flowing fluid, B.t.u./(hr. ft. °F.)

k_e° = stagnant effective thermal conductivity of bed (that is bed filled with stationary fluid), B.t.u./(hr. ft. °F.)

k_e^* = effective thermal conductivity representing heat transfer through the solid particles, B.t.u./(hr. ft. °F.)

k_g = thermal conductivity of fluid in bed, B.t.u./(hr. ft. °F.)

k_s = thermal conductivity of solid particles in bed, B.t.u./(hr. ft. °F.)

L = distance between extreme thermocouples at the axis of the bed, ft.

N_{Nu} = Nusselt number, $h\,D_p/k_g$, dimensionless

N_{Pr} = Prandtl number, $C_p\,\eta/k_g$, dimensionless

N_{Re_m} = modified Reynolds number, $D_p\,G/\eta$, dimensionless

t = temperature, °F., $\overline{t}$ denotes mean temperature in bed as indicated by thermocouple readings

x = distance measured from downstream thermocouple in the bed, ft.

Subscripts

g = fluid phase

s = solid phase

o = position at downstream thermocouple in the bed

L = position at a distance L from downstream thermocouple

Greek Letters

α = $G\,C_p/k_e$, ft.$^{-1}$

ϵ = void fraction in the bed

ϕ = parameter measuring the heat transfer resistance in the stagnant fluid adjacent to the contact point of adjacent solid particles, dimensionless; ϕ_1 and ϕ_2 refer to the loose and dense packing arrangements for spherical particles (*13*)

η = viscosity of fluid, lb./(hr. ft.)

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A Quantitative Treatment of the Forgotten Effect in Liquid Thermal Diffusion

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The influence of the concentration gradient on the density gradient in a thermal-diffusion column is called the *forgotten effect*. A modification of the theory developed by Furry, Jones, and Onsager is proposed to include the effect of the horizontal concentration gradient. A sample calculation with the system toluene-cyclohexane shows that the equations predict the proper behavior for a forgotten-effect system.

In quantitative treatments of thermal diffusion column operation it is frequently assumed that the horizontal density gradient in a column $\partial\rho/\partial x$ is caused only by the horizontal temperature gradient (*1, 2, 3*); that is

$$\frac{\partial\rho}{\partial x} = \left(\frac{\partial\rho}{\partial T}\right)\left(\frac{\partial T}{\partial x}\right) \qquad (1)$$

However $\partial\rho/\partial x$ must also depend on the horizontal gradient in composition of the mixture in the column. The composition gradient arises from the separation of the mixture by thermal diffusion. The influence of the concentration gradient on the density gradient in a column has been called the *forgotten effect* (*4*). The horizontal density gradient including the forgotten effect then becomes

$$\frac{\partial \rho}{\partial x} = \left(\frac{\partial \rho}{\partial T}\right)\left(\frac{\partial T}{\partial x}\right) + \left(\frac{\partial \rho}{\partial C_1}\right)\left(\frac{\partial C_1}{\partial x}\right) \quad (2)$$

For systems where the density gradient can be expressed by Equation (1) the separation proceeds qualitatively as shown in Figure 1*a*. The variation in time of the concentration of one component, C_1, of a two-component mixture at the top of the column is shown. For systems in which the forgotten effect is important, the term $(\partial \rho/\partial C_1)(\partial C_1/\partial x)$ in Equation (2) can have the same or the opposite sign to the term $(\partial \rho/\partial T)(\partial T/\partial x)$. Thus the forgotten effect can act to reinforce or to oppose the normal separation. A variety of effects can be observed depending on the rates of the diffusion and convection processes in the column. Figure 1*b* shows the possible qualitative behavior when the forgotten effect reinforces the normal separation. Figures 1*c* and 1*d* show the possible behavior when the forgotten effect opposes the normal separation; in Figure 1*d* the magnitude of the effect is large enough to reverse the separation.

