

EFFECT OF CONCENTRATION ASYMMETRY ON THE EFFECTIVENESS OF THERMAL-DIFFUSION COLUMNS

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One reason (never hitherto presented) for the development of parasitic convection in thermal-diffusion columns due to concentration asymmetry around their perimeter is set out; a method is also proposed for taking this effect into consideration quantitatively when conducting this process in the liquid phase.

It is well known that the efficiency of thermal-diffusion columns (as devices for separating liquid and gas mixtures) depends very largely on the degree of interference on the part of the "remixing" flows which develop in the columns. These "parasitic" [10] flows are usually [1, 10, 11] associated with differences in the densities of the separating mixture at various points of the column cross section, while sole responsibility for the density change is assigned to the nonuniformity of the temperature field over the cross section of the column.

Although in the overwhelming majority of cases the reason for parasitic convection is undoubtedly temperature asymmetry, the density variations in the column arise from not only the changes in volume expansion but also the changes in concentration around the column perimeter.

Figure 1 gives a schematic representation of three possible cases of the development of parasitic convection; these are based on a simplified model which assumes that only two oppositely-directed parasitic flows exist in the column.

Case 1 (Fig. 1a) corresponds to an ideal geometry of the column, but nonuniform conditions of heating and cooling. Here the temperature nonuniformity in the column cross section causes a change of density, and parasitic currents result, i. e., each half of the column will operate as a column working selectively, and each of these halves will establish its own concentration distribution with respect to height. The difference between the average concentrations in each half of the column also causes changes in density, which may either coincide with or be oppositely directed to the changes arising from temperature asymmetry.

In case 2 (Fig. 1b) both surfaces of the column are ideally thermostated, but the column itself is geometrically imperfect, being characterized by the existence of eccentricity. The difference between the gaps in the two halves of the column again leads to both temperature and concentration asymmetry. Finally case 3 (Fig. 1c) unites the two previous cases, involving both nonuniform heating and cooling and also geometrical defects in the column.

Thus the difference between the densities of the separating mixture in the two halves of the column equals the sum of the changes due to the temperature and concentration nonuniformities, i. e.,

$$\Delta\rho = \rho [\beta (\delta T) + \gamma (\delta c)], \quad (1)$$

where (δT) and (δc) are the temperature and concentration asymmetries, defined as the differences between the mean temperatures and concentrations in the left and right parts of the column.

In the subsequent presentation we make the following simplifying assumptions. 1) Both parasitic flows have the same cross section. The basis for this assumption in the presence of temperature asymmetry only was given in [2]. 2) The parasitic flows have only one (vertical) velocity component, i. e., we neglect the

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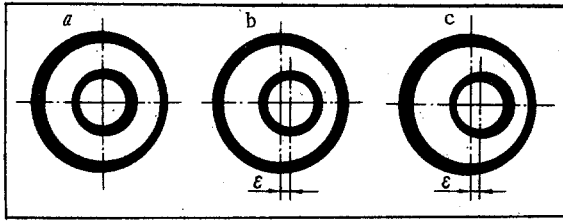


Fig. 1. Schematic representation of possible cases of temperature and concentration asymmetry in thermal-diffusion columns. The varying thickness of the black line qualitatively indicates the nonuniformity of heat transfer.

In contrast to [1] we shall take the coefficients H and K in the two parasitic flows as being identical, so as to simplify the final equations; this will not introduce any serious error if the ratio ε/δ is of the order of 1-2%.

