

# Thermal Separation Column with Vertical Barriers. I. Analytical Studies on the Thermal Separation Column Having Vertical Barriers

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The effect of installing a couple of vertical flow barriers inside a double cylinder-type thermal separation column has been studied analytically. The method of analysis is entirely similar to that developed by Jones and Furry. The only change is that an additional parameter, which specifies the location of the barriers, has been introduced. The results indicated that the installation of barriers retards the convective velocity and leads to a remarkable increase in the equilibrium separation. With the optimum design, which is achieved by locating barriers at 5/8 parts of the half-width of the wall spacing, the logarithmic equilibrium separation will become 4 times greater than that in the open column.

The use of thermal diffusion phenomena is practical enrichment of one component from a gaseous mixture was established by Clusius and Dickel<sup>1)</sup> in 1938. The method is often useful in certain restricted purposes; however, it is still necessary to improve the operational characteristics of the Clusius-Dickel column for a wider application of the method.

Accordingly, various ideas have been introduced for improving the column efficiency. For instance, tilting the column,<sup>9-11)</sup> use of column packing<sup>12,13)</sup> or spiral wire<sup>14,2)</sup> inside the annular space, and columns with moving wall<sup>10,13)</sup> have been examined with more or less satisfactory results. It is interesting that almost all of these attempts for improving the thermal separation unit were made with liquid mixtures.

In contrast, most experiments with gaseous mixtures so far reported have been conducted by using the Clusius-Dickel column in its original form. In many papers of gaseous separation, attention was focused on the effectiveness of spacers, which are usually installed in a Clusius-Dickel type column in order to fix the central heating wire at a proper position. The first use of such spacers is seen in the work of Clusius and Dickel themselves in 1939.<sup>3)</sup> In the experiment of separating an isotopic mixture of hydrogen chloride, they found that the introduction of spacers resulted in an increase of the separation efficiency. Later, Donaldson and Watson<sup>4)</sup> found that simple wire crosses which were welded perpendicularly on the heating wire at given regular distances significantly increased the separation efficiency. According to Corbett and Watson,<sup>5)</sup> however, the effect of spacers was insignificant and even disadvantageous when too many spacers were installed. Although discussion on the effectiveness of spacers have continued up to the present,<sup>6,7)</sup> no unified theory seems to have been accepted.

In such a situation, the work of Treacy and Rich<sup>15)</sup> seems exceptional. They examined the effect of installing several types of flow barriers inside the working space of the separation column. According to them, introduction of barriers usually resulted in the increase of both the magnitude and the rate of separation. Their work seems, however, unsystematic and lacks theoretical support.

In the following part of this paper, we shall examine the possibility of improving a thermal separation column by installing a couple of vertical flow barriers

inside the column space.

## Basic Idea and Analytical Approach

**Basic Idea.** As is well known, in a typical Clusius-Dickel apparatus, the velocity profile of vertical current has a maximum at a point having a finite distance from the wall surface. On the other hand, the maximum temperature difference, which is the driving force of the thermal diffusion, is attained undoubtedly at the wall surface. It is apparent that the convective flow arises in an inefficient way. If we are able to move the position of maximum convective velocity towards the wall surface, the efficiency will increase. This was our basic idea for the present study. We further thought that a vertical barrier will serve to separate functionally the column space to allow convection and thermal diffusion separately and this will lead to improvement of the column efficiency.

**Analytical Approach-Modification of the Theory of Jones and Furry.**<sup>8)</sup> Transport Equation by Jones and Furry: Jones and Furry<sup>8)</sup> developed a theory for the thermal separation in a Clusius-Dickel type column. Assuming the separation column to be a plane parallel type, they derived the following fundamental equations for the total transport of the lighter component,  $\tau_1$ , in the upward direction:

$$\tau_1 = wB\rho\bar{v}(\alpha\Delta T/2\bar{T})c_1c_2 - \{wB\rho\bar{v}(\bar{v}w^2/D) + 2wB\rho D\} \frac{\partial c_1}{\partial z} \quad (1), \text{ (Eq. (52), Ref. 8)}$$

