CALCULATION OF MULTISTAGE THERMAL DIFFUSION IN A CENTRIFUGAL FORCE FIELD

B. P. Konstantinov and L. S. Kotousov

Inzhenerno-Fizicheskii Zhurnal, Vol. 11, No. 4, pp. 455–462, 1966

UDC 621.039.341.6

An examination is made of the theory and design of a thermocentrifugal column with a small gap, which may be used for relatively rapid, high-rate isotope concentration in the presence of a finite amount of initial gas mixture.

The Clausius column, used to separate gaseous isotope mixtures, has a relatively large optimum gap between the hot and cold faces, for a gas pressure of $\sim 10.1 \times 10^4$ N/m^2. The HETP value of such a column is several centimeters, and, if $\alpha_T = 0.01$–0.02, we require a total length of the column of the order of several tens of meters, to obtain high separation coefficients (10^2–10^3), while the time to reach a steady state is several tens of days.

In practice one meets the problem of concentrating an isotope from specimens containing at most several grams of material. For this purpose we may use the method of thermal diffusion of gases at pressures of the order of $10.1 \cdot 10^4$ N/m^2, by reducing the gap to a fraction of a millimeter, and keeping the circulation flux optimal by rotational forces. This method gives a sharp reduction in settling time, since the charge of material in the column decreases in proportion to the square of the gap, while the diffusion coefficient is kept high. Similar equipment was described in [2], but no calculation of the thermocentrifugal column was given.

In this paper we analyze the operation of the thermocentrifugal column and determine some optimal parameters for it, including the time to reach equilibrium. The calculation presented may be applied, with certain changes, to the equipment described in [3], in which one of the discs is fixed. The radial (circulation) flow of the column therefore increases due both to the large relative velocity of rotation of the discs and to the thermosiphon action.

The circulation flux. We shall examine the motion of a gas or of a liquid inside a hollow rotating disc whose walls are at different temperatures (Fig. 1).

The Navier-Stokes equation for steady motion is

$$\rho(\mathbf{v}, \nabla)\mathbf{v} = -\nabla p + \eta\Delta\mathbf{v} + (\xi + \eta/3)\ \mathrm{grad\ div}\ \mathbf{v}. \quad (1)$$

The components of velocity and its derivatives in the directions r, φ and z make up the table of values

$$v_r, \quad v_\varphi \simeq \omega r, \quad v_z \simeq 0;$$

$$\frac{\partial v_r}{\partial r}, \quad \frac{\partial v_r}{\partial \varphi} = 0, \quad \frac{\partial v_r}{\partial z};$$

$$\frac{\partial v_\varphi}{\partial r} \simeq \omega, \quad \frac{\partial v_\varphi}{\partial \varphi} = 0, \quad \frac{\partial v_\varphi}{\partial z} \simeq 0. \quad (2)$$

The remaining quantities are zero. Using (2), we rewrite Eq. (1) in components in the directions r (3) and φ (4):

$$\rho v_r \frac{\partial v_r}{\partial r} = -\frac{\partial p}{\partial r} + \rho\omega^2 r + \eta\left(\frac{\partial^2 v_r}{\partial r^2} + \frac{1}{r}\frac{\partial v_r}{\partial r} + \frac{\partial^2 v_r}{\partial z^2} - \frac{v_r}{r^2}\right) + \left(\xi + \frac{\eta}{3}\right)\left(\frac{\partial^2 v_r}{\partial r^2} + \frac{1}{r}\frac{\partial v_r}{\partial r} - \frac{v_r}{r^2}\right); \quad (3)$$

$$2\rho v_r \omega = -\frac{\partial p}{r\,\partial\varphi} + \eta\frac{\partial^2 \varphi}{\partial z^2}. \quad (4)$$

For simplicity the transfer coefficients are assumed to be independent of temperature. In the first approximation, we allow for three terms of Eq. (3)

$$-\frac{\partial p}{\partial r} + \rho\omega^2 r + \eta\frac{\partial^2 v_r}{\partial z^2} = 0. \quad (3')$$

