PERFORMANCE OF A LIQUID THERMAL-DIFFUSION COLUMN WITH TEMPERATURE ASYMMETRY OVER THE WIDTH

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The theory of thermal diffusion [1] employs an idealized model in which the temperature at all points on the hot or cold surface is assumed to be the same. In practice, however, such a model cannot be realized, since even the most careful design of the column will result in asymmetry in the temperature distributions on these surfaces. This asymmetry is more pronounced in liquid columns because of the small width of the annulus, and so the column acquires parasitic convection [1], which results in resolution worse than theory predicts. The origin of this effect, which is extremely undesirable in practical operation is explained in Fig. 1a [1]. The azimuthal temperature gradient in regions I and II causes the mean temperature of the liquid to vary, so the liquid as a whole moves in one direction in region I and in the opposite direction in region II; in each of these regions there are also two flows caused by the radial temperature gradient, which is incorporated in the theory of the column.

It is considered [1] that the parasitic convection alters the coefficients H and K in the transport equation:

$$j = Hc\bar{c} - K\frac{dc}{dz}, \tag{1}$$

and the temperature asymmetry affects most substantially K, while H is insensitive to this factor. The result [1] is

$$\frac{K_p}{K_c} = 8\left(\frac{B}{a}\right)^2\left(\frac{\delta T}{\Delta T}\right)^2, \tag{2}$$

where K_p is the component of K due to parasitic convection.

It is clear from (2) that K_p is very strongly dependent on the ratio of the annulus perimeter B to the annulus width a. If we assume for a liquid column that $\delta T = 5°C$, $\Delta T = 80°C$, B = 100 mm, a = 0.25 mm, then (2) gives $K_p/K_c = 0.5 \cdot 10^6$, and such a column would produce no separation, although in practice a real column with these parameters gives satisfactory enrichment of the components.

This feature indicates that the arguments giving (2) do not reflect the true picture of the separation in the presence of parasitic convection.

An attempt has been made [2] to solve this problem via formal mathematical formulation for a column having a continuous through flow σ; no detailed physical model for the parasitic convection is then considered.

The result is

$$\frac{K_p}{K_c} = \frac{47}{6}\left(\frac{\varepsilon}{a}\right)^2 + 78\left(\frac{\delta T_h + \delta T_c}{2\Delta T}\right)^2 + \frac{1}{12}\left(\frac{\delta T_h - \delta T_c}{\Delta T}\right)^2 + \frac{7}{6}\cdot\frac{\delta T_h - \delta T_c}{\Delta T}\cdot\frac{\varepsilon}{a} - 28\frac{\delta T_h + \delta T_c}{2\Delta T}\cdot\frac{\varepsilon}{a},$$

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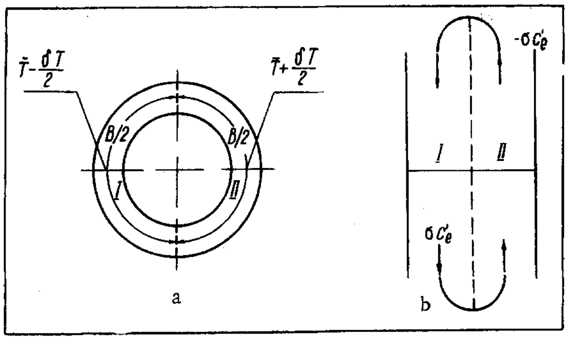

Fig. 1. Motion of the parasitic flows in a thermal-diffusion column with temperature asymmetry: a) horizontal section of the column; the broken line shows the boundary between the descending flow (region I) and the rising one (region II), these parasitic flows being due to the temperature difference δT between the volumes I and II of the liquid; b) vertical section. The flow of enriched component passes downwards from section I into section II but upwards from section II into section I.

which contrasts with (2) in giving K_p as too small; for instance, the conditions of the above example with $\varepsilon = 0$ would give $K_p/K_c = 0.3$.

Equation (3) takes no account of the geometrical dimensions of the column and the physical characteristics of the binary mixture, although we show below that these have substantial effects on the ratio K_p/K_c.

These features force us to be cautious about the results derived by Dickel.

Here we present an essentially different approach to the parasitic convection in such a column; as in [1], we assume that there are two parasitic flows (Fig. 1a). Let flow I sink and flow II rise; the flows reverse in direction at the top and bottom of the column, and flow I becomes flow II and vice versa. This is shown schematically in Fig. 1b. We envisage only liquid columns in what follows, in which diffusion transport in the azimuthal direction may be neglected without appreciable error, on account of the extremely small value of a/B. Then the column may be considered as consisting of two columns, one feeding the other with a flow σ equal to the flow caused by parasitic convection.

