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The Improvement of Separation Theory in a Continuous Thermal Diffusion Column

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Abstract

A more precise equation of separation applicable to the whole concentration range in a continuous thermal diffusion column with the feed introduced from any position has been derived. The results are also represented graphically and compared with those obtained by Powers in which he, as well as almost all previous investigators, considered the concentration at the feed position of the column to be approximately the feed concentration.

INTRODUCTION

Thermal diffusion is an unusual process which can be used to separate mixtures that are hard to separate by such conventional methods as distillation and extraction. This phenomena has been known for over one hundred years. Since the early studies of thermal diffusion were all carried out in a single-stage convective-free apparatus (*1*), the degree of separation obtained was so small that this process had nearly no practical value. In 1938, Clusius and Dickel (*3, 4*) devised a thermogravitational thermal diffusion column which acted as a multistage device and thus greatly enhanced the separation efficiency.

The complete theory of separation in a Clusius-Dickel column was first presented by Furry et al. (*5, 7*). However, the exact solution they obtained is so complicated and implicit that it makes analysis very difficult. For this reason, numerous investigators (*2, 5, 6, 8*) considered the product form of concentration, $c(1-c)$, in the transport equations to be constant. Hence, the separation equation derived is very simple and the results obtained under this assumption are valid only for $0.3 < c < 0.7$.

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Furry suggested that this quadratic form of concentration could be linearized to obtain a solution applicable to the whole range of concentration (*5*). Later, Yeh et al. (*14, 17, 18, 22*) derived simple but precise equations applicable to the whole range of concentration in various types of thermal diffusion columns, in a common form, by the linear approximation method coupled with the least-squares method.

Yet, all the previous investigators considered only cases with feed introduced from the center of the column. In 1962, Powers (*9*) derived the separation equation for the column with the feed introduced from any position, in which he, as well as almost all previous workers, considered the concentration at the feed position of the column to be approximately equal to the feed concentration. However, their results do not obey the law of conservation of mass for the entire column. To overcome this defect, Rabinovich (*10*) has derived a very complicated equation by solving two second-order differential equations. Since the solution is very tedious, he treated only some special cases, i.e., very dilute and high-concentration solutions, and nearly equifraction solution, $c(1 - c) \approx 0.25$.

It is the purpose of this work to improve the separation theory of continuous Clusius-Dickel columns for the whole range of concentration with feed introduced from any position of the column. The results obtained in this work can also be extended to concentric-tube thermal diffusion columns (*16*) or improved columns. (*2, 8, 11–13, 15, 19–21, 23, 24*).

COLUMN THEORY

Consider the Clusius-Dickel column with the feed introduced at the position L_s from the bottom of the column. Following the derivation presented by Furry et al. (*5, 7*), we obtain two transport equations at steady state:

$$H\left[c_e(1 - c_e) - \frac{\sigma}{H}(c_T - c_e)\right] = K\frac{dc_e}{dz} \tag{1}$$

for the enriching section and

$$H\left[c_s(1 - c_s) + \frac{\sigma}{H}(c_B - c_s)\right] = K\frac{dc_s}{dz} \tag{2}$$

for the stripping section (symbols are defined at the end of the text). The transport constants in above equations are defined by

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

$$K = \frac{\rho\beta_{\bar{T}}^2 g^2(2\omega)^7 B(\Delta T)^2}{9!\mu^2 D} + 2\omega D B\rho \tag{4}$$

Since Eqs. (1) and (2) are nonlinear, the simultaneous solution of this set of equations is complicated and inconvenient to analyze. Here, we linearize the quadratic terms in the form

$$c(1-c) = a + bc \tag{5}$$

and rewrite Eqs. (1) and (2) as

$$\frac{dc_e}{dz'} - (b_e + \sigma')c_e = a_e - \sigma' c_T \tag{6}$$

$$\frac{dc_s}{dz'} - (b_s - \sigma')c_s = a_s + \sigma' c_B \tag{7}$$

where

$$\sigma' = \sigma/H, \qquad z' = zH/K \tag{8}$$

Then, the solutions of Eqs. (6) and (7) are

$$\Delta_e = c_T - c_i = \frac{(a_e + b_e c_i)\{1 - e^{-(b_e+\sigma')L'(1-\zeta)}\}}{\{\sigma' + b_e e^{-(b_e+\sigma')L'(1-\zeta)}\}} \tag{9}$$