Putting $\rho \simeq \bar{\rho}(1 + z\Delta T/2a\bar{T})$, $\partial p/\partial r \approx \rho\omega^2 r$, we obtain

$$\frac{\partial^2 v_z}{\partial z^2} = -\frac{\bar{\rho}}{\eta}\omega^2 r\frac{\Delta T}{2a\bar{T}}z. \quad (3'')$$

Solving Eq. (3") under the conditions $v_r(\pm a) = 0$, we have

$$v_r = \frac{\bar{\rho}}{\eta}\frac{z(a^2 - z^2)}{12a}\frac{\Delta T}{\bar{T}}\omega^2 r. \quad (5)$$

Substitution of Eq. (5) into Eq. (3) gives, with $z = a/\sqrt{3}$, corresponding to $|v_{r,\max}|$

$$\bar{\rho}(k^2\omega^2)\omega^2 r = -\bar{\rho}\frac{\Delta T}{2\sqrt{3}\bar{T}}\omega^2 r + \eta\frac{\partial^2 v_r}{\partial z^2}, \quad (3''')$$

where

$$k \simeq \frac{\bar{\rho}}{\eta}\frac{a^2}{31}\frac{\Delta T}{\bar{T}}.$$

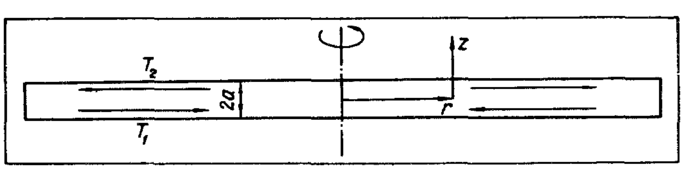

Fig. 1. Diagram of the thermocentrifugal column. The arrows along the lateral walls indicate the radial counterflow.

The column calculation made below shows that for $\Delta T/\bar{T} \sim 1, a \sim 10^{-1}-10^{-2}$; the optimal value of $k^2\omega^2$ is less than the expression $(1/2\sqrt{3})(\Delta T/\bar{T})$ by 2–3 orders.

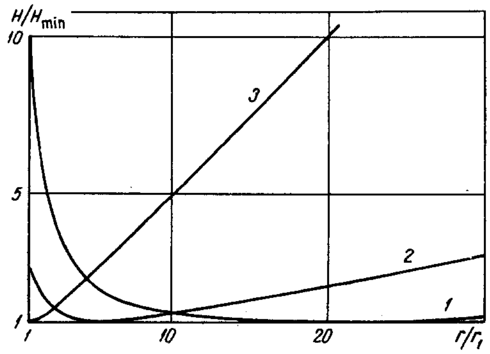

Fig. 2. Dependence of HETP on radius for a column with constant gap $\left(\frac{H}{H_{\min}} = \frac{1}{2}\left(\theta \frac{r}{r_1} + \frac{1}{\theta}\frac{r_1}{r}\right), \theta = \frac{r_1 \varkappa \sqrt{D}}{2\sqrt{2D_e}}\right)$: 1, 2, 3 are for θ = 0.05, 0.2, 1, respectively.

Therefore, Eq. (5) gives an almost exact value of the linear radial flow velocity. Further, Eq. (4) determines the conditions of excitation of the secondary azimuthal flows under the influence of the Coriolis acceleration. Solving Eq. (4) and using Eq. (5) and $(\partial p/\partial\varphi) = 0$, $\rho \simeq \bar{\rho}(1 + z\Delta T/2a\bar{T})$, $n \simeq$ const (z), we have

$$v_\varphi = \omega r + br\left[-\frac{7}{60}a^4 z + \frac{a^2 z^3}{6} - \frac{z^5}{20} + \frac{a z^4}{24}\frac{\Delta T}{\bar{T}} - \frac{z^6}{30}\frac{\Delta T}{2a\bar{T}} - \frac{a^5}{40}\frac{\Delta T}{\bar{T}}\right]. \quad (6)$$

Here

$$b = \left(\frac{\bar{\rho}}{\eta}\right)^2 \frac{\Delta T}{\bar{T}} \frac{\omega^3}{6a}.$$

