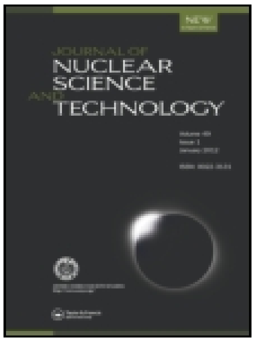

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Possibility of Remarkable Enhancement of Separation Factor by "Cryogenic-Wall" Thermal Diffusion Column

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From a view point that the maximum separation factor $(\alpha\beta)_{\max}$ attainable in total reflux operation of thermal diffusion column depends on temperature difference ΔT between hot T_h and cold T_c surfaces in the form of $\Delta T/T_c$ as

$$(\alpha\beta)_{\max}=\exp\left[\frac{\Delta T}{T_c}\cdot\frac{3\sqrt{6}\,(2+\ln\delta)\,\alpha_T Z^*}{2\ln\delta\sqrt{15(\ln\delta)^2+56\ln\delta+53}}\Big/\left\{1-\frac{\Delta T}{T_c}\cdot\frac{4+3\ln\delta}{4\ln\delta(1+\ln\delta)}\right\}\right],$$

where δ $(\equiv r_h/r_c)$ is the ratio of hot wire to cold-wall radii, α_T the thermal diffusion factor and Z^* $(\equiv Z/r_c)$ the normalized column height, remarkable enhancement of separation factor is expected through an adoption of "Cryogenic-Wall" (*ex.* 77.35 K) instead of "Ordinary Cold-wall" (*ex.* T_c=288.15 K). The effect of "Cryogenic-Wall" was analyzed on the basis of approximate formulae involving explicitly design parameters for the column constants. In the case where r_c=1.5 cm, Z=150 cm and ΔT=1,000 K, $(\alpha\beta)_{\max}$ for H_2-HT through "Ordinary Cold-wall" TD column is 63.4, while that through "Cryogenic-Wall" TD column is 885, for which ΔT= 2,430 K is required in the "Ordinary Cold-wall" TD column and it is impossible for the hot-wire to be at so high temperature because of its melting. The magnitude of enhancement is, however, smaller than that expected from the ratio of $\Delta T/T_c$, because (1) the value of α_T decreases in lower temperature region, and (2) the argument of the exponential function for $(\alpha\beta)_{\max}$ is not proportional directly to $\alpha_T\cdot\Delta T/T_c$ but to $\alpha_T\cdot\Delta T/\bar{T}$, where $\bar{T}$ is the reference mean temperature higher than T_c. The optimum pressure of "Cryogenic-Wall" TD column is considerably smaller (0.024 MPa for the case above) than that of "Ordinary Cold-wall" TD column (0.103 MPa).

KEYWORDS: ***thermal diffusion column, separation factor, cryogenic wall, temperature difference, temperature dependence, optimum pressure, thermal diffusion factor, tritium, nuclear fusion fuel cycle, tritium purification***

I. INTRODUCTION

Thermal diffusion(1)~(4) is not suitable for large scale production because of low thermodynamic efficiency. Gas phase thermal diffusion of hydrogen is, however, a convenient method for the separation of the isotopes of hydrogen on a small scale to moderate scale(5), because the process has advantages such as small tritium inventory, high separation factors in apparatus of moderate size and a relatively simple system. The process, therefore, attracts attention to purification of tritium in production system and recovery of tritium from used gas mixture of hydrogen isotopes in fusion fuel cycle. At the Tritium Process Laboratory (TPL) in the Japan Atomic Energy Research Institute, the thermal diffusion process is applied for recovery and enrichment of tritium, which is diluted and mixed with other hydrogen isotopes in the other experiments(6).

In previous papers(7)~(9), we have reported results for H_2-HT isotope separation; (1) an

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experimental apparatus[10] including flow and/or cut control, temperature regulation and measurement of tritium activity and results of stable separation with continuous feed and draw-offs[7], (2) rough estimate of thermal diffusion factor for H_2-HT isotopic mixture[8] through comparison of experimental results with analytical results based on a Newton iterative numerical solution of two-dimensional flow equations[11][12] as well as convection-diffusion equation[13], and (3) pressure dependences of separative characteristics[9].