The forgotten effect was first considered in detail by DeGroot (*4*). He gives a quantitative treatment of the effect for the special cases of columns with very large or very small spacing between walls. The importance of the forgotten effect in determining the separations of mixtures of hydrocarbons has been clearly shown by Prigogine, De Brouckere, and Amand (*5, 6*), Jones and Milberger (*7*), and Begeman and Cramer (*8*). Representative systems that show the qualitative types of behavior discussed above are given in Table 1. The contribution of the forgotten effect to the separation also depends on the starting concentrations of the components and on the geometry of the column (*4, 5, 6*).

In recent detailed treatments of column operation the forgotten effect has not been considered (3); however this effect can be very important in determining the separation obtained with a thermal diffusion column. It is the purpose of this treatment to show how the forgotten effect can be included in the general equations for column operation.

COLUMN THEORY NEGLECTING THE FORGOTTEN EFFECT

The solution of the convection and diffusion equation follows that of Furry, Jones, and Onsager (*1*). The mole fraction of component 1 of a binary mixture, C_1, at any point in the column is given by

$$\frac{\partial C_1}{\partial t} = D\left[\frac{\partial^2 C_1}{\partial x^2} + \frac{\partial^2 C_1}{\partial y^2}\right] - \frac{\alpha D}{T}\frac{dT}{dx}\frac{\partial}{\partial x}(C_1C_2) - v(x)\frac{\partial C_1}{\partial y} \quad (3)$$

As in earlier work (*1, 3*) it is assumed here that dT/dx is constant, independent of x and y, and that the average temperature can be used in the thermal diffusion term of Equation (3). For steady state conditions $\partial C_1/\partial t$ equals zero, and assuming as in earlier work that the term $D\,\partial^2 C_1/\partial y^2$ is negligible (*1*) one obtains

TABLE 1. HYDROCARBON MIXTURES SHOWING THE FORGOTTEN EFFECT

System	Reference
benzene—nitrobenzene	5
tetrabromoethane—tetrachlorethane	5
carbontetrachloride—bromobenzene	5
cyclohexanol—cyclohexane	6
ethyl alcohol—dioxane	6
toluene—cyclohexane	7
n-octane—2, 2, 4-trimethylpentane	7, 8
n-heptane—2, 2-dimethylpentane	8
toluene—methylcyclohexane	8
2, 2-dimethylpentane—2, 2-dimethylbutane	8
2, 2, 3-trimethylbutane—ethanol	8

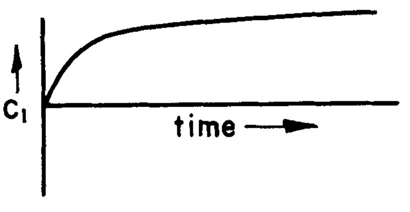

Fig. 1*a*. Normal separation where forgotten effect is not important.

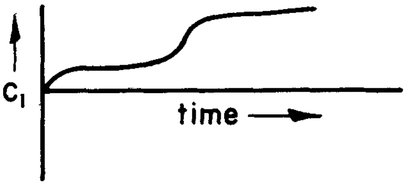

Fig. 1*b*. Forgotten effect reinforcing normal separation.

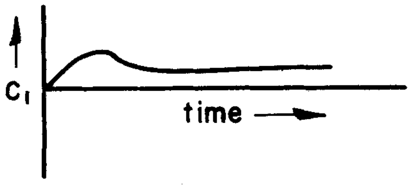

Fig. 1*c*. Forgotten effect opposing normal separation.

