The Separation of Isotopes by Countercurrent Gaseous Exchange Column. I. Transport Equation and Over-All Separation

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Introduction

Taylor and others(1)(2) have observed a very efficient isotope separation with experiments on the countercurrent gaseous exchange column. Bernstein and Taylor(2) have used a hot wire type thermal diffusion column with a top reservoir filled with carbon dioxide and a bottom reservoir in which a chemical converter reduces carbon dioxide to carbon monoxide. The column consisted of a water-jacketed glass tube 1.0 cm. in diameter and 110 cm. long. When it ran two or three days with hot wire temperature 1150°C., the heavy isotope C^{13} was concentrated 1.5 times to the initial concentration at the bottom reservoir. This overall separation is much greater than that of ordinary thermal diffusion.

To account for its high efficiency, Bernstein and Taylor(2) have treated the problem with the simplified flow pattern and idealized two cases; diffusion rate limited case and exchange rate limited case. However, the treatment of this problem is so simplified that it cannot explain the mechanism of isotope separation by this column. The exchange reaction between carbon dioxide and monoxide had been studied by Brandner and Urey,(3) and recently Norris and Ruben(4) reexamined this problem. Though their results are not entirely conclusive it may be said that the reaction velocity is very slow at the temperature below 700°K., and at the higher temperature the reaction velocity increases as the ordinary chemical reactions. Since the diffusion rate is proportional to the square of temperature, it may be reasonable to assume that above the certain temperature T_b the reaction is very rapid compared with the diffusion process. At the region where temperature is below T_b the exchange reaction may be considered very slow compared with the diffusion process, therefore we may treat the behavior of heavy isotope C^{13} in carbon dioxide and carbon monoxide separately. In the region where the temperature is higher than T_b, the equilibrium of the isotopic exchange reaction,

$$C^{12}O_2 + C^{13}O \rightleftarrows C^{13}O_2 + C^{12}O$$

holds with the equilibrium constant $\alpha(T)$ of this temperature. Under these situations the mechanism of the isotope enrichment by countercurrent gaseous exchange column can

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(1) T. I. Taylor and R. B. Bernstein, *J. Am. Chem. Soc.*, **69**, 2076 (1947); T. I. Taylor and W. Spindel, *J. Chem. Phys.*, **16**, 635 (1948).

(2) R. B. Bernstein and T. I. Taylor, *J. Chem. Phys.*, **16**, 903 (1948).

(3) J. Brandner and H. C. Urey, ibid., **13**, 351 (1945).

(4) T. H. Norris and S. Ruben, ibid., **18**, 1595 (1950).

be solved using no more serious assumptions and simplified cases.

Descriptions of Experimental Conditions and Notations

Suppose two different gases of species 1 and 2 each has the same element A, and there are two isotopes A^i and A^j. When we write AB for the gas molecule of species 1 and AC for species 2, the chemical exchange reaction,

$$A^iB+A^jC \rightleftarrows A^jB+A^iC \qquad (1)$$

takes place very rapidly compared with the diffusion process at the temperature higher than T_b and slowly enough at the temperature below T_b.

The apparatus is supposed to be an ordinary hot wire type thermal duffusion column, and its dimensions are as follows; r_1, r_2=radius of cooled cylinder and hot wire, Z=length of column, T_1 and T_2 are absolute temperatures of cooled cylinder and hot wire respectively. r and z are radial and vertical (along the column) coordinates; $r_2 \leq r \leq r_1$, $0 \leq z \leq Z$.

The gas in the apparatus is a mixture of AB and AC. c_1, c_2=fractional molar concentration of species 1(AB) and 2(AC); $c_1+c_2=1$. ν_1, ν_2= fractional molar concentration of isotope A^i of the species 1 and of species 2. ν=fractional molar concentration of isotope A^i as a whole; $\nu=c_1\nu_1+c_2\nu_2$. D_1, D_2=self diffusion coefficient of species 1 and 2. λ=thermal conductivity, η=viscosity, ρ=density of gas mixture. M_1, M_2=molecular weight of AB and AC, $\overline{M}$=mean molecular weight; $\overline{M}=c_1M_1+c_2M_2$.

