermal diffusion) is

$$J_x = \frac{Dp}{\omega} \left( -\frac{\partial C}{\partial \eta} + \frac{\alpha C \bar{C} dT}{T d\eta} \right) \quad (2)$$

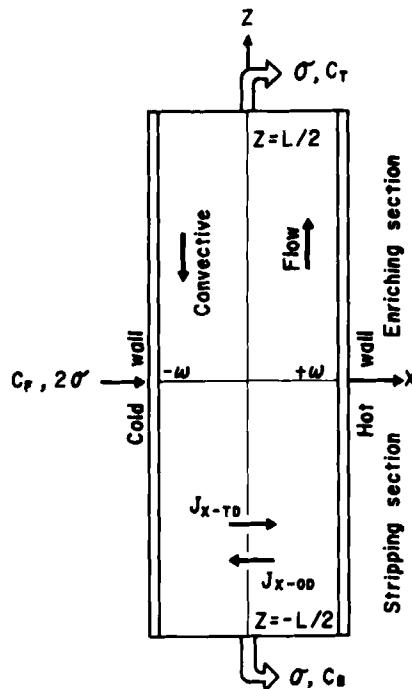


FIG. 1. Schematic diagram of a thermal diffusion column.

The concentration profile may be calculated by combining Eqs. (1) and (2) and integrating with the following boundary conditions:

$$\text{at } \eta = \pm 1, \quad J_x = 0 \quad (3)$$

$$\text{at } \eta = \eta, \quad C = C(\eta, z) \quad (4)$$

$$\text{at } \eta = 0, \quad C = C_0(0, z) \quad (5)$$

The solution is

$$C = C_0 + \frac{\alpha C \bar{C}(\Delta T)}{2\bar{T}}\eta + \frac{\omega^2 \partial C}{D \partial z} \int_0^{\eta} \int_1^{\eta} v_z d\eta d\eta \quad (6)$$

In obtaining the above solution,  $dT/d\eta = \Delta T/2$  as obtained from the energy equation was used, and it was assumed that  $(\partial C/\partial z) = f(z)$  for

moderate flow rates and that the quantity  $C\bar{C}/T$  appearing in the thermal diffusion term is independent of  $x$ .

The rate of mass transport of Component 1 in the  $z$ -direction is given by

$$\tau_1 = \int_{-\omega}^{+\omega} \bar{\rho} C v_z B dx - \int_{-\omega}^{\omega} \rho B dx - \int_{-\omega}^{\omega} \bar{\rho} D \frac{\partial C}{\partial z} B dx \quad (7)$$

Substituting Eq. (6) into Eq. (7) and using the relation

$$\sigma = \int_{-\omega}^{+\omega} \bar{\rho} v_z B dx \quad (8)$$

for the enriching section gives

$$\tau_1 = C_0 \sigma + H \bar{C} - K \frac{\partial C}{\partial z} \quad (9)$$

where the transport coefficients are calculated by

$$H = \frac{\bar{\rho} B \omega \alpha (\Delta T)}{2T} \int_{-1}^{+1} v_z d\eta d\eta \quad (10)$$

$$K = \frac{\bar{\rho} B \omega^3}{D} \int_{-1}^{+1} v_z \left( \int_0^{\eta} \int_1^{\eta} v_z d\eta d\eta \right) d\eta + 2\omega D B \bar{\rho} \\ = K_0 + K_i \quad (11)$$

In Eq. (9),  $\tau_1$  is the transport of Component 1 in the positive  $z$ -direction in the enriching section under steady, continuous operations. The term  $H$  represents the effectiveness of separation by thermal diffusion, and the term  $K$  represents the countereffect of remixing due to convection  $K_0$  and ordinary diffusion  $K_i$  in the vertical direction.