To estimate the value of the additional velocity of rotation of the gas $\Delta v_\varphi = v_\varphi - \omega r$, we put $z = \pm a/2$, r = 30 cm. Then $\Delta v_\varphi \cong (3/80)\, a^5 br$, the linear velocity of rotation decreasing by Δv_φ in the cold part, and increasing in the hot part. At a pressure of $10.1 \cdot 10^4$ N/m^2 and a gap of 0.5–1 mm, for methane under the conditions chosen above, the optimal value of v_r is 15–30 cm/sec, while the quantity Δv_φ is an order lower. The angular velocity of the secondary azimuthal flow is then only ~0.3% of ω_{opt}. Therefore, the sign of approximate equality may be omitted in (2). The additional velocity Δv_φ has a favorable influence on the operation of the column, smoothing out the azimuthal nonuniformities of concentration due to nonuniformities in the gap. The linear velocity averaged over z is

$$\bar{v}_r = \frac{\bar{\rho}}{\eta}\frac{a^2}{48}\frac{\Delta T}{\bar{T}}\omega^2 r = Br. \quad (7)$$

The circulation flux in the no-yield regime is

$$J = 2\pi r a n \bar{v}_r = 2n B \pi r^2 a. \quad (8)$$

In (8), J is proportional to r^2. As will be seen later, this relation is not the best from the viewpoint of maximum separation. Any change of J(r) is possible at the expense of $\Delta T/\bar{T} = f_1(r)$ and also $a = f_2(r)$. It may be seen from the foregoing that for the variables a and $\Delta T/\bar{T}$, the solutions (7) and (8) remain in the same form. We note only one special feature of the motion of the gas in the column. Above we put $v_z = 0$. We will show that this condition is well satisfied in practice for columns with a small gap. In principle, $v_z \neq 0$, since the transfer of fluid from the hot stream moving towards the center to the cold stream affords the possibility of increase of the radial velocity with increase of radius [2], because in practice the changes of gas density are small (see below). We will estimate the quantity $\bar{v}_z$ by the following simple method. For a constant mean density, we obtain $a(\partial v_r/\partial r) \cong \bar{v}_z$ from the condition of continuity div v = 0, and therefore $\bar{v}_z = 2aB$, and, correspondingly,

$$\bar{v}_z/\bar{v}_r = 2a/r. \quad (9)$$

Equation of the thermocentrifugal column. The transfer processes for one isotope with relative concentration c are described by the equations

$$J(c_{I} - c_{II}) = 4\pi r n D_e \frac{\partial c}{\partial r} = i_k (c_k - c), \quad (10)$$

$$nBr\frac{\partial c}{\partial r} = \frac{D}{a}\left[\frac{\alpha_T \Delta T}{2a\bar{T}}c(1-c) - \frac{c_I - c_{II}}{a}\right]. \quad (11)$$

Relation (11) for internal processes in the column is derived from the condition div j = 0, where j = nv, $v = v_r + v_\varphi + v_z$, derivatives with respect to z being approximated by a finite-difference relation.

Eliminating $(c_I - c_{II})$ from Eqs. (10) and (11), and using Eq. (8), we obtain

$$\frac{\partial c}{\partial r}\left(\frac{aBr}{D} + \frac{2D_e}{aBr}\right) = \gamma c(1-c) - \frac{i_k(c_k - c)}{2a^2 nB\pi r^2}. \quad (12)$$

The solution of Eq. (12) for the case $i_k = 0$ has the form

$$\ln q = \ln\frac{\beta(r_2)}{\beta(r_1)} = \frac{\gamma}{\varkappa}\ln\frac{\varkappa^2 D r_2^2 + 8D_e}{\varkappa^2 D r_1^2 + 8D_e}. \quad (13)$$

Here $\lambda = \alpha_T\Delta T/2a\bar{T}$; $\varkappa = 2aB/D$; $\beta = c/(1-c)$; B–see (7).

