

MASS EXCHANGE IN ISOTOPE SEPARATION BY
THE DISTILLATION METHOD

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We investigate the turbulent flow of a gas in a packed column; taking account of this flow, we obtain a general analytic expression for the height of the equilibrium stage in the separation of isotopes by the distillation method.

Today the separation of isotopes by the distillation method is carried out at low loads, at which the flow of gas and liquid in the columns is strictly laminar. However, the growing demand for stable isotopes of light elements has brought a need for intensifying separation processes by increasing the flow velocities and the dimensions of the packing elements. The hydrodynamic and diffusion processes leading to the separation effect will then take place in the turbulent region of the gas flow in the packing.

The investigation of the separation process in the laminar region has been dealt with in earlier studies [1-2], in which we proposed an approach that can be used for calculating the fundamental quantity determining the efficiency of the packed column, namely, the height of the equilibrium stage (TETS). Difficulties in describing the process in the turbulent region of gas flow were then related to the lack of experimental data concerning the real picture of gas flow in efficient packings at high loads. It was not clear even whether turbulent flow in channels of such small dimensions was theoretically possible.

It is practically impossible to use the gas-flow visualization method for obtaining such data because the dimensions of the packing channels are so small and because there may be blurring of the picture due to convective mixing. Only direct observation of turbulent pulsations and a description of their character could answer the question of whether turbulence existed in the flow of the gas in a column and what the threshold of appearance of such turbulence was as the load was increased.

In this study we observed the turbulent pulsations in the packing by means of a thermoanemometer whose sensor was constructed in the form of a packing element (a segment of a wire spiral of triangular or cylindrical shape) and was placed inside the column among the other similar packing elements. The filament of the thermoanemometer was placed within the element in the plane of its cross section perpendicularly to the gas flow. The coils of the element were glued together in order to provide rigidity and also to keep the wire stretched, preventing it from vibrating when acted upon by the gas flow. The placement of the active segment of the thermoanemometer sensor inside the packing element not only made it possible to make measurements in the separating channel itself but also protected the filament from being damaged when the sensor was oriented in various directions within the packing. The sensor filament was a copper-covered platinum wire with an outer diameter of 0.03 mm and an etched working segment 0.7 mm long. Its thermal inertia made it possible to measure pulsations with frequencies up to 5 kHz.

The measurements were carried out on a packing with element dimensions of 6.5×5.5 mm, with air blown through the column. It was found that in spiral wire packings, turbulent flow of the gas appears in the region $Re \approx 1000-1200$, depending on the configuration at the junctions of the elements. Below this limit, no turbulent pulsations were observed in the packing. After the pulsations appeared, their amplitude increased rapidly, and as the load was further increased, the pulsation amplitude remained practically unchanged. The characteristic shape of the pulsations, as seen on an oscillograph, is shown in Fig. 1. The bulk of the pulsations lie in the frequency range of 500-1000 Hz. No large low-frequency pulsations were observed in practice. Simultaneously with the appearance of turbulent pulsations, we

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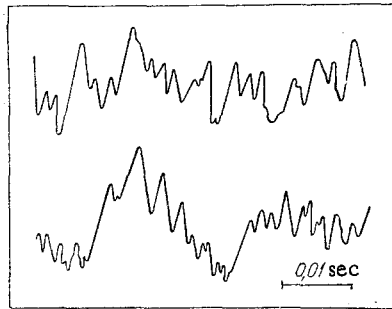


Fig. 1. Oscillogram of turbulent pulsations in packing made of segments of triangular wire (above, at $Re = 1500$; below, at $Re = 3000$).

observed a break in the curve of the hydrodynamic resistance of the packing as a function of the Reynolds number.

The picture we observed enabled us to draw the following conclusions concerning the nature of turbulent gas flow in the packing. The turbulence appears in the packing at the junctions between the elements and, since the channel segment is so short, cannot be extinguished within the elements, despite the nearness of the walls and the strong frictional effect. There is no fully developed turbulent flow in the packings, and the nature of the flow is similar to that of flow in a turbulent boundary layer, where the scale of the pulsations is determined by the distance to the channel wall.