Since  $|C(-1, z) - C(+1, z)|$  is small as compared with the concentration difference between the top and bottom ends, we make the approximation  $C_0(0, z) \cong C_b(z)$ , in which  $C_b(z)$  is defined by

$$C_b(z) = \frac{\rho B}{\sigma} \int_{-\omega}^{\omega} C v_z dx \quad (12)$$

By making this approximation, Eq. (9) can be written as

$$\tau_1 = \sigma C_b + H \bar{C} - K \frac{dC_b}{dz} \quad (13)$$

Since  $\tau_1$  and  $\sigma$  are constant at steady state, and  $(\tau_1/\sigma) = C_T$  everywhere in the enriching section, Eq. (13) becomes

$$H[\bar{C} - \frac{\sigma}{H}(C_T - C_b)] = K \frac{dC_b}{dz} \quad (14)$$

By making a similar analysis in the stripping section, one obtains

$$H[\bar{C} + \frac{\sigma}{H}(C_B - C_b)] = K \frac{dC_b}{dz} \quad (15)$$

The degree of separation, defined by

$$\Delta = C_T - C_B \quad (16)$$

may be obtained from Eqs. (14) and (15) associated with the boundary conditions:

$$C = C_F, \quad \text{at } z = 0 \quad (17)$$

$$C = C_T, \quad \text{at } z = L/2 \quad (18)$$

$$C = C_B, \quad \text{at } z = -L/2 \quad (19)$$

The solution is

$$\Delta = \frac{H}{2\sigma} \left[ 1 - \exp \left( -\frac{\sigma L}{2K} \right) \right] \quad (20)$$

### VELOCITY PROFILE IN THE CLUSIUS-DICKEL COLUMN

Applying the appropriate equations of motion and energy to the Clusius-Dickel column gives the following steady-state velocity profiles (3)

$$v_z = A_0(1 - \eta^2) + A_1(n - \eta^3) \quad (21)$$

where, for the enriching section,

$$A_0 = \frac{3\sigma}{4\bar{\rho}\omega B} \quad (22)$$

and

$$A_1 = \frac{\beta\omega^2(\Delta T)g}{12\mu} \quad (23)$$

In the stripping section, the above equations apply with  $\sigma$  replaced by  $-\sigma$ . The first term in the right-hand side of Eq. (21) represents the flow due to continuous feed, while the second term represents the flow due to natural convection. Consequently, the transport coefficients in Eq. (20) are obtained by substituting Eq. (21) into Eqs. (10) and (11) as

$$H = \frac{\alpha\bar{\beta}^2\bar{\rho}g(2\omega)^3B(\Delta T)^2}{6!\mu\bar{T}} \quad (24)$$

$$\begin{aligned} K &= K_0 + K_1 \\ &= \frac{\bar{\beta}^2\bar{\rho}g^2(2\omega)^7B(\Delta T)}{9!D\mu^2} + 2\omega DB\beta \end{aligned} \quad (25)$$

### VELOCITY PROFILE IN THE IDEAL COLUMN

The generalized velocity profile in all types of thermal diffusion column may be expressed as

$$v_z = A_0(1 - \eta^2) + A_1(\eta - \eta^3) + f(\eta) \quad (26)$$

Since

$$\bar{\rho}B\omega \int_{-1}^{+1} v_z d\eta = +\sigma \text{ in enriching section} \quad (27)$$

$$= -\sigma \text{ in stripping section} \quad (28)$$

the following restriction for  $f(\eta)$  is required:

$$\int_{-1}^{+1} f(\eta) d\eta = 0 \quad (29)$$

If the polynomial form is taken for  $f(\eta)$ , with the restriction of Eq. (29), one obtains

$$f(\eta) = \sum_{n=1}^N a_n \eta^{2n-1} \quad (30)$$

in which  $N$  is a positive integer and the constants  $a_n$  are specified for a certain case of thermal diffusion with the operating and design conditions fixed. When all the  $a_n$ 's are set equal to zero, Eq. (26) reduces to Eq. (21), the velocity profile in the C-D column.

Since in an ideal column the velocity profile is the best and the degree of separation is the highest, the constants  $a_n$  in Eq. (30) for such a column should be selected by the principle of optimization as

$$\frac{\partial \Delta}{\partial a_n} = 0, \quad n = 1, 2, 3, \dots \quad (31)$$

After solving for the  $a_n$ 's from Eq. (31), the velocity profile and the degree of separation in the ideal column are, respectively,

$$v_{z,\text{best}} = A_0(1 - \eta^2) + A_1(\eta - \eta^3) + \sum_{n=1}^N a_{n,\text{opt}} \eta^{2n-1} \quad (32)$$

$$\Delta_{\text{ideal}} = \frac{H(a_{1,\text{opt}}, a_{2,\text{opt}}, \dots, a_{N,\text{opt}})}{2\sigma} \times \left[ 1 - \exp \left\{ \frac{-\sigma L}{2K(a_{1,\text{opt}}, a_{2,\text{opt}}, \dots, a_{N,\text{opt}})} \right\} \right] \quad (33)$$

It should be noted that the value of  $N$  taken in Eq. (32) must be large enough so that  $v_{z,\text{best}}$  is the same when  $N$  is replaced by  $(N + 1)$ .