This model can be used to analyze the operation of columns differing in mean temperature and working in parallel in one stage of a thermal-diffusion system.

It has been shown [1] that the takeoff σ causes the flow of enriched component to the positive end to the column to be

$$j_z = Hc\bar{c} \pm \sigma c - K\frac{dc}{dz}, \tag{4}$$

where the upper sign refers to take off from the positive end and the lower to takeoff from the negative end.

In the steady state

$$\operatorname{div} j_z = 0. \tag{5}$$

In principle, the problem can be solved from an arbitrary variation in the concentration c, but in that case (5) becomes nonlinear, and to obtain results capable of analysis we consider only two linear approximations.

1. $c \ll 1$. Then the above statement about the signs in (4) give us from (5) in terms of dimensionless variables for columns I and II that

$$\frac{d^2c'}{dy'^2} - (1+\varkappa')\frac{dc'}{dy'} = 0, \tag{6}$$

$$\frac{d^2c''}{dy''^2} - (1-\varkappa'')\frac{dc''}{dy''} = 0. \tag{7}$$

The solution to (6) and (7) is

$$c' = A' + B'\exp\left[(1+\varkappa')y'\right], \tag{8}$$

$$c'' = A'' + B''\exp\left[(1-\varkappa'')y''\right], \tag{9}$$

which contains four arbitrary constants, which have to be determined via four suitable conditions.

It is clear that, in the steady state, we will have not only σ the same for columns I and II but also the flow of the enriched component; if we denote by c_e' the concentration of this component at the positive end of column I, then we should have for column I

$$j'|_{z=0} = j'_z|_{z=L} = \sigma c'_e, \tag{10}$$

and for column II

$$j''_z|_{z=0} = j''_z|_{z=L} = -\sigma c'_e, \tag{11}$$

where the minus sign indicates that withdrawal occurs at the negative end of the column. We replace the flows by the expressions for (4) with the condition $cc \approx c \ll 1$ and replace the concentrations by the values from (8) and (9) to get, after conversion to dimensionless form, instead of (10) and (11) that

$$A' = \varkappa' B' \exp\left[(1+\varkappa')\,y'_e\right], \tag{12}$$

$$(1-\varkappa'')\,A'' + \varkappa'' A' + \varkappa'' B' \exp\left[(1+\varkappa')\,y'_e\right] = 0. \tag{13}$$

The component concentration at the withdrawal point should be equal to the concentration in the withdrawal product, so we have

$$c'|_{z=L} = c''|_{z=0}, \tag{14}$$

which from (8) and (9) leads to

$$A' + B' \exp\left[(1+\varkappa')\,y'_e\right] = A'' + B''. \tag{15}$$

The pair of columns I and II is a closed system for which we have to apply the law of conservation of mass for a given component.

On this basis we get an integral condition for the pair of columns I and II:

$$c_0 = \frac{c'_{av} + c''_{av}}{2}. \tag{16}$$

Equation (16) assumes that the masses of liquid in columns I and II are equal; here c'_{av} and c''_{av} are the mean concentrations in these columns respectively, which are defined by

$$c'_{av} = \frac{1}{y'_e}\int_0^{y'_e} c'\,dy, \tag{17}$$

$$c''_{av} = \frac{1}{y''_e}\int_0^{y''_e} c''\,dy. \tag{18}$$

From (16) with (8), (9), (17), and (18) we get

$$A' + \frac{B'}{y'_e(1+\varkappa')}\left\{\exp\left[(1+\varkappa')\,y'_e\right] - 1\right\} + A'' + \frac{B''}{y''_e(1-\varkappa'')}\left\{\exp\left[(1-\varkappa'')\,y''_e - 1\right\} = 2c_0. \tag{19}$$

From system (12), (13), (15), and (19) we get the constants A', B', A", B", and substitute their values into (8) and (9) to get equations for the concentration variation in each column in relation to the dimensionless length. For column I

$$c' = 2c_0(1-\varkappa'')^2\left\{\varkappa' + \exp\left[-(1+\varkappa')(y'_e - y')\right]\right\}\left\{\frac{1+\varkappa'}{y''_e}\left(\exp\left[(1-\varkappa'')\,y''_e\right] - 1\right) - \varkappa''(1-\varkappa'')(1+\varkappa') + \varkappa'(1-\varkappa'')^2 + \frac{\varkappa'(1-\varkappa'')^2}{y'_e(1+\varkappa')}\left(1 - \exp\left[-(1+\varkappa')\,y'_e\right]\right)\right\}^{-1}; \tag{20}$$