$$\Delta_s = c_i - c_B = \frac{(a_s + b_s c_i)\{1 - e^{(b_s-\sigma')L'\zeta}\}}{\{\sigma' - b_s e^{(b_s-\sigma')L'\zeta}\}} \tag{10}$$

under the boundary conditions:

$$z' = 0, \qquad c_e = c_s = c_i \tag{11}$$

$$= L_e' = (1-\zeta)L', \qquad c_e = c_T \tag{12}$$

$$= -L_s' = -\zeta L', \qquad c_s = c_B \tag{13}$$

Combining Eqs. (9) and (10) gives the separation equation for the whole column

$$\Delta = c_T - c_B = \Delta_e + \Delta_s \tag{14}$$

$$= \frac{(a_e + b_e c_i)\{1 - e^{-(b_e+\sigma')L'(1-\zeta)}\}}{\{\sigma' + b_e e^{-(b_e+\sigma')L'(1-\zeta)}\}} + \frac{(a_s + b_s c_i)\{1 - e^{(b_s-\sigma')L'\zeta}\}}{\{\sigma' - b_s e^{(b_s-\sigma')L'\zeta}\}} \tag{15}$$

where the concentration at the feed position of the column can be determined by writing a material balance around the entire column, i.e.,

$$2\sigma c_f = \sigma c_T + \sigma c_B \tag{16}$$

or

$$c_i = c_f - \frac{(\Delta_e - \Delta_s)}{2} \tag{17}$$

The appropriate values of a_e, a_s, b_e, and b_s may be determined by the method of least squares suggested by Yeh et al. However, the separations obtainable from the enriching and stripping sections are generally different. Therefore, the calculation of these constants must be substantially integrated from c_i to c_T for the enriching section and from c_B to c_i for the stripping section, instead of integrating from $(c_i - \Delta/2)$ to $(c_i + \Delta/2)$ for the entire column as performed by Yeh et al. Accordingly, set

$$\min R_e = \int_{c_i}^{c_T} [c_e(1 - c_e) - (a_e + b_e c_e)]^2 dc_e \tag{18}$$

and

$$\min R_s = \int_{c_B}^{c_i} [c_s(1 - c_s) - (a_s + b_s c_s)]^2 dc_s \tag{19}$$

We obtain

$$a_e = c_i(c_i + \Delta_e) + \frac{1}{6}(\Delta_e)^2 \tag{20}$$

$$b_e = 1 - 2c_i - \Delta_e \tag{21}$$

$$a_s = c_i(c_i - \Delta_s) + \frac{1}{6}(\Delta_s)^2 \tag{22}$$

$$b_s = 1 - 2c_i + \Delta_s \tag{23}$$

Substituting the above equations into Eqs. (9) and (10) and eliminating c_i by Eq. (17) yield

$$\Delta_e = \frac{\left\{c_f(1-c_f) - \frac{(1-2c_f)}{2}(\Delta_e - \Delta_s) - \frac{(\Delta_e - \Delta_s)^2}{4} + \frac{\Delta_e^2}{6}\right\}\{1 - e^{-(1-2c_f-\Delta_s+\sigma')L'(1-\zeta)}\}}{\{\sigma' + (1-2c_f-\Delta_s)e^{-(1-2c_f-\Delta_s+\sigma')L'(1-\zeta)}\}} \tag{24}$$

$$\Delta_s = \frac{\left\{c_f(1-c_f) - \frac{(1-2c_f)}{2}(\Delta_e - \Delta_s) - \frac{(\Delta_e - \Delta_s)^2}{4} + \frac{\Delta_s^2}{6}\right\}\{1 - e^{(1-2c_f+\Delta_e-\sigma')L'\zeta}\}}{\{\sigma' - (1-2c_f+\Delta_e)\, e^{(1-2c_f+\Delta_e-\sigma')L'\zeta}\}} \tag{25}$$

Although Eqs. (24) and (25) are in implicit form, they converge very rapidly by the successive iteration method. Once the values of Δ_e and Δ_s are calculated from Eqs. (24) and (25) simultaneously, the degree of separation obtainable in the whole column may be evaluated from Eq. (14).

