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Solution of the Diffusion Equations in a Gas Centrifuge for Separation of Multicomponent Mixtures

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ABSTRACT

The demand for stable isotopes in physical and chemical research and in medical diagnostics is growing, and the gas centrifuge process is able to provide large quantities of stable isotopes. A set of diffusion equations describing separation in a gas centrifuge for a multicomponent mixture is established. These equations involve general diffusion coefficients. Using the radial averaging method and the simplified diffusion transport vector for a multicomponent isotopic mixture, nonlinear partial differential equations are transformed to a set of nonlinear ordinary differential equations. An iteration method for the solution is presented. The relationship between the separation factor and the mass difference, $\gamma_{ij} = \gamma_0^{M_j - M_i}$, is shown to be in agreement with both the computational and the experimental results with very high precision.

Key Words. Gas centrifuge; Diffusion equation; Multicomponent mixture; Stable isotopes; Separation

1. INTRODUCTION

Over the past years, many countries have developed the gas centrifuge to separate the binary mixture of $^{235}UF_6$ and $^{238}UF_6$ for producing uranium enriched in the fissionable isotope ^{235}U for fuel in nuclear reactors. Recently, with the growing demand for stable isotopes in physical and chemical research and in medical diagnostics, the use of the gas centrifuge process has made it possible to produce many different isotopes, especially when large quantities are needed. Many countries [such as the United States (1, 2), Russia (3–5), and China (7)] and organizations [URENCO (6)] have reported their activities in the public literature in the field of multicomponent separation by gas centrifuge.

In a recent paper, Wood et al. (8) discussed multi-isotope separation in a gas centrifuge. In that paper they solved Onsager's pancake equation for the countercurrent flow field and the diffusion equation for each isotope. This solution method was connected to an algorithm which optimizes the centrifuge's performance. In the analysis, the diffusion coefficient was assumed to be the same throughout the centrifuge, a good assumption for gases with large molecular weights and small differences in the molecular weights of the isotopes. However, when these conditions are not met, variations in the diffusion coefficient may be important.

The object of the present paper is to establish a general set of diffusion equations describing the separation phenomena in a gas centrifuge for multicomponent mixtures. The diffusion equations in a gas centrifuge for multicomponent mixtures are different from those for binary mixtures because general diffusion coefficients are involved. These equations are a set of nonlinear partial differential equations. Using the radial averaging method (9–12) and the simplified diffusion transport vector for multicomponent isotopic mixtures (13), nonlinear ordinary differential equations are obtained. An iteration method of the solution is presented. The computational and experimental results show the separation factor, γ_{ij}, may be expressed as $\gamma_{ij} = \gamma_0^{M_j - M_i}$, where γ_0 is the overall separation factor for the unit mass difference, and M_i and M_j are the molecular weights of the ith and the jth component, respectively. This relationship is compared with the experimental data and is found to agree with very high precision.

2. DIFFUSION EQUATIONS AND DIFFUSION COEFFICIENTS FOR MULTICOMPONENT MIXTURES

For a mixture of n components, the diffusion transport vector $\mathbf{J}_i$ of the ith isotope is (14)

$$\mathbf{J}_i = -\rho \frac{M_i}{\overline{M}} C_i \left(\sum_{j=1}^{n} D_{ij}\mathbf{d}_j + D_i^T \nabla \ln T \right); \qquad i = 1, 2, \ldots, n \tag{2.1}$$

where ρ is the density of the mixture, C_i is the concentration of the ith component, M_i is the molecular weight of the ith component, $\overline{M}$ is the average molecular weight of the mixture, i.e., $\overline{M} = \sum_{i=1}^{n} M_i C_i$, D_{ij} are the general multicomponent diffusion coefficients (GMDC), and $\mathbf{d}_j$ is the diffusion driving force, which could be written as (15)

$$\mathbf{d}_j = \nabla C_j + C_j\left(1 - \frac{M_j}{\overline{M}}\right)\nabla \ln p - \frac{M_j}{\overline{M}}\frac{C_j}{p}\left(\rho \mathbf{F}_j - \sum_{k=1}^{n} \rho_k \mathbf{F}_k\right); \quad j = 1, 2, \ldots, n \tag{2.2a}$$

$$\sum_{j=1}^{n} \mathbf{d}_j = 0 \tag{2.2b}$$

where p is the pressure, $\mathbf{F}_k$ is the external body force per unit mass of the kth component, and ρ_k is the density of the kth component. If we consider the process gas is rotating in the gas centrifuge with angular velocity Ω, then $\mathbf{F}_j = \Omega^2 \mathbf{r}$. Because $\rho = \sum_{k=1}^{n} \rho_k$, the last two terms in Eq. (2.2a) are eliminated and we obtain

$$\mathbf{d}_j = \nabla C_j + C_j\left(1 - \frac{M_j}{\overline{M}}\right)\nabla \ln p; \qquad j = 1, 2, \ldots, n \tag{2.3}$$