For column II

$$c'' = 2c_0(1-\varkappa'')\left\{\exp\left[(1-\varkappa'')\,y''\right] - \varkappa''\right\}\left\{\frac{1}{y''_e}\left(\exp\left[(1-\varkappa'')\,y''_e\right] - 1\right) - \varkappa''(1-\varkappa'') + \frac{\varkappa'(1-\varkappa'')^2}{1+\varkappa'} + \frac{\varkappa'}{y'_e}\left(\frac{1-\varkappa''}{1+\varkappa'}\right)^2\left(1 - \exp\left[-(1+\varkappa')\,y'_e\right]\right)\right\}^{-1}. \tag{21}$$

If $\varkappa'' = 1$, the last two equations become indeterminate in the form 0/0, which is resolved to give

$$c'|_{\varkappa''=1} = \frac{2c_0}{\varkappa'}\cdot\frac{\varkappa' + \exp\left[-(1+\varkappa')(y'_e - y')\right]}{1 + \dfrac{1}{y'_e(1+\varkappa')}\left\{1 - \exp\left[-(1+\varkappa')\,y'_e\right]\right\}}, \tag{22}$$

$$c''|_{\varkappa''=1} = 2c_0 \frac{1+\varkappa'}{\varkappa'} \cdot \frac{1+y''}{1+\frac{1}{y_e'+(1+\varkappa')}\{1-\exp[-(1+\varkappa')y_e']\}}. \quad (23)$$

A real column consists of these columns I and II, so the concentrations at the ends are the half sums of the concentrations at the corresponding ends of the component columns, i.e.,

$$c|_{y=y_e} = \frac{1}{2}(c'_{y'=y_e'} + c''_{y''=y_e''}),$$
$$c|_{y=0} = \frac{1}{2}(c'_{y'=0} + c''_{y''=0}). \quad (24)$$

We substitute in (24) for the quantities in parentheses the values given by (20) and (21) to get

$$c_{y=y_e} = c_0\{1-2\varkappa''+\exp[(1-\varkappa'')y_e'']\}\left\{\frac{1}{y_e''(1-\varkappa'')}(\exp[(1-\varkappa'')y_e'']-1)\right.$$
$$\left.-\varkappa''+\frac{\varkappa'(1-\varkappa'')}{1+\varkappa'}+\frac{\varkappa'(1-\varkappa'')}{y_e'(1+\varkappa')^2}(1-\exp[-(1+\varkappa')y_e'])\right\}^{-1}, \quad (25)$$

$$c_{y=0} = c_0\frac{1-\varkappa''}{1+\varkappa'}\{1+2\varkappa'+\exp[-(1+\varkappa')y_e']\}$$
$$\times\left\{\frac{1}{y_e''(1-\varkappa'')}(\exp[-(1-\varkappa'')y_e'']-1)-\varkappa''\right.$$
$$\left.+\frac{\varkappa'(1-\varkappa'')}{1+\varkappa'}+\frac{\varkappa'(1-\varkappa'')}{y_e'(1+\varkappa')^2}(1-\exp[-(1+\varkappa')y_e'])\right\}^{-1}. \quad (26)$$

We get the separation coefficient for a column with parasitic convection as follows when $c \ll 1$, if we separate the left and right sides of (25) and (26):

$$\frac{c_{y=y_e}}{c_{y=0}} \equiv q = \frac{1+\varkappa'}{1-\varkappa''} \cdot \frac{1-2\varkappa''+\exp[(1-\varkappa'')y_e'']}{1+2\varkappa'+\exp[-(1+\varkappa')y_e']}. \quad (27)$$

If $\varkappa'' = 1$

$$q_{\varkappa''=1} = \frac{(2+y_e'')(1+\varkappa')}{1+2\varkappa'+\exp[-(1+\varkappa')y_e']}. \quad (28)$$

If we put $\varkappa' = \varkappa'' = 0$ in (27), which corresponds to absence of parastic convection, we get $y_e' = y_e''$ and the separation coefficient becomes

$$q^* = \exp(y_e). \quad (29)$$

Simplicity of analysis leads us to replace (27) by the simplified form with

$$y_e' = y_e'' = y_e, \quad \varkappa' = \varkappa'' = \varkappa.$$

Then the resulting expression is

$$q = \frac{1+\varkappa}{1-\varkappa} \cdot \frac{1-2\varkappa+\exp[(1-\varkappa)y_e]}{1+2\varkappa+\exp[-(1+\varkappa)y_e]} \quad (30)$$

which relates the partition coefficient to the two parameters $\varkappa$ and y_e.