Alternatively,

$$\tau_1 = Hc_1(1-c_1) - (K_c + K_d) \frac{dc_1}{dz} \quad (2), \text{ (Eq. (53), Ref. 8)}$$

with the following abbreviations,

$$H = \frac{\alpha\rho^2 g w^3 B}{96\eta} \left( \frac{\Delta T}{\bar{T}} \right)^2 \quad (3), \text{ (Eq. (54), Ref. 8)}$$

$$K_c = \frac{\rho^3 g^2 w^7 B}{2304\eta^2 D} \left( \frac{\Delta T}{\bar{T}} \right)^2 \quad (4), \text{ (Eq. (55), Ref. 8)}$$

and

$$K_d = 2wB\rho D \quad (5), \text{ (Eq. (56), Ref. 8)}$$

**Modification of the Theory:** An entirely similar treatment can be applied to the present case, where a couple of plane parallel screens are installed vertically inside the column. We shall describe the procedure shortly. Prior to developing this analysis, we suppose the conditions of column operation to be as follows.

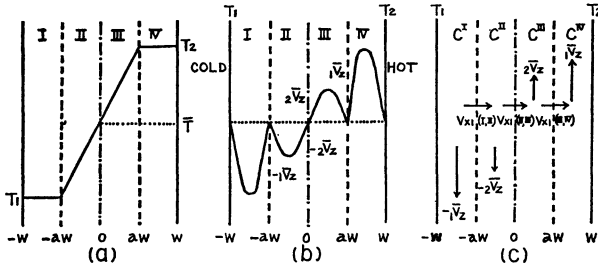


Fig. 1. Assumptions and notations for a column which equipped a couple of screens.

(a): Temperature gradient. (b) Convective flow velocity as a function of  $x$ . (c) Notations.

1. The apparatus is of the plane parallel type having an interwall distance of  $2w$ .

2. Two screens are placed at positions  $\pm aw$  parallel to the column wall.

3. A temperature gradient is set up between the pair of screens and the two gaps produced outside the screens,  $|aw| < x < |w|$ , are kept isothermal.<sup>17)</sup>

4. The velocity of vertical gas flow is zero at  $x = \pm aw$  where the two screens are located.

Accordingly, the space in the column are divided vertically into four sub-columns: column I ( $-w \leq x \leq -aw$ ), II ( $-aw \leq x \leq 0$ ), III ( $0 \leq x \leq aw$ ), and IV ( $aw \leq x \leq w$ ). The symbols and coordinates necessary for the analytical treatment are given in Fig. 1 and are also listed at the end of this article. Unless otherwise stated, the symbols used by Jones and Furry will be used.

On account of the above assumptions, the basic equation to be solved for the region of  $-aw < x < aw$  becomes

$$\eta \frac{d^2 v_z(x)}{dx^2} = -\rho g \frac{x \Delta T}{2aw \bar{T}} \quad (6)$$

Under the boundary condition

$$\text{at } x = \pm aw; \quad v_z(x) = 0$$

Eq. (6) is solved to give

$$v_z = \frac{\rho g \Delta T}{12 \eta aw \bar{T}} \times (a^2 w^2 - x^2) \quad (7)$$

Denoting the average velocity of convective flow in columns II and III as  $\bar{v}_z$ , we obtain

$$\bar{v}_z = \frac{1}{aw} \int_0^{aw} v_z(x) dx = \frac{\rho g w^2}{48 \eta} \frac{\Delta T}{\bar{T}} a^2 \quad (8)$$

On the other hand, in column IV, where the region is specified by  $aw \leq x \leq w$ , the temperature must be the same everywhere, by assumption, and is given by

$$T = T_2 = \bar{T} + \Delta T/2 \quad (9)$$

The convective flow in column IV may be expressed as

$$\eta \frac{d^2 v_z(x)}{dx^2} = -\rho g \frac{\Delta T}{2 \bar{T}} \quad (10)$$

This equation is solved under the boundary condition

$$v_z = 0 \text{ at } x = aw \text{ and } x = w$$

to give the average vertical velocity in column IV (and also in column I)

$$\bar{v}_z = \frac{\rho g w^2}{48 \eta} \frac{\Delta T}{\bar{T}} \cdot 2(1-a)^2 \quad (11)$$