The GMDC are determined by (14)

$$\sum_{k=1}^{n} \frac{C_i C_k}{\mathfrak{D}_{ik}}(D_{ij} - D_{kj}) = \delta_{ij} - \frac{M_i}{\overline{M}} C_i; \qquad j = 1, 2, \ldots, n-1 \tag{2.4a}$$

$$\sum_{i=1}^{n} M_i C_i D_{ij} = 0 \tag{2.4b}$$

here δ_{ij} is the Kronecker delta and $\mathfrak{D}_{ik}$ is the binary diffusion coefficient (15)

$$\mathfrak{D}_{ik} = 2.628 \times 10^{-7} \frac{T^{3/2}\sqrt{(M_i + M_k)/(2M_i M_k)}}{p\sigma_{ik}^2 \Omega_{ik}^{(1,1)}} \ [\mathrm{m^2/s}] \tag{2.5}$$

where p (atm) is the pressure, σ_{ik} (10^{-10} m) is the molecular interaction diameter; $\Omega_{ik}^{(1,1)}$ is the integral of interaction for molecular mass transfer; M_i and M_k (mole) are the molecular weights of the ith and kth component, respectively; and T (K) is the temperature. $\mathfrak{D}_{ik}$ does not depend on the concentration C_i or C_k, but from Eq. (2.4) one finds that D_{ij} is a function of the concentration.

Using Eqs. (2.1) to (2.5) to obtain the diffusion transport vector in the gas centrifuge, we assume that the term of thermal diffusion $D_i^T \nabla \ln T$ in Eq. (2.1) is negligible.

As for the diffusion coefficients, Levin and Ying (13) showed that if any transformation such as

$$\overline{D}_{ij} = D_{ij} + A_i^* \tag{2.6}$$

gives a new value of the diffusion coefficient $\overline{D}_{ij}$, the diffusion transport vector of $\mathbf{J}_i$ is unchanged. This is because

$$\sum_{j=1}^{n} A_i^* \mathbf{d}_j = A_i^* \sum_{j=1}^{n} \mathbf{d}_j = 0 \tag{2.7}$$

Reference 13 also shows that for an isotopic mixture with large molecular weights and a small difference in the molecular weights of the isotopes, the diffusion coefficient $C_i D_{ij}^{is} \approx \mathcal{D}_i(\delta_{ij} - M_i/\overline{M})$ is diagonally dominant, where

$$\mathcal{D}_i = \left(\sum_{k=1}^{n} \frac{C_k}{\mathcal{D}_{ik}} \right)^{-1}$$

We may take

$$C_i \overline{D}_{ij}^{is} = \mathcal{D}_i \delta_{ij} \tag{2.8}$$

Then the approximate diffusion transport vector $\mathbf{J}_i$ in the gas centrifuge for an isotopic mixture is obtained as

$$\mathbf{J}_i = -\rho \frac{M_i}{\overline{M}} \mathcal{D}_i \left[\nabla C_i + \left(1 - \frac{M_i}{\overline{M}}\right) C_i \nabla \ln p \right] \tag{2.9}$$

The diffusion equations in steady state are a set of mass conversation equations for each component in the mixture. They are

$$\nabla \cdot \left(\rho \mathbf{V} \frac{M_i}{\overline{M}} C_i + \mathbf{J}_i \right) = 0; \qquad i = 1, 2, \ldots, n - 1 \tag{2.10a}$$

$$C_n = 1 - \sum_{i=1}^{n-1} C_i \tag{2.10b}$$

where $\mathbf{V}$ is the velocity of the mixture. The first term in Eq. (2.10a) is the convection vector of the ith component and the second term is the diffusion transport vector. By substituting Eq. (2.9) into Eq. (2.10), a set of concentration equations is obtained:

$$\rho V_r \frac{\partial C_i}{\partial r} - \frac{\rho \mathcal{D}_i}{r} \frac{\partial}{\partial r} \left[r \frac{\partial C_i}{\partial r} + \frac{\Omega^2 r^2}{RT} (\overline{M} - M_i) C_i \right] + \rho V_z \frac{\partial C_i}{\partial z} - \rho \mathcal{D}_i \frac{\partial^2 C_i}{\partial z^2} = 0; \qquad i = 1, 2, \ldots, n - 1 \tag{2.11a}$$

$$C_n = 1 - \sum_{i=1}^{n-1} C_i \tag{2.11b}$$

where V_r is the radial component of the velocity and V_z is the axial component of the velocity. The boundary conditions are as follows:

a. There is no radial transport at the rotor wall and on the axis.