Figure 2 shows this relationship in the coordinates $\ln q/\ln q^* - \varkappa y_e$, where q^* is defined by (29); note that for $y_e \le 1$ the curves practically coincide, i.e., there is no effect from y_e on the product $\varkappa y_e$.

Figure 2 gives also the important conclusion that the ratio $\ln q/\ln q^*$ is reduced not only when $\varkappa$ increases but also when y_e increases with $\varkappa$ unchanged.

One can increase $\varkappa = \sigma/H$ either by increasing σ or by reducing H; this means, for example, that $\varkappa$ will be considerably larger for a binary mixture with a small thermal-diffusion constant than it will be for a readily separated mixture with a larger α, and consequently the effects of parasitic convection will be less in the latter case for the same parasitic flow rate σ, while the separation coefficient will be larger.

The effects of y_e may be examined via the expanded form of the expression derived from the definition of H and K_c [3]:

$$y_e = \frac{HL}{K_c} = 504\,\frac{\alpha\eta D}{g\beta\rho}\cdot\frac{L}{a^4}. \tag{31}$$

We see that y_e is the product of two quantities, one of which is dependent on the physical constants of the mixture, while the other is dependent on the geometrical characteristics of the column. Then, increase in column length L should lead for the given binary mixture to increase in y_e and for fixed $\varkappa$ to reduction in $\ln q/\ln q^*$.

A similar conclusion can be drawn from the effects of reducing the annulus width, as may be seen by the following argument.

It has been shown [1] that the mean speed of the parasitic flow is

$$\bar{w}_p = \frac{g\beta\rho a^2\delta T}{48\eta}. \tag{32}$$

As $\sigma = (1/2)\rho a B\bar{w}_p$, we have

$$\varkappa = \frac{\sigma}{H} = 7.5\,\frac{\bar{T}\delta T}{\alpha(\Delta T)^2}, \tag{33}$$

i.e., $\varkappa$ is not explicitly dependent on the gap width, although there is an inexplicit dependence, since reduction in a may increase the temperature asymmetry δT, and $\varkappa$ will increase.

The result given by (30) may be related to the K_p by incorporating parasitic convection into (2).

We put

$$\frac{HL}{K_c + K_p} = \frac{y_e}{1+\dfrac{K_p}{K_c}} = y_e^*, \tag{34}$$

which enables us to put that

$$\frac{1+\varkappa}{1-\varkappa}\cdot\frac{1-2\varkappa+\exp\left[(1-\varkappa)\,y_e\right]}{1+2\varkappa+\exp\left[-(1+\varkappa)\,y_e\right]} = e^{y_e^*},$$

whence from (34) we have

$$\frac{K_p}{K_c} = \frac{y_e}{\ln\dfrac{1+\varkappa}{1-\varkappa}\cdot\dfrac{1-2\varkappa+\exp\left[(1-\varkappa)\,y_e\right]}{1+2\varkappa+\exp\left[-(1+\varkappa)\,y_e\right]}} - 1, \tag{35}$$

i.e., K_p is strongly dependent on y_e, which is not reflected in the results of [2].

2. $\overline{cc} = b$. In this case, (6) and (7) are replaced from (5) by

$$\frac{d^2c'}{dy'^2} - \varkappa'\,\frac{dc'}{dy'} = 0, \tag{36}$$

$$\frac{d^2c''}{dy''^2} + \varkappa''\,\frac{dc''}{dx''} = 0. \tag{37}$$

The solutions are

$$c' = A' + B'\exp(\varkappa' y'), \tag{38}$$

$$c'' = A'' + B''\exp(-\varkappa'' y'') \tag{39}$$

which should, as in the previous case, satisfy conditions (10), (11), (14), and (17), from which we find the values of the arbitrary constants.

Then the concentrations in each of the columns are given by

$$c' = c_0 - \frac{b}{2\varkappa''}\left(1+\frac{\varkappa''}{\varkappa'}\right) + \frac{b}{2\varkappa''^2 y_e''}\left[1-\exp(-\varkappa'' y_e'')\right] - \frac{b}{2\varkappa'^2 y_e'}\left[1-\exp(-\varkappa' y_e')\right] + \frac{b}{\varkappa'}\exp\left[-\varkappa'(y_e'-y')\right], \tag{40}$$

$$c'' = c_0 + \frac{b}{2\varkappa''}\left(1+\frac{\varkappa''}{\varkappa'}\right) + \frac{b}{2\varkappa''^2 y_e''}\left[1-\exp(-\varkappa'' y_e'')\right] - \frac{b}{2\varkappa'^2 y_e'}\left[1-\exp(-\varkappa' y_e')\right] - \frac{b}{\varkappa''}\exp(-\varkappa'' y'').$$

