TRANSIENT IN A THERMODIFFUSION COLUMN WITH TEMPERATURE ASYMMETRY

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On the basis of an already known physical model, quantitative relations are derived which describe the transient process in thermodiffusion columns with temperature asymmetry.

The theory of thermodiffusion columns developed by Jones and Ferry in [1] is based on an idealization of the process, which disregards any temperature asymmetry due to the unattainability of both a uniform heating of the active column surfaces and a constant clearance between them.

For this reason, the heat-transfer process in a real thermodiffusion apparatus cannot be described by the equation

$$j = Hc\bar{c} - (K_c + K_d)\frac{dc}{dz}, \tag{1}$$

since the latter does not account for perturbations due to temperature asymmetry.

In view of this, the authors of [1] have added inside the parentheses in (1) another term K_s which represents the so-called stray transfer. It has been shown in [2] that, in the physical sense, the premise on which the same term $K_s(dc/dz)$ is introduced into (1) contradicts the experimental data on the partition of liquid isotope mixtures and, therefore, another physical model has been proposed instead, in which a column with stray convection is treated as an aggregate of column operating in the extraction mode and with the stray convection current acting as the extractor. The main parameter which determines the deviation of a column performance from the performance of the ideal column according to Eq. (1) is, to the first approximation,

$$\varkappa = \frac{15}{2}\cdot\frac{\bar{T}(\delta T)}{\alpha(\Delta T)^2}. \tag{2}$$

The larger is this parameter, the larger appears the effect of stray convection. Relation (2) explains why the test results in the partition of molecular liquid and gaseous mixtures confirm the Jones–Ferry theory based on Eq. (1). Indeed, for mixtures of different gases and liquids and magnitude of α, of the order of 10^{-1}, is rather high and, consequently, even at a relatively high degree of temperature asymmetry the parameter $\varkappa$ is sufficiently small to make the deviation between a real column and an ideal column performance slight. The thermal diffusivity for isotopic mixtures is by one order of magnitude smaller than that and, therefore, in thermodiffusion plants designed for isotope partitions the parameter $\varkappa$ may become sufficiently high to make the performance of a column deviate quite significantly from the theoretically predicted performance. This fact has been emphasized, particularly in [3]. In the meantime, the quantity K_s introduced by the authors of [1] is independent of α and, therefore, identical hydrodynamic conditions should, according to [1], produce equal results in the partition of molecular and isotopic mixtures.

In this way, the equation of transfer in a real thermodiffusion apparatus is characterized, as has been shown in [2], by the additional term which accounts for stray convection. Obviously, the transient processes in a real and in an ideal apparatus will differ and the time to reach the enriched-product extraction mode will also be different.

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In this article we consider the case of temperature asymmetry when the column contains two stray currents, equal in magnitude but opposite in direction and characterized by equal transfer coefficients H and K.

According to [2], in this case

$$j' = Hc'\overline{c'} - K\frac{dc'}{dz} + \sigma c', \tag{3}$$

$$j'' = Hc''\overline{c''} - K\frac{dc''}{dz} - \sigma c''. \tag{4}$$

For the transient state we have, evidently,

$$m'\frac{\partial c'}{\partial \tau} = -\operatorname{div} \mathbf{j}'; \qquad m''\frac{\partial c''}{\partial \tau} = -\operatorname{div} \mathbf{j}''. \tag{5}$$

Assuming, for simplicity, that $m' = m''$, we obtain from (5), taking into account (3) and (4) and changing to dimensionless variables:

$$\begin{gathered}\frac{\partial c'}{\partial \theta} = \frac{\partial^2 c'}{\partial y^2} - \frac{\partial (c'\overline{c'})}{\partial y} - \varkappa\frac{\partial c'}{\partial y};\\ \frac{\partial c''}{\partial \theta} = \frac{\partial^2 c''}{\partial y^2} - \frac{\partial (c''\overline{c''})}{\partial y} + \varkappa\frac{\partial c''}{\partial y}.\end{gathered} \tag{5a}$$