Since

$$\Delta_e = \Delta_s\Big|_{\substack{c_f\to\bar{c}_f \\ \zeta\to\bar{\zeta}}} \qquad \Delta_s = \Delta_e\Big|_{\substack{c_f\to\bar{c}_f \\ \zeta\to\bar{\zeta}}} \tag{26}$$

thus, we obtain

$$\Delta\Big|_{\substack{c_f\to\bar{c}_f \\ \zeta\to\bar{\zeta}}} = \Delta_s + \Delta_e = \Delta \tag{27}$$

Consequently, we conclude from Eq. (27) that Δ is symmetric with $c_f = 0.5$ and $\zeta = 0.5$, simultaneously. For example, once the degree of separation for $c_f = 0.3$ and $\zeta = 0.6$ is calculated, the same result will be obtained for $c_f = 0.7$ and $\zeta = 0.4$, i.e.,

$$\Delta\Big|_{\substack{c_f=0.3 \\ \zeta=0.6}} = \Delta\Big|_{\substack{c_f=0.7 \\ \zeta=0.4}}$$

Hence, we only consider the cases with the feed concentration not larger than 0.5. Some graphical results of Eq. (14) are calculated and represented in Figs. 1 through 6 by taking feed concentration, dimensionless column length,

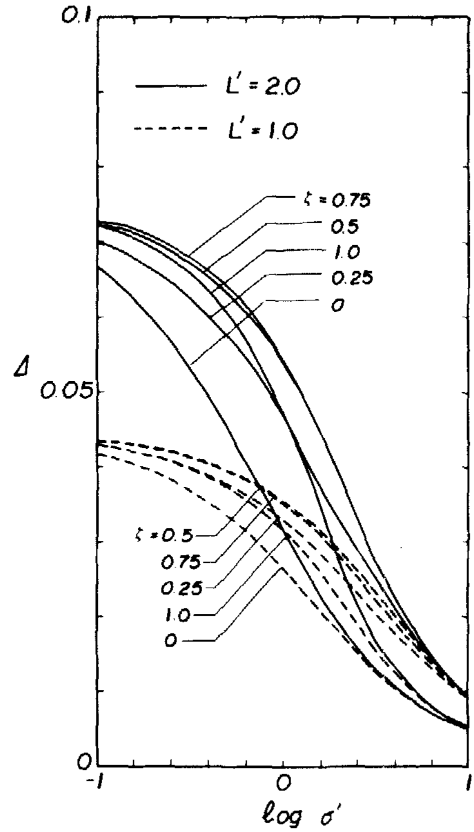

FIG. 1. The degree of separation for $c_f = 0.05$ at various feed positions. For $c_f = 0.95$, ζ is replaced by $\bar{\zeta}$.

and dimensionless feed position as parameters. There are three special cases which will be discussed as follows.

1) If the concentration at the feed position of the column is considered approximately as the feed concentration, i.e., $c_i = c_f$, Eqs. (9) and (10) become

$$\Delta_{e,1} = c_T - c_f = \frac{(a_{e,1} + b_{e,1}c_f)\{1 - e^{-(b_{e,1}+\sigma')L'(1-\zeta)}\}}{\{\sigma' + b_{e,1}e^{-(b_{e,1}+\sigma')L'(1-\zeta)}\}} \tag{28}$$

$$\Delta_{s,1} = c_f - c_B = \frac{(a_{s,1} + b_{s,1}c_f)\{1 - e^{(b_{s,1}-\sigma')L'\zeta}\}}{\{\sigma' - b_{s,1}e^{(b_{s,1}-\sigma')L'\zeta}\}} \tag{29}$$