The solution of Eq. (13) is obtained under the condition B = const(r), i.e., $\Delta T/\bar{T}$ and a are chosen to be independent of r. We will examine the optimal parameters of the thermocentrifugal column from the viewpoint of maximum separation in a given section of the radius. We assume that the flux J as the form (8) and that solution (13) is applicable. We write Eq. (13) in the form

$$\Delta r = r_2 - r_1 = \sqrt{\varkappa^2 D r_1^2 q^{\varkappa/\gamma} + 8D_e(q^{\varkappa/\gamma} - 1)} - r_1. \quad (14)$$

From the equations $(\Delta r)'_{r_1} = 0$, $(\Delta r)''_{r_1 r_1} > 0$ we find that $\Delta r = \Delta r_{\min}$ if

$$r_{1\,\text{extr}}^2 = \frac{8}{\varkappa^2}\frac{D_e}{D}q^{-\varkappa/\gamma}; \quad (15)$$

we substitute Eq. (15) into Eq. (13) to obtain

$$r_2^2 = \frac{8}{\varkappa^2}\frac{D_e}{D}q^{\varkappa/\gamma}.$$

Hence

$$\Delta r = \frac{2\sqrt{2}}{\varkappa}\sqrt{\frac{D_e}{D}}\,(q^{\varkappa/2\gamma} - q^{-\varkappa/2\gamma}). \qquad (16)$$

From the equation $(\Delta r)'_{\varkappa} = 0$ we find that $\Delta r \to \Delta r_{min}$, if $\varkappa \to 0$:

$$\Delta r_{min} = \frac{2\sqrt{2}}{\gamma}\sqrt{\frac{D_e}{D}}\ln q, \qquad (17)$$

but it is seen, from Eq. (15), that then $r_1 \to \infty$. Thus, absolutely minimum values of Δr for given γ and q are practically unattainable in a column with constant gap. We note, that Δr_{min} corresponds to minimum HETP. It is seen from Eq. (12) that here the HETP is the variable quantity

$$H = \frac{a^2 B}{D} r + \frac{2D_e}{B}\frac{1}{r} =$$

$$= H_{min}\frac{1}{2}\left(\frac{\varkappa r}{2\sqrt{2}}\sqrt{\frac{D}{D_e}} + \frac{2\sqrt{2}}{\varkappa r}\sqrt{\frac{D_e}{D}}\right). \qquad (18)$$

We see that H is a minimum if conditions (15) and (17) are satisfied. Then

$$H_{min} = \frac{\varepsilon e}{\gamma} 2\sqrt{2}\sqrt{\frac{D_e}{D}} = 2\sqrt{2}\,a\sqrt{\frac{D_e}{D}}.$$

Finally, we will point out the main deficiency of the column with constant gap. It follows from Eq. (16) that Δr varies as a power of q. This dependence is not favorable, since, if we choose the r_1 small enough, i.e., $\varkappa$ large enough, then the condition $H \simeq H_{min}$ is satisfied only on the small section Δr. The separation diminishes appreciably with change of column radius (Fig. 2).

Optimization of circulation flux to obtain maximum separation on an arbitrary section of the radius. We will find the circulation flux function J(r) which gives the maximum separation on a given section of the radius. We put γ = const; this satisfies the requirement that the relative temperature gradient be constant as a function of radius as a and $\Delta T/\overline{T}$ vary. To the calculations we consider that $i_k = 0$.

From Eqs. (10) and (11) under these conditions we have

$$\frac{\partial c}{\partial r} = \frac{J\nu r}{J^2 + \mu r^2} c(1-c), \qquad (19)$$

where $\nu = 2\pi n D\gamma$, $\mu = 8\pi^2 n^2 D D_e$. We integrate Eq. (19) on the given section Δr of the radius

$$\ln q = \int_{r_1}^{r_2} \frac{J\nu r}{J^2 + \mu r^2} dr. \qquad (20)$$

From the Euler equation, we find for the integrand in Eq. (20)

$$J_{opt} = \sqrt{\mu}\, r. \qquad (21)$$

With condition (21) it follows from Eq. (19) that

$$\Delta r = \delta \ln q, \qquad (22)$$

where

$$\delta = \frac{2\sqrt{2}}{\gamma}\sqrt{\frac{D_e}{D}}.$$

The value of Δr in Eq. (22) coincides with Eq. (17), but now the condition $H = H_{min}$ is satisfied for all values of r. Of course, in practice r always satisfies condition (9) as well.

From expression (8), with γ = const, we have

$$a = a_1\sqrt[4]{r_1/r}, \qquad (23)$$

i.e., to obtain a linear dependence of J(r) a weak (e.g., step-by-step) decrease of the gap with increase of radius is sufficient.