Let us consider the simplest case of a transient process in a real column connected at one end to a large reservoir, with $c\overline{c}$ = const. Instead of the last two equations we have now:

$$\frac{\partial c'}{\partial \theta} = \frac{\partial^2 c'}{\partial y^2} - \varkappa\frac{\partial c'}{\partial y}; \tag{6}$$

$$\frac{\partial c''}{\partial \theta} = \frac{\partial^2 c''}{\partial y^2} + \varkappa\frac{\partial c''}{\partial y}. \tag{7}$$

The solution to (6) and (7) must satisfy the following boundary conditions:

$$c'|_{\theta=0} = c''|_{\theta=0} = c_0; \tag{8}$$

$$\left(aH + \sigma c' - K\frac{\partial c'}{\partial z}\right)_{z=L} = \sigma c'|_{z=L}; \tag{9}$$

$$\left(aH - \sigma c'' - K\frac{\partial c''}{\partial z}\right)_{z=L} = -\sigma c'|_{z=L}; \tag{10}$$

$$c'|_{z=0} = c_0; \qquad c''|_{z=0} = c_0. \tag{11}$$

Conditions (9) and (10) indicate that the stray current is flowing from one half of the column to the other.*

A solution of (6) and (7) with the conditions (8)-(11) by the method of integral Laplace–Carson transformations yields the following image functions:

$$\overline{u}' = a\exp\left[-\frac{\varkappa}{2}(y_e - y)\right]\frac{\lambda \operatorname{sh}\lambda y}{\lambda^2 \operatorname{ch}\lambda y_e + \frac{\varkappa}{2}\lambda\operatorname{sh}\lambda y_e}; \tag{12}$$

$$\overline{u}'' = a\exp\left[-\frac{\varkappa}{2}(y_e - y)\right]\left[\frac{\lambda \operatorname{sh}\lambda y}{\lambda^2 \operatorname{ch}\lambda y_e + \frac{\varkappa}{2}\lambda\operatorname{sh}\lambda y_e} + \varkappa\frac{\lambda^2\operatorname{sh}^2\lambda y_e}{\left(\lambda^2 \operatorname{ch}\lambda y_e + \frac{\varkappa}{2}\lambda\operatorname{sh}\lambda y_e\right)^2}\right], \tag{13}$$

where $\lambda = \sqrt{\frac{\varkappa^2}{4} + p}$ and $\overline{u}$ is the image of the difference $c - c_0$. Inverting to the original functions and considering that the concentration at the positive end of the column is, for this pattern of stray fluxes, equal to the arithmetic mean of the concentrations at both, i.e., $c = (c' + c'')/2$, we have

*A rigorous formulation of the boundary conditions is given in [2].

$$\Delta c = c - c_0 = a\left\{\frac{2}{\varkappa}\exp\left(-\frac{\varkappa}{2}y_e\right)\operatorname{sh}\frac{\varkappa}{2}y_e + \frac{2}{\varkappa}\exp(-\varkappa y_e)\operatorname{sh}^2\frac{\varkappa}{2}y_e\right.$$

$$-8y_e\sum_{n=1}^{\infty}\frac{\mu_n^2\exp\left(-\frac{4\mu_n^2+\varkappa^2y_e^2}{4y_e^2}\theta\right)}{(4\mu_n^2+\varkappa^2y_e^2)\left[\frac{\varkappa}{2}y_e\left(1+\frac{\varkappa}{2}y_e\right)+\mu_n^2\right]}$$

$$-32\varkappa y_e^2\sum\frac{\mu_n^4\exp\left(-\frac{4\mu_n^2+\varkappa^2y_e^2}{4y_e^2}\theta\right)}{(4\mu_n^2+\varkappa^2y_e^2)^2\left[\frac{\varkappa}{2}y_e\left(1+\frac{\varkappa}{2}y_e\right)+\mu_n^2\right]^2}$$