It was established earlier that only the initial portion of the hydrodynamic gas-velocity profile is realized in the packing elements, and until turbulent flow appears at $Re \approx 1,000$, the central region of the flat velocity profile occupies most of the cross section of the channel, except for a narrow annular region, in which frictional forces have a marked effect [3]. Therefore it is natural to identify this central region with the region of turbulent flow, since the turbulence in this case is an incoming turbulence, not one that arises in the channel itself. The frictional forces which lead to the formation of the parabolic velocity profile near the wall will at the same time extinguish the turbulent motion near the wall. Since there is no fully developed turbulent motion, we can also assume that the turbulent pulsations do not arise in the laminar boundary layer and are attenuated on its boundary. Under these conditions, the turbulent flow is determined by the local values of ρ and τ_0 , and its scale is of the order of the distance measured from the boundary of the laminar layer. On the basis of the localization hypothesis and dimensionality considerations [4, 5], the coefficient of turbulent diffusion in the interior channels of the packing can be represented in the form

$$D_t = \beta v_* (r_0 - r), \quad (1)$$

where $v_* = \sqrt{\tau_0 / \rho}$, β is a proportionality constant, and the total diffusion coefficient is

$$D(r) = D_g + \beta v_* (r_0 - r). \quad (2)$$

By introducing the notation

$$D_g + \beta v_* r_0 = a, \quad -\beta v_* = b, \quad (3)$$

we can reduce the expression for the total diffusion coefficient to the form

$$D(r) = a + br. \quad (4)$$

The expressions (2) and (4) are determined in the region $0 \leq r \leq r_0$ when $Re > 1000$. When $Re < 1000$, β is considered to be zero over the entire length of the separating channel.

In [1] we showed that the efficiency of the column can be calculated by using a cylindrical bottle with radius R , equal to the average length of the diffusion path in the gaseous phase occurring in the packing. Such a model includes liquid and gas flows averaged over the entire column, and the gas-flow profiles, just as in the packing elements, have a shape characteristic of the initial segment of the channels, i.e., they are subdivided into an annular region, in which the distribution is parabolic, and a central region, in which the velocity is constant irrespective of the nature of the hydrodynamic conditions. Therefore, in the central region of the gas channel of the model, where there is turbulent gas flow, we used a flat velocity profile rather than a logarithmic profile. The radius of the central region is satisfactorily approximated by an empirical function of the Reynolds number:

$$r_0 = R \frac{\ln(\text{Reg} - 200)}{\ln(\text{Reg} + 6 \cdot 10^6 / \text{Reg})} \quad (5)$$

In the same study, we showed that the diffusion equations for the gaseous and liquid phases in the general case, having the form

$$(\mathbf{V} \cdot \nabla) y = (\nabla D \nabla) y, \quad (\mathbf{L} \cdot \nabla) x = D_{\text{liq}} \Delta x, \quad (6)$$

can be reduced to ordinary differential equations by introducing a parameter h , whose physical significance is the height of the equilibrium stage in the channel of the model. For our case, which takes account of turbulent gas flow in the central region of the separating channel, the diffusion equations can be written as follows:

$$\frac{d}{dr} \left(r \frac{dy}{dr} \right) - (c_1 r^3 + c_2 r) y = 0, \quad r_0 \leq r \leq R, \quad (7)$$

$$\frac{d}{dr} \left[r D(r) \frac{dy}{dr} \right] - c_3 r y = 0, \quad 0 \leq r \leq r_0, \quad (8)$$

$$\frac{d^2 x}{dn^2} - (c_4 n^2 + c_5 n) x = 0, \quad 0 \leq n \leq m, \quad (9)$$

where

$$\begin{aligned} c_1 &= -\frac{\varepsilon}{h D_g} \frac{2q_h}{\pi(R^2 - r_0^2)^2}; \quad c_2 = -c_1 R^2; \\ c_3 &= \frac{\varepsilon V_{\text{liq}}}{h}; \quad c_4 = \frac{\varepsilon}{h D_{\text{liq}}} \left(\frac{3\Gamma}{2m^3} + \frac{3\tau_0}{4\mu_{\text{liq}} m} \right); \\ c_5 &= -\frac{\varepsilon}{h D_{\text{liq}}} \left(\frac{3\Gamma}{m^2} + \frac{\tau_0}{2\mu_{\text{liq}}} \right). \end{aligned} \quad (10)$$

The velocity profiles $V(r)$ and $L(n)$ used in expressions (6)-(10) were obtained by solving the boundary-layer equations for the flow of thin films of liquid and the Poiseuille equation for an annular laminar region of gas flow.