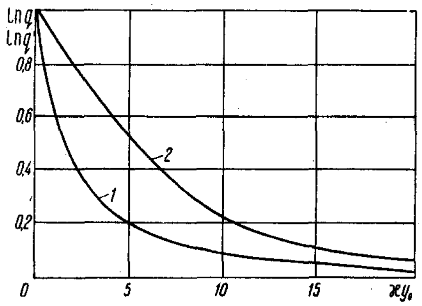

Fig. 2. Dependence of $\ln q/\ln q^*$ on $\varkappa y_e$ for various dimensionless lengths of the column y_e: 1 – $y_e = 0.01$; 0.1; 1.0; 2 – $y_e = 10$.

(41)

From (24) we get the concentrations at the bottom and top of the real column

$$c_{y=y_e} = c_0 + \frac{b}{2\varkappa''^2 y_e''}\left[1-\exp(-\varkappa'' y_e'')\right] - \frac{b}{2\varkappa'^2 y_e'}\left[1-\exp(-\varkappa' y_e')\right] + \frac{b}{2\varkappa'}\left[1-\frac{\varkappa'}{\varkappa''}\exp(-\varkappa'' y_e'')\right], \qquad (42)$$

$$c_{y=0} = c_0 + \frac{b}{2\varkappa''^2 y_e''}\left[1-\exp(-\varkappa'' y_e'')\right] - \frac{b}{2\varkappa'^2 y_e'}\left[1-\exp(-\varkappa' y_e')\right] - \frac{b}{2\varkappa''}\left[1-\frac{\varkappa''}{\varkappa'}\exp(-\varkappa' y_e')\right]. \qquad (43)$$

If for simplicity, we put $\varkappa' = \varkappa'' = \varkappa$, $y_e' = y''_e = y_e$ we get for separation coefficient the expression

$$q \equiv \frac{c_{y=y_e}}{1-c_{y=y_e}} \cdot \frac{1-c_{y=0}}{c_{y=0}} = \frac{c_0 + \frac{b}{2\varkappa}\left[1-\exp(-\varkappa y_e)\right]}{1-c_0-\frac{b}{2\varkappa}\left[1-\exp(-\varkappa y_e)\right]} \cdot \frac{1-c_0+\frac{b}{2\varkappa}\left[1-\exp(-\varkappa y_e)\right]}{c_0-\frac{b}{2\varkappa}\left[1-\exp(-\varkappa y_e)\right]}, \qquad (44)$$

which for $c_0 = 0.5$ becomes

$$q = \left\{\frac{1+\frac{b}{\varkappa}\left[1-\exp(-\varkappa y_e)\right]}{1-\frac{b}{\varkappa}\left[1-\exp(-\varkappa y_e)\right]}\right\}^2. \qquad (45)$$

The calculated points lie almost exactly on curve 1 in Fig. 2 for the range of values of y_e in which we have $\overline{cc}$ = const ($y_e < 3.4$); this indicates that the above analysis is correct for any concentration provided that $\bar{c} \ll 1$.

We consider that these results are of vital importance to the design of liquid thermal-diffusion apparatus and indicate ways of raising performance.

NOTATION

$H = \beta g \rho^2 (\Delta T)^2 B a^3 \alpha / 720 \eta \bar{T}$, $K = K_c + K_p$, $K_c = \beta^2 g^2 \rho^3 (\Delta T)^2 B a^7 / 362880 \eta^2 D$ are transfer coefficients;

K_p is the parasitic component of the convective transfer coefficients;

B is the perimeter of the separating slot;

a is the gap;

δT is the temperature difference due to temperature asymmetry;

ΔT is the temperature difference between hot and cold column surfaces;

$\bar{T}$ is the mean temperature of liquid in column;

jz is the flow of enriching component;

σ is the selection;

c is the concentration;

L is the column length;

$y = Hz/K_c$;

$\varkappa = \sigma/H$;

w is the velocity;

q is the separation coefficient;

η is the viscosity;

ρ is the density;

D is the diffusion coefficient;

β is the volume expansion coefficient;

α is the thermodiffusion constant.

Subscripts

e denotes to positive and of column;

h denotes hot;

c denotes cold;

p denotes parasitic;

0 denotes reference.

One prime and two primes refer, respectively, to the column parts shown in Fig. 1 as I and II.

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