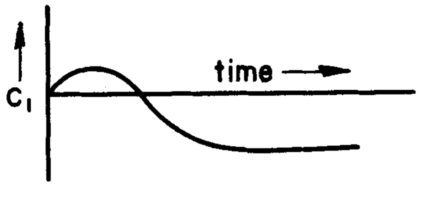

Fig. 1*d*. Forgotten effect reversing normal separation.

$$\frac{\partial}{\partial x}\left[D\frac{\partial C_1}{\partial x} - \frac{\alpha D}{T}\frac{dT}{dx}(C_1C_2)\right] = v(x)\frac{\partial C_1}{\partial y} \quad (4)$$

A function $G(x, y)$ is defined as follows:

$$\left(\frac{\partial C_1}{\partial y}\right)G(x,y) \equiv D\left(\frac{\partial C_1}{\partial x}\right) - \frac{\alpha D}{T}\frac{dT}{dx}(C_1C_2) \quad (5)$$

The net vertical transport of species 1, τ_1, is then given by

$$\tau_1 = B\int_{-\omega}^{\omega}\rho\, C_1\frac{\partial}{\partial x}G(x)dx - B\int_{-\omega}^{\omega}\left(\frac{\partial C_1}{\partial y}\right)\rho\, D\, dx \quad (6)$$

In earlier work it has been assumed that the variation of $\rho(x)$ with x in Equation (6) can be neglected (*1, 3*). Furthermore since it has been assumed that $\partial C_1/\partial y$ is independent of x, the first term of Equation (6) can be integrated by parts to give

$$\tau_1 = -B\int_{-\omega}^{\omega}\rho\left(\frac{\partial C_1}{\partial y}\right)\frac{[G(x)]^2}{D}dx - B\int_{-\omega}^{\omega}\rho\frac{\alpha}{T}\frac{dT}{dx}(C_1C_2)G(x)dx - B\int_{-\omega}^{\omega}\left(\frac{\partial C_1}{\partial y}\right)\rho\, D\, dx \quad (7)$$

For steady state conditions τ_1 equals zero, and assuming the product C_1C_2 and $\partial C_1/\partial y$ are independent of x one gets

$$\frac{\partial C_1}{\partial y}(K + K_d) + H\, C_1C_2 = 0 \quad (8)$$

TABLE 2. VALUES OF PHYSICAL CONSTANTS USED IN CALCULATIONS

Property	Toluene (15)	Cyclohexane (15)
Density ρ (35°C.), g./cc.	0.85307	0.76465
$-\partial\rho/\partial T$, $\beta \times 10^3$, g./cc.°C.	0.924	0.927
Coefficient of viscosity, η, (35°C.), centipoise	0.4776	0.732
Index of refraction, η_D^{25}	1.49413	1.42354

Column length, l, 152.3 cm.
Column width, B, 5.03 cm.
Spacing between walls, 2 ω, 0.0292 cm.
Temp. diff. between walls, ΔT, 30°C.
Average temp., $\overline{T}$, 35°C.

$$\ln q = -\left[\frac{H}{K+K_d}\right] l \qquad (9)$$

where

$$K = -B\int_{-\omega}^{\omega} \rho \frac{[G(x)]^2}{D} dx \qquad (9a)$$

$$K_d = -B\int_{-\omega}^{\omega} \rho D \, dx \qquad (9b)$$

$$H = -\frac{B}{\overline{T}}\frac{dT}{dx}\int_{-\omega}^{\omega} \rho\, \alpha\, G(x) dx \qquad (9c)$$

Furry, Jones, and Onsager (*1*) show that the convection flow in the column is laminar and that the Navier-Stokes equation is applicable. Thus

$$\eta \frac{d^2v}{dx^2} = g(\rho(x) - \overline{\rho}) \qquad (10)$$

If it is assumed that the density of the mixture in the column depends only on the temperature, then the density gradient is given by Equation (1). If it is further assumed that dT/dx is constant, then

$$\rho(x) - \overline{\rho} = -\beta \frac{x}{2\omega} \Delta T \qquad (11)$$

Using the relationship between $v(x)$ and $G(x)$ given by Equations (4) and (5), together with Equations (10) and (11), one obtains

$$\eta \frac{d^3G(x)}{dx^3} = -g\beta\frac{x}{2\omega}\Delta T \qquad (12)$$

Equation (12) can be integrated by the use of the boundary conditions of Furry, Jones, and Onsager (*1*) to give $G(x)$. Equations (9*a*) and (9*c*) can then be integrated by the use of $G(x)$ to give the constants K and H. The equations for K and H obtained in this way are given by Wilke (*3*).