Substituting into (7) the expression for $D(r)$ from (4) and solving these equations by the method of successive approximations with the boundary conditions

$$\text{for } r = 0 \quad y = y_c, \quad \frac{dy}{dr} = 0, \quad (11)$$

$$\text{for } r = r_0 \quad y_{\text{T}} = y_{\text{1am}}, \quad \frac{dy_{\text{T}}}{dr} = \frac{dy_{\text{1am}}}{dr},$$

$$\text{for } n = 0 \quad x = x_c, \quad \frac{dx}{dn} = 0, \quad (12)$$

we find

$$y_{\text{1am}} = y_c \left[1 + \frac{c_3 r_0}{2b} + \frac{c_3 a}{2b^2} \ln \frac{a}{D_g} + \frac{c_3 r_0^2}{2D_g} \ln \frac{r}{r_0} + \frac{c_1}{16} (r^4 - r_0^4) + \frac{c_2}{4} (r^2 - r_0^2) - \left(\frac{c_1 r_0^4}{4} + \frac{c_2 r_0^2}{2} \right) \ln \frac{r}{r_0} \right], \quad (13)$$

$$y_{\text{T}} = y_c \left[1 + \frac{c_3}{2b^2} \left(br + a \ln \frac{a}{a + br} \right) \right], \quad (14)$$

$$x = x_c \left(1 + \frac{c_4}{12} n^4 + \frac{c_5}{6} n^3 \right). \quad (15)$$

From Eqs. (13) and (15), replacing r by R and n by m , we can obtain the values of the concentrations at the interface between the phases — y_{R} and x_{m} . Now we determine the average values of the concentrations in each of the regions:

$$\bar{y}_{\text{1am}} = y_c \left\{ 1 + \frac{c_3 r_0}{2b} + \frac{c_3 a}{2b^2} \ln \frac{a}{D_g} + \frac{2}{R^2 - r_0^2} \left[\frac{c_1}{96} (R^6 - r_0^6) \right. \right.$$

$$+ \frac{c_2}{16} (R^4 - r_0^4) + \frac{c_1}{32} r_0^4 (R^2 - r_0^2) - \frac{c_3}{8D_g} r_0^2 (R^2 - r_0^2) - \left(\frac{c_1}{8} r_0^4 R^2 + \frac{c_2}{4} r_0^2 R^2 - \frac{c_3}{4D_g} r_0^2 R^2 \right) \ln \frac{R}{r_0} \Bigg], \quad (16)$$

$$\bar{y}_r = y_c \left[1 - \frac{5c_3 r_0}{12b} + \frac{5c_3 D_g}{4b^2} - \frac{c_3 D_g^2}{2b^3 r_0} + \frac{c_3 D_g a}{2b^4 r_0^2} (2br_0 - D_g) \ln \frac{a}{D_g} \right], \quad (17)$$

$$\bar{x} = x_c \left(1 + \frac{c_4}{60} m^4 + \frac{c_5}{24} m^3 \right), \quad (18)$$

and also the average concentration in the gaseous phase

$$\bar{y} = \left(1 - \frac{r_0^2}{R^2} \right) \bar{y}_{lam} + \frac{r_0^2}{R^2} \bar{y}_t, \quad (19)$$

$$\bar{y} = y_c \left\{ 1 + \frac{2}{R^2} \left[\frac{c_1}{96} (R^6 - r_0^6) + \frac{c_2}{16} (R^4 - r_0^4) + \frac{c_1}{32} r_0^4 (R^2 - r_0^2) - \frac{c_3 r_0^2}{8D_g} (R^2 - r_0^2) - \left(\frac{c_1}{8} r_0^4 R^2 + \frac{c_2}{4} r_0^2 R^2 - \frac{c_3}{4D_g} r_0^2 R^2 \right) \ln \frac{R}{r_0} \right] - \frac{c_3 r_0}{12b^3 R^2} (11b^2 r_0^2 - 15D_g b r_0 + 6D_g^2 - 6b^2 R^2) - \frac{c_3 a}{2b^4 R^2} (a^2 - b^2 R^2) \ln \frac{a}{D_g} \right\}. \quad (20)$$

In order to eliminate the unknown concentrations y_c and x_c , we use the condition of balance in the cross section of the channel, which in a nonsampling regime leads to equality of the average concentrations in the liquid and the gas:

$$\bar{x} = \bar{y}, \quad (21)$$

and also use the relation which holds on the interface between the phases:

$$\frac{y_R}{1 - y_R} = \alpha \frac{x_m}{1 - x_m}. \quad (22)$$

For the case of small values of ϵ and small concentrations, the expression (22) can be expanded in series and reduced to the form

$$y_R - x_m = \epsilon y_c. \quad (23)$$

It should be noted that the value of product sampling from the column, including the case of a nonsampling regime, and also the absolute values of the concentrations, by their very nature, do not affect the height of the equilibrium stage, while they substantially simplify the calculation of h .