Isotopic Transport

The consideration about the distribution of different gases in the thermal diffusion column treated here is based on the theory which had been developed by Furry, Jones and Onsager.(5) Suppose species 1 concentrates to the hot part of the column by thermal diffusion, the transport τ_1 (mol./sec.) of species 1 along the column is given by Furry and Jones(6)(7) as follows,

$$\overline{M}\tau_1=Hc_1c_2-(K_c+K_d)(dc_1/dz) \qquad (2)$$

where H, K_c and K_d are given by expressions (52), (40) and (47). Regarding the flow of gas, the equation of flux (mol./sec. cm.) of species 1 was given by Furry, Jones and Onsager(5) as follows,

$$\boldsymbol{J}_1=(\rho/\overline{M})[\boldsymbol{v}c_1+D'_1(-\mathrm{grad}\, c_1 + \alpha_t c_1 c_2\, \mathrm{grad}\, \ln T)] \qquad (3)$$

and a corresponding equation holds for $\boldsymbol{J}_2$. Where $\boldsymbol{v}$ is velocity vector of convective flow, D'_1 is diffusion coefficient of species 1 in the gas mixture and α_t is thermal diffusion constant for the mixture of two different gases. The isotopic exchange reaction and the separation of isotope in the column are treated here at the situation in which the steady state was attained about the distribution of two gases of species 1 and 2. Then we have

$$\mathrm{div}\, \boldsymbol{J}_1=\mathrm{div}\, \boldsymbol{J}_2=0 \qquad (4)$$

Regarding the flow of isotope in the gas of species 1, the equation of flux of A^i is

$$\boldsymbol{I}_1=\nu_1\boldsymbol{J}_1-(\rho D_1 c_1/\overline{M})(\mathrm{grad}\, \nu_1 - \alpha_1(1-\nu_1)\nu_1\, \mathrm{grad}\, \ln T) \qquad (5)$$

and the flux of A^i in the gas of species 2 is

$$\boldsymbol{I}_2=\nu_2\boldsymbol{J}_2-(\rho D_2 c_2/\overline{M})(\mathrm{grad}\, \nu_2 - \alpha_2(1-\nu_2)\nu_2\, \mathrm{grad}\, \ln T) \qquad (6)$$

where α_1 and α_2 are isotopic thermal diffusion constants for gases of species 1 and 2 respectively. We assume that the time variation of ν_1 and ν_2 at each point in the tube is negligible, i. e. stationary or quasistationary state with respect to the isotopic distribution in the column. This assumption holds not only at the final equilibrium but also during the approach to equilibrium when both reservoirs are sufficiently large. From these conditions we have

$$\mathrm{div}\, \boldsymbol{I}_1=\mathrm{div}\, \boldsymbol{I}_2=0 \quad (T<T_b) \qquad (7)$$

at the point where the temperature is below T_b. In the region where the temperature is higher than T_b, the exchange reaction (1) takes place. Then in this region we have only one equation,

$$\mathrm{div}\, (\boldsymbol{I}_1+\boldsymbol{I}_2)=0 \quad (T \geqq T_b) \qquad (8)$$

and in this region ν_1 and ν_2 are related by equilibrium canstant

$$\nu_2/\nu_1=\alpha(T) \quad (T \geqq T_b) \qquad (9)$$

The application of equations (5), (6) and (2) on equation (7) gives

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(5) W. H. Furry, R. C. Jones and L. Onsager, *Phys. Rev.*, **55**, 1083 (1939).

(6) R. C. Jones and W. H. Furry, *Rev. Mod. Phys.*, **18**, 151 (1946).