Since there will be a transport down the tube because of ordinary diffusion, the total transport of the lighter component up the tube will be

$$\tau_1' = \{(c_1^{IV} - c_1^I)(1-\alpha)_1 \bar{v}_z + (c_1^{III} - c_1^I) a_2 \bar{v}_z\} \omega B \rho - 2DwB\rho \frac{\partial c_1}{\partial z} \quad (12)$$

where  $c_1^J$  stands for the average concentration of the lighter component in column  $J$ .

For the transverse flow, we have the following set of approximate equations:

$$\rho c_1 v_{x1}(I, II) = -\rho D \text{ grad } c_1 = -\frac{2(c_1^{II} - c_1^I) \rho D}{w} \quad (13)$$

$$\begin{aligned} \rho c_1 v_{x1}(II, III) &= \rho D (\alpha c_1 c_2 \text{ grad } \ln T - \text{grad } c_1) \\ &= \rho D \left( \alpha c_1 c_2 \frac{\Delta T}{2aw \bar{T}} - \frac{c_1^{III} - c_1^{II}}{aw} \right) \end{aligned} \quad (14)$$

$$\rho c_1 v_{x1}(III, IV) = -\rho D \text{ grad } c_1 = -\frac{2(c_1^{IV} - c_1^{III}) \rho D}{w} \quad (15)$$

where  $v_{x1}(H, J)$  represents the rate of the horizontal transport of the lighter component from column  $H$  to  $J$ .

If we assume the concentration change from column to column is so small that  $c_1^I$ ,  $c_1^{II}$ ,  $c_1^{III}$ , and  $c_1^{IV}$  can be safely replaced by  $c_1$ , the terms  $(c_1^{IV} - c_1^I)$  and  $(c_1^{III} - c_1^{II})$  appearing in Eq. (12) can be evaluated by the aid of Eqs. (13) through (15) to give

$$c_1^{III} - c_1^I = \alpha c_1 c_2 \frac{\Delta T}{2 \bar{T}} + \frac{w^2}{D} \{(a^2 - a)_1 \bar{v}_z - a^2 \bar{v}_z\} \frac{\partial c_1}{\partial z} \quad (16)$$

$$c_1^{IV} - c_1^I = \alpha c_1 c_2 \frac{\Delta T}{2 \bar{T}} + \frac{w^2}{D} \{(a^2 - 1)_1 \bar{v}_z - a^2 \bar{v}_z\} \frac{\partial c_1}{\partial z} \quad (17)$$

Accordingly, Eq. (12) is now rewritten in the form of Eq. (2)

$$\tau_1' = H' c_1 c_2 - (K_c' + K_d') \frac{dc_1}{dz} \quad (18)$$

Where the constants are defined as follows:

$$\begin{aligned} H' &= \frac{\alpha \rho g w^3 B}{96 \eta} \left( \frac{\Delta T}{\bar{T}} \right)^2 \{2(1-a)^3 + a^3\} \\ &= \{2(1-a)^3 + a^3\} H \end{aligned} \quad (19)$$

$$\begin{aligned} K_c' &= \frac{\rho^3 g^2 w^7 B}{2304 \eta^2 D} \left( \frac{\Delta T}{\bar{T}} \right)^2 \{4(1-a)^6(1+a) + 4a^4(1-a)^3 + a^7\} \\ &= \{4(1-a)^6(1+a) + 4a^4(1-a)^3 + a^7\} K_c \end{aligned} \quad (20)$$

$$K_d' = K_d \quad (21)$$

### Operational Characteristics and Remarks

Several parameters which are important for describing the operational characteristics of the separation column have already been given by Jones and Furry.<sup>8)</sup> We shall reproduce some of the results necessary in the following discussion.