For the purpose of illustration, a numerical example for the separation of water and deuterium oxide in various columns is given:  $\alpha = 0.0184$ ,  $\bar{\beta} = 5 \times 10^{-3} \text{ g} \cdot \text{cm}^{-3} \cdot \text{K}^{-1}$ ,  $g = 10^3 \text{ cm/s}^2$ ,  $\bar{\rho} = 1 \text{ g/cm}^3$ ,  $D = 5 \times 10^{-5} \text{ cm}^2/\text{s}$ ,  $\Delta T = 60 \text{ K}$ ,  $L = 500 \text{ cm}$ ,  $B = 10 \text{ cm}$ ,  $\mu = 1.2 \text{ cP}$ ,  $2\omega = 0.1 \text{ cm}$ ,  $\bar{T} = 330 \text{ K}$  (15).

Using these values, the optimal velocity profiles and the maximum separations were calculated for  $N = 0, 1, 2$ , and  $4$ . The results are presented in Tables 1, 2, and 3. It was found that the best velocity profile for the highest degree of separation in an ideal column was readily obtained when

TABLE 1  
Separation When  $N = 0$  (special case: the Clusius-Dickel column) and Maximum Separation When  $N = 1$  (special case: the vertical moving-wall column)

$\sigma \times 10^4$ (g/s)	$\Delta_0(\Delta_c)$ (%)	$ \Delta_1 _{\max}( \Delta_m _{\max})$ (%)
12	0.420	1.582
9	0.421	1.612
6	0.421	1.643
3	0.421	1.675
1	0.421	1.698

$N = 4$  was taken in Eq. (30). The same results were also obtained when  $N > 4$  was taken. For lower flow-rate operation, however, it was observed that a larger number for  $N$  is needed to achieve high precision.

### VELOCITY PROFILES IN SOME IMPROVED COLUMNS

It is interesting to find that the optimal velocity profiles in some improved columns may be written in the following form, similar to Eq. (32):

$$v_{z,\text{opt}} = A_0(1 - \eta^2) + A_1(\eta - \eta^3) + \sum_{n=1}^M a_{n,\text{opt}} \eta^{2n-1}, \quad M < N \quad (34)$$

Let us take the flat-plate columns for example (13).

TABLE 2  
Optimal Velocity Profiles and Maximum Separations When  $N = 2$  (special case: the inclined moving-wall column),  $v_{z,\text{opt}} = 1.5\sigma(1 - \eta^2) + 5.21(\eta - \eta^3) + a_{1,\text{opt}}\eta + a_{2,\text{opt}}\eta^3 = 1.5\sigma(1 - \eta^2) + (5.21 \cos \theta_{\text{opt}})(\eta - \eta^3) + V_{\text{opt}}\eta$

$\sigma \times 10^4$ (g/s)	$a_{1,\text{opt}}$ (cm/s)	$a_{2,\text{opt}}$ (cm/s)	$\theta_{\text{opt}}$ (degree)	$V_{\text{opt}}$ (cm/s)	$ \Delta_2 _{\max}$ (%)	$ \Delta_{i-m} _{\max}$ (%)
12	-4.96	4.46	81.65	-0.50	3.328	3.326
9	-5.00	4.57	82.66	-0.44	3.843	3.840
6	-5.04	4.69	84.08	-0.36	4.706	4.703
3	-5.09	4.84	85.70	-0.26	6.654	6.651
1	-5.14	5.00	87.61	-0.15	11.513	11.520

TABLE 3

The Optimal Velocity Profiles and Maximum Separations When  $N = 4$  (the ideal column),  
 $v_{z,\text{opt}} = 1.5\sigma(1 - \eta^2) + 5.21(\eta - \eta^3) + a_{1,\text{opt}}\eta + a_{2,\text{opt}}\eta^3 + a_{3,\text{opt}}\eta^5 + a_{4,\text{opt}}\eta^7$