We will examine the question of relative angular velocity. For the optimal circulation flux, it follows from Eqs. (8), (21), and (23) that

$$\omega_{opt}^2 = \frac{48\alpha_T D\eta}{\overline{\rho}\gamma a_1^4 r_1} = \frac{96 D\eta}{\overline{\rho}\, a_1^3 r_1}\left(\frac{\overline{T}}{\Delta T}\right)_1. \qquad (24)$$

Finally, using the value ω_{opt}, we will calculate the pressure drop on the section Δr:

$$\Delta p = \int_{r_1}^{r_2} \overline{\rho}\omega_{opt}^2 r\,dr,$$

ω_{opt} being $\leqslant 10^2$ rad/sec for the majority of gases, when $\overline{p} \approx (10.1\text{–}100.1)\cdot 10^4$ N/m², $2a$ = 0.05–0.1 cm, $\Delta T/\overline{T} \sim 1$, r_1 = 10 cm, r_2 = 50 cm. Therefore, $\Delta p/p \leqslant 10^{-2}$, i.e., we may neglect the variations in density along the radius.

Unsteady operation of column, and settling time. The variation of the concentration of one of the components in the space bounded by radii r and r_2 is equal, in the no-yield regime, to

$$[J(c_I - c_{II})]_r - 4\pi\, a n D_e r\frac{\partial c}{\partial r} = \frac{\partial}{\partial t}\int_r^{r_2} 4\pi\, n a c\rho\, d\rho; \qquad (25)$$

the continuity equation for the light component is

$$\operatorname{div} \mathbf{j}_c = -n\frac{\partial c}{\partial t}. \qquad (26)$$

We write the flux j_c in the general form:

$$\mathbf{j}_c = c\mathbf{j} + \mathbf{j}_D;\quad \mathbf{j}_D = \mathbf{j}_{D,r} + \mathbf{j}_{D,z};\quad \mathbf{j} = n\mathbf{v}. \qquad (27)$$

From Eqs. (26) and (27), with the conditions div j = 0 and $v_z \ll v_r$ (see Eq. (9)) and $j_{d,r} = nD_e(\partial c/\partial r) \ll cj_r$ (the latter follows from the fact that $nD_e(\partial c/\partial r) < j_r \cdot (c_I - c_{II})$ and $(c_I - c_{II}) \ll c_{I,II}$), we obtain

$$j_r\frac{\partial c}{\partial r} - \frac{nD}{a}\left[\gamma c(1-c) - \frac{c_I - c_{II}}{a}\right] = -n\frac{\partial c}{\partial t}. \qquad (28)$$

Later on we will put $c \ll 1$ in Eqs. (25) and (28); J and a are determined from Eqs. (21) and (23):

$$\sqrt{\frac{DD_e}{2}}(c_I - c_{II}) - aD_e\frac{\partial c}{\partial r} = \frac{\partial}{\partial t}\int_r^{r_2}\frac{ac\rho}{r}d\rho; \qquad (25')$$

$$\sqrt{2DD_e}\frac{\partial c}{\partial r} - D\left(\gamma c - \frac{c_I - c_{II}}{a}\right) = -a\frac{\partial c}{\partial t}. \qquad (28')$$

Eliminating $c_I - c_{II}$, we find

$$\frac{\partial^2 c}{\partial r^2} - \delta \frac{\partial c}{\partial r} - \frac{1}{4r}\left(\frac{\partial c}{\partial r} - \delta c\right) = \frac{1}{2D_e}\frac{\partial c}{\partial t}. \quad (29)$$

Here we have omitted terms of order a^2 in Eq. (29), this being equivalent to the quasi-steady condition div $j_c = 0$ ordinarily assumed. We note that in the steady regime this equation has the solution (22), owing to the linear independence of the two terms on the left of Eq. (29).

Under the condition $\delta \gg 1/4r$ we may neglect the second term on the left of Eq. (29). The solution of the equation in that event is given, for example, in [1].

