## INFLUENCE OF THE LONGITUDINAL TEMPERATURE GRADIENT ON SEPARATION IN A PLANE THERMAL DIFFUSION COLUMN

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The problem of determining the separation coefficient of a plane diffusion column with a wall temperature which varies along the height is solved.

We will examine the problem of thermal diffusion separation of isotopes in the liquid phase in a column for which

T = T(x, z),

the axis z is directed along the column, and the axis x is directed across the gap. The thermal diffusion process taking into account the condition of quasi-steady state is described by the equation

$$\operatorname{div} \mathbf{j} = \mathbf{0},\tag{1}$$

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where

$$\mathbf{j} = \rho \mathbf{v} c - \rho D \left( \nabla c - \frac{\alpha}{T} c \overline{c} \nabla T \right).$$
(2)

Substituting (2) into (1) and introducing simplifications, based on the condition  $\frac{L}{\delta} \gg 1$ ,  $\frac{\partial^2 c}{\partial x^2} \gg \frac{\partial^2 c}{\partial z^2}$ ,  $\frac{\partial T}{\partial x} \cdot \frac{\partial (c\overline{c})}{\partial x} \gg \frac{\partial T}{\partial z} \cdot \frac{\partial (c\overline{c})}{\partial z}$ , and paying attention that  $\Delta T = 0$  (the heat exchange process in the column – the thermal conductivity process), we will obtain

$$\rho u \frac{\partial c}{\partial x} + \rho v \frac{\partial c}{\partial z} - \rho D \frac{\partial^2 c}{\partial x^2} + \frac{\rho D \alpha}{T} \cdot \frac{\partial T}{\partial x} \cdot \frac{\partial (c \bar{c})}{\partial x} = 0.$$
(3)

On deriving equation (3) it is assumed that  $\rho$ , D,  $\alpha$  do not depend on the temperature and concentration.

As usual [1], the current of the light component towards the top of the column (with the condition that the axis z is directed upwards) is given by the equation

$$\tau = B\left(\int_{0}^{b} \frac{\partial c}{\partial x} \Phi(x, z) dx - \int_{0}^{b} D\rho \frac{\partial c}{\partial z} dx\right) + \sigma c, \qquad (4)$$

where  $\Phi(\mathbf{x}, \mathbf{z}) = -\int_{0}^{x} \rho v d\mathbf{x}$  is what is known as the function of the flow.

The value  $\partial c/\partial x$  is determined from equation (3) by consecutive approximation. For the first approximation we will adopt the value  $\partial c/\partial x$  for a case in which there is no longitudinal temperature gradient in the thermal diffusion column and consequently u = 0.

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From [1] we have

$$\frac{\partial c}{\partial x} = -\frac{1}{\rho D} \cdot \frac{\partial c}{\partial z} \Phi + \frac{\alpha}{T} \cdot \frac{\partial T}{\partial x} c\bar{c}.$$
(5)

Substituting the value  $\partial c/\partial x$  into the first term of equation (3) and integrating it with respect to the coordinate x, assuming that  $\overline{cc}$  and  $\partial c/\partial z$  do not depend on x, we will obtain a new value for  $\partial c/\partial x$ :

$$\frac{\partial c}{\partial x} = -\frac{1}{\rho D} \cdot \frac{\partial c}{\partial z} \Phi \left( 1 + \frac{f}{D} \right) + \frac{\alpha}{T} \cdot \frac{\partial T}{\partial x} \overline{c} \left( 1 + \frac{f}{D} \right), \tag{6}$$

where  $f = \int_{0}^{x} udx$  is the function of the flow in the lateral direction, if  $f \ll D$  for  $\partial c/\partial x$  we obtain the expression of the form (5). Expression (6) can be used for the second approximation according to the determination  $\partial c/\partial x$  etc.

