

Present Status of Hydrogen Isotope Separation at Nagoya University

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In Isotope Separation Laboratory at Nagoya University, we have been studying water distillation and thermal diffusion for hydrogen isotope separation. The present paper describes some recent developments of the separative analyses on these technologies.

1 Introduction

The technology of hydrogen isotope separation is a most essential field for fusion energy, and successful operations of a fusion reactor cannot be realized without an establishment of its fuel cycle. Among various methods of hydrogen isotope separation, water distillation (WD) and thermal diffusion (TD) have been studied analytically and experimentally in Isotope Separation Laboratory at Nagoya University.

Water distillation is attractive in spite of its small separation factor because the process has inherent advantages such as simplicity of apparatus and its operation, safety due to absence of explosive or corrosive gasses, and large flow rate. Water distillation is, therefore, quite promising for the de-tritiation of drainage from a nuclear fuel reprocessing plant and for the volume reduction of tritiated water from the cooling and safety systems of a nuclear fusion reactor. In ITER, a combination of WD and vapor phase catalytic exchange columns is applied for the separation process of hydrogen isotopes.

Gas-phase TD column is a convenient way of separation on a small scale hydrogen isotope separation. In view of its simplic-

ity of apparatus, the small inventory inherent in gas phase operation and a relatively large separation factor, especially when enhanced by "cryogenic-wall" (CW) [1], [2], the method is applicable to tritium handling systems, such as the tritium purification process in fusion nuclear fuel cycle.

The purpose of the present paper is to summarize some recent developments of the separative analyses on WD and TD.

2 Water Distillation

A packed column will be used in a practical plant of WD for tritium isotope separation. Separative performances of the packed distillation column are affected by many parameters such as (1) structure and dimension of the column, (2) pressure and temperature within the column, (3) flow rates of vapor and liquid, (4) physical properties of vapor and liquid, (5) dimension and material of packings, and (6) packing condition. The dependences on these parameters, however, have not sufficiently been studied for separative performances of the column.

In order to estimate effects of the parameters above, we have developed "Channeling Stage Model". This model requires "Va-

por/Liquid Passing Coefficients". They are defined as the rates for vapor and liquid not taking part in the reaction of vapor-liquid exchange at virtual stages, which are set in the column for calculation.

2.1 Vapor/Liquid Passing Coefficients Estimated Through Concentration Profiles in a Vertical Circular Tube

Complex fluid paths in the packed distillation column are simplified an assembly of many short vertical circular tubes. "Vapor/Liquid Passing Coefficients" are estimated through concentration profile of HTO in a circular tube obtained by numerical calculation for HTO-H₂O binary system. In this section, the symbols *A* and *B* refer to HTO and H₂O, respectively.

Figure 1 shows a longitudinal section of a circular fluid path. Liquid flows down along the inner wall of the tube forming an uniform thin film and contacts its vapor countercurrently. Each component is transferred through vapor-liquid interface by vapor-liquid exchange reaction and then diffuses from the interface to the both phases owing to concentration gradient.

The equation of continuity which describes HTO mass transfer is

$$0 = -\nabla \cdot \{\rho_A \mathbf{v} + \mathbf{j}_A^{(x)}\} + r_A, \quad (1)$$

where ρ_A is a mass density of *A*, \mathbf{v} is a mass average velocity, $\mathbf{j}_A^{(x)}$ is a mass flux of *A* due to concentration gradient, and r_A is the production rate of *A* which describes HTO transfer through the vapor-liquid interface.

A *r*-directional distribution of v_z is obtained by analytical calculation under the following assumptions. (a) The flow field is independent of time and axisymmetric (so that the cylindrical coordinates are used). (b) The entrance and exit disturbances are ignored. (c) The fluids are Newtonian and

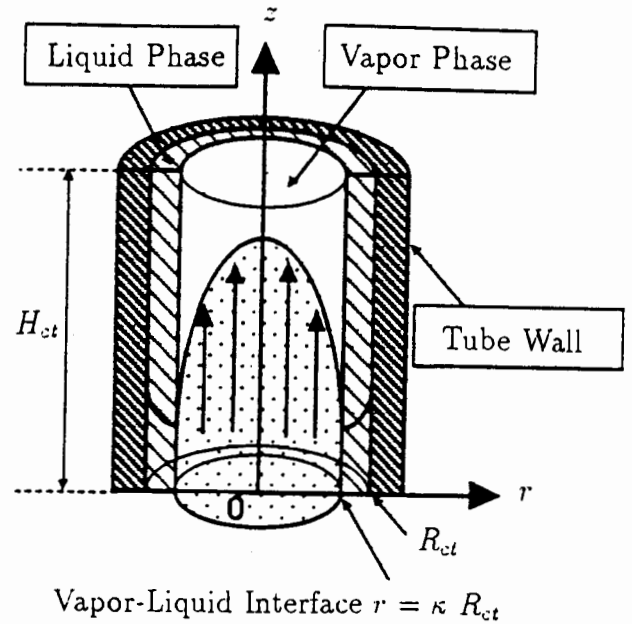


Figure 1: Longitudinal section of a circular fluid path.

incompressible. (d) The flow is laminar with straight streamlines parallel to *z*-axis. (e) There is no slip at vapor-liquid and liquid-solid interfaces. (f) Vapor and liquid flow rates are equal to each other.