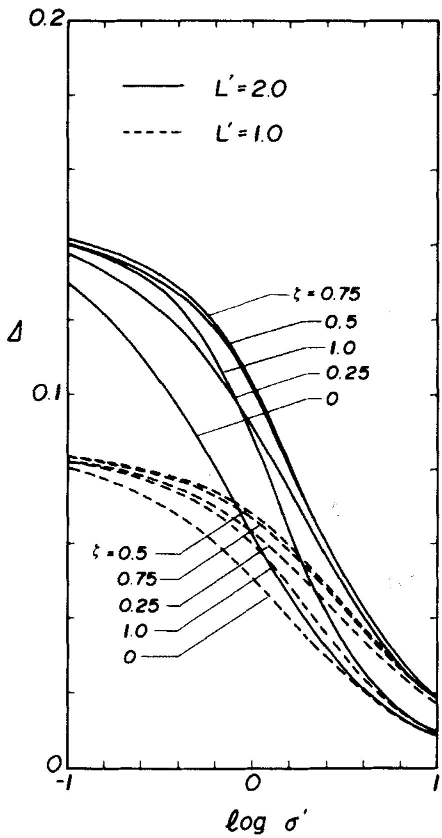

FIG. 2. The degree of separation for $c_f = 0.1$ at various feed positions. For $c_f = 0.9$, ζ is replaced by $\bar{\zeta}$.

The appropriate constants $a_{e,1}$, $a_{s,1}$, $b_{e,1}$, and $b_{s,1}$ may be obtained by following the same procedure performed in Eqs. (18) and (19), except that c_i is replaced by c_f. The results are

$$a_{e,1} = c_f(c_f + \Delta_{e,1}) + \frac{1}{6}(\Delta_{e,1})^2 \tag{30}$$

$$b_{e,1} = 1 - 2c_f - \Delta_{e,1} \tag{31}$$

$$a_{s,1} = c_f(c_f - \Delta_{s,1}) + \frac{1}{6}(\Delta_{s,1})^2 \tag{32}$$

$$b_{s,1} = 1 - 2c_f + \Delta_{s,1} \tag{33}$$

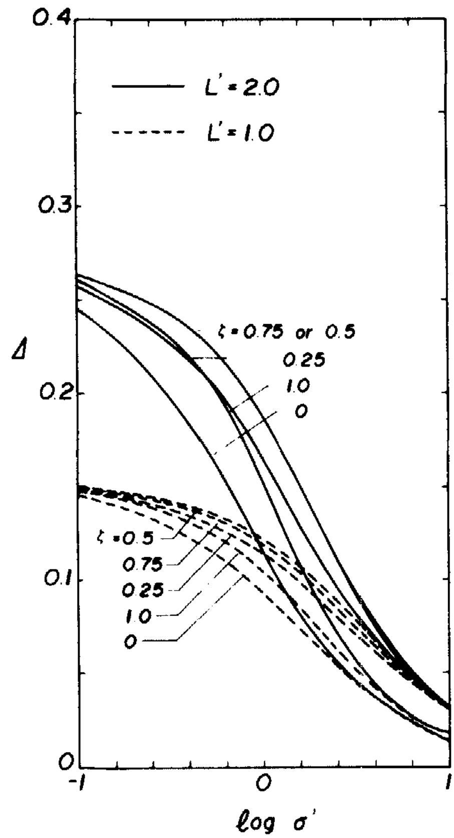

FIG. 3. The degree of separation for $c_f = 0.2$ at various feed positions. For $c_f = 0.8$, ζ is replaced by $\bar{\zeta}$.

Substituting the above equations into Eqs. (28) and (29) results in

$$\Delta_{e,1} = \frac{\left(c_f(1-c_f) + \frac{(\Delta_{e,1})^2}{6}\right)\{1 - e^{-(1-2c_f-\Delta_{e,1}+\sigma')L'(1-\zeta)}\}}{\{\sigma' + (1-2c_f-\Delta_{e,1})e^{-(1-2c_f-\Delta_{e,1}+\sigma')L'(1-\zeta)}\}} \tag{34}$$

$$\Delta_{s,1} = \frac{\left(c_f(1-c_f) + \frac{(\Delta_{s,1})^2}{6}\right)\{1 - e^{(1-2c_f+\Delta_{s,1}-\sigma')L'\zeta}\}}{\{\sigma' - (1-2c_f+\Delta_{s,1})e^{(1-2c_f+\Delta_{s,1}-\sigma')L'\zeta}\}} \tag{35}$$