In the n-th approximation the expression for  $\partial c/\partial x$  will have the form

$$\frac{\partial c}{\partial x} = -\frac{1}{\rho D} \cdot \frac{\partial c}{\partial z} \Phi \left[ 1 + \sum_{n=1}^{n} \frac{f^{n}}{D^{n}} \right].$$
$$+ \frac{\alpha}{T} \cdot \frac{\partial T}{\partial x} \frac{c \overline{c}}{c \overline{c}} \left[ 1 + \sum_{n=1}^{n} \frac{f^{n}}{D^{n}} \right].$$
(6')

Substituting (6') into (4) for  $\tau$  we will obtain the expression

$$\tau = Hc\bar{c} - K \frac{dc}{dz} + \sigma c, \tag{7}$$

where

$$H = B \int_{0}^{0} \left\{ \frac{\alpha}{T} \cdot \frac{\partial T}{\partial x} \Phi(x; z) \left[ 1 + \sum_{1}^{n} \frac{f^{n}}{D^{n}} \right] \right\} dx,$$

$$K = B \int_{0}^{0} \left\{ \frac{1}{\rho D} \Phi^{2}(x, z) \left[ 1 + \sum_{1}^{n} \frac{f^{n}}{D^{n}} \right] \right\} dx + B \int_{0}^{0} D\rho dx.$$
(8)

For the case of a column operating without separating, the equation for the flow in the steady state will be

$$\tau = Hc\bar{c} - K \frac{dc}{dz} = 0.$$
<sup>(9)</sup>

As distinct from the Jones and Ferry equation here H = H(z), K = K(z).

Equation (9) is the Bernoulli equation which can be integrated by squares. For the case of a column operating with separation we will obtain the Rikkati equation

$$H(z)c\overline{c} - K(z)\frac{dc}{dz} + \sigma(c_e - c) = 0.$$
<sup>(10)</sup>

This equation is not usually integrated by squares, as the solution can be obtained for the linear approximation  $\overline{cc} = a + bc$ .

We will examine the thermal diffusion plane column in which the temperature on the walls is assigned in the following way:

 $x=0, T=T_0, x=\delta, T=T_1+kz,$ 

where k is constant.

Solution of the problem does not vary in principle in the case of linear variation of the temperatures on both walls and the given system is chosen only for simplification of the calculations. The equation of movement for a steady state case

$$\frac{\partial \Psi}{\partial z} \cdot \frac{\partial (\Delta \Psi)}{\partial x} - \frac{\partial \Psi}{\partial x} \cdot \frac{\partial (\Delta \Psi)}{\partial z} = v\Delta \Delta \Psi + \beta g \frac{\partial T}{\partial x} , \qquad (11)$$

where, as usually,

$$u = -\frac{\partial \Psi}{\partial z}; \ v = \frac{\partial \Psi}{\partial x}; \ \beta = -\frac{1}{\rho} \left(\frac{\partial \rho}{\partial T}\right)_{\rho}.$$

The energy equation

$$v \ \frac{\partial T}{\partial z} + u \ \frac{\partial T}{\partial x} = a \left( \frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial z^2} \right).$$
(12)

For the case of the thermal diffusion column used for separation of isotopes in the liquid phase considerable simplifications are possible in the equations of movement and energy. We will write the equation of movement in the coordinate form:

$$v \frac{\partial v}{\partial z} + u \frac{\partial v}{\partial x} = -\frac{1}{\rho} \cdot \frac{\partial p}{\partial z} + g\beta\theta + v \left[ \frac{\partial^2 v}{\partial x^2} + \frac{\partial^2 v}{\partial z^2} \right], \qquad (13)$$

$$v \ \frac{\partial u}{\partial z} + u \ \frac{\partial u}{\partial x} = -\frac{1}{\rho} \cdot \frac{\partial p}{\partial x} + v \left[ \frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial z^2} \right].$$
(14)

From the balance of liquid in the gap

$$u \approx \frac{\delta}{2} \cdot \frac{\partial v}{\partial z} . \tag{15}$$

Taking into account (15), we will carry out an evaluation of the terms in equations (13) and (14):

$$v \frac{\partial v}{\partial z} \sim \frac{v^2}{L}$$
;  $u \frac{\partial v}{\partial x} \sim \frac{v^2}{2L}$ ;  $v \frac{\partial^2 v}{\partial x^2} \sim v \frac{v}{\delta^2}$ 

hence

$$\frac{v \frac{\partial v}{\partial z}}{v \frac{\partial^2 v}{\partial x^2}} \sim \frac{\delta^2 v}{Lv}; \quad \frac{u \frac{\partial v}{\partial x}}{v \frac{\partial^2 v}{\partial x^2}} \sim \frac{\delta^2 v}{2Lv} \quad (16)$$