(7) W. H. Furry and R. C. Jones, *Phys. Rev.*, **69**, 459 (1946).

$$\boldsymbol{J}_1 \operatorname{grad} \nu_1 - \operatorname{div}(\rho D_1 c_1 \operatorname{grad} \nu_1)/\bar{M} = 0 \qquad (T<T_b) \qquad (10)$$

$$\boldsymbol{J}_2 \operatorname{grad} \nu_2 - \operatorname{div}(\rho D_2 c_2 \operatorname{grad} \nu_2)/\bar{M} = 0 \qquad (T<T_b) \qquad (11)$$

in the same way we have from equations (8) and (9)

$$\boldsymbol{J}_1 \operatorname{grad} \nu_1 + J_2 \operatorname{grad}(\alpha\nu_1) - \operatorname{div}(\rho D_1 c_1 \operatorname{grad} \nu_1 + \rho D_2 c_2 \operatorname{grad}(\alpha\nu_1))/\bar{M} = 0 \quad (T \geqq T_b) \quad (12)$$

where we neglected the third terms of equations (5) and (6) which are derived from thermal diffusion of isotope in each of two gases. Because we are now interested in the case having a large separation factor which would never be explained by thermal diffusion. We can see from equation (3) that the z-component j_{iz} of the vector $\boldsymbol{J}_i$ ($i=1, 2$) is sufficiently large compared with other components except the small region where v_z becomes zero. Then from equations (10), (11) and (12), neglecting the radial components of $\boldsymbol{J}_i$ we have

$$j_{iz}(\partial\nu_1/\partial z) - r^{-1}(\partial/\partial r)\rho D_i c_i r(\partial\nu_i/\partial r)/\bar{M} = 0 \qquad (i=1, 2) \ (T<T_b) \qquad (13)$$

$$j_{1z}(\partial\nu_1/\partial z) + j_{2z}(\partial\nu_2/\partial z) - r^{-1}(\partial/\partial r)\rho r[D_1 c_1(\partial\nu_1/\partial r) + D_2 c_2(\partial\nu_2/\partial r)]/\bar{M} = 0 \quad (T \geqq T_b) \quad (14)$$

In the above equations the terms containing $(\partial^2\nu_1/\partial z^2)$ and $(\partial^2\nu_2/\partial z^2)$ are omitted. These terms represent the effects of diffusion of isotope along the column which are separately treated in the equation of isotopic transport. The term $D_1\rho(\partial c_1/\partial z)(\partial\nu_1/\partial z)\bar{M}$ and corresponding term of species 2 are also omitted in (13) and (14), since the diffusion along the column is small enough compared with the convective flow. The neglect of these terms in equations (13) and (14) gives only a slight effect as much as the neglect of radial components of $\boldsymbol{J}_i$ in equations (10) and (11).

Now we introduce the functions L_i and L which are defined as follows,

$$\int_{r_1}^{r} 2\pi r j_{iz} dr = L_i(r) \qquad (i=1, 2) \qquad (15)$$

$$L_1(r) + L_2(r) = L(r) \qquad (16)$$

Using the above equations we can integrate (13) and (14),

$$(\partial\nu_1/\partial z)L_i(r) = 2\pi\rho D_i c_i r(\partial\nu_i/\partial r)/\bar{M} \qquad (i=1, 2) \quad (T<T_b) \qquad (17)$$

$$(\partial\nu_1/\partial z)L_1(r) + (\partial\nu_1/\partial z)L_2(r) = 2\pi\rho r[D_1 c_1(\partial\nu_1/\partial r) + D_2 c_2(\partial\nu_1\alpha/\partial r)]/\bar{M} \qquad (T \geqq T_b) \qquad (18)$$

since $\partial\nu_1/\partial r$ and $\partial\nu_2/\partial r$ must vanish at the walls and we neglect small differences between $\partial\nu_1/\partial z$ and $\partial\nu_2/\partial z$, and their dependence on radial coordinate. By means of equation (15) the total flow of gases of species 1 and 2 are given by

$$-\tau_i = L_i(r_2) \quad (i=1, 2) \qquad (19)$$

As the total flow of gas as a whole is zero, we get

$$\tau_1 + \tau_2 = 0 \qquad (20)$$

At any point $\operatorname{div}(\boldsymbol{I}_1 + \boldsymbol{I}_2)$ is zero then, total transport (τ = mol./sec.) of isotope A^i along the tube is independent of z, i. e.,

$$\tau = \int_{r_2}^{r_1} 2\pi r(j_{1z}\nu_1 + j_{2z}\nu_2)dr - (\partial\nu_1/\partial z)\int_{r_2}^{r_1} 2\pi\rho r(D_1 c_1 + D_2 c_2)dr/M = \text{const.} \qquad (21)$$