As an example we will examine the concentration of radio-carbon, starting from the isotope mixture $C^{12,14}H_4$. We put $r_1 = 5$ cm, $\Delta r = 50$ cm, $2a_1 = 0.04$ cm, $p = 50.6 \cdot 10^4$ N/m², $(\Delta T/\overline{T})_1 = 1$, $\overline{T} = 273°$ K; $\overline{D} \simeq 0.05$ cm²/sec, $\rho/\eta \simeq 6.5$ sec/cm², $\alpha_T = 0.015$ m² (for the mixture $C^{12,13}H_4$, according to the data of [4], $\alpha_T = 0.0077$). We assume the condition $D_e = 2D$. It is possible that in a well-designed column $D_e < 2D$, since the parameter Re for $\overline{v}_{r\ extr}$ corresponding to $H = H_{min}$ (see Eq. (18)), is equal to $\frac{\overline{v}_r \overline{\rho}\, 2a}{\eta} = \frac{2\sqrt{2DD_e}}{\eta}\overline{\rho} \cong 6$, i.e., is relatively small. Moreover, nonuniformities of the gap will be partly compensated for by the additional azimuthal flow Δv_φ. Then $H = 0.08$ cm, $\delta = 0.094$ cm^{-1} and $q = 110$. The value of ω_{opt}, according to Eq. (24), is equal to 61 rad/sec, i.e., $\nu \simeq 600$ min^{-1}. The relaxation time for a column fed with material from a central reservoir ($r \le r_1$) and closed at the end $r = r_2$, is

$$\tau \simeq \frac{2\Delta r^2}{D_e[(\delta\Delta r)^2 + 2u]}, \quad (30)$$

where $u \sim 1$ [1]. Hence $\tau \simeq 2 \cdot 10^3$ sec.

The example given provides quite a good illustration of the advantages of the thermocentrifugal column with a small gap. For a thermogravitational column with the same gap, the optimal pressures are several tens of atmospheres, and the settling time and the flow rate of the material increase correspondingly. Furthermore, it is considerably easier, with a small gap, to satisfy the condition that the gap be uniform in a plane column, and in this case the rotating system will be completely free of azimuthal temperature nonuniformities. Of course, the basic difficulties in making up the column lie in obtaining a uniform gap around the azimuth. An accuracy of machining the metal of the order of 10 microns is required, over a substantially large area ($r \sim 50$ cm). In the case of a gap $2a_1 = 0.3 - 0.5$ mm, $\Delta r = 50$ cm for relatively small values of $\alpha_T = 0.01 - 0.02$ the quantity q is 10^2–10^3.

NOTATION

p is the pressure; ρ is the mass density; n is the mole density; ΔT is the temperature drop between faces; c is the mole fraction of isotope being concentrated; c_I and c_{II} are the mean concentrations in cold and hot phases; c_0 is the initial relative concentration; η is the dynamic viscosity; r_1, r_2 are the minimum and maximum radii of working part of column; $\Delta r = r_2 - r_1$; φ is the azimuthal coordinate; z is the axial coordinate; a is the half width of gap between faces; $a_1 = a(r_1)$; ω is the angular velocity of rotation; ν is the frequency of rotation; ω_{opt} is the corresponds to maximum separation; v_r is the radial linear velocity; Δv_φ is the velocity of rotation of gas under action of Coriolis forces; v_z is the velocity of motion between faces; D is the diffusion coefficient; D_e is the equivalent diffusion coefficient; α_T is the thermodiffusion coefficient; J is the radial circulation flux; j is the gas flux density; j_c is the flux density of isotope being concentrated; i_k, c_k are the flux and relative concentration in sample; $\varepsilon_e = H_{min}\delta$ is the equivalent enrichment coefficient; H is the HETP of column; $\gamma = \varepsilon_e/a$; q is the column separation factor; τ is the relaxation time.

REFERENCES

1. K. Jones and V. Ferri, Separation of Isotopes by Thermal Diffusion [Russian translation], IL, 1947.
2. M. Farber and W. E. Libbey, J. Chem. Phys. 8, 965, 1940.
3. F. Schulz-Grunow, AZMP, 9, 628, 1958.
4. A. O. Nier, Phys. Rev., 57, 30, 1940.

15 June 1966 Leningrad Polytechnic Institute