Thus, the validity of the two-dimensional separative analysis is proved, but computational time required for the analysis is so large that the analysis is not suitable for parametrical survey for conceptual design. An approximate one-dimensional analysis[2]~[4] has usually been used for such a problem, but the analysis is not so simple and requires inevitably numerical calculations with computers. Recently, we have, therefore, derived approximate column constants[14] involving explicitly design parameters of hot-wire thermal diffusion column through further approximating traditional analysis at the sacrifice of minute accuracy. We have pursued a simpler analysis that is easy to operate analytically as the second step of the study[15] for obtaining a "rule of thumb" allowing an immediate estimate of an optimal dimension and/or operational condition of the column in terms of physical properties of gases to be separated. As the first application, optimum operating pressure at total reflux[16] was obtained in addition to the maximum separation factor attainable at the optimum pressure. Observation of the expression of maximum separation factor leads to a possibility of remarkable enhancement of separation factor in the case where the temperature of the cold wall is very low. It is because the temperature difference ΔT between hot T_h and cold T_c surfaces affects separation factor in the form of $\Delta T/T_c$. The present concept of "Cryogenic-Wall" thermal diffusion column is the first fruitful finding from the simple analysis with the approximate column constants.

II. Optimum Pressure and Maximum Separation Factor

Optimum pressure P_{opt} in the sense that the pressure maximize the total separation factor is expressed in the form:

$$P_{\mathrm{opt}}^2=384\sqrt{6}\,\frac{\mu D'R}{r_c^3 Mg}\cdot\frac{T_c^2}{\Delta T}\cdot\left[1-\frac{\Delta T}{T_c}\cdot\frac{4+3\ln\delta}{4\ln\delta(1+\ln\delta)}\right]^2\cdot\sqrt{\frac{(\ln\delta)^2(1+\ln\delta)^2}{15(\ln\delta)^2+56\ln\delta+53}}, \tag{1}$$

where
- μ: Viscosity (kg/m·s)
- D': Product of diffusion coefficient at standard pressure by standard pressure (m²·Pa/s)
- R: Gas constant, or 8.314×10^3 (kg·m²/s²·kg-mole·K)
- M: Molecular weight (kg/kg-mole)
- g: Gravitational acceleration, or 9.80665 (m/s²)
- δ: Ratio of hot wire radius, r_h, to cold wall radius r_c.

Physical properties are estimated at the reference mean temperature[15]:

$$\bar{T}=T_c\left[1-\frac{\Delta T}{T_c}\cdot\frac{4+3\ln\delta}{4\ln\delta(1+\ln\delta)}\right]. \tag{2}$$

Moreover, the maximum total separation factor attainable in the total reflux operation in the form:

$$(\alpha\beta)_{P_{\mathrm{opt}}}^{\max}=\exp\left[\frac{\Delta T}{T_c}\cdot\frac{3\sqrt{6}(2+\ln\delta)\alpha_T Z^*}{2\ln\delta\sqrt{15(\ln\delta)^2+56\ln\delta+53}}\Big/\left\{1-\frac{\Delta T}{T_c}\cdot\frac{4+3\ln\delta}{4\ln\delta(1+\ln\delta)}\right\}\right], \tag{3}$$

where
- α_T: Thermal diffusion factor
- Z^*: Normalized column height, *i.e.* Z/r_c.

Note that the temperature difference appears in Eq. (3) only in the form of $\Delta T/T_c$. Equations (1) and (3) are not always applicable to all the range of design parameters, because (1) the equations were obtained for the case[16] where the term $x(1-x)$ (where x: Mole fraction of the species to be separated) is linearized to x

in the transport equation[2], *i.e.* for the case where $x \ll 1$, and (2) the approximate column constants tend to deviate from the traditional column constants in the larger range[14] of $\Delta T/T_c$ probably due to an adoption[14] of constant temperature for easier calculation of the column constants. The accuracy of the column constants has not, however, been checked strictly because the present column constants were obtained by further approximation of the traditional approximate one-dimensional analysis. The role of the approximate column constants is to give insight into extensive survey of effective thermal diffusion column. When high accuracy is required, we will adopt the two-dimensional separative analysis[11]~[13] rather than studying the accuracy of the approximation.

III. Results and Discussion

Computations are performed for H_2-HT (tracer level) hydrogen gas mixture, whose physical properties are summaried as follows:

$$\mu=1.60\times10^{-7}T+4.39\times10^{-5},\quad (\mathrm{g/cm\cdot s}) \qquad (4)$$

$$D_{H_2\cdot HT}=\sqrt{3/4}[1.459\times10^{-5}T^2+7.423\times10^{-4}T],\quad (\mathrm{cm^2/s}) \qquad (5)$$

$$\alpha_T=0.0945-0.16337\cdot\exp(-0.01024T),\quad (T>100\,\mathrm{K}), \qquad (6)$$

where the expression for α_T is an approximate fitting of the rough estimate of temperature dependence of α_T based on the Monchick-Sandler-Mason formula with the loaded sphere molecular model[8], and the expressions for μ and D are fittings of measured data[16].