Substituting into (21) the expressions for \bar{x} from (18) and for \bar{y} from (20), and substituting into (23) the expression y_R and x_m defined by (13) and (15), and eliminating y_c and x_c from these, we obtain an expression for the height of the equilibrium stage in the separating channel of the model of a packed column:

$$h = \frac{1}{6} \frac{q_{ann}}{\pi D_g} \left(1 - \frac{r_0^2}{R^2} \right) + \frac{1}{4} \frac{q_{cent}}{\pi D_g} \left(1 - \frac{r_0^2}{R^2} \right) + \frac{1}{12} \frac{q_{cent}}{\pi \beta^4 v_*^4 R^2 r_0^2} \left[6(D_g + \beta v_* r_0)^3 \ln \frac{D_g + \beta v_* r_0}{D_g} - 11\beta^3 v_*^3 r_0^3 - 15D_g \beta^2 v_*^2 r_0^2 - 6D_g^2 \beta v_* r_0 \right] + \frac{11}{40} \frac{\Gamma m}{D_{liq}}. \quad (24)$$

In the limiting case $\beta \rightarrow 0$, which occurs when $Re < 1000$, formula (24) takes the form

$$h = \frac{1}{6} \frac{q_{ann}}{\pi D_g} \left(1 - \frac{r_0^2}{R^2} \right) + \frac{1}{4} \frac{q_{cent}}{\pi D_g} \left(1 - \frac{r_0^2}{2R^2} \right) + \frac{11}{40} \frac{\Gamma m}{D_{liq}}, \quad (25)$$

which was obtained earlier for the laminar regime of gas flow. In the case of a turbulent regime, for practical calculations we may with satisfactory accuracy disregard the annular laminar sublayer. By passing to the limit as $r_0 \rightarrow R$, we obtain from (24) the relation

$$h = \frac{1}{12} \frac{q}{\pi \beta^4 v_*^4 R^4} \left[6(D_g + \beta v_* R)^3 \ln \frac{D_g + \beta v_* R}{D_g} \right]$$

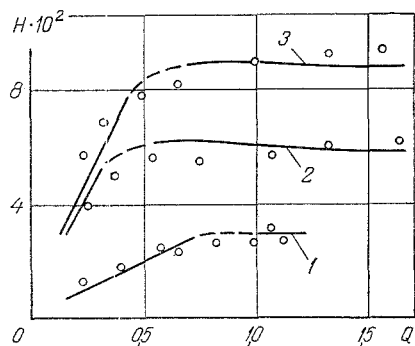


Fig. 2. Experimental and calculated graphs of the height of the equilibrium stage as a function of the load: 1) for packing made of segments of triangular spiral, 3.5 × 3.0 mm; 2) for the same packing, 6.5 × 5.5 mm; 3) for packing made of segments of cylindrical spiral, 6.0 × 5.0 mm. H, m; Q, kg/m²·sec.

$$- 11\beta^3 v_*^3 R^3 - 15D_g \beta^2 v_*^2 R^2 - 6D_g^2 \beta v_* R \left] + \frac{11}{40} \frac{\Gamma m}{D_{\text{liq}}} \right. \quad (26)$$

The height of the equilibrium stage (HETS) for a packed column is calculated, according to [2], by the formula

$$H = \frac{i\epsilon l}{\ln[(1 - \lambda) \exp(\epsilon l/h) + \lambda]} \quad (27)$$

where h is given by the expression (24).

An experimental check of the expressions obtained was carried out in the distillation of a calibrating mixture of C₆H₆ and CCl₄ in a column with a diameter of 70 mm and a height of 1 m in the packed section, on packings made of segments of triangular and cylindrical wire spirals.

The calculated values of h for Re < 1000 were found from formula (25), and those for Re > 1200 were found directly from formula (24). In the transition region 1000 < Re < 1200 the value of h was not calculated, but the curves corresponding to laminar and turbulent regions were joined by a line on the graph. The value of β appearing in the coefficient of turbulent diffusion was determined in preliminary experiments from the known value of H and found to be equal to 0.25.