Subsequently in order to simplify the calculations and present a clear final picture we shall confine ourselves to a binary mixture; we shall take: 1) $c(1-c) \approx c$, i. e., $c \ll 1$; 2) $c(1-c) \approx p$, where p is a constant. Then for case 1 (if we remember that in the steady-state condition $\text{div } j = 0$) we obtain the following instead of (2) and (3):

$$\frac{d^2 c'}{dy^2} - (1 + \kappa) \frac{dc'}{dy} = 0, \quad (4)$$

$$\frac{d^2 c''}{dy^2} - (1 - \kappa) \frac{dc''}{dy} = 0. \quad (5)$$

Let the thermal-diffusion column be closed at both ends. For each of the column halves under consideration we then obtain the obvious equations

$$j_1|_{y=y_e} = \sigma c'_e, \quad j_2|_{y=0} = -\sigma c''_i,$$

which on allowing for (2) and (3) and the condition $c \ll 1$ lead to the following two boundary conditions:

$$\left(\frac{dc'}{dy} - c' \right)_{y=y_e} = 0; \quad \left(\frac{dc''}{dy} - c'' \right)_{y=0} = 0. \quad (6)$$

On the other hand, since the column as a whole operates in the nonselective mode, the total flow of the component under consideration integrated over the height of the column should be equal to zero for any arbitrary cross-section under steady state conditions, i. e.,

$$j_1 + j_2 = 0,$$

or according to (2) and (3)

$$\frac{dc'}{dy} + \frac{dc''}{dy} - (1 + \kappa) c' - (1 - \kappa) c'' = 0. \quad (7)$$

Finally we must remember that the average concentration over the whole volume of the column should be equal to the initial value, i. e.,

$$\frac{1}{2y_e} \int_0^{y_e} (c' + c'') dy = c_0. \quad (8)$$

The integration constants found from conditions (6), (7), (8) entering into the solution of Eqs. (4) and (5)

$$c' = A_1 + B_1 e^{(1+\kappa)y}, \quad c'' = A_2 + B_2 e^{(1-\kappa)y} \quad (9)$$

take the form

$$A_1 = 2c_0 R \kappa (1 - \kappa) e^{(1+\kappa)y_e}, \\ B_1 = 2c_0 R (1 - \kappa),$$

column, regarding it as a small quantity. 3) We neglect diffusion in the azimuthal direction through the interface between the two flows; for thermal diffusion in the liquid phase this assumption is perfectly well-based [2].

Allowing for the foregoing assumptions, and considering each of the halves as a column in its own right, working with parasitic selection, we obtain the following transfer equations [1]

$$j_1 = Hc'(1 - c') - K \frac{dc'}{dz} + \sigma c', \quad (2)$$

$$j_2 = Hc''(1 - c'') - K \frac{dc''}{dz} - \sigma c''. \quad (3)$$

$$A_2 = -2c_0 R \kappa (1 + \kappa) e^{(1+\kappa)y_e}, \quad (10)$$

$$B_2 = 2c_0 R (1 + \kappa) e^{(1+\kappa)y_e},$$

where

$$R = \frac{y_e (1 - \kappa^2) e^{-(1+\kappa)y_e}}{(1 - \kappa)^2 [1 - e^{-(1+\kappa)y_e}] + (1 + \kappa)^2 [e^{(1-\kappa)y_e} - 1] - 2\kappa^2 y_e (1 - \kappa^2)}. \quad (11)$$

Substitution of (10) and (11) into (9) gives the following expressions for the concentration in each half of the column:

$$c' = 2c_0 R (1 - \kappa) [\kappa e^{(1+\kappa)y_e} + e^{(1+\kappa)y_e}], \quad (12)$$

$$c'' = 2c_0 R (1 + \kappa) e^{(1+\kappa)y_e} [e^{(1-\kappa)y_e} - \kappa]. \quad (13)$$

The average concentration in any arbitrary cross section will be equal to the half sum of the concentrations in each of the halves, i. e., on allowing for (12) and (13)

$$c \equiv \frac{1}{2} (c' + c'') = c_0 R \{ (1 - \kappa) [\kappa e^{(1+\kappa)y_e} + e^{(1+\kappa)y_e}] + (1 + \kappa) e^{(1+\kappa)y_e} [e^{(1-\kappa)y_e} - \kappa] \}. \quad (14)$$

Putting $\kappa = 0$ in (14) we obtain the well-known expressions for the change in concentration over the height of a column closed at both ends in the absence of parasitic convection.