The mass flux $\mathbf{j}_A^{(x)}$ is expressed as

$$\mathbf{j}_A^{(x)} = -\left(\frac{c^2}{\rho}\right) M_A M_B D_{AB} \nabla x_A, \quad (2)$$

where *c* is a molar density of the mixture, ρ is a mass density of the mixture, *M* is a molecular weight, D_{AB} is a diffusion coefficient of the binary system, and x_A is a mole fraction of *A*. The source term r_A can be written as

$$r_A = M_A k_r \Delta S \left\{ \frac{1}{\alpha_0} \frac{[\text{H}_2\text{O}]_v}{[\text{H}_2\text{O}]_l} [\text{HTO}]_l - [\text{HTO}]_v \right\}, \quad (3)$$

where k_r is a reaction rate constant, ΔS is a contacting area of vapor and liquid, α_0 is an equilibrium separation factor, and subscripts *v*, *l* indicate vapor and liquid, respectively.

HTO concentration profile in the circular tube is solved by use of the Newton iterative solution of the finite difference expression of Eq.(1) with HTO mole fraction in the vapor phase y_A and in the liquid phase x_A as variables. This calculation is performed with

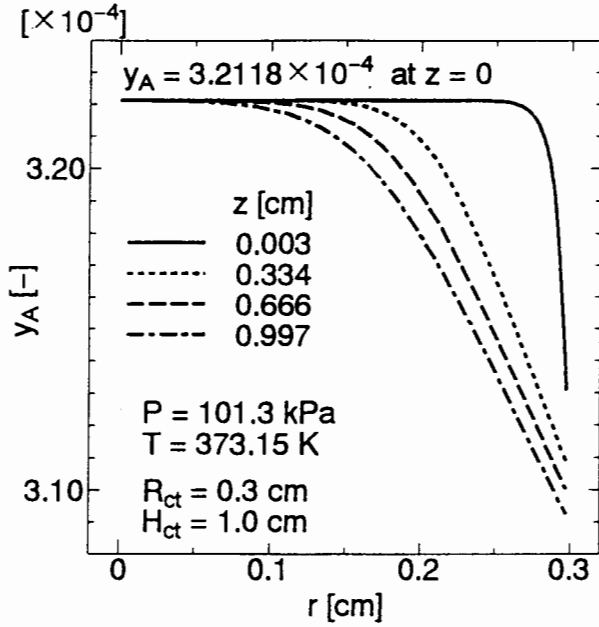


Figure 2: HTO mole fraction profile in the vapor phase.

boundary conditions $y_A = \bar{x}_A$ at $z = 0$ and $x_A = \bar{y}_A$ at $z = H$, where overlines indicate the value averaged for r -direction. Figure 2 shows an example of calculational results of HTO mole fraction profile for the vapor phase.

The rate of HTO transfer through the interface is assumed to be proportional to the volum of fluids which take part in the vapor-liquid exchange reaction. The vapor passing coefficient is, then, expressed as

$$e_V = 1 - \frac{v_A^{in} - v_A^{out}}{v_A^{in} - v_A^{eq}}, \quad (4)$$

$$v_A^{in} = \frac{\rho}{M} y_A^{in} \cdot 2\pi \int_0^{\kappa R} r \cdot v_z(r) dr, \quad (5)$$

$$v_A^{out} = 2\pi \int_0^{\kappa R} r \cdot v_z(r) \frac{\rho}{M} y_A^{out}(r) dr, \quad (6)$$

$$v_A^{eq} = \frac{\rho}{M} y_A^{eq} \cdot 2\pi \int_0^{\kappa R} r \cdot v_z(r) dr, \quad (7)$$

where v_A is a mole flow rate of A , and superscripts *in*, *out*, *eq* indicate inflow, outflow and equilibrium state, respectively.

Liquid passing coefficient e_L is also calculated by the same manner as e_V . It becomes equal to e_V under the assumptions in this study.

Figure 3 shows vapor/liquid passing coefficients calculated for 1.0 cm-height tube with

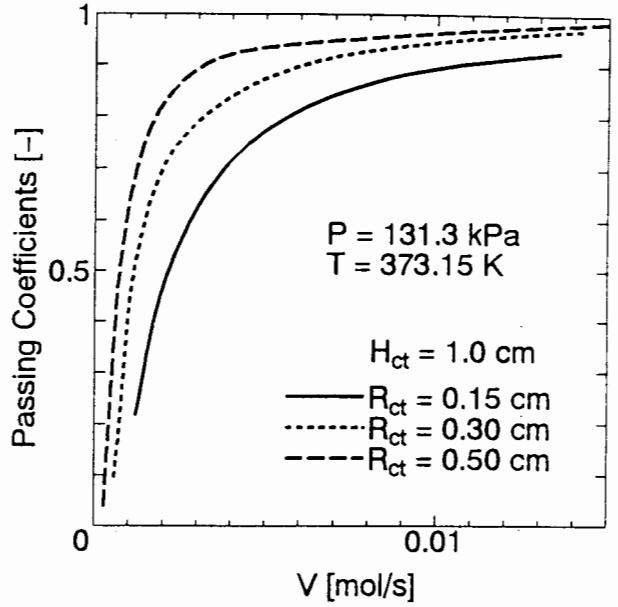


Figure 3: Vapor/liquid passing coefficients.