In the case of thermal diffusion separation in the liquid phase for columns, the characteristic magnitudes will be [1]  $\delta = 2.5 \cdot 10^{-4} \text{ m}$ , L  $\approx 0.5 \text{ m}$ ,  $\nu = 0.5 \cdot 10^{-6} \text{ m}^2/\text{sec}$ , the longitudinal component of the speed of the convective flow in the case of  $\Delta T_x \approx 100^{\circ}$ C,  $v \approx 10^{-3} \text{ m/sec}$ .

Substituting these values into (16), we will obtain

$$\frac{v \frac{\partial v}{\partial z}}{v \frac{\partial^2 v}{\partial x^2}} \approx 10^{-4}; \quad \frac{u \frac{\partial v}{\partial x}}{v \frac{\partial^2 v}{\partial x^2}} \approx 10^{-4},$$

according to the same considerations in equation (14)

$$\frac{v \frac{\partial u}{\partial z}}{v \frac{\partial^2 u}{\partial x^2}} \approx 10^{-4}; \quad \frac{u \frac{\partial u}{\partial x}}{v \frac{\partial^2 u}{\partial x^2}} \approx 10^{-4}.$$

Consequently, in equations (13) and (14) it is possible to disregard the nonlinear terms. Hence, by introducing the stream function  $\Psi$  and eliminating from the equation the pressure term, we will write the equation of movement in the form

$$v\Delta\Delta\Psi + \beta g \ \frac{\partial T}{\partial x} = 0 \tag{17}$$

with the boundary conditions x = 0,  $\delta: \partial \Psi / \partial x = \partial \Psi / \partial z = 0$ .

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On analyzing equation (12) we will note that in the examined problem the temperature variation along the axis z has the character of temperature nonuniformities; hence  $\Delta T_Z \leq T_X$ , where  $\Delta T_Z$ ,  $\Delta T_X$  are the maximum temperature differences along the length of the column and across the gap respectively.

We will carry out evaluation of the terms in (12), paying attention to the relationship:

$$v \frac{\partial T}{\partial z} \sim v \frac{T}{L}; \quad u \frac{\partial T}{\partial x} \sim \frac{vT}{2L}; \quad a \frac{\partial^2 T}{\partial x^2} \sim a \frac{T}{\delta^2},$$
$$\frac{v \frac{\partial T}{\partial z}}{a \frac{\partial^2 T}{\partial x^2}} \sim \frac{v\delta^2}{aL}; \quad \frac{u \frac{\partial T}{\partial x}}{a \frac{\partial^2 T}{\partial x^2}} \sim \frac{v\delta^2}{2aL} \cdot$$
(18)

hence

Substituting the magnitudes characteristic for thermal diffusion in the liquid phase (for the liquids  $a = 10^{-4}$  to  $5 \cdot 10^{-4}$  m<sup>2</sup>/h) into (18) we will obtain

$$\frac{v \frac{\partial T}{\partial z}}{a \frac{\partial^2 T}{\partial x^2}} \sim \frac{u \frac{\partial T}{\partial x}}{a \frac{\partial^2 T}{\partial x^2}} \sim 10^{-3}$$

Consequently it is possible to neglect the left hand part in equation (12) and equation (12) is transformed into the thermal conductivity equation:

$$\Delta T = 0. \tag{19}$$

The boundary conditions:

$$x=0, T=T_0, x=\delta, T=T_1+kz$$

The solution (19) taking the boundary conditions into account will be the function

$$T = T_0 + (m + nz) x_{,}$$

where

$$m = \frac{T_1 - T_0}{\delta}$$

Substituting the value T into equation (13), we will obtain the equation of movement in the form

$$v\Delta\Delta\Psi + \beta g (m + nz) = 0. \tag{20}$$

We will look for the solution (20), as usual, in the form

$$\psi = \varepsilon(x) + \varphi(x)(m + nz). \tag{21}$$

Substituting (21) into (20) gives two equations for determining  $\varepsilon(x)$  and  $\varphi(x)$ :

$$\varepsilon(x)^{\rm IV}=0, \tag{22}$$

$$\varphi^{\rm IV} + \frac{\beta g}{\nu} = 0 \tag{23}$$

with the boundary conditions

-

x=0, 
$$\delta$$
:  $\varphi=0$ ;  $\frac{d\varphi}{dx}=0$ ;  $\frac{d\varepsilon}{dx}=0$ ;  $\varepsilon(\delta)-\varepsilon(0)=\frac{\sigma}{B\rho}$ . (24)