The first term of equation (21) represents the transport of isotope by the convective flow of gases and the second term is derived from the vertical diffusion of isotope. When the small radial variations of c_1 and c_2 are neglected, the second term becomes the same as the case of thermal diffusion column, and we can use the results of Furry and Jones(6)(7) as follows,

$$(\partial\nu_1/\partial z)\int_{r_2}^{r_1} 2\pi\rho r(D_1 c_1 + D_2 c_2)dr/\bar{M} = [c_1(K_d)_1/\bar{M} + c_2(K_d)_2/\bar{M}](\partial\nu_1/\partial z) \qquad (22)$$

where $(K_d)_i = 2\pi(\rho D_i)_1 r^2{}_1 k_d \quad (i=1, 2) \qquad (23)$

They tabulated the numerical values of k_d for the different values of (T_2/T_1) and (r_1/r_2) in the case of a Maxwellian gas. The subscript 1 of (ρD_i) emphasizes that the quantity in parethesis is to be evaluated at T_1. Using equation (15), partial integration of the fir term of equation (21), gives

$$\int_{r_2}^{r_1} 2\pi r(j_{1z}\nu_1 + j_{2z}\nu_2)dr$$

$$= \int_{r_2}^{r_b} 2\pi r\nu_1(j_{1z} + \alpha j_{2z})dr - \int_{r_1}^{r_b} 2\pi r(\nu_1 j_{1z} + \nu_2 j_{2z})dr$$

$$= \tau_1\nu_1(r_b) + \tau_2\nu_2(r_b) - \int_{r_2}^{r_b} [(\partial\nu_1/\partial r)(\tau_1 + L_1)$$

$$+(\partial\nu_2/\partial r)(\tau_2+L_2)\}dr$$
$$+\int_{r_1}^{r_b}((\partial\nu_1/\partial r)L_1+(\partial\nu_2/\partial r)L_2)dr$$

where r_b is radial coordinate, the temperature of which is T_b. According to (15), (9), (20), (17) and (18) the above expression becomes

$$\begin{aligned}&\nu_1\tau_1(1-\alpha(T_b))\\&+\int_{r_2}^{r_b}\frac{D_2c_2(\tau_1+L_1)-D_1c_1(\tau_2+L_2)}{D_1c_1+D_2c_2}\nu_1\frac{d\alpha}{dr}dr\\&+\int_{r_1}^{r_b}\frac{\overline{M}}{2\pi\rho r}\left(\frac{L^2{}_1}{D_1c_1}+\frac{L^2{}_2}{D_2c_2}\right)\frac{\partial\nu_1}{\partial z}dr\\&+\int_{r_b}^{r_2}\frac{\overline{M}}{2\pi\rho r}\left(\frac{L^2}{D_1c_1+D_2c_2}\right)\frac{\partial\nu_1}{\partial z}dr\end{aligned}\tag{24}$$

We may regard in expression (24) the functions ν_1 and $\partial\nu_1/\partial z$ depend only on z. By expressions (22) and (24), the isotopic transport given by (21) becomes as follows,

$$\tau=A\nu_1-(B_c+B_d)\partial\nu_1/\partial z\tag{25}$$

where

$$\begin{aligned}A=&\tau_1(1-\alpha(T_b))\\&+\int_{r_2}^{r_b}\frac{D_2c_2(\tau_1+L_1)-D_1c_1(\tau_2+L_2)}{D_1c_1+D_2c_2}\frac{d\alpha}{dr}dr\end{aligned}\tag{26}$$