Since the degree of separation in the column for low concentrations is practically equal to the ratio of the concentrations at the ends, at which $y = y_e$ and $y = 0$, remembering (14) we have

$$q \equiv \frac{c_e}{c_i} = \frac{(1 + \kappa) [1 - 2\kappa + e^{(1-\kappa)y_e}]}{(1 - \kappa) [1 + 2\kappa + e^{-(1+\kappa)y_e}]}. \quad (15)$$

This result coincides with that obtained in [1]. However, certain errors were committed in the latter for the case under consideration ($c \ll 1$), and the expression for R and hence Eqs. (12) and (13) accordingly assumed different forms.

We thus see from (12) and (13) that the imperfection of the column geometry inevitably creates a difference in concentrations between the two parasitic flows, and hence an additional change in density, accounted for by the second term in Eq. (1). Figure 2 shows how the concentrations vary along the column in each of the parasitic flows. We particularly notice that in one of the column halves the concentration is even lower than the initial value along the whole length for the same values of the parameters indicated in the figure caption.

This means that, if, in taking a sample for analysis, that sample is, by pure chance, taken from this particular half of the column, the research worker will observe, as it were, a reversal of the sign of the Soret coefficient.

For the case $c(1-c) \approx p$ the concentrations [1] are given by

$$c' = c_0 - \frac{p}{\kappa} + \frac{p}{\kappa} e^{-\kappa(y_e - y)}, \quad (16)$$

$$c'' = c_0 + \frac{p}{\kappa} - \frac{p}{\kappa} e^{-\kappa y}. \quad (17)$$

Let us now find the velocity of the parasitic flow. Let us assume (as in [3]) that the interface between the two parasitic flows is equivalent to a solid wall. This enables us to use the well-known formula for calculating the loss of pressure head associated with the flow of liquid in a channel in order to determine the velocity

$$\frac{\Delta P}{L} = \frac{\lambda}{\delta} \frac{\bar{w}^2}{2} \rho, \quad (18)$$

where the coefficient of friction in the narrow slotted channel for low Reynolds numbers is $\lambda = 96/\text{Re}$.

The loss of head defined by (18) equals the motive force of the parasitic convection, i. e., the difference between the specific gravities of the two flows. Thus remembering that $\text{Re} = \bar{w}\delta\rho/\eta$ we find that

$$\bar{w} = \frac{g\delta^2\Delta\rho}{48\eta},$$

or allowing for (1)

$$\bar{w} = \frac{g\delta^2\rho}{48\eta} [\beta(\delta T) + \gamma(\delta c)]. \quad (19)$$

Since the parasitic flow σ is related to the mean velocity by

$$\sigma = \frac{1}{2} \rho B \delta \bar{w},$$

on allowing for (19) we have

$$\sigma = \frac{g\rho^2\delta^3 B}{96\eta} [\beta(\delta T) + \gamma(\delta c)],$$

while the parasitic selection coefficient $\kappa = \sigma/H$ takes the following form on replacing the coefficient H of the transfer equation by its actual value

$$\kappa = \frac{15}{\beta_s (\Delta T)^2} [\beta(\delta T) + \gamma(\delta c)]. \quad (20)$$

Let us express the Soret coefficient in terms of the characteristics of the column and the mixture undergoing separation by using the equation

$$y_e = 504 \frac{s\eta DL}{g\rho\beta\delta^4}. \quad (21)$$

After introducing the nomenclature

$$7560 \frac{\eta DL(\delta T)}{g\rho\beta(\Delta T)^2\delta^4} = a, \quad 7560 \frac{\eta DL\gamma}{g\rho\beta^2(\Delta T)^2\delta^4} = b \quad (22)$$