COLUMN THEORY INCLUDING THE FORGOTTEN EFFECT

Equation (2) gives a more complete description of the density gradient than Equation (1). For a two component system

$$\left(\frac{\partial\rho}{\partial C_1}\right)_T = \rho_1(T) - \rho_2(T) = \Delta\rho(T) \qquad (13)$$

$$\left(\frac{\partial\rho}{\partial T}\right)_{C_1} = C_1(x)\,(\beta_2 - \beta_1) - \beta_2 \qquad (14)$$

Then from Equation (2)

$$d\rho(x,T) = [\Delta\rho(T)]\left(\frac{\partial C_1}{\partial x}\right)dx + [C_1(x)\,(\beta_1-\beta_2) - \beta_2]\left(\frac{dT}{dx}\right)dx \qquad (15)$$

For mixtures of organic liquids $\Delta\rho$ is about independent of T over small temperature ranges. If it is assumed that $\Delta\rho$ is independent of T, or β_1 equals β_2, then Equation (15) becomes

$$d\rho = \Delta\rho\left(\frac{\partial C_1}{\partial x}\right)dx - \beta\left(\frac{dT}{dx}\right)dx \qquad (16)$$

Finally if it is assumed that $\partial C_1/\partial x$ and dT/dx are independent of x, Equation (16) can be integrated to give

$$\rho(x) - \overline{\rho} = \left[\Delta\rho\left(\frac{\partial C_1}{\partial x}\right) - \frac{\beta\,\Delta T}{2\omega}\right]x \qquad (17)$$

From the work of De Groot (*4*) $\partial C_1/\partial x$ is probably not independent of x; however the assumption that $\partial C_1/\partial x$ is constant is not serious here.

A function $G(x)$ is defined as in Equation (5), and from Equations (4), (10), and (17)

$$\frac{d^3G(x)}{dx^3} = \left[a\left(\frac{\partial C_1}{\partial x}\right) + b\right]x \qquad (18)$$

where

$$a = g\frac{\Delta\rho}{\eta}, \quad b = -g\frac{\beta\,\Delta T}{\eta\, 2\omega}$$

For convenience a new function $F(x)$ is defined as follows:

$$\left[a\left(\frac{\partial C_1}{\partial x}\right) + b\right]F(x) \equiv G(x) \qquad (19)$$

The new form of $G(x)$ is then substituted in Equation (6) to give a new expression for the net vertical transport of species 1:

$$\tau_1 = -B\int_{-\omega}^{\omega}\rho a\left(\frac{\partial C_1}{\partial x}\right)^2 F(x)dx - B\int_{-\omega}^{\omega}\rho\left(\frac{\partial C_1}{\partial x}\right)b\,F(x)dx - B\int_{-\omega}^{\omega}\left(\frac{\partial C_1}{\partial y}\right)\rho\, D\, dx \qquad (20)$$

From Equations (19) and (5)

$$\frac{\partial C_1}{\partial x} = \left(\frac{\partial C_1}{\partial y}\right)\frac{1}{D}\left[a\left(\frac{\partial C_1}{\partial x}\right) + b\right] F(x) + \frac{\alpha}{\overline{T}}\frac{dT}{dx}(C_1C_2) \qquad (21)$$

When one assumes that the term $\frac{a}{D}\left(\frac{\partial C_1}{\partial y}\right)F(x)$ is much less than 1, Equation (21) becomes

$$\frac{\partial C_1}{\partial x} = \frac{b}{D}\left(\frac{\partial C_1}{\partial y}\right)F(x) + \frac{\alpha}{\overline{T}}\frac{dT}{dx}(C_1C_2) \qquad (22)$$