$\sigma \times 10^4$ (g/s)	$a_{1,\text{opt}}$ (cm/s)	$a_{2,\text{opt}}$ (cm/s)	$a_{3,\text{opt}}$ (cm/s)	$a_{4,\text{opt}}$ (cm/s)	$ \Delta_{\text{ideal}} $ (%)
12	-5.10	4.02	4.03	-3.98	3.392
9	-4.95	1.92	9.89	-8.01	3.933
6	-5.14	4.08	4.11	-3.90	4.808
3	-5.17	4.13	4.17	-3.84	6.805
1	-5.14	4.14	3.35	-2.77	11.782

### (1) Vertical Column with Moving Walls

The optimal velocity is

$$v_{z,\text{opt}} = A_0(1 - \eta^2) + A_1(\eta - \eta^3) + V_{\text{opt}}\eta \quad (35)$$

Hence, comparing Eq. (35) with Eq. (34), we have  $M = 1$  and

$$a_{1,\text{opt}} = V_{\text{opt}} \quad (36)$$

### (2) Inclined Column with Moving Walls

The optimal velocity can be rewritten as

$$\begin{aligned} v_{z,\text{opt}} &= A_0(1 - \eta^2) + A_1 \cos \theta_{\text{opt}}(\eta - \eta^3) + v_{\text{opt}}\eta \\ &= A_0(1 - \eta^2) + A_1(\eta - \eta^3) + [A_1(\cos \theta_{\text{opt}} - 1) + V_{\text{opt}}]\eta \\ &\quad + [-A_1(\cos \theta_{\text{opt}} - 1)]\eta^3 \end{aligned} \quad (37)$$

Thus, comparing Eq. (37) with Eq. (34), one obtains  $M = 2$  and

$$a_{1,\text{opt}} = A_1(\cos \theta_{\text{opt}} - 1) + V_{\text{opt}} \quad (38)$$

$$a_{2,\text{opt}} = -A_1(\cos \theta_{\text{opt}} - 1) \quad (39)$$

By using the same numerical values given in the preceding section, the optimal conditions,  $\theta_{\text{opt}}$  and  $V_{\text{opt}}$ , and maximum separations obtained in

an inclined moving-wall column were also calculated by the following equations (13):

$$V_{\text{opt}} = 0.516 \left( \sqrt{\frac{630D}{2}} \right) \sqrt{\frac{\sigma L}{2Kd}} \quad (40)$$

$$\theta_{\text{opt}} = \cos^{-1} \left[ 1.55 \sqrt{\frac{\sigma L}{2K_0}} \right] \quad (41)$$

$$|\Delta_{i-m}|_{\text{max}} = 0.184 \sqrt{\frac{2H_0^2 L}{\sigma K_0}} \quad (42)$$

The results are also presented in Table 2 for comparison. It is seen from the table that Eqs. (38) and (39) do hold, and that  $|\Delta_2|_{\text{max}} \cong |\Delta_{i-m}|_{\text{max}}$ .

### DISCUSSION

It is shown in Tables 1 and 2 that the vertical moving-wall column is a special case of the quasi-ideal column with  $N = M = 1$  for  $|\Delta_m|_{\text{max}} = |\Delta_1|_{\text{max}}$ , while the inclined moving-wall column is another special case of the quasi-ideal column with  $N = M = 2$  for  $|\Delta_{i-m}|_{\text{max}} = |\Delta_2|_{\text{max}}$ . The separation efficiency of various columns based on the ideal column may be defined as

$$E = \frac{|\Delta|}{|\Delta_{\text{ideal}}|} \times 100\% \quad (43)$$

Table 4 shows the separation efficiencies calculated from Eq. (43). Since the separation efficiencies of the inclined moving-wall column reach approximately 98%, the inclined moving-wall column may be considered as

TABLE 4  
The Separation Efficiencies Based on an Ideal Column,  $E = |\Delta|/|\Delta_{\text{ideal}}|$

$\sigma \times 10^4$ (g/s)	$E_0$ (%)	$E_m$ (%)	$E_{i-m}$ (%)
12	12.4	46.6	98.1
9	10.7	41.0	97.6
6	8.8	34.2	97.8
3	6.2	24.6	97.7
1	3.6	14.4	97.8

an ideal column for the conditions given in the present example, and it therefore indicates that there exists no better improved column. However, for systems other than  $H_2O-D_2O$ , or for other operating and design conditions, some better improved columns may still be developed.

### CONCLUSIONS

On the basis of the results of this study, the following conclusions are reached.