$$\begin{aligned}-B_c=\frac{\overline{M}}{2\pi}\Bigg[&\int_{r_1}^{r_b}\left(\frac{L_1{}^2}{D_1c_1}+\frac{L_2{}^2}{D_2c_2}\right)\frac{dr}{\rho r}\\&+\int_{r_b}^{r_2}\frac{L^2}{D_1c_1+D_2c_2}\cdot\frac{dr}{\rho r}\Bigg]\end{aligned}\tag{27}$$

$$B_d=(c_1(K_d)_1+c_2(K_d)_2)/\overline{M}\tag{28}$$

Evaluation of integrals in B_c is simply performed when some approximations are introduced for L_1, L_2 and diffusion coefficients. In expression (27) diffusion coefficients D_1 and D_2 are contained in positive terms, therefore we can substitute these coefficients by mean value $\overline{D}$ without great error.

The main part of j_{1z} is convective flow along the tube. Then we may put

$$j_{1z}=\rho c_i v/\overline{M}\quad(i=1,2)\tag{29}$$

in equation (15), where v is z-component of convective flow $\boldsymbol{v}$. This is very close approximation since the effects of diffusion are small enough compared with convective flow. We can remove c_i from under the integral sign of (15), because c_i varies only slightly with T. Then we may substitute L_i by c_iL. Using above approximation we obtain

$$-B_c=\frac{\overline{M}}{2\pi}\int_{r_1}^{r_2}\frac{L^2}{\rho\overline{D}r}dr\tag{30}$$

In order to evaluate the second term of expression (26) we use the following approximation which is similar to one applied to L_i in the above case;

$$\tau_i+L_i=\int_{r_2}^{r}2\pi rj_{iz}dr\cong c_iL$$

By this approximation we can rewrite equation (26) as follows,

$$A=\tau_1(1-\alpha(T_b))+\frac{c_1c_2(D_2-D_1)}{c_1D_1+c_2D_2}\int_{r_2}^{r_b}L\frac{d\alpha}{dr}dr\tag{31}$$

Convective Flow

Except the both ends of the tube, temperature gradient is determined by conduction alone, and in this region we have

$$2\pi Q=2\pi r\lambda(-dT/dr)\tag{32}$$

where $2\pi Q$ is heat flow by conduction per unit length of the tube, in cal./cm. sec.

The hydrodynamical equation of steady viscous flow in cylindrical coordinates is

$$r^{-1}(\partial/\partial r)r\eta(\partial v/\partial r)=dp/dz+\rho g\tag{33}$$

where p is pressure and g is acceleration of gravity. We assume that the velocity is entirely in z direction, and it is independent of z. Using equation (33) we have

$$\begin{aligned}Q^2(d/dT)\lambda^{-1}r^{-2}(d/dT)\eta\lambda^{-1}(dv/dT)\\=g(d\rho/dT)\end{aligned}\tag{34}$$

Convective flow v must vanish at the walls;

$$v(T_1)=v(T_1)=0\tag{35}$$

We have from equations (15), (16) and (29),

$$L=\int_{r_1}^{r}2\pi r\rho vdr/\overline{M}\tag{36}$$

From this equation we can see that L is the total flow of convection at the outside of the cylindrical part of radius r. Then we have

$$L(r_1)=L(r_2)=0\tag{37}$$

Differentiate equation (36) with respect to r, and using equation (32) we have

$$v=-(\overline{M}Q/2\pi\lambda r^2)(dL/dT)\tag{38}$$

Substitution of equation (38) in (34) gives a differetial equation for L,

$$\frac{\bar{M}\alpha^3}{2\pi}\frac{d}{dT}\frac{1}{r^2\lambda}\frac{d}{dT}\frac{\eta}{\lambda}\frac{d}{dT}\frac{1}{r\lambda\rho}\frac{dL}{dT}=g\frac{d\rho}{dT} \tag{39}$$