Substitution of this expression for $\partial C_1/\partial x$ into Equation (20) for conditions when τ_1 equals zero leads finally to the relationship

$$L\left(\frac{\partial C_1}{\partial y}\right)^2 + [M\,C_1C_2 + K' + K'_d]\left(\frac{\partial C_1}{\partial y}\right) + N\,(C_1C_2)^2 + H'C_1C_2 = 0 \qquad (23)$$

where

$$L = -B\int_{-\omega}^{\omega}\rho\, a\left(\frac{b}{D}\right)^2 [F(x)]^3 dx \qquad (23a)$$

$$M = -B\int_{-\omega}^{\omega}\rho\, a\frac{b\alpha}{D\overline{T}}\frac{dT}{dx}[F(x)]^2 dx \qquad (23b)$$

$$N = -B\int_{-\omega}^{\omega}\rho\, a\left(\frac{\alpha}{\overline{T}}\frac{dT}{dx}\right)^2 F(x)\, dx \qquad (23c)$$

$$K' = -B\int_{-\omega}^{\omega}\rho\frac{b^2}{D}[F(x)]^2 dx \qquad (23d)$$

$$H' = -B\int_{-\omega}^{\omega}\rho\frac{b\alpha}{\overline{T}}\frac{dT}{dx}F(x)\, dx \qquad (23e)$$

$$K'_d = -2B\,\rho\, D\omega \qquad (23f)$$

As previously, it is assumed that C_1C_2, and $\partial C_1/\partial y$ are independent of x and can thus be taken from under the integral. If it is further assumed that C_1C_2 is about constant and independent of both y and x (*2, 3*), then Equation (23) can be solved to give

$$\Delta C_1 = \frac{-R \pm \sqrt{R^2 - 4LS}}{2L}\, l \qquad (24)$$

where

$$\Delta C_1 = C_T - C_B$$
$$R = M\,C_1C_2 + K' + K'_d$$
$$S = N\,(C_1C_2)^2 + H'\,C_1C_2$$

Equation (18) can be integrated to give $F(x)$ with the boundary conditions of Jones, Furry, and Onsager (*1*) used. With $F(x)$ thus obtained the integrals (23*a*) to (23*e*) can be evaluated. In final form these parameters are

$$L = -\frac{75}{13!}\,\frac{\Delta\rho\,\beta^2(\Delta T)^2\beta\,\rho\,(2\omega)^{11}}{2\eta^3\,D^2} \qquad (25)$$

$$M = \frac{g^2\,\Delta\rho\,\beta(\Delta T)^2(2\omega)^7\,B\,\rho\,\alpha}{\eta^2\,D\overline{T}\,9!} \qquad (26)$$

$$N = -\frac{g\,\Delta\rho\,(2\omega)^3\,B\,\rho\,\alpha^2\,(\Delta T)^2}{\eta\,\overline{T^2}\,6!} \qquad (27)$$

$$K' = -\frac{\beta^2\,\rho\,g^2\,(2\omega)^7\,B\,(\Delta T)^2}{\eta^2\,D\,9!} \qquad (28)$$

$$H' = \frac{\alpha\,\beta\,\rho\,g\,(2\omega)^3\,B\,(\Delta T)^2}{\eta\,\overline{T}\,6!} \qquad (29)$$

COMPARISON WITH EXPERIMENT

The results of Jones and Milberger (*7*) on the system toluene-cyclohexane are used to show that Equation (24) correctly describes the unusual behavior of a system where the forgotten effect is important. There are two times for which $\partial C_1/\partial t$ and τ_1 equal zero; steady state conditions exist after time t_f, when the influence of the horizontal concentration gradient begins to reverse the direction of concentration of the components, and after very long times. From the data of Jones and Milberger (*7*) ΔC_1 can be obtained for both these conditions. The two values of ΔC_1 used with the two solutions of Equation (24) enable both α and D to be calculated for the system.