1. Effect of "Cryogenic-Wall"

Here, we show the effect of "Cryogenic-Wall (for example, liquid nitrogen temperature 77.35 K)" by comparing with "Ordinary Cold-wall (288.15 K)" thermal diffusion (TD) column with the same dimension (*i.e.* the same cold-wall radius (1.5 cm), the same δ (0.01), the same column height (150, 300, 450 cm)) and the same temperature difference ΔT.

Figure 1 shows the optimum pressure which maximize the total separation factor as a function of ΔT. Note that the optimum pressure of "Cryogenic-Wall" TD column is

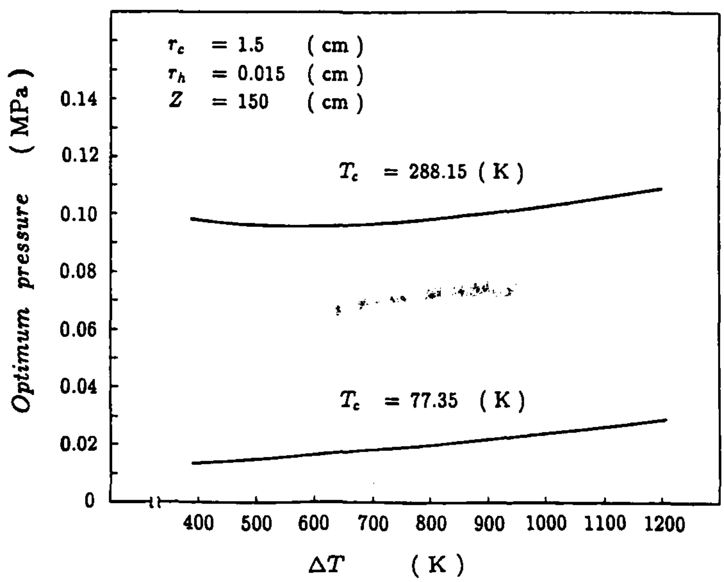

Fig. 1 Comparison of optimum pressure, maximizing total separation factor in total reflux operation, between "Cryogenic-Wall" and "Ordinary Cold-wall" TD columns as a function of temperature difference between hot wire and cold wall

considerably smaller (for example, 0.024 MPa at ΔT=1,000 K) than that of "Ordinary Cold-wall" TD column (0.103 MPa at ΔT=1,000 K). In addition, the optimum pressure is almost constant respectively for each cold-wall temperature and increases gradually in larger ΔT region as ΔT increases.

Figure 2 shows the maximum separation factor $(\alpha\beta)_{max}$ attainable in total reflux operation as functions of ΔT. The maximum separation factors are different by more than one digit between "Cryogenic" and "Ordinary" cold walls. In the case where ΔT=1,000 K, for example, the separation factors of "Cryogenic-Wall" TD column are 8.85×10^2, 7.83×10^5 and 6.93×10^8 respectively for Z=150, 300 and 450 cm, while those of "Ordinary Cold-wall" TD column are 6.34×10^1, 4.02×10^3 and 2.55×10^5. When the limit of ΔT is determined by the hot-wire temperature, we must compare separation factors between "Cryogenic-Wall" column with ΔT=1,000 K and "Ordinary Cold-wall" column with ΔT=1,000−(288.15−77.35)≃800 K, latter of which gives maximum separation factors of 3.43×10^1, 1.18×10^3 and 4.04×10^4 respectively for Z=150, 300 and 450 cm.