From (35), (37) and (38) the boundary conditions are

$$L(r_1)=L(r_2)=L'(r_1)=L'(r_2)=0 \qquad (39)'$$

The function L obtained from (39) and (39)′ is identical to

$$(2\pi\rho\bar{D}/\lambda Q^3\bar{M})G$$

where G is the function used by Furry and Jones (6) (7). Therefore we can use their results of calculation about the integral contained in equation (30), and we have

$$B_c=K_c/\bar{M}=(2\pi/9!)(\rho^3g^2/\eta^2\bar{D})_1\cdot r^8{}_1k_c/\bar{M} \tag{40}$$

where values of k_c are tabulated for Maxwellian gases by Furry and Jones (6) for various combinations of (T_2/T_1) and (r_1/r_2). In order to evaluate the integral in equation (31), we have to solve the differential equation (39). Using the assumption that the quantities λ, η and ρD are proportional to T, an assymptotic solution of equation (39) for extreme cylindrical case $(r_2/r_1\to 0)$ has been given by Furry and Jones (7);

$$L(T)=2\pi\bar{M}(\rho g/\eta)_1\cdot r^4{}_1t^3{}_1\exp(2t^2{}_1)\gamma(t) \tag{41}$$

$$\gamma(t)\cong\frac{\gamma_\infty-(\gamma''/t_2)}{1-(\delta''/t_2)},$$

$$\delta''=\left[\exp(-t^2{}_1)\Big/\int_{t_1}^{\infty}\exp(-x^2)dx\right]-t_1$$

Where variable t is defined by

$$t=(\lambda/QT)^{1/2}T \tag{42}$$

and t_1 and t_2 are values of t at $T=T_1$ and T_2 respectively. The numerical values of γ_∞ and γ'' as the functions of t were tabulated by Furry and Jones. (7) Using these results we obtain the expression for the integral in (31) in terms of known functions as follows,

$$\begin{aligned}H_b&=\int_{r_2}^{r_b}L(d\alpha/dr)dr\\&=(2\pi/\bar{M})(\rho g/\eta)_1r^4{}_1t^3{}_1\exp(2t^2{}_1)\\&\quad\times[h_b-(h''_b/t_2)]/[1-(\delta''/t_2)]\end{aligned} \tag{43}$$

$$h_b=-\int_{t_b}^{\infty}\gamma_\infty(d\alpha/dt)dt \tag{44}$$

$$h''_b=-\int_{t_b}^{\infty}\gamma''(d\alpha/dt)dt$$

Equilibrium Separation Factor

We proceed from the equation for transport of isotope A up to the tube:

$$\tau=A\nu_1-(B_c+B_d)(d\nu_1/dz) \tag{45}$$

where A is given by (31) and (43);

$$A=\tau_1[1-\alpha(T_b)]+[c_1c_2(D_2-D_1)/\bar{D}]H_b \tag{46}$$

B_c is given by equation (40), and B_d is given by equation (28);

$$B_d=K_d/\bar{M}=2\pi(\rho\bar{D})_1r^2{}_1k_d/\bar{M} \tag{47}$$

The approximation was used to derive above equations that we put D_1 and D_2 are equal to $\bar{D}$ except the case which contains their difference.

In the final equilibrium state τ is zero all along the tube. Putting $\tau=0$ in (45) we obtain

$$A\nu=(B_c+B_d)(d\nu/dz) \tag{48}$$

The integral of (48) gives a ratio of the concentration of isotope A in the top and the bottom reservoirs. In general case the distinction between ν_1 and ν_2 is not necessary when we consider the large separation of isotope in both reservoirs. The functions of z in equation (48) are ν and A which depends z through c_1c_2 in (46). Then integral of (48) is

$$\begin{aligned}(1-\alpha(T_b))Z+\frac{D_2-D_1}{\bar{D}}H_b\int_0^Z c_1c_2dz\\=(B_c+B_d)\ln\left(\frac{\nu(Z)}{\nu(0)}\right)\end{aligned} \tag{49}$$

where $\nu(0)$ and $\nu(Z)$ are values of ν at the bottom and the top reservoirs respectively.