$$\left.\times\left[\frac{3}{2}+\frac{\varkappa}{2}y_e+\frac{\varkappa^2y_e^2(\varkappa y_e-1)}{8\mu^2}-\frac{\varkappa y_e}{2\mu^2+\varkappa y_e\left(1+\frac{\varkappa}{2}y_e\right)}+\frac{4\mu^2+\varkappa^2y_e^2}{4y_e^2}\theta\right]\right\}, \tag{14}$$

where μ_n are the roots of the characteristic equation

$$\operatorname{tg}\mu_n = -\frac{2\mu_n}{\varkappa y_e}. \tag{15}$$

In the steady state there will remain only the first two terms in (14) and, when the hyperbolic sine is replaced with its exponential equivalents, we have

$$\Delta c_\infty = \frac{a}{2\varkappa}(1-e^{-\varkappa y_e})(3-e^{-\varkappa y_e}), \tag{16}$$

which is the same results as in [2]. A linear approximation $c\bar{c} = a + bc$ yields, instead of (5a),

$$\begin{aligned}\frac{\partial c'}{\partial\theta} &= \frac{\partial^2c'}{\partial y^2}-(b+\varkappa)\frac{\partial c'}{\partial y},\\ \frac{\partial c''}{\partial\theta} &= \frac{\partial^2c''}{\partial y^2}-(b-\varkappa)\frac{\partial c''}{\partial y}.\end{aligned} \tag{17}$$

For the positive end of the column the solution to these equations with conditions (8)-(11) is

$$c_e - c_0 = \frac{1}{2}(a+bc_0)\left\{\frac{\operatorname{th}k_1y_e}{k_1-k_2\operatorname{th}k_1y_e}+\frac{[k_1-(k_2-\varkappa)\operatorname{th}k_1y_e]\operatorname{th}k_2y_e}{k_2(k_1-k_2\operatorname{th}k_1y_e)(1-\operatorname{th}k_2y_e)}+2y_e\sum_{n=1}^{\infty}\frac{r_n^2}{k_2y_e(1-k_2y_e)+r_n^2}\right.$$

$$\left.\times\left[\frac{[\lambda_1\operatorname{ch}\lambda_1-y_e(k_2-\varkappa)\operatorname{sh}\lambda_1]\exp\left(-\frac{r_n^2-k_1y_e^2}{y_e^2}\theta\right)}{(r_n^2-k_1^2y_e^2)(\lambda_1\operatorname{ch}\lambda_1-k_2y_e\operatorname{sh}\lambda_1)}+\frac{[\lambda_2\operatorname{ch}\lambda_2-y_e(k_2-\varkappa)\operatorname{sh}\lambda_2]\exp\left(-\frac{r_n^2-k_2^2y_e^2}{y_e^2}\theta\right)}{(r_n^2-k_2^2y_e^2)(\lambda_2\operatorname{ch}\lambda_2-k_2y_e\operatorname{sh}\lambda_2)}\right]\right\},$$

where

$$\begin{aligned}\lambda_1 &= \sqrt{r_n^2-\varkappa by_e^2}; \quad \lambda_2 = \sqrt{r_n^2+\varkappa by_e^2};\\ k_1 &= \frac{1}{2}(b+\varkappa); \quad k_2 = \frac{1}{2}(b-\varkappa),\end{aligned} \tag{18}$$

and the roots r_n are determined from the equation:

$$\operatorname{th}r_n = \frac{2r_n}{(b-\varkappa)y_e}, \tag{19}$$

when $c_0 \geq 0.5, b < 0$ and Eq. (19) has purely imaginary roots. When $c_0 < 0.5$, then $b > 0$ and Eq. (18) may have, in addition to the imaginary roots, one real root, if $(b-\varkappa)/2 > 1$.

It follows from (14) that the change in concentration at the positive end of a column depends on two parameters: $\varkappa$ and y_e.

The character of this relation is shown in Fig. 1 for $y_e = 0.3$. As can be seen here, the most intensive partition occurs when $\varkappa = 0$ (curve 1 plotted for $c_0 = 0.5$). As the parameter $\varkappa$ increases, the transient time becomes shorter and the final effect of the partition process is diminished.