various radii ($R_{ct} = 0.15, 0.3, 0.5$ cm). As in Fig.3, passing coefficients increase with vapor flow rate V and approach unity asymptotically. It is also shown in Fig.3 that passing coefficients increase with the radius R_{ct} .

2.2 Separative Analyses of Packed Distillation Column by "Channeling Stage Model"

Separative analyses of a 1.6 cm-inner diameter, 100 cm-height packed column are performed by the "Channeling Stage Model" for H_2O -HTO binary distillation at total reflux conditions. The results in the previous section are used for the "Vapor/Liquid Passing Coefficients". The height adopted for a channeling stage in the present study is 1 cm and the total number of channeling stages is 100.

Table 1 summarizes the basic equations of the model which are conservations of material and heat, and phase equilibrium relations. A HTO mole fraction profile in the column is obtained by solving simultaneously all the nonlinear equations in Table 1, with the LU factorization technique and a successive iteration procedure.

A total separation factor α_T of the column

Table 1: Basic equations of the "Channeling Stage Model"

 Total mass balance equation of the j -th stage

$$(1 - e_j^V) \sum_{k=1}^{N-j} \prod_{n=1}^{k-1} e_{j+n}^V V_{j+n} + (1 - e_j^L) \sum_{k=1}^{j-1} \prod_{m=1}^{k-1} e_{j-m}^L L_{j-m} - (V_j + L_j) = 0 \quad (8)$$

 The i -th component mass balance equation of the j -th stage

$$(1 - e_j^V) \sum_{k=1}^{N-j} \prod_{n=1}^{k-1} e_{j+n}^V v_{i,j+n} + (1 - e_j^L) \sum_{k=1}^{j-1} \prod_{m=1}^{k-1} e_{j-m}^L l_{i,j-m} - (v_{i,j} + l_{i,j}) = 0 \quad (9)$$

 Heat balance equation of the j -th stage

$$(1 - e_j^V) \sum_{k=1}^{N-j} \prod_{n=1}^{k-1} e_{j+n}^V V_{j+n} H_{j+n} + (1 - e_j^L) \sum_{k=1}^{j-1} \prod_{m=1}^{k-1} e_{j-m}^L L_{j-m} h_{j-m} - (V_j H_j + L_j h_j) = 0 \quad (10)$$

 Stoichiometric equation of the j -th stage

 Equilibrium equation of the i -th component j -th stage

$$\sum_{i=1}^6 K_{i,j}(T_j) x_{i,j} = 1 \quad (11) \quad y_{i,j} = K_{i,j} x_{i,j} \quad (12)$$

is defined by liquid HTO mole fraction at the bottom and vapor HTO mole fraction at the top as

$$\alpha_T \equiv \frac{x_{\text{bottom}}/(1 - x_{\text{bottom}})}{y_{\text{top}}/(1 - y_{\text{top}})} \quad (13)$$

Because an equilibrium separation factor α_0 of water distillation is expressed in terms of the vapor pressure p of H_2O and T_2O by

$$\alpha_0 = (p_{\text{H}_2\text{O}}/p_{\text{T}_2\text{O}})^{1/2}, \quad (14)$$

the HETP value and the number of theoretical plates n are calculated in the following manner :

$$n = (\ln \alpha_T / \ln \alpha_0) - 1, \quad (15)$$

$$\text{HETP} = H_p / n, \quad (16)$$

where H_p is a height of packed section of the column. Vapor pressure ratio in Eq.(14) obtained by Jones [3] is expressed in terms of temperature T [K] :

$$(p_{\text{H}_2\text{O}}/p_{\text{T}_2\text{O}}) = \exp \left(-\frac{103.87}{T} + \frac{46.480}{T^2} \right). \quad (17)$$

The equilibrium separation factor at 100 °C is 1.0281.