$$\text{at} \quad r = r_a, \qquad \frac{\partial C_i}{\partial r} + \frac{\Omega^2 r_a}{RT}(\overline{M} - M_i)C_i = 0 \tag{2.12a}$$

$$\text{at} \quad r = 0, \qquad \frac{\partial C_i}{\partial r} = 0 \tag{2.12b}$$

where r_a is the radius of the rotating cylinder.

b. The axial transport over the end caps equals the withdrawal flow rate.

$$\text{at} \quad z = 0, \qquad \int_0^{r_a} \left(-\rho \mathfrak{D}_i \frac{\partial C_i}{\partial z} + \rho V_z C_i\right) 2\pi dr = -F(1 - \theta)C_i^W; \quad i = 1, 2, \ldots, n \tag{2.13a}$$

$$\text{at} \quad z = Z_H, \qquad \int_0^{r_a} \left(-\rho \mathfrak{D}_i \frac{\partial C_i}{\partial z} + \rho V_z C_i\right) 2\pi dr = -F\theta C_i^P; \quad i = 1, 2, \ldots, n \tag{2.13b}$$

where Z_H is the length of the gas centrifuge. F is the feed flow rate, and θ is the "cut," i.e., the product flow rate equals θF.

In addition, the feed concentration C_i^F of the ith component is related to product concentration C_i^P and waste concentration C_i^W of the ith component by the overall balance equation for the ith component:

$$C_i^F = \theta C_i^P + (1 - \theta)C_i^W; \qquad i = 1, 2, \ldots, n \tag{2.14}$$

It is obvious that Eq. (2.11) are a set of nonlinear partial differential equations. The coefficient $\mathfrak{D}_i$ and average molecular weight $\overline{M}$ in Eq. (2.11a) are dependent on the concentration.

3. RADIAL AVERAGING APPROXIMATION METHOD FOR THE SOLUTION OF THE DIFFUSION EQUATIONS

An averaged concentration of the ith isotope $\overline{C}_i$ is defined as

$$\overline{C}_i = \frac{1}{\pi r_a^2} \int_0^{r_a} C_i 2\pi r dr; \qquad i = 1, 2, \ldots, n \tag{3.1}$$

The variable $\overline{C}_i$ depends only on the axial coordinate z.

The stream function ψ is defined as

$$\psi(r, z) = \int_0^{r_a} \rho V_z 2\pi r dr \tag{3.2}$$

Then we have

$$\partial\psi/\partial r = 2\pi r \rho V_z \tag{3.3}$$

The radial convection term $\rho V_r(\partial C_i/\partial r)$ in Eq. (2.11) can be neglected because the radial component V_r of the velocity is predominant over the axial component V_z only in the very thin Ekman layers near the end caps. The diffusion term $-\rho\mathfrak{D}_i(\partial^2 C_i/\partial z^2)$ is negligible also. Integrating Eq. (2.11a) over r, we obtain

$$\begin{aligned} \frac{\partial C_i}{\partial r} &= -\frac{\Omega^2 r}{RT}(\overline{M} - M_i)C_i + \frac{1}{r\rho\mathfrak{D}_i}\frac{d\overline{C}_i}{dz}\int_0^r \rho V_z r' dr' \\ &= -\frac{\Omega^2 r}{RT}(\overline{M} - M_i)C_i + \frac{\psi}{2\pi r\rho\mathfrak{D}_i}\frac{d\overline{C}_i}{dz}; \qquad i = 1, 2, \ldots, n \end{aligned} \tag{3.4}$$

We introduce the net axial flow flux of the ith component P_i^*, as

$$P_i^* = \int_0^{r_a}\left(J_{iz} + \rho C_i V_z \frac{M_i}{\overline{M}}\right) 2\pi r dr; \qquad i = 1, 2, \ldots, n \tag{3.5}$$

The net axial flow flux of mixture, P^*, is

$$P^* = \sum_{i=1}^{n} P_i^* \tag{3.6}$$

Using integration by parts, we obtain

$$\begin{aligned} \int_0^{r_a} 2\pi r\rho V_z C_i dr &= \int_0^{r_a}\frac{\partial\psi}{\partial r} C_i dr = \psi C_i\Big|_{z,r=r_a} \\ &- \int_0^{r_a}\psi\frac{\partial C_i}{\partial r}dr = P^*\overline{C}_i(z) - \int_0^{r_a}\psi\frac{\partial C_i}{\partial r}dr \end{aligned} \tag{3.7}$$