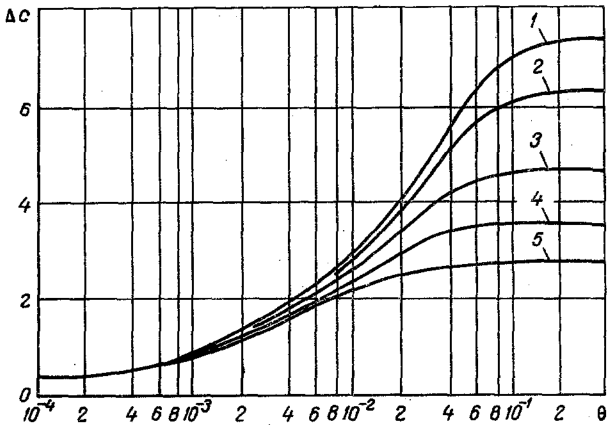

Fig. 1. Curves representing the percent increment of concentration at the positive end of a column ($\Delta c = c_e - c_0$), as a function of time θ (dimensionless), for $y_e = 0.3$ and $\varkappa = 0$ (1), 3.33 (2), 6.7 (3), 10 (4), 13 (5).

It is particularly noteworthy that, up to $\theta = 10^{-3}$ (initial period), stray convection plays a practically insignificant role in the process, which is very important for the experimental determination of thermal diffusivity.

In liquid thermodiffusion columns it is hardly feasible to ensure thermostatic protection at the cold and at the hot surface carefully enough to reduce the temperature asymmetry to less than 0.2°C. With this estimate, one can now determine the effectiveness of liquid thermodiffusion columns.

For an example we will use the data in [4] on bromide partition in bromobenzene: $\alpha = 0.04$, $\bar{T} = 340°K$, $\Delta T = 130°C$. Equation (2) yields $\varkappa = 0.755$. This value of $\varkappa$ corresponds to a steady-state $\Delta c = 7.2\%$ in Fig. 1 Taking the ratio $(\Delta c)/(\Delta c)_0$ as the measure of effectiveness, where $(\Delta c)_0$ denotes the increment of concentration at the positive end of the ideal column, we find here $(\Delta c)/(\Delta c)_0 = 0.96$. The value thus obtained may, evidently, be seen as the upper limit of technical feasibility.

When $\delta T = 1°C$, we already have only $(\Delta c)/(\Delta c)_0 = 0.8$.

Indeed, as is well known, the thermodiffusive partition process is never carried to a complete steady state and extraction from the column begins when the concentration at the end of the column is below its equilibrium level. In this case, as is evident from the diagram, stray convection has less influence on the effectiveness of partition.

Thus, for example, for $\varkappa = 3.33$ after a time $\theta = 4 \cdot 10^{-2}$ the effectiveness will still be sufficiently high and equal to $(\Delta c)/(\Delta c)_0 = 0.92$, while $(\Delta c)/(\Delta c)_0 = 0.83$ in the steady state.

The results obtained here may be used as a basis for an efficient design and operation of thermodiffusion apparatus.

NOTATION

H, K	are transfer coefficients;
σ	is the extraction;
c	is the concentration;
$\bar{c} = 1 - c$;	
L	is the column length;
$y = Hz/K$;	
$y_e = HL/K$;	
$\theta = H^2\tau/mK$;	
m	is the mass of liquid per unit column length;
z	is the longitudinal coordinate;
$a = \bar{c}c$;	
α	is the thermal diffusivity;
$\bar{T}$	is the mean temperature in a column;
ΔT	is the temperature difference between the hot and the cold wall;
δT	is the temperature difference between the two stray fluxes;
p	is an operator;
'	refers to the first stray flux;
"	refers to the second stray flux.

Superscripts

' refers to the first stray flux;
" refers to the second stray flux.

Subscripts

0 denotes the initial state;

e denotes the value at the positive end of the column;

∞ denotes the equilibrium value, steady state.

LITERATURE CITED

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