The equilibrium separation factor,  $q_e$ , is given by

$$q_e = e^{2AL} \quad (22)$$

where the parameter  $A$  is defined as

$$A = H/2(K_c + K_d) \quad (23)$$

and  $L$  is the length of the separation column.

We now examine the separation characteristics of

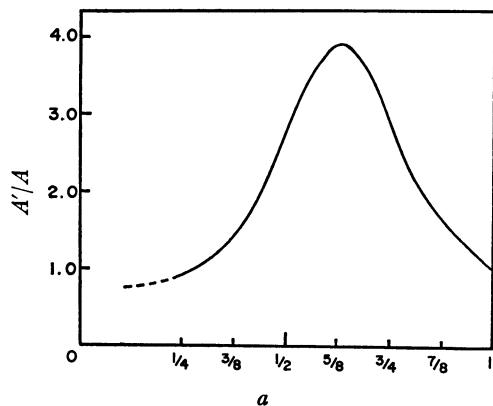


Fig. 2. The relative performance,  $A'/A$ , against design parameter  $a$ .

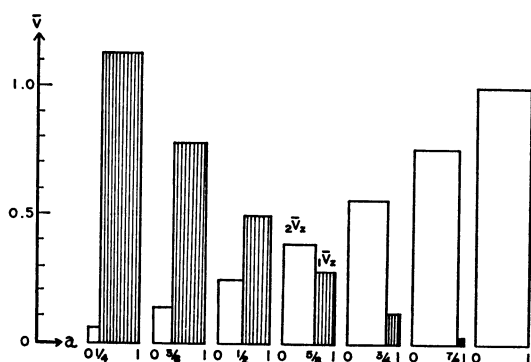


Fig. 3. Change in average convective velocity in sub-columns I and IV ( ${}_1\bar{v}_z$ ) and II and III ( ${}_2\bar{v}_z$ ) with various value of design parameter  $a$ . Hatched bar represents  ${}_1\bar{v}_z$  and empty bar  ${}_2\bar{v}_z$ , respectively.

columns with and without screens. For simplicity, we suppose that the value of  $K_c$  is sufficiently greater than  $K_d$ , so that the contribution of the latter can be ignored in the expression of  $A$  in Eq. (23). As has been done already in Eqs. (18) through (21), the symbols corresponding to the column with screens will be primed to distinguish them from the ordinary column symbols. When  $K_d$  is ignorable, the change in  $A'$  with varying  $a$  can be expressed by a simple analytical function

$$\frac{A'}{A} = \frac{H/2K_c'}{H/2K_c} = \frac{2(1-a)^3 + a^3}{4(1-a)^6(1+a) + 4a^4(1-a)^3 + a^7} \quad (24)$$

this function is graphically represented in Fig. 2. It is seen that the use of screens significantly increases the equilibrium separation factor. In order to see what was the cause of this improvement, we prepared Fig. 3, where the average velocity of convective flow is indicated as a function of the design parameter  $a$ . It is indisputable that the vertical movement of gas is suppressed by the installation of screens and the suppression becomes maximum at about  $a=5/8$ , where the separation becomes also maximum. This is predictable analytically also in the following manner. Eqs. (3) and (4) can be rewritten in terms of  $\bar{v}$

$$H = \frac{1}{2} \alpha \bar{\rho} w B \bar{v} \left( \frac{\Delta T}{T} \right)$$

$$K_c = \frac{\bar{\rho} w^2 B \bar{v}^2}{D}$$

Accordingly, if  $K_d$  is ignored,

$$\ln q_0 = \frac{HL}{K_c} = \frac{\alpha DL(\Delta T/T)}{2w^2 \bar{v}}$$

The last equation indicates that  $q_0$  will increase when  $\bar{v}$  decreases. Accordingly, our original idea, which anticipated an improvement by shifting the position of maximum vertical velocity towards the column wall, must be said to have been not wholly correct.