Eq. (20) may be rewritten in the following form:

$$\kappa y_e = a + b(\delta c). \quad (23)$$

At the beginning of this article we drew attention to the fact that, in general (Fig 1c), temperature asymmetry arose both as a result of the nonuniformity of the heating and cooling processes and as a result of the geometrical imperfection of the column, i. e., the quantity (δT) entering into the parameter a may be expressed as a sum

$$(\delta T) = (\delta T)_T + (\delta T)_e, \quad (24a)$$

in which the temperature asymmetry due to the nonuniformity of heat transfer at the surfaces of the column (δT) and the temperature asymmetry due to the existence of an eccentricity $(\delta T)_e$ may naturally assume different signs. These quantities may be determined on the basis of elementary thermotechnical calculations. Thus in the case of a cylindrical column

$$(\delta T)_e = \frac{\varepsilon\Delta T}{\delta} \cdot \frac{h_1/h_2 - 1}{1 + h_1/h_2 + h_1\delta/d\lambda_1}, \quad (24b)$$

where the index 1 relates to the hotter heat carrier.

As regards the expression for $(\delta T)_T$, we shall not present this here since the heat-transfer coefficients which it contains (relating to the two halves of the column) cannot in practice be calculated, being functions of many random, uncontrollable factors. Hence $(\delta T)_T$ can only be found experimentally.

An analysis of the calculations leading to Eq. (24b) shows that for $h_1 > h_2$ the mean temperature of the liquid will be higher in the narrower part of the gap (the right-hand side in Fig. 1) than in the wider part, and owing to the difference in densities the liquid in the left half of the column will tend to move downward in the positive direction of the z axis. If $h_1 < h_2$ the picture will be the opposite, and the liquid will tend to move upward in the left half of the column. On the other hand, in the narrower part of the gap the concentration of the target component will always be higher than in the wider part, in accordance with the general theory of thermal-diffusion columns, i. e., $c'' - c' > 0$.

Hence the difference between the concentrations in the two parasitic flows may either accentuate the parasitic convection due to the temperature nonuniformity, or else weaken the over-all parasitic flow.

The original equations (4) and (5) were written on the assumption that the flux of concentration c' arising as a result of the combined action of the temperature and concentration asymmetry was directed in the positive z sense, i. e., $\kappa > 0$. However, if $\kappa < 0$, we must replace c' by c'' by c' in Eqs. (4) and (5) and also in the boundary conditions (6). This substitution has no effect on Eq. (15), determining the degree of separation, but the change in the sign of κ means that instead of (23) for $\kappa < 0$ we shall have

$$-\kappa y_e = a + b(\delta c), \quad (25)$$

while the difference in concentrations should be written $c' - c'' > 0$.

Thus the difference in concentrations between the two parasitic flow averaged over the height of the column is

$$(\delta c) = \pm \frac{1}{y_e} \int_0^{y_e} (c'' - c') dy. \quad (26)$$

Hence for $c(1-c) \approx p$ we obtain the following instead of (26) on substituting the corresponding values from (16) and (17)

$$(\delta c) = \pm \frac{2p}{\kappa y_e} \left[y_e - \frac{1}{\kappa} (1 - e^{-\kappa y_e}) \right], \quad (27)$$

while for the case $c \ll 1$, remembering (11)-(13), we have

$$(\delta c) = \pm 2c_0 \{ (1 + \kappa)^2 [e^{(1-\kappa)y_e} - 1] - (1 - \kappa)^2 [1 - e^{-(1+\kappa)y_e}] - 2\kappa y_e (1 - \kappa^2) \} \{ (1 + \kappa)^2 [e^{(1-\kappa)y_e} - 1] + (1 - \kappa)^2 [1 - e^{-(1+\kappa)y_e}] - 2\kappa^2 y_e (1 - \kappa^2) \}^{-1}, \quad (28)$$

where the upper sign corresponds to values $\kappa > 0$ and the lower to $-\kappa < 0$.