The last condition is the condition for conservation of mass in the gap of the thermal diffusion column. Solution of equations (22) and (23) taking into account the boundary conditions (24) gives the following expressions for u and v:

$$v = -\frac{\beta g}{6\nu} x \left(x - \frac{\delta}{2}\right) (x - \delta) (m + nz) + \frac{6\sigma}{\delta^3 B\rho} x (\delta - x),$$
$$u = \frac{\beta gn}{24\nu} x^2 (x - \delta)^2.$$

In order to determine the thermal diffusion coefficients H and K from (8) we will evaluate the magnitudes f: for  $\delta = 0.25$  mm and the parameters of the liquid  $\beta = 10^{-3} \text{ deg}^{-1}$ ;  $\nu = 0.5 \cdot 10^{-2} \text{ cm}^2/\text{sec}$ ;  $D \approx 10^{-5} \text{ cm}^2/\text{sec}$  and k = 1,  $f = 10^{-7}$ , hence  $f/D \approx 10^{-2} \ll 1$ .

Consequently, even for large longitudinal gradients (k = 1)  $f/D \ll 1$ , in equation (8)

$$\sum_{1}^{n} \frac{f^{n}}{D^{n}} \ll 1.$$

The function of the flow in the longitudinal direction  $\Phi(x, z)$  will be

$$\Phi = \frac{\beta g \rho}{12\nu} (m+nz) \left[ \frac{x^4}{2} - \delta x^3 + \frac{\delta x^2}{2} \right] - \frac{6\sigma}{\delta^3 B} \left[ \frac{\delta^2 x^2}{2} - \frac{x^3}{3} \right].$$
(25)

Substituting (25) into (8) and taking into account that  $\sum_{n=1}^{n} f^n/D^n \ll 1$ , we will obtain

$$H = H_0 \left[ 1 - \frac{(m+nz) \alpha \varkappa_0 \delta}{2T} \right], \qquad (26)$$

where

$$H_{0} = \frac{B\beta g\rho^{2} (m+nz)^{2} \delta^{5} \alpha}{720 \eta T}; \quad \varkappa_{0} = \frac{\upsilon}{H_{0}};$$

$$K = K_{c} + K_{d}; \quad K_{c} = K_{c}^{0} \left[ 1 - \frac{7 (m+nz) \alpha \varkappa_{0} \delta}{2T} + \frac{13 \alpha^{2} \varkappa_{0} (m+nz)^{2} \delta^{2}}{50T^{2}} \right].$$
(27)

Here

$$K_{c}^{0} = rac{eta^{2}g^{2}
ho^{3}B\delta^{9}(m+nz)^{2}}{362880\eta^{2}D}$$
,  $K_{d} = B\delta\rho D$ .

For the case of thermal diffusion of isotopes the terms in the square brackets (26) and (27), after unity are much smaller than unity, and therefore, as in [1],  $H \approx H_0$ ;  $K_d \ll K_c$ ;  $K \approx K_c^0$ .

It is seen from expressions (20) and (21) that H and K differ for our case from the transfer coefficients for a column with a temperature which is constant over the height by the fact that  $(\Delta T)^2$  is replaced by  $(m + nz^2)$ . It is then clear that for a column operating without separation, in the steady state, we will obtain

$$q = \exp\left(\frac{H}{K} L\right),$$

where H/K will be accurately equal to H/K for a column with a constant temperature over the height. However, this means that even marked linear variation of the temperature over the height does not lead to variation Q in an equilibrium state and, consequently, for a column operating without separation it cannot be the cause of deterioration of the separation coefficient. It is necessary to stress most clearly that this conclusion is accurate only when the temperature over the width of the column is completely constant, since in the opposite case the column can no longer be regarded as operating without separation.