- (1) An ideal thermal diffusion column in which the velocity profile is adjusted properly for securing the highest degree of separation has been defined.
- (2) The separation theory of the ideal column has been derived, and the velocity profile and the equation of separation are presented in Eqs. (32) and (33), respectively.
- (3) Because the ideal gas and the ideal stage do not exist in reality, the ideal column may be employed to examine if there is the possibility of better columns which have not yet been developed for certain operating and design conditions.
- (4) In the given illustration, the best velocity profile for the highest degree of separation in an ideal column is obtained when  $N = 4$  is used in Eq. (30). For a relatively low flow-rate operation, however, a higher number for  $N$  is needed to achieve high precision.

### SYMBOLS

$A_0, A_1$	constant defined by Eqs. (22) and (23)
$a_n$	constant defined by Eq. (30)
$B$	column width of flat-plate column (cm)
$C$	fractional mass concentration of Component 1 in a binary mixture
$\bar{C}$	fractional mass concentration of Component 2, $1 - C$
$C_B, C_F, C_T$	$C$ in bottom product, feed stream, and top product
$C_b$	bulk concentration defined by Eq. (12)
$C_0(0, Z)$	fractional mass concentration of Component 1 at $x = 0$
$D$	mass diffusivity ( $cm^2/s$ )
$f(\eta)$	the excess velocity over that in the Clusius-Dickel column
$g$	gravitational acceleration ( $cm/s^2$ )
$H$	transport coefficient defined by Eq. (10) or Eq. (24) ( $g/s$ )
$J_x$	mass flux of Component 1 in the $x$ -direction [ $g/(s \cdot cm^2)$ ]
$J_{x-OD}$	mass flux of Component 1 in the $x$ -direction due to ordinary diffusion [ $g/(s \cdot cm)$ ]

$J_{x\text{-TD}}$	mass flux of Component 1 in the $x$ -direction due to thermal diffusion [ $\text{g}/(\text{s}\cdot\text{cm})$ ]
$J_{z\text{-OD}}$	mass flux of Component 1 in the $z$ -direction due to ordinary diffusion [ $\text{g}/(\text{s}\cdot\text{cm})$ ]
$K, K_0, K_1$	transport coefficients defined by Eq. (11) or Eq. (25) [ $(\text{g}\cdot\text{cm})/\text{s}$ ]
$L$	length of flat-plate column (cm)
$M, N$	positive integer in Eqs. (30) and (34), $M < N$
$T$	absolute temperature (K)
$\bar{T}$	reference temperature (K)
$\Delta T$	difference in temperature of hot and cold surfaces (K)
$v_z(x)$	general velocity distribution function of fluid flowing in the $z$ -direction (cm/s)
$v_{z,\text{opt}}$	optimal velocity profile in improved column (cm/s)
$v_{z,\text{best}}$	the best velocity profile in ideal column (cm/s)
$v_{\text{opt}}$	optimal wall velocity of moving-wall column (cm/s)
$x$	axis normal to hot and cold surface (cm)
$z$	axis parallel to transport direction (cm)

### Greek Letters

$\alpha$	thermal diffusion constant
$\beta$	$-(\partial \rho / \partial T)_p$ evaluated at $\bar{T}$ [ $\text{g}/(\text{cm}^3\cdot\text{K})$ ]
$\Delta$	$C_T - C_B$
$\Delta_c, \Delta_0$	$\Delta$ obtained in the Clusius-Dickel column, or in the column when all $a$ 's are set equal to zero
$ \Delta_n _{\text{max}}$	maximum $ \Delta $ , $n = 1, 2, \dots, N$
$\Delta_{\text{ideal}}$	$\Delta$ obtained in ideal column
$ \Delta_m _{\text{max}}$	maximum $\Delta$ obtained in vertical moving-wall column
$ \Delta_{i-m} _{\text{max}}$	maximum $\Delta$ obtained in inclined moving-wall column
$\eta$	$x/W$
$\theta_{\text{opt}}$	optimal angle of inclination in flat-plate column (degree)
$\mu$	absolute viscosity (cP)
$\bar{\rho}$	mass density evaluated at $\bar{T}$ ( $\text{g}/\text{cm}^3$ )
$\tau_1$	transport of Component 1 along $z$ -direction (g/s)
$\omega$	one-half the distance between hot and cold walls (cm)
$\sigma$	mass flow rate (g/s)

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