Treacy and Rich<sup>15)</sup> studied the effect of barriers introduced in a double-cylinder type separation column. They found that the use of compound barriers made by cutting the vertical into sections of length suitable for insertion between horizontals exhibits a pronounced increase of column efficiency. Although they state that use of single vertical barriers is still effective for increasing efficiency, the data shown in the paper are not convincing. This is quite natural because they ignored the importance of the location of the barriers. In order to prove the result of the foregoing analysis to be acceptable, we need some experimental verification. This will be reported in a following paper.<sup>16)</sup>

### Conclusion

It was confirmed analytically that the introduction of a couple of vertical barriers in a thermal separation column should increase the separation significantly. The improvement is rather sensitive to the design parameter  $a$  and the maximum improvement is expected at about  $a=5/8$ , where a four times greater value can be expected in  $\ln q_0$  as compared with the ordinary column.

At the end of this article, it should be noted that in the foregoing treatment we have used somewhat particular boundary conditions according to which the temperature gradient exists only over the region specified by  $-aw \leq x \leq aw$ . This was necessary because of mathematical simplification. In an actual column, however, a finite temperature gradient may be unavoidable over the regions outside the screens. Errors due to this assumption will be larger for smaller values of design parameter  $a$ . This point will be examined in a future paper.

### Table of Notation

$A = H/2(K_c + K_d)$ , [cm <sup>-1</sup> ].
$a$ = design parameter, $0 < a < 1$ .
$B$ = mean circumference, [cm].
$c$ = either $c_1$ or $c_2$ .
$\bar{c} = 1 - c$ .
$c_1$ = fractional molar concentration of the lighter component.
$c_2$ = fractional molar concentration of the heavier component. $c_1 + c_2 = 1$ .
$c_1^J$ = average concentration of the lighter component in column $J$ .
$D$ = coefficient of self-diffusion, [cm <sup>2</sup> /s].
$g$ = acceleration of gravity, [cm/s <sup>2</sup> ].
$H$ , [g/s], $K_c$ , $K_d$ , [g cm/s] = transport coefficient.
$L$ = length of column, $0 \leq z \leq L$ , [cm].

- $q$ =separation factor.  
 $q_0=e^{2AL}$ =equilibrium separation factor.  
 $T$ =absolute temperature, [K].  
 $T_1$ =temperature of cold wall [K].  
 $T_2$ =temperature of hot wall,  $T_2>T_1$ , [K].  
 $\bar{T}=(T_1+T_2)/2$ , [K].  
 $\Delta T=T_2-T_1$ , [K].  
 $v$ =velocity of convectional flow, [cm/s].  
 $\bar{v}$ =average velocity of convectional flow, [cm/s].  
 ${}_1\bar{v}_2$ =average velocity of convectional flow in columns I and IV, [cm/s].  
 ${}_2\bar{v}_2$ =average velocity of convectional flow in columns II and III, [cm/s].  
 $v_{x1}(H,J)$ =rate of the horizontal transport of the lighter component from column  $H$  to  $J$ , [cm/s].  
 $w$ =one-half of the distance between the hot and cold walls, [cm].  
 $x$ =coordinate perpendicular to the walls,  $-w\leq x\leq w$ , [cm].  
 $z$ =coordinate along the column,  $0\leq x\leq L$ , [cm].  
 $\alpha$ =thermal diffusion constant.  
 $\eta$ =viscosity, [P].  
 $\rho$ =density, [g/cm<sup>3</sup>].  
 $\tau_1$ =transport of species 1 along the column, mass per unit time, [g/s].

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- 16) K. Sasaki, N. Miura, and T. Yoshitomi, *This Bulletin*, **49**, 367 (1976).
- 17) This assumption is necessary only for mathematical simplicity. In an actual column, however, there may be a finite temperature gradient in the gaps concerned. Because of this assumption, the question may arise whether "The comparison should be made with an open column having width of  $2aw$  but not of  $2w$ ". In fact, there may be two different views with regard to the installation of barriers: 1) screens are merely inserted in the column space of width of  $2w$  and 2) additional spaces are produced outside the pair of screens. If, for an actual column, a finite temperature gradient is unavoidable over spaces outside the screens, the former view must be accepted. In the following part of this paper and also in the successive paper, the discussion has been developed on this standpoint.