For a column operating with separation, the equation of the flow

$$Hc\overline{c} - K \frac{dc}{dz} - \sigma (c_e - c) = 0, \qquad (28)$$

with the condition on the boundary z = 0,  $c = c_0$ .

We will introduce the designations:  $H = A (m + nz)^2$ ,  $K = N (m + nz)^2$  where A and N are constant. The solution of equation (28) will be:

$$c = \exp\left\{\frac{1}{N}\left[AbLz - \frac{\sigma}{n(m+nLz)}\right]\right\}\left\{\frac{L}{N}\int\left[A\delta - \frac{\sigma c_e}{(m+nLz)^2}\right] \times \exp\left(-\frac{1}{N}\left[AbLz - \frac{\sigma}{n(m+nLz)}\right]\right)dz + P\right\},$$
(29)

where  $0 \le z \le 1$ . The integration constant is determined from the condition z = 0;  $c = c_0$ .

The expression under the integral sign cannot be integrated to the end. An expression suitable for calculation can be obtained for the case  $\overline{cc} = const = a$ . We will note, by the way, that for the individual column in the case of thermal diffusion separation of isotopes, this limitation does not appear rigorous.

The expression which establishes the connection between the initial  $c_0$ , the final  $c_e$ , the concentration and the separation  $\sigma$  for the case  $\overline{cc} = const = a$  has the form

$$\frac{y_e a}{nL} \left[ \xi \ln \omega_L + \xi \sum_{n=1}^{\infty} \frac{\xi^n \left( \omega_L - \frac{1}{m^n} \right)}{nn!} + \exp \left( \frac{\xi}{m} \right) m - \frac{\exp \left( \xi \omega_L \right)}{\omega_L} \right] = (c_0 - c_e) \exp \left( \frac{\xi}{m} \right), \qquad (30)$$

where  $\xi = \sigma/Nn$ ;  $\omega_{T} = 1/(m + nLz)$ ;  $0 \le z \le 1$ .

Since equation (30) as well as the equation for the column with a constant temperature over the height is transcendental, then a direct explanation of the influence of the longitudinal temperature gradient on the separation in the column is not possible. In order to illustrate this influence we will compare the operation of the thermal diffusion column with temperatures which are constant and variable over the height. We will carry out the comparison for temperature fields which guarantee equal discharges of energy. The parameters of the column are L = 40 cm; B = 9.4 cm;  $\Delta T = 100^{\circ}C$ .

In the case of a temperature which is variable over the height, we will examine two systems: a)  $T_1 = 50^{\circ}C$ , k = 2.5 deg/cm;  $T_0 = 0^{\circ}C$ ; 2)  $T_1 = 150^{\circ}C$ , k = -2.5 deg/cm;  $T_0 = 0^{\circ}C$ .

For a case of constant temperature  $T_1 = 100^{\circ}C$ ,  $T_0 = 0^{\circ}C$ , k = 0. Bromobenzene [1]  $c^0 = 0.5$ ,  $c_e = 0.6$  is taken as the initial substance.

As a result of calculation we obtain: for a temperature which is constant over the height  $\sigma = 10 \text{ g/}24$  hours, for the case k = 2.5 deg/cm  $\sigma = 6 \text{ g/}24$  hours, for the case k = -2.5 deg/cm  $\sigma = 3.8 \text{ g/}24$  hours.

Consequently the linear variation of the temperature over the height has a negative effect on the operation of the thermal diffusion column in a separating system.

## NOTATION

- T temperature, °C;
- $\rho$  density of the mixture
- $\beta$  coefficient of volume expansion;
- $\nu$  coefficient of kinematic viscosity;
- g gravitiational acceleration;

- u, v velocity components of convective flows;
- $\alpha$  coefficient of thermal diffusitivity;
- D concentration diffusion coefficient;
- L length of the column;
- $\delta$  width of the gap;
- $\sigma$  magnitude of separation.

## LITERATURE CITED

1. K. Aleksander, Uspekhi Fizicheskikh Nauk, 26, No. 4 (1962).