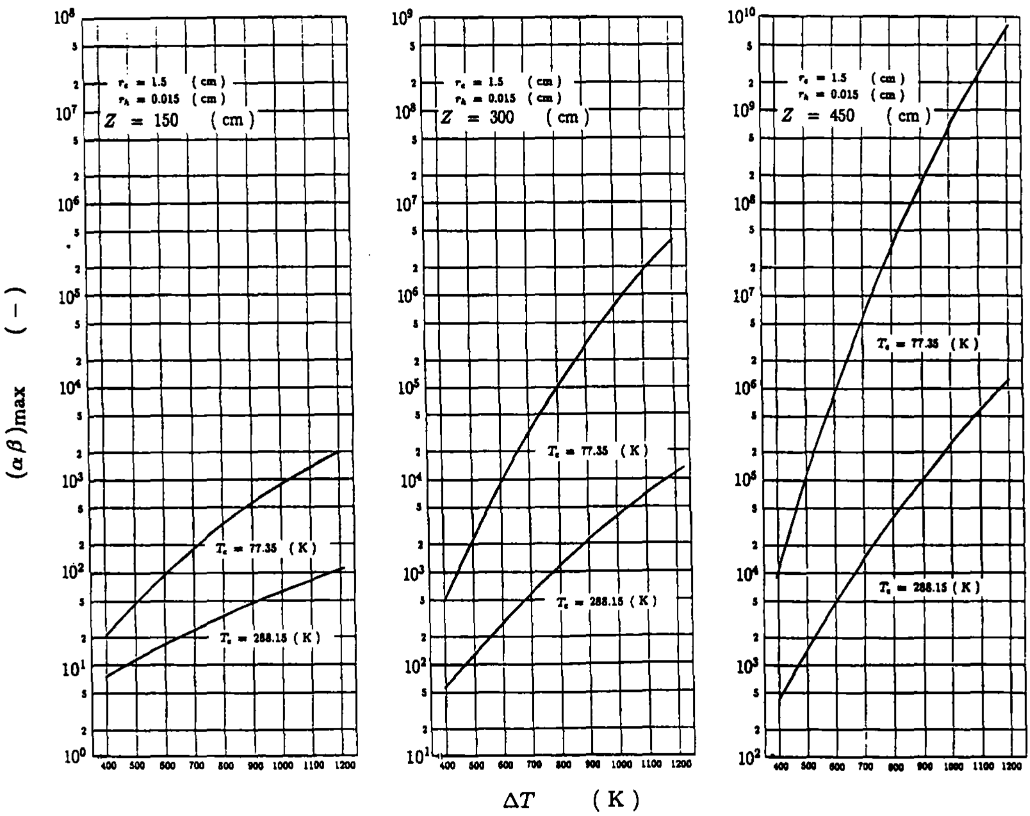

Fig. 2 Comparison of maximum total separation factor attainable in total reflux operation between "Cryogenic-Wall" and "Ordinary Cold-wall" TD columns as a function of temperature difference between hot wire and cold wall

Figure 3 shows the pressure dependence of maximum separation factors in the case of ΔT=1,000 K. The results are obtained by the use of the approximate formulae of the column constants(14). The pressure affects much the maximum separation factors.

2. Points to Examine in "Cryogenic-Wall" TD Column

There are many points to check concerning the "Cryogenic-Wall" TD column. Theoretical

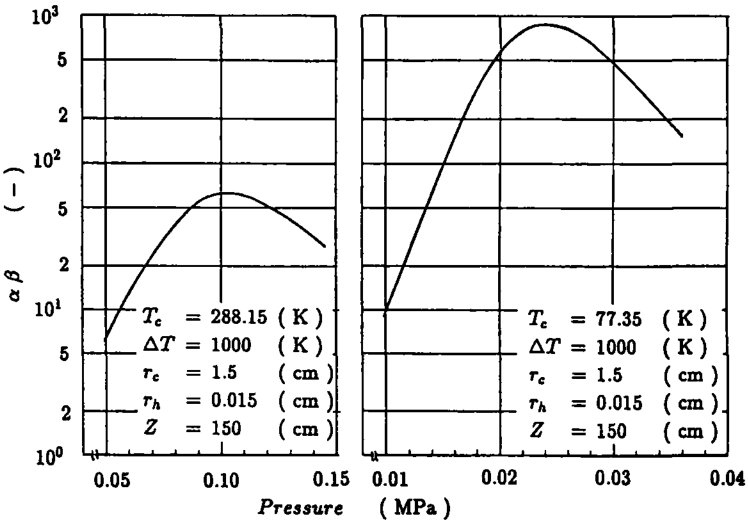

Fig. 3 Comparison of pressure dependence of maximum total separation factor attainable in total reflux operation between "Cryogenic-Wall" and "Ordinary Cold-wall" TD columns in case of ΔT=1,000 K

points are summarized as follows:

(1) Value of α_T in Lower Temperature

There is no data available for α_T in lower temperature. In the calculations above, we use Eq. (6), or the approximate fitting of our rough estimate of temperature dependence of α_T. In lower temperature region, the value of α_T usually decreases, for example, in the approximate fitting above, α_T for the reference mean temperature $\bar{T}$ of the "Cryogenic-Wall" TD column is 84.4% of that of "Ordinary Cold-wall" TD column:

	T_c(K)	ΔT(K)	$\bar{T}$(K)	α_T
"Cryogenic-Wall"	77.35	1,000	225.15	0.0782
"Ordinary Cold-wall"	288.15	1,000	435.95	0.0926
	288.15	800	406.39	0.0919

If the value of α_T decreases more rapidly than our rough estimate in the lower temperature region, the expected values of the maximum separation factor would be smaller. Thus, the study of α_T in the lower temperature region is indispensable for the "Cryogenic-Wall" TD column.

In addition, it is important to pay attention to the fact that the argument of the exponential function of Eq. (3) is not proportional directly to $\alpha_T \cdot \Delta T/T_c$ but to $\alpha_T \cdot \Delta T/\bar{T}$. The magnitude of the argument for the "Cryogenic-Wall" TD column with ΔT=1,000 K is, therefore, only 1.635 (435.95/225.15×0.844) times larger than that for "ordinary cold-wall" TD column, while the value of $\Delta T/T_c$ is 3.725 times larger. It is, however, difficult for the "Ordinary Cold-wall" TD column to multiply the value of the argument by the factor of 1.635; the value of ΔT must be 2,430 K, then, $\bar{T}$=647.31 K and $\Delta T/\bar{T}$=3.754, which is 1.635 times larger than that for ΔT=1,000 K. It is impossible for the hot wire to be (2,430+288.15) K, at which temperature the wire would melt down. Decreasing the cold wall temperature by 210.8 K (from 288.15 to 77.35 K) has the same effect as increasing the hot-wire temperature by impossible 1,430 K. Consequently, the effect of "Cryogenic-Wall" is large, and the prospects for application of "Cryogenic-Wall" TD column are good.

The present estimate is based on the approximate formulae of the column constants and is limitted to the case of total reflux operation. Detailed calculations based on the two-dimensional separative analysis, therefore, are required to verify the separative performances of the "Cryogenic-Wall" TD column also for the case with continuous feed and draw-offs.

(2) Possibility of Onset of Turbulence

In the thermal diffusion columns laminar free convection plays very important role to multiply the small separation effect due to thermal diffusion into substantial degree of separation. The one-dimensional separative analysis(2)~(4) of TD column is performed under an assumption of existence of laminar free convection within the column, and becomes meaningless when the laminar free convection could not exist and shifts to turbulent, because turbulent flow will inherently mix gases and destroy concentration gradient within the column. Hence, the detailed flow analysis is necessary to study whether the free convection remains laminar even when the cold-wall being kept in cryogenic temperature.

Moreover, problems relating practical application are:

(3) Loss of Coolant Accident

Even when the coolant is stopped by some accident, there is no problem because the total amounts of tritium within the TD column operated in gas phase in low pressure is very small compared with those of cryogenic distillation column operated in two-phase (gas and liquid).

(4) Increase of Fixed Cost

Indeed, the fixed cost of coolant systems for the "Cryogenic-Wall" TD column is higher than that of "Ordinary Cold-wall" TD column. The liquid nitrogen system is, however, relatively cheaper than liquid helium system required for cryogenic distillation column for hydrogen isotope separation.

IV. CONCLUSION

Possibility of enhancement of separation factor in total reflux operation of "Cryogenic-Wall" thermal diffusion column was discussed on the basis of approximate formulae involving explicitly design parameters for the column constants of TD column. Conclusions are summarized as follows:

(1) The total separation factor is fairly enhanced by the adoption of "Cryogenic-Wall". The magnitude of enhancement is, however, smaller than that expected from the ratio of $\Delta T/T_c$, because (i) the value of α_T decreases in lower temperature region, and (ii) the argument of the exponential function expressing the maximum value of the separation factor is not proportional directly to $\alpha_T \cdot \Delta T/T_c$ but to $\alpha_T \cdot \Delta T/\bar{T}$, where $\bar{T}$ is the reference mean temperature higher than T_c.

(2) The optimum pressure of "Cryogenic-Wall" TD column is considerably smaller than that of "Ordinary Cold-wall" TD column. It means smaller inventory of tritium within the "Cryogenic-Wall" TD column than that of "Ordinary Cold-wall" TD column which operated higher pressure.

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