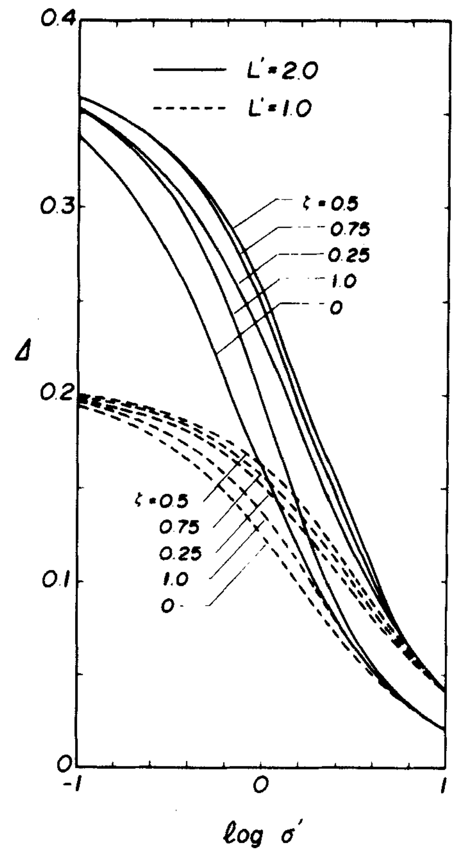

FIG. 4. The degree of separation for $c_f = 0.3$ at various feed positions. For $c_f = 0.7$, ζ is replaced by $\bar{\zeta}$.

and the separation equation for the entire column is

$$\Delta_1 = \Delta_{e,1} + \Delta_{s,1} \tag{36}$$

It is evident from Eq. (17) that Eq. (36) is valid only when $\Delta_e \approx \Delta_s$. It is also shown from Eqs. (34) and (35) that $\Delta_{e,1} \approx \Delta_{s,1}$ as both c_f and ζ approach 0.5. Accordingly, the concentration at the feed position of the column may be considered as the feed concentration if the feed concentration is close to 0.5 and the feed is introduced near the center of the column. Under this circumstance, $\Delta_1 \approx \Delta$. It is easy to show that Eq. (27) holds also for Δ_1.

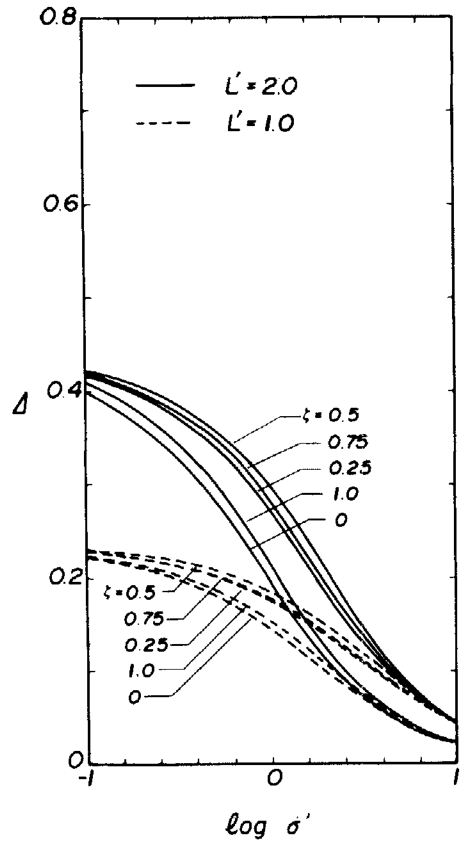

FIG. 5. The degree of separation for $c_f = 0.4$ at various feed positions. For $c_f = 0.6$, ζ is replaced by $\bar{\zeta}$.