For $\kappa = 1$ after resolving the indeterminacy we obtain

$$(\delta c)_{\kappa=1} = \pm 2c_0 \frac{2y_e(y_e + 1) - 1 + e^{-2y_e}}{2y_e(y_e + 3) + 1 - e^{-2y_e}}.$$

For very large values of the parameter κ from Eq. (28) we find $(\delta c)_{\kappa \rightarrow \infty} \rightarrow 0$, which corresponds to the physical essence of the phenomenon under consideration, since a large value of κ indicates an intensive agitation, a reduction in the degree of separation as implied by Eq. (15), and hence a balancing of the concentrations in the two parasitic flows. It is also interesting to note that (δc) does not vanish when $\kappa = 0$. In this case Eqs. (27) and (28) give:

$$[(\delta c)_{\kappa=0} = \pm p y_e, \quad (30)$$

$$(\delta c)_{\kappa=0} = \pm 2c_0 \operatorname{th} \frac{1}{2} y_e, \quad (31)$$

i. e., concentration asymmetry will even exist when the same degree of separation is reached as in an ideal thermal-diffusion column. This phenomenon may occur when

$$a = \mp 2c_0 b \operatorname{th} \frac{1}{2} y_e, \quad (32)$$

as may clearly be seen from (23), i. e., for a specific value of the temperature asymmetry determined by Eq. (24a).

We see from (32), for example, that, if $b > 0$, then, in order that the parameter a should assume a negative value, it is essential that (putting $(\delta T)_T = 0$) the heat-transfer coefficient on the cold surface of the column should be greater than on the hot side.

Thus the removal of the temperature nonuniformity $[(\delta T) = 0]$ cannot be regarded as a procedure necessarily reducing the parasitic convection. This compensating effect will occur if both terms in (23) have the same signs; otherwise any reduction in the temperature asymmetry will worsen the conditions of separation in the column.

An analysis of Eqs. (27) and (28) leads to the conclusion that any increase in the dimensionless length of the column should produce an increase in the concentration asymmetry, the maximum value of which is $(\delta c)_{y_e \rightarrow \infty} = 2c_0$ for the case of $c \ll 1$. Hence a reduction in y_e enables us to reduce the harmful influence of parasitic convection.

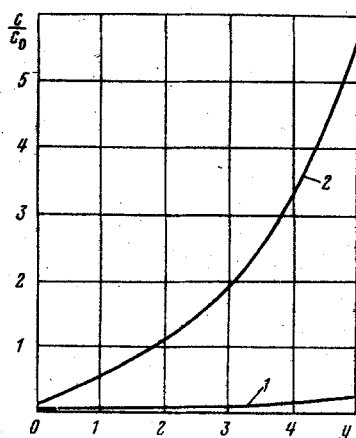


Fig. 2. Ratio c/c_0 as a function of the dimensionless length of the column in the left and right halves for $y_e = 5$; $\kappa = 0.5$: 1) c'/c_0 ; 2) c''/c_0 .

Figure 3a and b shows how, in the case of mixtures with $c \ll 1$, the parameter b , constituting the cause of the concentration asymmetry, influences the efficiency of the thermal-diffusion column for large and small values of $y \equiv \ln q^*$.

The value of $y_e = 10$ is typical of many organic mixtures usually separated in columns with $\delta = 0.25$ mm and a height of the order of 30–40 cm. We see from Fig. 3a that, even in the complete absence of temperature nonuniformity around the column perimeter ($\alpha = 0$), the concentration nonuniformity is an extremely important factor, sharply reducing the efficiency of the column.

The value of the concentration expansion coefficient entering into the parameter b for a mixture of nonpolar liquids may be determined reasonably accurately from the relation

$$\gamma = \frac{1}{\rho} \left(\frac{\partial \rho}{\partial c} \right)_T \approx \frac{\rho_1 - \rho_2}{\rho}, \quad (33)$$

where the indices 1 and 2 respectively refer to the pure components of the mixture.