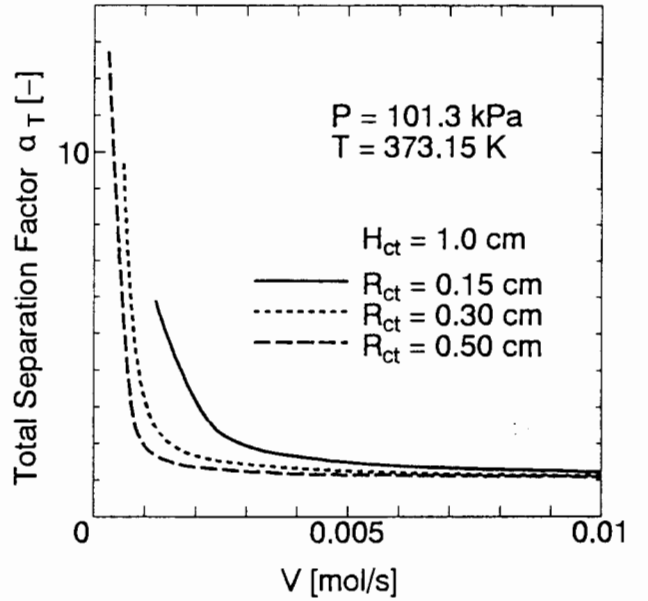


Figure 4: Total separation factor of the 1.6 cm-inner diameter, 100 cm-long packed column at total reflux conditions.

Figure 4 shows total separation factor of the column calculated for various radii of the circular tube ($R_{ct} = 0.15, 0.3, 0.5$ cm). As in Fig.4, total separation factor decreases with increasing vapor flow rate and approaches unity asymptotically. Figure 5 shows the

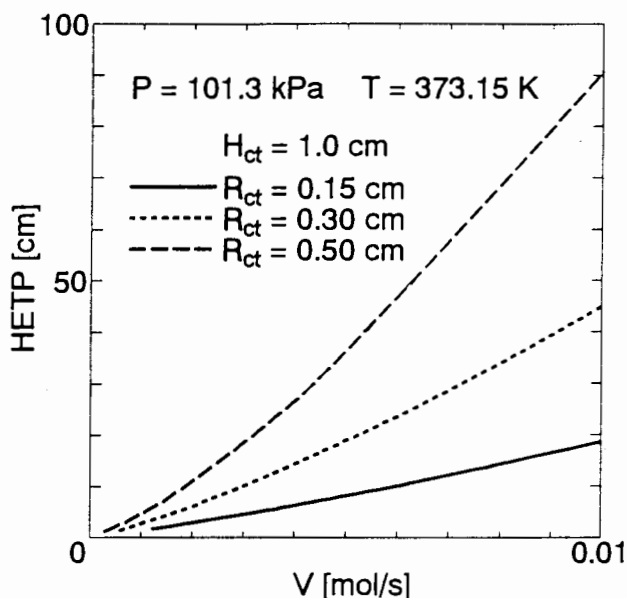


Figure 5: HETP values of the 1.6 cm-inner diameter, 100 cm-long packed column at total reflux conditions.

HETP values of the column by Eqs.(15), (16). As in Fig.5, the HETP values increase with vapor flow rate. It is also shown in Fig.5 that the HETP value increases with the radius of the circular tube R_{ct} .

3 Cryogenic-Wall Thermal Diffusion

We developed a rigorous 2-dimensional 2-component separative analysis [4], [5], [6], for two cases [a] a mixture of isotopes with large masses and resultant relatively-small mass difference, or [b] for a mixture, one of which constituents is predominant even when others are enriched. However, this analysis of the CW-TD assumes that isotope separation causes negligible changes in the gas properties such as the density: the convection flows are first calculated on the basis of thermal convection of a homogeneous gas, and then the separation effects are deduced from the flow, temperature and density distribution. This approach cannot deal with hydrogen isotope separation because the den-

sity changes due to composition differences are no longer negligible compared to those due to thermal expansion. Hence, we have been developing a system of separative analysis which can meet separation of multi-component hydrogen isotope molecules (H_2 , HD, D_2 , HT, DT, T_2) in the CW-TD column.

3.1 Simultaneous Solution of Flow and Concentration Fields for 2-Component Isotope Separation

The simultaneous equations [7] describing the flow field and concentration profiles were solved [8] simultaneously using the Newton iterative solution of finite difference expression of equations of change with 5 variables — density ρ , mole fraction of the 1-st component x , radial velocity u , axial velocity v and temperature T —. This numerical procedure enables us to analyze separation phenomena where the degree of separation is so large that values of physical properties (viscosity, thermal conductivity etc.) of the mixture depend on the location within the column.

Because of the dependence of thermal conductivity k_{mix} , temperature of surface of hot-wire is not constant. Hence, boundary condition on the hot-wire is not a constant temperature but a constant radial heat flux q_r .

$$q_r = -k_{mix} \left. \frac{\partial T}{\partial r} \right|_{\text{hot-wire}} = \text{Const.} \quad (18)$$

Basic equation for concentration profile analysis are the convection-diffusion equation [7], which require thermal diffusion factor $\alpha_{ij}^{(2)}$. Monchick *et al.* have derived an expression [9] of $\alpha_{ij}^{(2)}$ (MSM formula) for inelastic molecules such as hydrogen molecules. The MSM formula requires experimental values such as rotational relaxation times, however experimental values of the properties for hydrogen isotopes are not available. Hence, the MSM formula was approximated [10] so