Substituting Eq. (3.7) into Eq. (3.5), we have

$$\begin{aligned} P_i^* &= \frac{M_i}{\overline{M}}\left[P_i^*\overline{C}_i(z) - \int_0^{r_a}\psi\frac{\partial C_i}{\partial r}dr - \int_0^{r_a}\rho\mathfrak{D}_i\frac{d\overline{C}_i}{dz}2\pi r dr\right]; \\ i &= 1, 2, \ldots, n \end{aligned} \tag{3.8}$$

Using Eq. (3.4) from Eq. (3.8), we obtain

$$\left(\frac{1}{2\pi\rho\mathcal{D}_i}\int_0^{r_a}\frac{\psi^2}{r}\,dr + \pi\rho\mathcal{D}_i r_a^2\right)\frac{d\overline{C}_i}{dz} = \frac{\Omega^2}{RT}(\overline{M} - M_i)C_i\int_0^{r_a}\psi r dr - \left(\frac{\overline{M}}{M_i}P_i^* - P^*\overline{C}_i\right); \qquad i = 1, 2, \ldots, n \tag{3.9}$$

Equation (3.9) is a set of concentration equations.

We define the following parameters:

$$L \equiv \frac{1}{2}\int_0^{r_a}|\rho V_z|2\pi r dr$$

$$\epsilon_i \equiv \frac{\Omega^2}{RT}(\overline{M} - M_i)\int_0^{r_a}\frac{\psi r}{L}\,dr$$

$$\varphi_{Pi} \equiv \frac{\theta F}{\pi r_a \rho\mathcal{D}_i}; \qquad \varphi_{Wi} \equiv \frac{(1 - \theta)F}{\pi r_a \rho\mathcal{D}_i} \tag{3.10}$$

$$Y_{1i} \equiv \frac{1}{r_a^2 \pi r_a \rho\mathcal{D}_i}\int_0^{r_a}\psi r dr$$

$$Y_{2i} \equiv \frac{1}{2(\pi r_a \rho\mathcal{D}_i)^2}\int_0^{r_a}\frac{\psi^2}{r}\,dr$$

Using a procedure similar to that used by Soubbaramayer (12) for the binary mixture, the concentration equations in the enriching section are obtained:

$$(1 + Y_{2i})\frac{dC_i}{ds} = (2\epsilon_i Y_{1i} + \varphi_{Pi})C_i - \varphi_{Pi}C_i^P; \qquad i = 1, 2, \ldots, n - 1$$

$$C_n = 1 - \sum_{i=1}^{n-1} C_i \tag{3.11}$$

where $s = z/r_a$. We drop the overbar of C from Eq. (3.11).

The concentration equations in the stripping section are

$$(1 + Y_{2i})\frac{dC_i}{ds} = (2\epsilon_i Y_{1i} - \varphi_{Wi})C_i + \varphi_{Wi}C_i^W; \qquad i = 1, 2, \ldots, n - 1$$

$$C_n = 1 - \sum_{i=1}^{n-1} C_i \tag{3.12}$$

The coefficients ϵ_i, Y_{1i}, Y_{2i}, φ_{Pi}, and φ_{Wi} are dependent on concentration. Equations (3.11) and (3.12) are two sets of nonlinear ordinary differential equations. Before we start to solve Eqs. (3.11) and (3.12), we need to know the velocity distribution in the gas centrifuge, i.e., the V_z or ψ. The velocity field can be obtained in a variety of ways, such as with the Onsager pancake model reported by Wood and Morton (16). However, here we use a simplified model for the purpose of illustration. We assume that ψ has the following pattern (17):

$$\psi(\xi, \eta) = R_W[e^{-b_1\xi} - (1 + b_1\xi)e^{-2b_1\xi}][4\eta(1 - \eta)]^{2/3} + R_S[e^{-b_2\xi} - (1 + b_2\xi)e^{-2b_2\xi}]e^{-2\eta} \quad (3.13)$$

where

$$\xi = A^2\left(1 - \frac{r^2}{r_a^2}\right); \qquad \eta = \frac{z}{Z_H}; \qquad A^2 = \frac{\overline{M}\Omega^2 r_a^2}{2RT}$$

The term with R_W represents the wall-driven pattern; the term with R_S represents the mechanical-driven pattern.