For example, in the case of a mixture consisting of *n*-heptane and a trace of *n*-hexadecane, for an average gap temperature of 40°C obtain the value $\gamma = +0.14$. If the separation is carried out in a column with $\delta = 2.5 \cdot 10^{-4}$ m and $L = 0.3$ m, and if we remember that such physical characteristics as β , ρ , η should be equated to those of heptane on substituting into Eq. (22), then, remembering that for the mixture under consideration $D = 1.24 \cdot 10^{-9}$ m²/sec, for $\Delta T \approx 40^\circ\text{C}$ we obtain the value $b = +1.9 \cdot 10^3$. Let the initial concentration of *n*-hexadecane in the mixture be $c_0 = 10^{-3}$. Then $bc_0 = +1.9$. Since according to [4] this mixture has $s = 5.2 \cdot 10^{-3}$ deg⁻¹, from Eq. (20) we find $y_e = 11.4$. If we now make use of the data of Fig. 3a, we see that, depending on the degree of temperature asymmetry, the efficiency of the column may fluctuate over an extremely wide range. This example is typical in that it establishes the strong influence of concentration inhomogeneity, even for comparatively low initial concentrations. We may well consider that for $y_e \approx 10$ the influence of this factor only becomes negligibly small when $c_0 \leq 10^{-4}$.

For small y_e , such as are characteristic of liquid isotope mixtures, concentration asymmetry should only appear to a marked degree for large values of bc_0 . For these mixtures experimental data as to the densities only exist for certain deuterized organic compounds [5].

However, to an accuracy sufficient for the problem under consideration, we may calculate the concentration expansion coefficient from the equation

$$\gamma = \frac{\Delta M}{M}, \quad (33a)$$

where ΔM is the difference between the molecular weights of the two isotopic compounds.

For example, in the case of bromine isotopes in butyl bromide for $\bar{T} = 333^\circ\text{K}$, $\gamma = 0.012$ in a column with the dimensions just considered $b \approx 80$. Hence for isotope mixtures in the range $c \ll 1$ concentration asymmetry need not be taken into account in short columns. However, on using longer columns the effect of this factor may be very considerable, as witnessed by the experiments of Abelson and Hoover [6].

Whereas the parameter b may be calculated from the known physical characteristics of the mixture being separated and the geometrical dimensions of the column, the determination of parameter a encounters

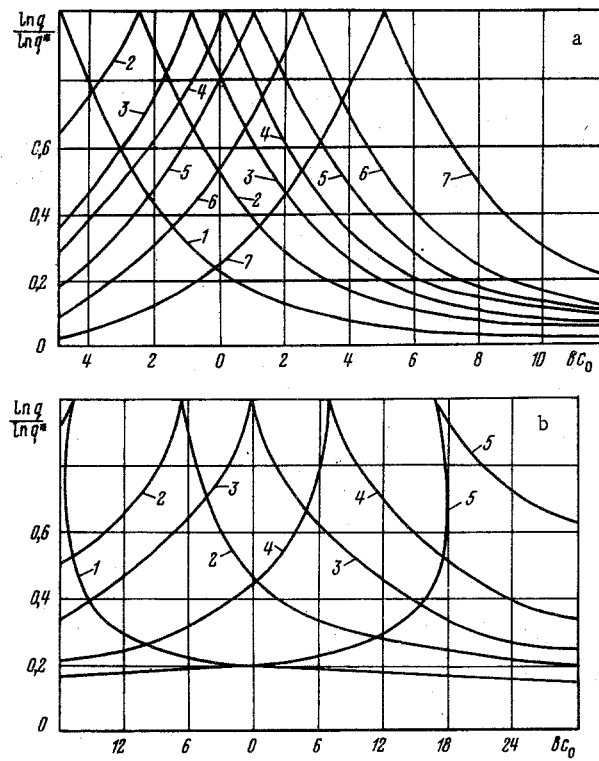


Fig. 3. Efficiency of the thermal-diffusion column $\ln q/\ln q^*$ as a function of the concentration asymmetry bc_0 for various temperature asymmetries a) ($y_e = 10$ and $c \ll 1$) 1) $a = +10$; 2) $+5$; 3) $+2$; 4) 0 ; 5) -2 ; 6) -5 ; 7) -10 ; b) ($y_e = 0.3$ and $c \ll 1$) 1) $a = +5$; 2) $+2$; 3) 0 ; 4) -2 ; 5) -5 .