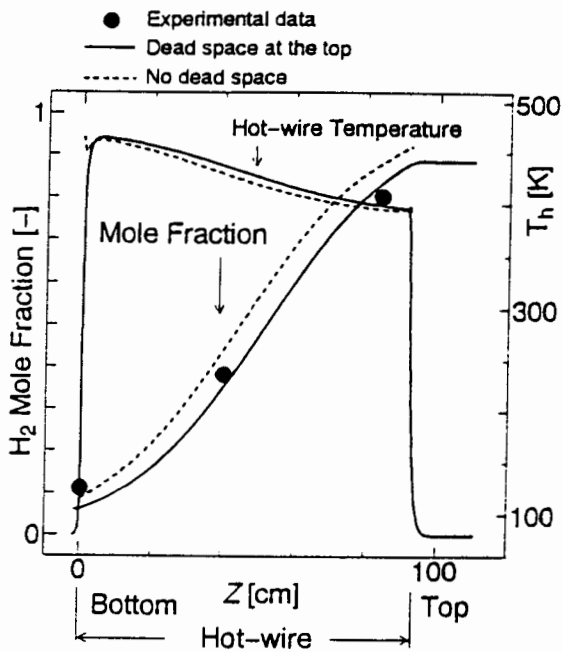


Figure 6: Profiles of hot-wire temperature and comparison of mole fraction between analytical and experimental data ($q_r = 4.8645 \text{ W/cm}^2$)

as to be “roughly estimated” even when input data for the formula are not available. Application of the MSM formula could have reduced discrepancies between experimental and analytical results of separation of TD column for $\text{H}_2\text{-HT}$ (tracer level) [11] although the estimates of the formula and the inelastic collision integral ratios were rough.

For convenience of calculation, the temperature dependences of the rough estimate of thermal diffusion factors have been fitted [12] to exponential and inverse-power functions for all pairs of $\alpha_{ij}^{x_i \approx 0.0}$ and $\alpha_{ij}^{x_j \approx 0.0}$. Fitting of $\alpha_{ij}^{x_j \approx 0.0}$ between H_2 and D_2 (tracer), for example,

$$\begin{aligned} \alpha_{ij}^{x_j \approx 0.0} = & 0.90145 \times 10^{-1} \\ & + 0.9626 \times 10^{-5} T - 0.6051 \times 10^{-8} T^2 \\ & - \exp(-0.7713 - 0.2862 \times 10^{-1} T \\ & + 0.7988 \times 10^{-4} T^2 - 1.5999 \times 10^{-7} T^3) \end{aligned} \quad (19)$$

Moreover, composition dependences of thermal diffusion factor have been approximated to

$$\alpha_{ij}^{(2)} = x_i \alpha_{ij}^{x_j \approx 0.0} + x_j \alpha_{ij}^{x_i \approx 0.0}. \quad (20)$$

Concentration distributions of H_2 and D_2

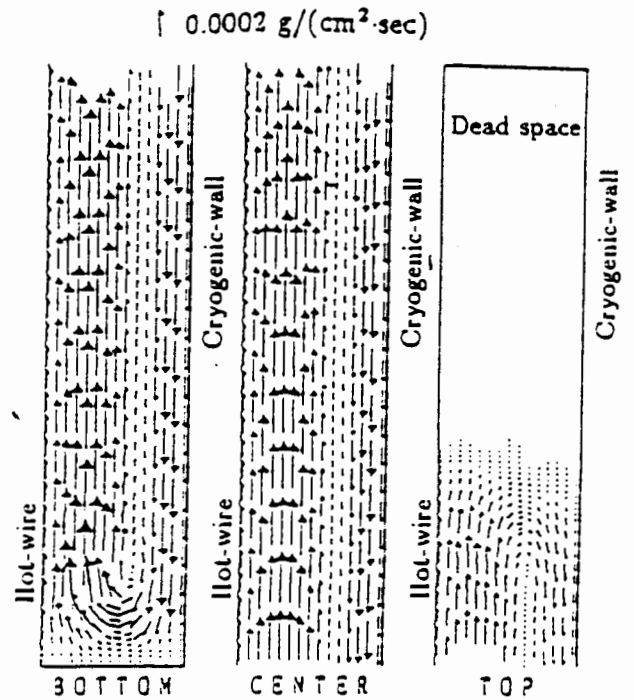


Figure 7: Mass flow vectors in top, center and bottom of TD column with dead space at the top

in the CW-TD column with a hot-wire of low temperature, such that isotopic exchange does not occur, were analyzed and compared [13] with experimental data [14]. Axial profiles of hot-wire temperature and mole fraction are shown in Fig.6, and mass flow vectors in Fig.7.