The constants b_1 and b_2 vary for different process gases, and we take $12 \le b_1A^2 \le 25$, $8 \le b_2A^2 \le 15$, and $b_1A^2 > b_2A^2 > 7.2$.

When parameters F, θ, ψ, and C_i^F are given, using our iteration method we obtain the solution of Eqs. (3.11) and (3.12), i.e., the concentration distribution of each component in the gas centrifuge and the C_i^P, C_i^W. The solution of the concentration $C_i^{(k+i)}$ for the $(k + 1)$th iteration in the enriching section from Eq. (3.11) is

$$\frac{C_i^{(k+1)}(s)}{C_{i0}^{(k+1)}} = \exp[B_{Pi}^{(k)}(s)]\left[1 - \frac{C_i^{P(k+1)}}{C_{i0}^{(k+1)}}\varphi_{Pi}^{(k)}\int_{S_F}^{s}\frac{\exp[-B_{Pi}^{(k)}(s')]}{1 + Y_{1i}^{(k)}}ds'\right];$$

$$i = 1, 2, \ldots, n - 1 \quad (3.14)$$

$$C_n^{(k+1)} = 1 - \sum_{i=1}^{n-1} C_i^{(k+1)}$$

where

$$B_{Pi}^{(k)} \equiv \int_{S_F}^{s}\frac{\varphi_{Pi}^{(k)} + 2\epsilon_i^{(k)}Y_{1i}^{(k)}}{1 + Y_{2i}^{(k)}}ds'$$

C_{i0} is the concentration of the ith component at the feed point in the gas centrifuge, $S_f = Z_f/r_a$, and Z_F is the feed position.

The solution of the concentration $C_i^{(k+i)}$ for the $(k + 1)$th iteration in the stripping section from Eq. (3.12) is

$$\frac{C_i^{(k+1)}(s)}{C_{Wi}^{(k+1)}} = \exp[B_{Wi}^{(k)}(s)]\left[1 + \varphi_{Wi}^{(k)} \int_0^s \frac{\exp[-B_{Wi}^{(k)}(s')}{1 + Y_{2i}^{(k)}} ds'\right];$$

$$i = 1, 2, \ldots, n - 1 \qquad (3.15)$$

$$C_n^{(k+1)} = 1 - \sum_{i=1}^{n-1} C_i^{(k+1)}$$

where

$$B_{Wi}^{(k)} \equiv \int_0^s \frac{-\varphi_{Wi}^{(k)} + 2\epsilon_i^{(k)} Y_{1i}^{(k)}}{1 + Y_{2i}^{(k)}} ds'$$

From Eq. (3.14) we obtain

$$\frac{C_i^{P(k+1)}}{C_{i0}^{(k+1)}} = \frac{1}{\exp[-B_{Pi}^{(k)}(S_H)] + \varphi_{Pi}^{(k)} \int_{S_F}^{S_H} \frac{\exp[-B_{Pi}^{(k)}(s')]}{1 + Y_{2i}^{(k)}} ds'} \qquad (3.16)$$

From Eq. (3.15) we obtain

$$\frac{C_{i0}^{(k+1)}}{C_i^{W(k+1)}} = \exp[B_{Wi}^{(k)}]\left[1 + \varphi_{Wi}^{(k)} \int_0^{S_F} \frac{\exp[-B_{Wi}^{(k)}(s')]}{1 + Y_{2i}^{(k)}} ds'\right] \qquad (3.17)$$

The right sides of Eqs. (3.16) and (3.17) are known. Combining Eqs. (3.16), (3.17), and (2.14), the $C_i^{P(k+1)}$, $C_i^{W(k+1)}$, and $C_{i0}^{(k+1)}$ are obtained. At the same time, we obtain the concentration distribution $C_i^{(k+1)}$ for the $(k + 1)$th iteration in the gas centrifuge. The criterion of the convergence of the iteration is

$$\max_{i=1}^{n} \left|\frac{C_i^{(k+1)} - C_i^{(k)}}{C_i^{(k+1)}}\right| < \epsilon$$

where ϵ is the allowable error.

We calculated many examples, and some of them are shown in Figs. 1 to 4. The figures show the concentration distribution along the axis. Figure 1 shows the concentration of ^{234}U, ^{235}U, and ^{236}U increasing along the axis from the bottom to the top. The concentration of ^{238}U is nearly constant through the whole gas centrifuge because the concentration of ^{238}U in the feed flow is very large, 0.9925. Figure 2 shows a similar result, only the dominant isotope is the light one instead of the heavy one. Figure 3 shows the concentration distribution of the tungsten isotopes in the gas centrifuge. Figure 4 gives the results for OsO_4 separation. Note that the maximum concentration of ^{190}Os is not at the ends of the gas centrifuge. For long gas centrifuges, this phenomenon often appears.