2) For the cases with equifraction feed (i.e., $c_f = 0.5$) and small degree of separation, the quadratic term $c(1 - c)$ is approximately equal to 0.25 and we obtain

$$a_{e,2} = a_{s,2} = 0.25 \tag{37}$$

$$b_{e,2} = b_{s,2} = 0 \tag{38}$$

Thus, Eq. (36) reduces to

$$\Delta_2 = \frac{\{2 - e^{-\sigma' L'(1-\zeta)} - e^{-\sigma' L' \zeta}\}}{4\sigma'} \tag{39}$$

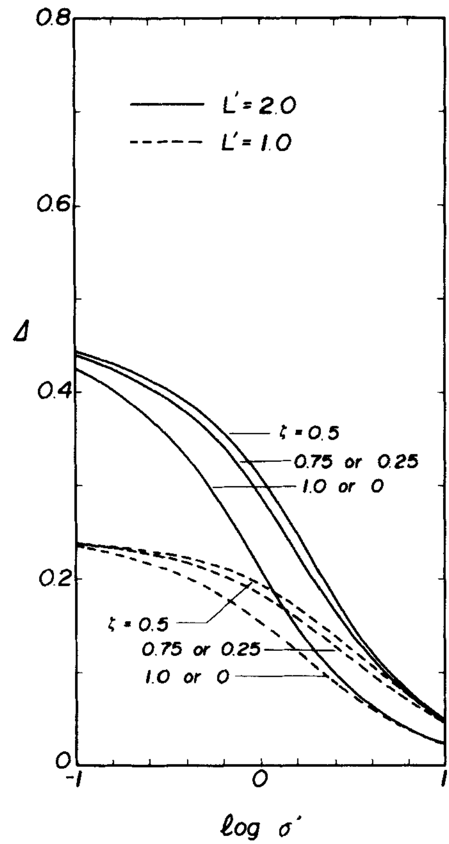

FIG. 6. The degree of separation for $c_f = 0.5$ at various feed positions.

3) In addition, if the feed is introduced from the center of the column, i.e., $\zeta = 0.5$, Eq. (39) can be further simplified as

$$\Delta_3 = \frac{\{1 - e^{-\sigma' L/2}\}}{2\sigma'} \tag{40}$$

which is the separation equation appearing frequently in the literature (*2, 5, 7, 8*).

COMPARISON OF SEPARATION

Since

$$c_T - c_B < c_T + c_B \tag{41}$$

we find, from Eqs. (14) and (16), that

$$\Delta < 2c_f \tag{42}$$

However, when using Eq. (36) to evaluate the degree of separation, we have found a conflict for some cases. That is to say, the degree of separation will be greater than $2c_f$, which indeed violates the conservation of mass for the entire column.

The deviation of Δ_I from Δ is defined as

$$E = \frac{\Delta_I - \Delta}{\Delta} \times 100\% \tag{43}$$

Again, some results of Eq. (43) are calculated and represented graphically in Figs. 7 through 10.

DISCUSSION AND CONCLUSION

On the basis of this study, some discussions and conclusions are reached:

1) The generalized equation of separation, Eq. (14), for thermal diffusion columns with the feed introduced from any position over the whole concentration range has been derived by taking the overall mass balance into consideration.

2) Some graphical solutions of the degree of separation are represented in Figs. 1 through 6 by the successive iteration method. From these figures we found that the effect of feed position on the degree of separation is evident, especially when the dimensionless column length is increased. However, when the dimensionless flow rate σ' is greater than 10, the feed position has nearly no influence on the degree of separation, except for the extreme cases with feed introduced from the bottom or the top of the column.

3) The deviation of Δ_I from Δ is evident as shown in Figs. 7 through 10. This is due to the incorrect assumption that the concentration at the feed position of the column is considered to be approximately the feed concentra-