3.2 3-Component Concentration Profile Analysis

Explicit Approximation of Thermal Diffusion Factor for Isotopic 3-Component Mixture Three component concentration profiles are obtained through solution of 3-component diffusion equations, which require diffusion coefficients (D_{ij}) and thermal diffusion coefficients (D_i^T) for a 3-component mixture. In a binary mixture, the diffusion coefficient is given experimentally and theoretically and the thermal diffusion coefficient is obtained from the binary diffusion coefficient and the binary thermal diffusion factor ($\alpha_{ij}^{(2)}$) [15], [16]:

$$D_i^T = -\frac{c^2}{\rho} M_i M_j D_{ij}^{(2)} x_i x_j \alpha_{ji}^{(2)}. \quad (21)$$

In the case where molecules collide elastically, the thermal diffusion factor for a bi-

nary isotopic mixture was obtained by Chapman and Cowling (CC formula) [15]

$$\alpha_{ij}^{(2)} = \frac{M_i - M_j}{M_i + M_j} \frac{15(2A^* + 5)(6C^* - 5)}{2A^*(16A^* - 12B^* + 55)} \quad (22)$$

where A^* , B^* , C^* are collision integral ratios which are given as functions of temperature.

In a 3-component mixture, an explicit expression [16] of diffusion coefficient ($D_{ij}^{(3)}$) is given:

$$D_{ij}^{(3)} = D_{ij}^{(2)} \left[1 + \frac{x_k \left((M_k/M_j) D_{ik}^{(2)} - D_{ij}^{(2)} \right)}{x_i D_{jk}^{(2)} + x_j D_{ik}^{(2)} + x_k D_{ij}^{(2)}} \right] \quad (23)$$

Rigorously, D_i^T s in a ν -component mixture are solutions of a set of 2ν -simultaneous equations whose coefficients are given in terms of binary diffusion coefficients and collision integral ratios [16]. These equations are complicated to deal with, and no explicit expression in terms of the binary thermal diffusion coefficients has been yet demonstrated. Being equivalent to Eq.(21) in binary mixture, $D_i^{T(\nu)}$ can be expressed in terms of diffusion coefficients and thermal diffusion factors in ν -component mixture. [17]

$$D_i^T = -\frac{c^2}{\rho} M_i \sum_{j \neq i} M_j D_{ij}^{(\nu)} \sum_{k \neq j} \alpha_{jk}^{(\nu)} x_j x_k \quad (24)$$

Thermal diffusion factor $\alpha_{Tjk}^{(\nu)}$ is obtained from a set of equations in terms of the matrix of thermal diffusion coefficients [17]. The isotope approximation of the 3-component thermal diffusion factor ($\alpha_{ij}^{(3)}(I.A.)$) was proved to be equal to the approximation of binary thermal diffusion factor ($\alpha_{ij}^{(2)}(I.A.)$) [18]. that is,

$$\begin{aligned} \alpha_{ij}^{(3)}(I.A.) &\approx \frac{M_i - M_j}{M_i + M_j} \frac{15(2A^* + 5)(6C^* - 5)}{2A^*(16A^* - 12B^* + 55)} \\ &= \alpha_{ij}^{(2)}(I.A.) \end{aligned} \quad (25)$$

From Eqs.(24) and (25), we can obtain thermal diffusion coefficients in 3-component mixture.

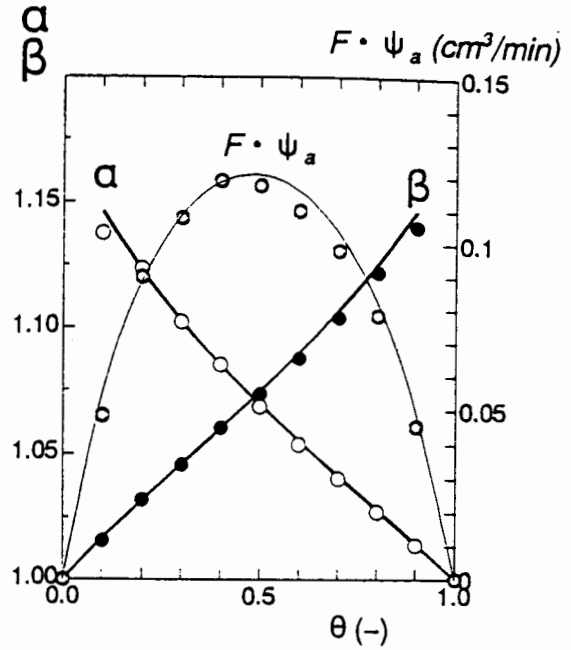


Figure 8: Analytical and experimental data of separation factor and separative power of ^{36}Ar ($F = 50 \text{ cm}^3/\text{min}$). $\varphi_a \equiv [\beta(\alpha - 1) \ln \beta - (\beta - 1) \ln \alpha]/(\alpha\beta - 1)$.