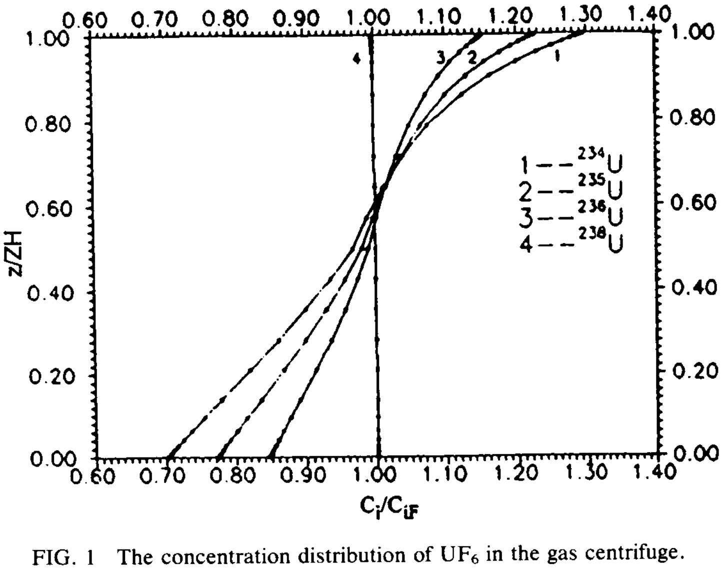

FIG. 1 The concentration distribution of UF_6 in the gas centrifuge.

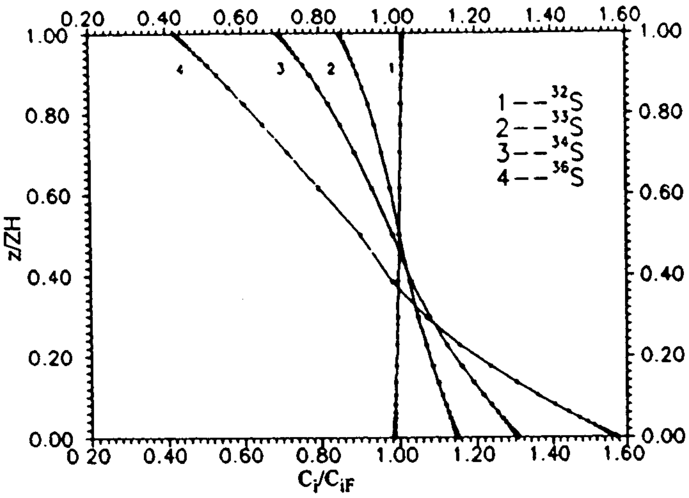

FIG. 2 The concentration distribution of SF_6 in the gas centrifuge.

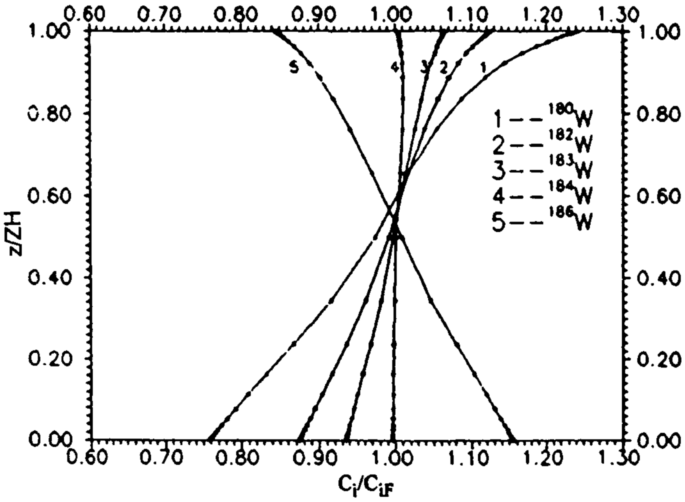

FIG. 3 The concentration distribution of WF_6 in the gas centrifuge.