Toluene is taken as component 1, while cyclohexane is component 2. The data of Jones and Milberger (*7*) show that ΔC_1 equals 0.0071 after time t_f and that ΔC_1 approaches -0.0566 for very long times. The dimensions of the column used by Jones and Milberger (*7*) and the physical constants of the hydrocarbons used are given in Table 2. When one uses these data and Equation (24), values of α equal to 1.6 and D equal to 0.13×10^{-5} are calculated for the system toluene-cyclohexane for an initial C_1 equal to 0.5312. The value of α is of the order of magnitude expected from the measurements of other workers on similar systems (*3, 5, 6, 9, 10, 11, 12, 13, 14*). The value of D however is low by about a factor of 10.

SUMMARY

The importance of the forgotten effect in determining thermal diffusion column operation has been clearly demonstrated by the measurements of De Groot (*4*), Prigogine, De Brouckere, and Amand (*5, 6*), Jones and Milberger (*7*), and Begeman and Cramer (*8*). The effect causes separations far different from what would be expected with treatments based on Equation (1). Often the separation is the complete reverse of what is calculated; the component expected to appear at the top of the column concentrates at the bottom. In the design of thermal diffusion columns for practical separations the possibility of a large forgotten effect must be considered.

Equation (24) incorporates the more complete description of the horizontal density gradient given in Equation (2). Equation (24) predicts the proper behavior for systems where the forgotten effect is important, and it reduces to the results of Furry, Jones, and Onsager (*1*) for systems with low values of $\Delta\rho(\partial C_1/\partial x)$.

Even with the added refinement of considering the horizontal concentration gradient, it does not seem likely that thermal-diffusion columns can compete with static methods for the determination of thermal-diffusion ratios. However for the practical purposes of column design the influence of the concentration gradient on the horizontal density gradient can not be neglected.

NOTATION

a, b = general constants
B = column width
C_1, C_2 = mole fraction of component 1, 2 in a binary solution
C_T, C_B = mole fraction of component 1 at the top and bottom of the column
D = ordinary diffusion coefficient of component 1 with respect to component 2
$F(x,y)$ = general function of x and y
$G(x,y)$ = general function of x and y
$G'(x,y)$ = $\partial G(x,y)/\partial x$
g = acceleration due to gravity
K, K_d, H = parameters defined by Equations (9*a*), (9*b*), and (9*c*)
K', H', K_d' = parameters defined by Equations (23*d*), (23*e*), and (23*f*)
l = length of the column
L, M, N = parameters defined by Equations (23*a*), (23*b*), and (23*c*)
q = equilibrium separation factor $= \dfrac{C_T(1-C_B)}{C_B(1-C_T)}$
R = $MC_1C_2 + K' + K_d'$
S = $N(C_1C_2)^2 + H'C_1C_2$
T = absolute temperature
$\overline{T}$ = arithmetic average of the temperatures of the hot and cold wall
t = time
t_f = time at which $\partial C_1/\partial t$ equals zero due to reversal of the direction of concentration of the components
$v(x)$ = velocity distribution function
x = axis normal to the column walls
y = vertical axis

Greek Letters

α = thermal diffusion constant
β = $-\partial\rho/\partial T$
β_1, β_2 = $-\partial\rho_1/\partial T$, $-\partial\rho_2/\partial T$ for components 1, 2
ΔC_1 = $C_T - C_B$
$\Delta\rho(T)$ = $\rho_1(T) - \rho_2(T)$ = density difference between component 1 and 2 at temperature T
ΔT = difference in temperature of the hot and cold wall
η = coefficient of viscosity
η_D^{25} = index of refraction, sodium D line at 25°C.
ρ = density of liquid mixture at any point in the column
$\overline{\rho}$ = average density
τ_1 = net vertical transport of species 1
ω = one-half the distance between walls of a thermal-diffusion column

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