Although there is a formal expression in matrix form for the multi-component thermal diffusion coefficient of molecules suffering inelastic collision [19], almost all elements of the matrix cannot be evaluated without knowledge of the inelastic collision. Hence, we assumed that the thermal diffusion factor for multi-component inelastic molecules can be defined with Eq.(24) and those $\alpha_{ij}^{(\nu)}$ s are approximately equal to $\alpha_{ij}^{(2)}$ s. In order to apply Eq.(20) to a multi-component mixture, Eq.(20) is modified to [18].

$$\alpha_{ij}^{(\nu)} = \frac{x_i}{x_i + x_j} \alpha_{ij}^{x_j \approx 0.0} + \frac{x_j}{x_i + x_j} \alpha_{ij}^{x_i \approx 0.0} \quad (26)$$

3-Component Concentration Profile Analysis for Separation of Argon Isotopes As the second step, 3-component concentration distributions were analyzed (— *not simultaneously*—) with flow vectors, temperature and density profiles as inputs [20]. Analytical results were compared with experimental data of 3-component natural abundance argon 36-38-40 isotopes separated in a 950 mm-height, inner 15 mm ϕ tube column with a 0.3 mm ϕ hot wire in

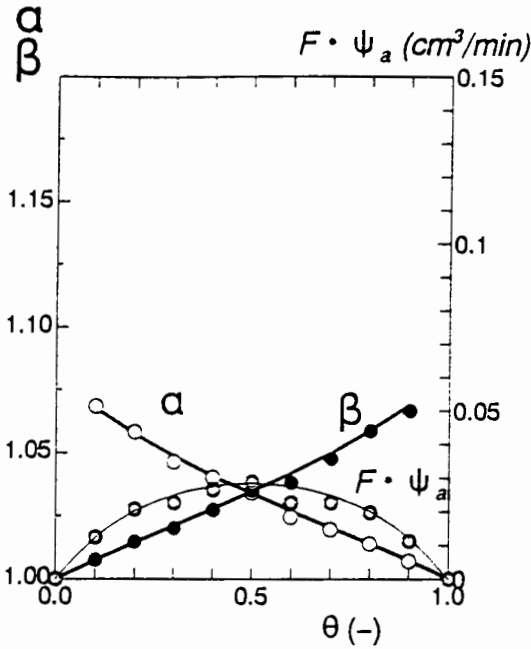


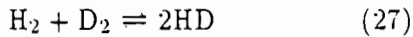
Figure 9: Analytical and experimental data of separation factor and separative power of ^{38}Ar ($F = 50 \text{ cm}^3/\text{min}$)

the axis. This analytical system could explain experimental data (Fig.8~Fig.9) quantitatively [20].

3.3 Simulations Solution of Flow and Concentration Fields for 3-component Hydrogen Isotope Separation

Three-component isotope separation has been analyzed by a newly developed code which solves simultaneously a set of equations governing flow fields and concentration profiles. This code incorporates the isotope exchange reaction [21] on the hot-wire. Thus, we can analyze 3-component hydrogen isotope separation in the case where the degree of separation is very large and that the third component is generated on the surface of the hot-wire.

When the feed gas is composed of H_2 and D_2 , HD is generated by the isotope exchange reaction



$$K(T) \equiv \frac{k_f}{k_r} = \frac{[\text{HD}]^2}{[\text{H}_2][\text{D}_2]} \quad (28)$$

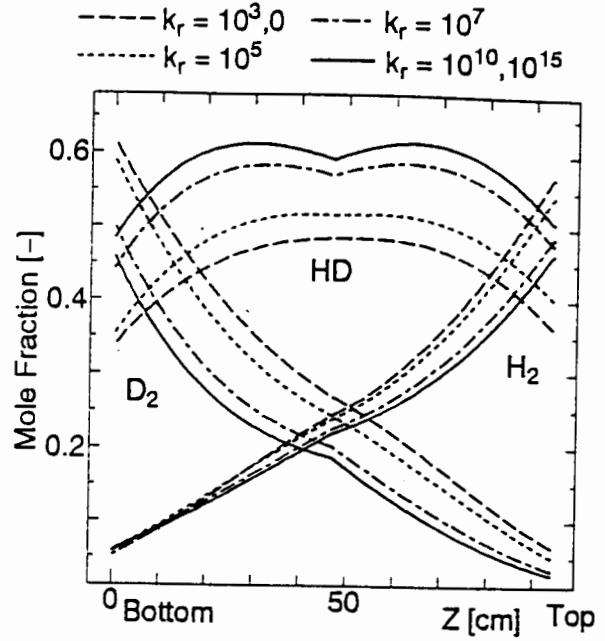


Figure 10: Effect of k_r on axial concentration profiles in TD column

where $K(T)$ is equilibrium constant, k_f is the forward reaction rate and k_r is the backward rate. When the hot-wire surface functions as a catalyst, the rates of formation and disappearance of HD, H_2 and D_2 are expressed as

$$\frac{d[\text{HD}]}{dt} = 2k_r \{ K(T)[\text{H}_2][\text{D}_2] - 2[\text{HD}]^2 \} \Delta S \quad (29)$$

$$\frac{d[\text{H}_2]}{dt} = -k_r \{ K(T)[\text{H}_2][\text{D}_2] - [\text{HD}]^2 \} \Delta S \quad (30)$$

$$\frac{d[\text{D}_2]}{dt} = -k_r \{ K(T)[\text{H}_2][\text{D}_2] - [\text{HD}]^2 \} \Delta S \quad (31)$$

where ΔS is the surface area of the hot-wire. A temperature dependence of $K(T)$ is available but the values of k_r and k_f are unknown, so the value of k_r is varied from 10^0 to $10^{15} \text{ cm}^3/\text{mol}\cdot\text{s}$ parametrically (Fig.10) [21].