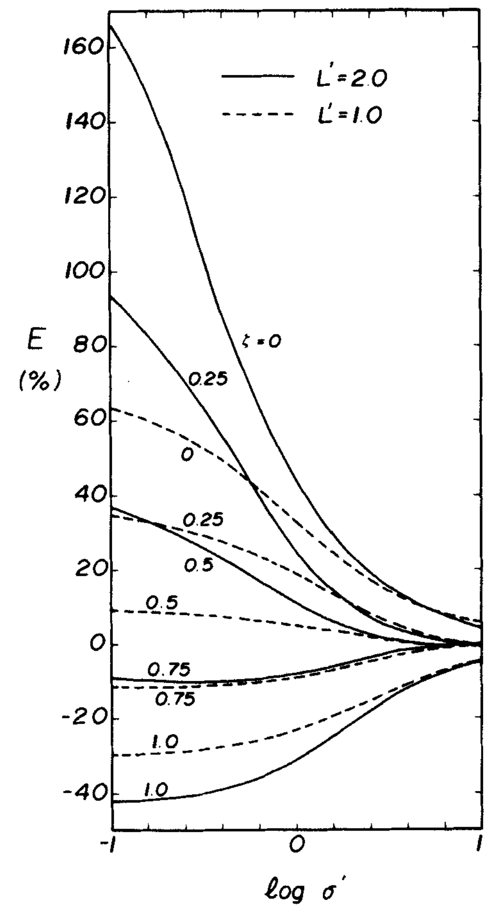

FIG. 7. The deviation of Δ_1 from Δ defined by Eq. (43) for $c_f = 0.05$ or 0.95 at various feed positions.

tion. It is found from these figures that the deviation increases as the dimensionless column length does, or the dimensionless flow rate decreases, or the feed concentration as well as the dimensionless feed position departs from 0.5. In one case, the deviation is more than 160%.

4) Basically, the separation equations for concentric-tube thermal diffusion columns (*7, 16*) or the improved columns (*2, 8, 11–13, 15, 19–22, 23, 24*) are in the same form as Eq. (14) except that transport constants H and K are multiplied by modified factors. Thus, the results obtained in this work

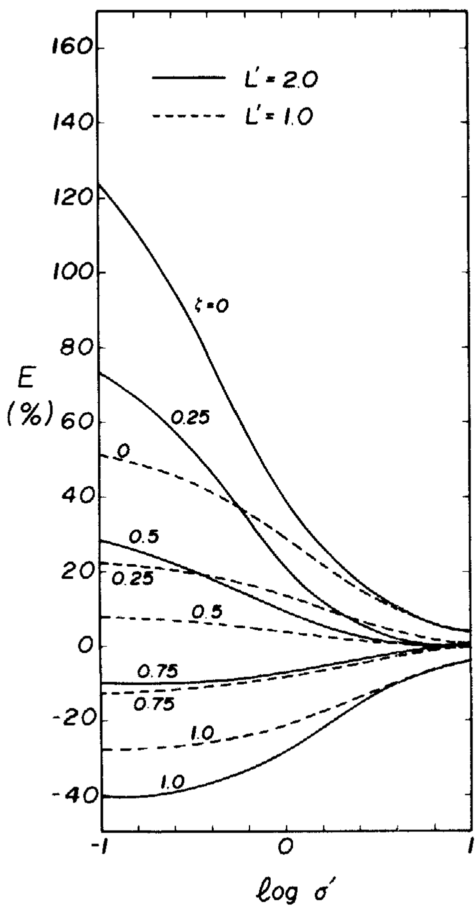

FIG. 8. The deviation of Δ_1 from Δ defined by Eq. (43) for $c_f = 0.1$ or 0.9 at various feed positions.