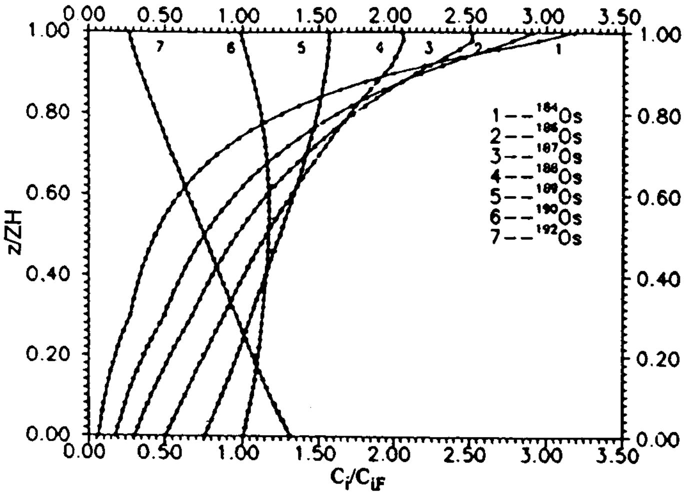

FIG. 4 The concentration distribution of OsO_4 in the gas centrifuge.

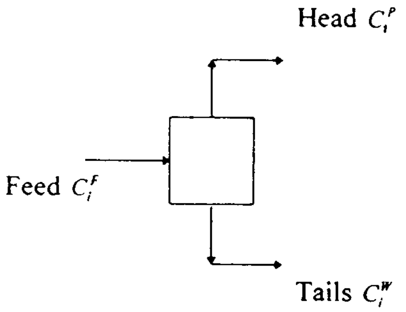

FIG. 5 A schematic of a gas centrifuge.

4. RELATIONSHIP BETWEEN SEPARATION FACTORS AND MASS DIFFERENCE

There are several definitions of the separation factors for multicomponent mixtures, and we use the following definitions (see Fig. 5):

$$\alpha_{ij} = \frac{C_i^P}{C_i^F}\bigg/\frac{C_j^P}{C_j^F}; \qquad \beta_{ij} = \frac{C_i^F}{C_i^W}\bigg/\frac{C_j^F}{C_j^W}; \qquad \gamma_{ij} = \alpha_{ij}*\beta_{ij} = \frac{C_i^P}{C_i^W}\bigg/\frac{C_j^P}{C_j^W} \tag{4.1}$$

The computational results show that the following relationship holds with very high precision:

$$\gamma_{ij} = \gamma_0^{M_j - M_i} \tag{4.2}$$

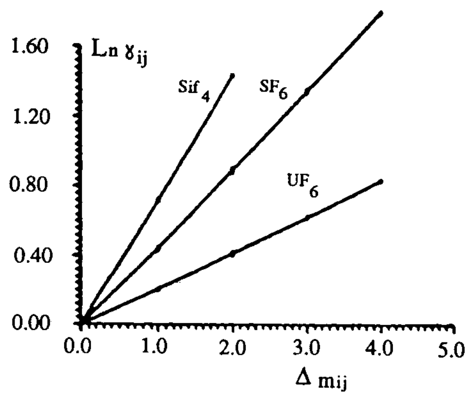

FIG. 6 The relationship between $\ln \gamma_{ij}$ and $(M_j - M_i)$.

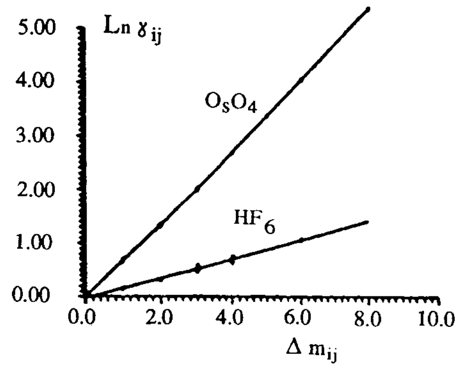

FIG. 7 The relationship between ln γ_{ij} and $(M_j - M_i)$.

where γ_0 is the overall separation factor with unit mass difference. γ_0 depends on the flow pattern in the gas centrifuge, the size and the operating parameters, etc.

The relationship between ln γ_{ij} and $(M_j - M_i)$ for the computational results is shown in Figs. 6 and 7. In the figures the points are the calculated results. Almost all of them are on the straight lines which represent the relationship.

Some authors, such as Von Halle (18) and Raichura et al. (19), define the separation factors differently. They made assumptions for the relationship between their separation factor and mass difference. Here, we use the definition of the separation factor as (4.1), and we find that the relationship of (4.2) is kept with very high precision. This is useful for the calculation of the concentration distribution in a cascade of gas centrifuges. Our work about the cascade theory will be published in the near future.

5. COMPARISON OF THE EXPERIMENTAL RESULTS WITH THE RELATIONSHIP

A variety of isotopes have been enriched using gas centrifuges in the laboratory of Tsinghua University, and some experimental results are shown in Figs. 8 to 10. In Figs. 9 and 10 the feed concentrations are different because they were obtained from different areas.