With this value of β, formula (24) satisfactorily describes all the experimental results irrespective of the shape of the packing, the dimension of the element, and the magnitude of the flow in the column (Fig. 2). It also enables us to clarify why the height of the equilibrium stage is practically independent of the load in the turbulent region. The behavior of H in the turbulent region is determined by the simultaneous action of four factors which depend on the load: the convective flows of gas and liquid leading to an increase in H and the compensating influence of the coefficient of turbulent diffusion and the relative amount of stagnation zones. The contribution of each of these factors can be determined from formulas (24) and (27). As a result, the value of H is only slightly dependent on the load, although this dependence itself, as can be seen from Fig. 2, is complicated in nature.

It should be pointed out that the height of the equilibrium stage in the turbulent region is determined in practice by the value it has when the turbulent gas flow first appears. This indicates that it would be desirable to design new and more efficient packings with a lower threshold of turbulence. The weak dependence of the height of the equilibrium stage on the load in the turbulent region of gas flow also leads to the important practical conclusion that the productivity of columns can be substantially increased without increasing their volume and length.

NOTATION

D_g, D_{liq}, coefficients of molecular diffusion; D_t, coefficient of turbulent diffusion in the gas; R, radius of the gaseous region of the model channel; r₀, radius of the central region of gas flow; m, thickness of the liquid film; n, r, transverse coordinates in the liquid and gaseous regions; L, V, linear velocities of liquid and gas; q, volumetric gas flow rate; Γ, volumetric flow rate of liquid per unit perimeter; x, y, molar concentrations of the desired isotope in the liquid and the gas; H, height of the equilibrium stage (HETS); ε, coefficient of enrichment; τ₀, frictional stress on the interface between the phases; μ, viscosity; λ, fraction of stagnation zones in the packing. Subscripts: t, turbulent; lam, laminar; liq, liquid; g, gas; ann, annular region; cent, central region.

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CHARACTERISTICS OF CENTRIFUGAL SEPARATOR

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An approximate method is proposed for the calculation of the integral separational characteristics of a centrifugal separator.

The process of separating two-phase (two-component) mixtures forms the basis of many production technologies [1]. One of the possible models for calculating the separational characteristics of a centrifugal separator (CS) of the standard scheme shown in Fig. 1 is considered in the present work. It has been established experimentally [2] that, in the central part of the separation zone (SZ), there arises an axial column of carrier gas that is practically free from inclusions of the second phases; "particles" of gas in the region of the boundaries of this column move around the axis of the system at some angular velocity. The main simplification of the model proposed is that the axial column of carrier gas is represented as a rigid impermeable cylinder with a radius R_2 that is constant over the SZ height. Flow of the mixture under the action of the applied pressure difference is then considered in a channel bounded by two coaxial cylinders of radii R_1 and R_2 ; the inner cylinder, modeling the axial column of carrier gas, moves in the axial direction at a velocity U_0 and rotates about its axis at constant angular velocity ω . It is assumed that the phase which is to be separated is present, and moves, in the form of undeformable, indivisible spherical particles. In addition, only the region of "dilute" mixtures, with $nR_1 a^2 \ll 1$, is considered; in the first approximation, according to [3], this allows: a) the influence of collisions between particles on the overall character of the disperse-phase motion to be neglected; b) the assumption to be made that the presence of particles has no influence on the velocity-field distribution of the carrier gas. Note here that satisfying the condition of a "dilute" mixture imposes sufficiently rigorous constraints on the magnitude of the mass content of particles in the flow at the SZ inlet, b_0 . Thus, for example, estimation of the limiting value of b_0 for a mixture of air (carrier gas) and water (disperse phase) gives (taking into account that $\kappa b_0 \sim n a^3$)

$$b_0 \ll \left(\frac{\bar{a}}{R_1} \frac{\rho'}{\rho} \right) \sim \frac{10\sigma}{\rho \omega_*^2} \frac{1}{\kappa R_1} \sim 20\%. \quad (1)$$

Within the framework of the given model, the velocity distribution of the carrier-gas flow w is determined from the solution of the steady equations of motion of a single-phase incompressible medium [4] between two coaxial cylinders (the effect of spatial variation of the flow density may be neglected in view of the smallness of the carrier-gas characteristic velocity in the SZ $w_* = 10-30$ m/sec in comparison with the velocity of sound). In this system, the cylinders are regarded as infinitely extended in the axial direction, so that edge effects

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