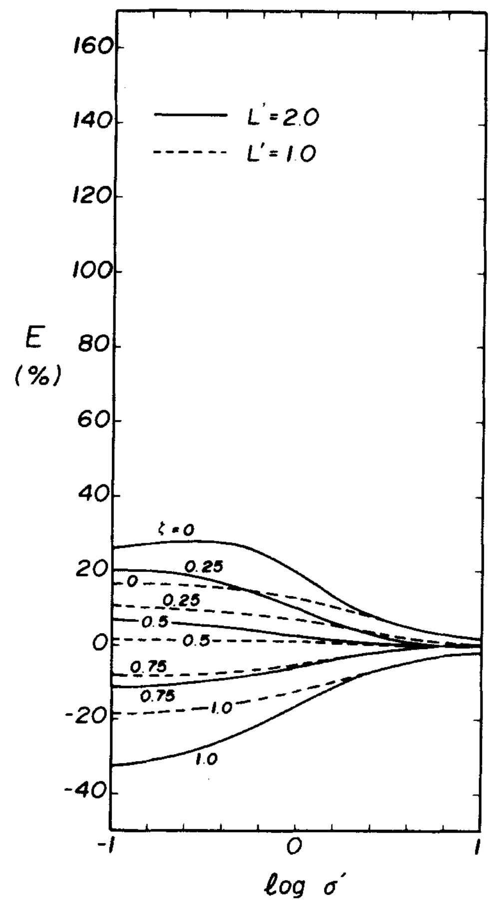

FIG. 9. The deviation of Δ_1 from Δ defined by Eq. (43) for $c_f = 0.3$ or 0.7 at various feed positions.

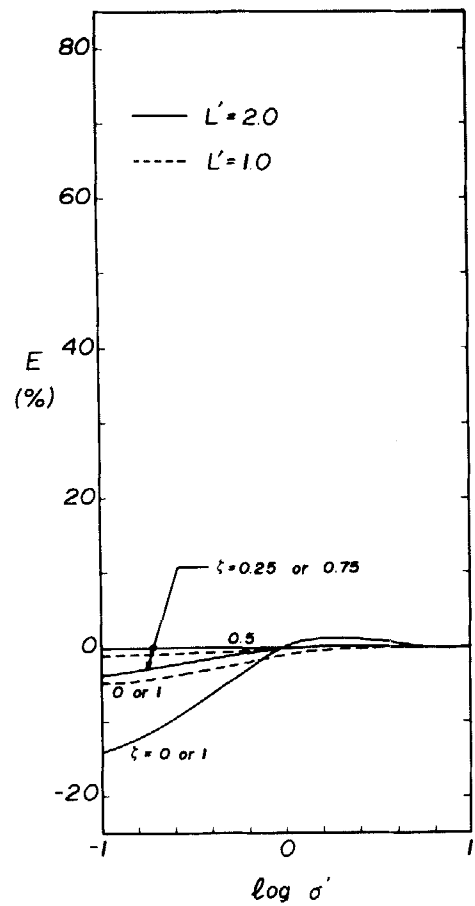

FIG. 10. The deviation of Δ_1 from Δ defined by Eq. (43) for $c_f = 0.5$ at various feed positions.

may be extended to all these columns even if the curvature effect is considered in the concentric-tube columns (*19–21*).

SYMBOLS

a	constant defined by Eq. (5)
B	column width
b	constant defined by Eq. (5)
c	concentration of component 1 in a binary solution

c_B, c_f, c_i, c_T	c in bottom product, feed stream, at the feed position of the column, and in the top product, respectively
$\bar{c}_f$	$= (1 - c_f)$
D	ordinary diffusion coefficient
E	deviation defined by Eq. (43)
g	gravitational acceleration
H	transport constant defined by Eq. (3)
K	transport constant defined by Eq. (4)
L	total column length
L'	dimensionless column length defined as LH/K
R	residue defined in least-squares method
$\bar{T}$	reference temperature
z	axis parallel to the plates in the direction of convective flow
z'	dimensionless coordinate defined by Eq. (8)

Greek Letters

α	thermal diffusion constant
$\beta_{\bar{T}}$	$= (-\partial\rho/\partial T)$ evaluated at $\bar{T}$
Δ	degree of separation defined by Eq. (14)
Δ_1	degree of separation defined by Eq. (36) when the concentration at the feed position of the column is approximately the same as the feed concentration
Δ_2	degree of separation defined by Eq. (39) for equifraction solutions
Δ_3	degree of separation defined by Eq. (40)
ΔT	difference in temperature of the hot and the cold plates
μ	viscosity of the solution
ρ	density of the solution
σ	mass flow rate of the top or the bottom product
σ'	dimensionless mass flow rate defined by Eq. (8)
ζ	dimensionless feed position defined as L_s/L
$\bar{\zeta}$	$= (1 - \zeta)$
ω	one-half of the distance between the plates of a thermal diffusion column

Subscript

e	for the enriching section
s	for the stripping section

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