By the similar way we obtain from equation(2)

$$\begin{aligned}(H/\bar{M})\int_0^Z c_1c_2dz=\tau_1Z\\+(K_c+K_d)(c_1(Z)-c_1(0))/\bar{M}\end{aligned} \tag{50}$$

where $c_1(Z)$ and $c_1(0)$ are c_1 at the bottom and the top reservoirs. Substitution of (50), (40) and (47) into (49) leads to the expression for over-all separation S as follows,

$$\begin{aligned}\ln S=\ln\left(\frac{\nu(Z)}{\nu(0)}\right)=\Big[(1-\alpha(T_b))\\+\frac{D_2-D_1}{\bar{D}}\frac{H_b}{H/\bar{M}}\Big]\frac{\tau_1Z}{B_c+B_d}\\+\frac{D_2-D_1}{\bar{D}}\frac{H_b}{H/\bar{M}}(c_1(Z)-c_1(0))\end{aligned} \tag{51}$$

When we use the approximate formula (41), H becomes

$$H=2\pi(\rho^2 g/\eta)_1 r^4{}_1 t^3{}_1 \exp(2t^2{}_1)\frac{h_\infty-(h''/t_2)}{1-(\delta''/t_2)} \tag{52}$$

where
$$h_\infty=\int_{t_1}^{\infty}\alpha_t(\gamma_\infty/t)dt$$
$$h''=\int_{t_1}^{\infty}\alpha_t(\gamma''/t)dt \tag{53}$$

The expressions (53) and (43) give

$$\frac{H_b}{H/\bar{M}}=\frac{h_b-(h_b''/t_2)}{h_\infty-(h''/t_2)} \tag{54}$$

Discussion

From rather surprisingly simple results given by equation (51) we can easily calculate overall separation S, since B_c, B_d and $(c_1(Z)-c_1(0))$ may be determined from experimental conditions, and H_b/H may be numerically calculated by expression (54) when we know the temperature T_b.

The details of experimental verification of equation (51) will be reported in part 2 on this Bulletin.

We shall now discuss the functioning of gaseous exchange column. The remixing effects of the diffusion of isotope along the tube and of the countercurrent in the tube are represented by $B_d d\nu_1/dz$ and $B_c d\nu_1/dz$ respectively in the transport equation (45). It is reasonable that an appropriate approximation leads these terms to the expressions which are identical for the ordinary thermal diffusion as shown by equation (40) and (47). The initial isotopic transport obtained by putting $d\nu_1/dz=0$ in equation (45) is rather complicated, but its meaning is clear. When the ratio ν_2/ν_1 is $\alpha(T_b)$ at any point in the tube,

$$\nu_1\tau_1(1-\alpha(T_b))=\tau_1\nu_1+\tau_2\nu_1\alpha(T_b)$$

is transport of isotope A along the tube. Whereas in the equilibrium region ($T\geqq T_b$) α varies with temperature. If we assume the self diffusion coefficient D_1 is larger than D_2, the radial variation of ν_1 may not be large, but the radial variation of α results in a strong variation of ν_2. The deviation of transport estimated in the first term of the expression (46) is corrected by the second term which is derived from such an asymmetric variation of ν_1 and ν_2 between r_2 and r_b. An application of (46) to the experiments of Bernstein and Taylor(2) shows that the first term is negligibly small compared with the second term. Therefore we may conclude that the transport of isotope in the equilibrium region results in the high efficiency of the gaseous exchange column. Perphaps there is another possibility to obtain the high efficiency by the mode of operation in which τ_1 takes large value by the thermal dissociation with suitable gas. In this case the transport of gases must be recalculated with the equilibrium constant of thermal dissociation.

Summary

The theoretical treatment of the countercurrent gaseous exchange method for the separation of isotopes is developed here. The equation of transport is derived with the assumption that the equilibrium of the isotopic exchange reaction holds only in the sharply defined region in which temperature is higher than T_b. An appropriate approximation leads the transport equation to one which is formally similar to the transport equation of thermal diffusion column. The mechanism of isotope enrichment by countercurrent gaseous exchange method is discussed using the transport equation. The equilibrium separation factor is also derived from the transport equation.

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