serious difficulties because of the indeterminacy of the temperature asymmetry, as expressed by Eq. (24a). With modern methods of calculating heat-transfer coefficients, and assuming careful preparation of the column, the second term in this equation may be calculated to an acceptable accuracy by Eq. (24b). As regards the first term, as already noted, this has to be determined experimentally. The possibility thus arises of using the thermal-diffusion column as an instrument for determining the Soret coefficient in mixtures for which the traditional method (based on the use of a cell) is ineffective. To this end we have to carry out an experiment with a standard mixture the Soret coefficient of which is already known, so that the quantity y_e is known also [see Eq. (21)]. The degree of separation q is determined by experiment. Then by using Eq. (15) we find the parasitic selection parameter κ , and from Eq. (23) the unknown parameter a . The experiments with the test mixture should be carried out with the same column assembly, and the same flows of heating and cooling carriers; the new value of a will, on the basis of Eq. (2), take the form

$$a = a_{st} \left(\frac{\eta D}{g \rho \beta} \right) \left(\frac{g \rho \beta}{\eta D} \right)_{st} \frac{(\Delta T)_{st}^2}{(\Delta T)^2} \frac{(\delta T)}{(\delta T)_{st}}, \quad (34)$$

where the index "st" relates to the standard mixture. The experimental value of the degree of separation provides us with a system of two equations (15) and (23), containing two unknowns y_e and κ , the first of which contains the desired Soret coefficient.

In conclusion, it is useful to note that the new approach to the quantitative analysis of parasitic convection set out in this paper has no connection with the so-called "forgotten effect" mentioned elsewhere [7-9].

This effect arises as a result of changes in density with changing concentration, not in the parasitic flows, which the authors of the papers in question never considered, but in the main convective flows; as indicated in [7, 8] the influence of this effect in the region of low concentrations is negligibly small, as in the case of a small concentration expansion coefficient.

The special feature of our own theory lies in the fact that it establishes the substantial influence of the concentration asymmetry arising in parasitic convective flows on the efficiency of the column under standard operating conditions for small values of the initial concentrations. As indicated in [9], the "forgotten effect" has no influence on the final result of the separation process under steady-state conditions.

NOTATION

$\Delta\rho = \rho_1 - \rho_2$;	
ρ	is the density;
$\beta = (1/\rho)(\partial\rho/\partial T)$;	
$\gamma = (1/\rho)(\partial\rho/\partial c)T$;	
(δT)	is the temperature asymmetry;
(δc)	is the concentration asymmetry;
j	is the flow of the target component;
c	is the concentration;
$H = g\beta\rho^2 s\delta^3 (\Delta T)^2 B/6!\eta$;	
$K = g^2\rho^3\beta^2\delta^7 (\Delta T)^2 B/9!\eta^2 D$;	
δ	is the gap between the hot and cold walls;
B	is the perimeter of the separating gap;
ΔT	is the temperature difference;
s	is the Soret coefficient;
D	is the diffusion coefficient;
σ	is the parasitic flow;
z	is the vertical coordinate;
$y = Hz/K$;	
L	is the length of column;
$y_e \equiv \ln q^* = HL/K$;	
q^*	is the degree of separation in the column without parasitic convection;
q	is the degree of separation in the column with parasitic convection;
ΔP	is the pressure drop;
\bar{w}	is the mean velocity of the parasitic flow;
η	is the viscosity;
α_j	is the heat-transfer coefficient from the side of the j -th heat carrier;
$h_j = \alpha_j d_{ij}/[1 + \alpha_j d_{ij} \ln(d_{ex}/d_{in})_i/\lambda_{wi}]$, d_{ij}	are the diameter of the i -th cylinder from the side of the j -th heat carrier;
$(d_{ex}/d_{in})_i$	is the ratio of the external and internal diameters of the i -th cylinder;
d	is the diameter of the annular gap;
λ_{wi}	is the thermal conductivity of the wall of the i -th cylinder;
λ_l	is the thermal conductivity of the mixture being separated;
M	is the molecular weight;
ϵ	is the eccentricity.

Subscripts

one and two primes respectively refer to the first and second parasitic flows;
 e denotes positive end of the column;
 i denotes negative end of the column.

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