The analytical results explain well the experimental data of H_2 -HD- D_2 separation (Fig.11) [22].

3.4 Simulations Solution of Flow and Concentration Fields for Higher Component Hydrogen Isotope Separation

For separative analysis of ν -component isotope mixture, $D_{ij}^{(\nu)}$ is required. In

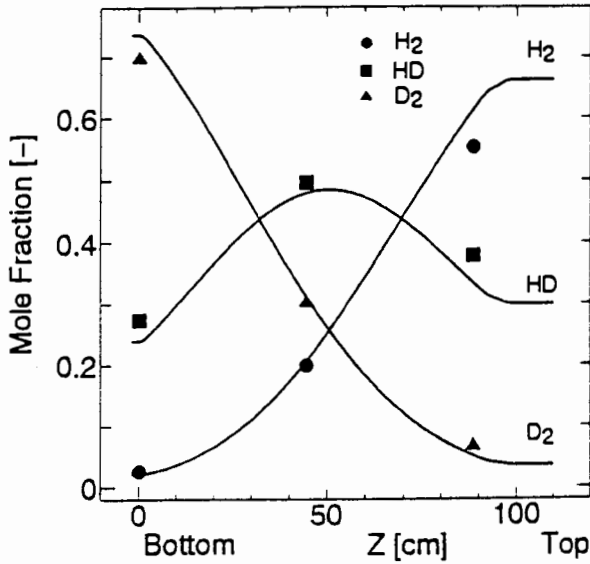


Figure 11: Analytical and Experimental data of axial concentration profiles in TD column ($k_r = 10^7$ cm/mol-s)

a ν -component mixture ($\nu \geq 3$), $D_{ij}^{(\nu)}$ in ν -component mixture is given by Hirschfelder *et al.* in a form [16] of a ratio of determinants whose elements are in terms of the binary diffusion coefficients ($D_{ij}^{(2)}$). The explicit expression [16] of $D_{ij}^{(3)}$ in Eq.(23) is given only in a 3-component mixture.

Thereupon, diffusion coefficients in 4-component mixture $D_{ij}^{(4)}$ were expressed [24] explicitly in terms of binary diffusion coefficients and mole fractions by solving the ratio of determinants. Explicit expressions of $D_{ij}^{(4)}$ were divided into two terms:

$$D_{12}^{(4)} = D_{12} \left[1 - \frac{A_{12}^{(4)}}{B^{(4)}} \right], \quad (32)$$

where $A_{12}^{(4)}$ is a term due to the i - j pairs of attention and $B^{(4)}$ a term common to all the pairs out of the 4 components. So that the form of the explicit expression had extended structures similar to corresponding those of $D_{ij}^{(3)}$ respectively, the terms are obtained in terms of binary diffusion coefficients. In addition, by similar procedure, the explicit expressions [25], [26] of $D_{ij}^{(5)}$ and $D_{ij}^{(6)}$ were obtained.

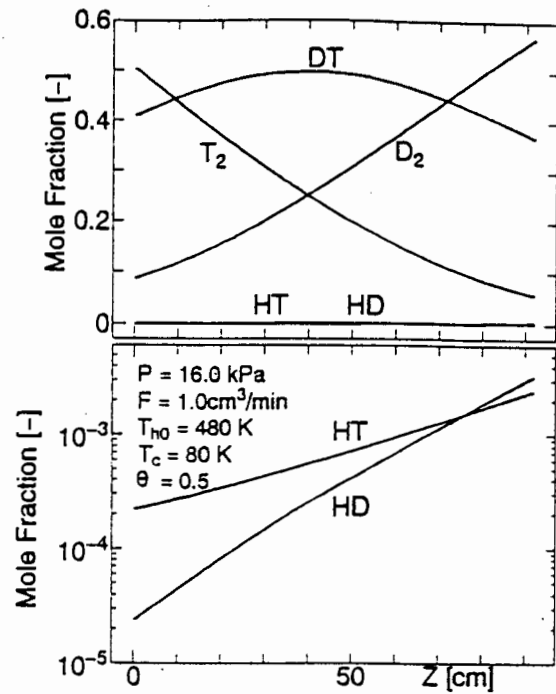


Figure 12: Axial distributions of profiles of mole fractions of 5-component hydrogen isotope mixture in CW-TD

With the expression of $D_{ij}^{(5)}$, separative performances [27] of CW-TD for 5-component hydrogen isotope (HD, D₂, HT, DT, T₂) mixture were analyzed by solving simultaneous equations governing flow field and concentration profiles. In the analysis, abundances of HD and HT were assumed to be tracer level so that generation of H₂ caused by isotope exchange reaction were negligible. An example of analytical results is shown in Fig.12. This code is expected to enable us to design CW-TD columns for tritium recovery system.

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