We checked our experimental results and the results published by Roberts (1) and Szady (2) with the relationship (4.2). The correlation coefficients r and the overall separation factors for unit mass difference, γ_0, are calculated. When the absolute value of the correlation coefficient $|r|$ is close to unity, it means the relationship (4.2) agrees with the experimen-

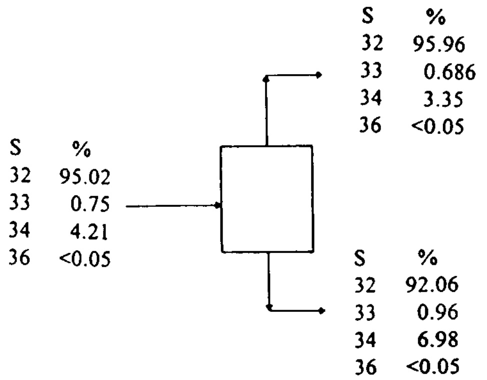

FIG. 8 Sulfur isotopes were separated in a gas centrifuge. SF_6 was the process gas.

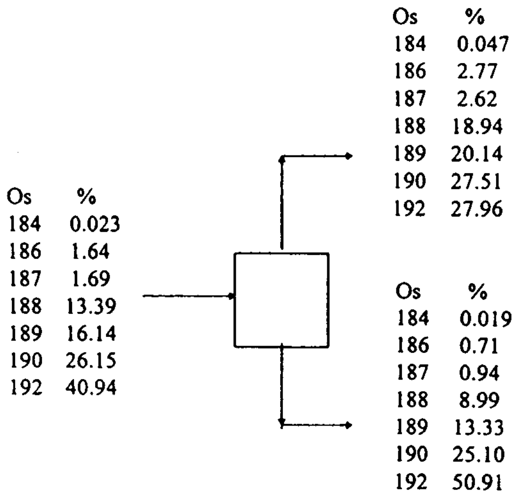

FIG. 9 Osmium isotopes were separated in a gas centrifuge. OsO_4 was the process gas.

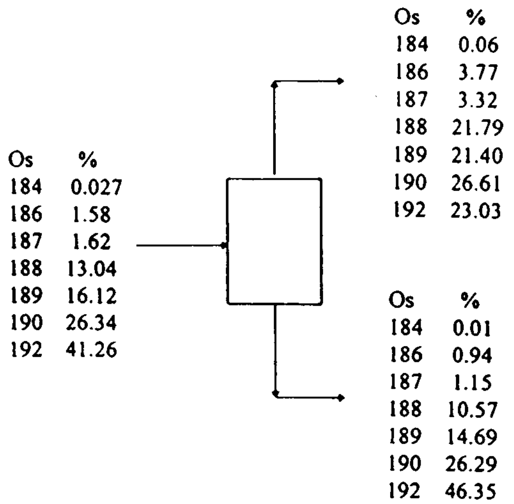

FIG. 10 Osmium isotopes were separated in a gas centrifuge. OsO_4 was the process gas.

tal data. If $|r| = 1$, it means all the data are completely satisfied with the relationship (4.2).

Table 1 lists the correlation coefficients for six experimental samples. The first two columns, i.e., sulfur and osmium, are calculated using the experimental data obtained at Tsinghua University; the other four columns are calculated using the experimental data from References 1 and 2. Most of the correlation coefficients are greater then .99 which confirms that relationship (4.2) is a good approximation for the separation factors. γ_0 is different for these six samples.

TABLE 1
The Correlation Coefficients r for the Experimental Data

Elements	S	Os	Cr	S	Kr	Xe
Process gas	SF_6	OsO_4	CrO_2F_2	SF_6	Kr	Xe
r	.9997	.9983	.9991	.9960	.9951	.9895
γ_0	1.47	1.40	2.43	2.06	6.09	2.28

6. CONCLUSIONS

A theory for the separation of multicomponent mixtures based on general diffusion coefficients has been developed. The method of radial averaging has been used to reduce the partial differential equations to ordinary differential equations, and an iterative method has been used to obtain solutions to these equations. A simplified model of the countercurrent flow has been used here to calculate the concentration distributions for a variety of isotopic mixtures. The relationship between the separation factor and mass difference, $\gamma_{ij} = \gamma_0^{M_j - M_i}$, is determined from the results of these calculations and compared to experimental results obtained at Tsinghua University and at Oak Ridge, Tennessee. The formula is found to be in good agreement.

In the future we plan to couple this separation theory with Onsager's pancake model for the centrifuge flow field. This will allow direct comparisons with the work reported by Wood et al. (8), and it will allow studies in which the isotopes do not necessarily have large molecular weights or small differences in molecular weights.

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