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Ho-Ming Yeh[a]
[a] DEPARTMENT OF CHEMICAL ENGINEERING, TAMKANG UNIVERSITY, TAMSUI, TAIWAN, REPUBLIC OF CHINA

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Enrichment of Heavy Water in Flat-Plate Thermal Diffusion Columns of the Frazier Scheme Inclined for Improved Performance

HO-MING YEH
DEPARTMENT OF CHEMICAL ENGINEERING
TAMKANG UNIVERSITY
TAMSUI, TAIWAN, REPUBLIC OF CHINA

ABSTRACT

A separation theory for the enrichment of heavy water in flat-plate thermal diffusion columns of the Frazier scheme inclined for improved performance has been developed. Equations for the best angle of inclination and maximum separation have been derived. Considerable improvement in separation is obtainable if the columns are inclined at the best angle, so that the convective strength can be properly reduced and controlled, resulting in suppression of the undesirable remixing effect while still preserving the desirable cascading effect.

INTRODUCTION

Nuclear energy plays an important role in fulfilling society's energy requirements. Long-term forecasts indicate that a growing proportion of this energy will be supplied by fission reactors and, in the distant futher, perhaps by thermonuclear fusion. In addition to being the resource of fusion energy, deuterium oxide (D_2O) is also the most feasible moderator and coolant for fission reactors. Lewis (7) concentrated a large quantity of water to a small amount of nearly pure D_2O by electrolysis at the University of California. Between 1940 and 1945, four heavy water production plants were placed in operation by the US Government under the Manhattan Program (1, 8).

Thermal diffusion is a well-established method for separating isotopes. It was used to separate uranium isotopes at Oak Ridge Laboratory in

World War II. This method is particularly attractive for the separation of hydrogen isotopes because of the large ratio in molecular weights. Verhagen (12) tried this method for routine enrichment of tritium samples. By suitable arrangement of two thermal-diffusion columns and a container, a ten-times enrichment with better than 95% recovery has been achieved in about 20 hours. It has been shown that heavy water can also be concentrated successfully in thermal diffusion columns (3, 4, 6). Later, the enrichment of heavy water in thermal diffusion columns was studied both theoretically and experimentally (14).

In industrial applications, however, thermal diffusion columns are connected in series such as that shown in Fig. 1, called the Frazier scheme (5). The feeding method of the Frazier scheme is different from that of a conventional column. In a conventional thermal diffusion column, the feed is introduced at the middle and products are withdraw from the top and bottom, while in the Frazier scheme the sampling streams do not pass through but move outside the columns, as shown in Fig. 1.

Figure 2 indicates the flows and fluxes in one of the flat-plate thermal diffusion columns of the Frazier scheme. This column consists essentially of two opposing parallel plates separated by a very narrow open space. One plate is heated and the other cooled, and the thermal diffusion effect produced by the temperature gradient causes one component of a mixture to diffuse toward the hot plate. At the same time, the density gradient which arises because of the temperature gradient causes smooth laminar convection currents to move up the hot plate and down the cold plate. Because of the concentration gradient set up by thermal diffusion, the

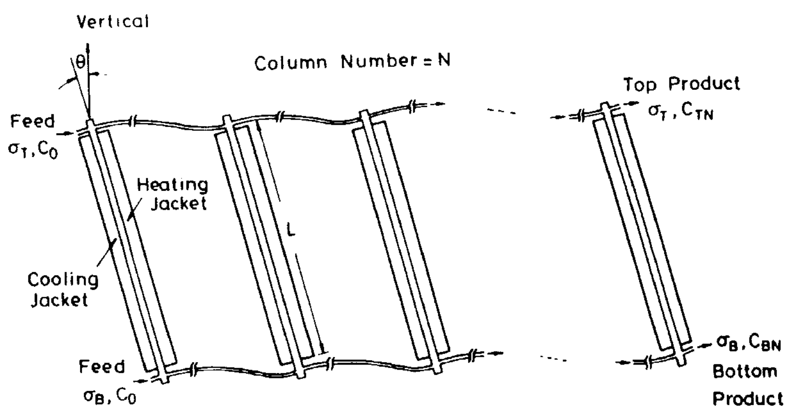

FIG. 1 Schematic diagram of the Frazier scheme for forward transverse flow.

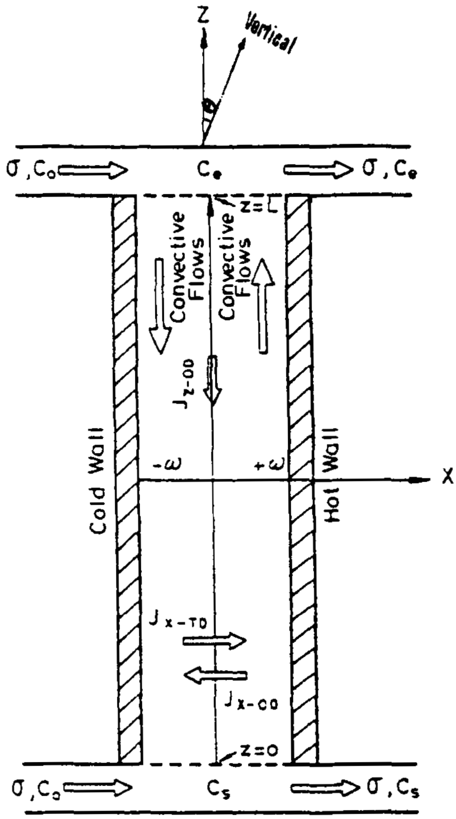

FIG. 2 The flows and fluxes in one of the flat-plate thermal diffusion columns of the Frazier scheme.

convective currents transport one component preferentially toward the top and thus create large concentration differences between the top and the bottom of the column.

A more detailed study of the mechanism of thermodiffusion separation indicates that the convective currents in thermal diffusion columns actually have two conflicting effects: the desirable cascading effect and the undesirable remixing effect. It appears, therefore, that proper control of the convective strength might effectively suppress the undesirable remixing effect while still preserving the desirable cascading effect, and thereby lead to improved performance. It has been shown that the undesirable remixing effect in a flat-plate thermal diffusion column can be effectively reduced and controlled by tilting the column, resulting in substantial improvement in separation efficiency (2, 9, 13). It is the purpose of this work to develop and investigate the separation theory for the enrichment of heavy water in flat-plate thermal diffusion columns of the Frazier scheme inclined for improved performance.

THEORY OF THE FRAZIER SCHEME

The scheme proposed by Frazier to connect several vertical thermodiffusion columns with forward transverse sampling streams is shown in Fig. 1 with θ set to zero. The delivery of the supply σ_T and σ_B with concentration C_0 is accomplished at the upper and lower ends in a plain thermodiffusion column with gap (2ω), length L, and width B, where both streams have the same direction. Sampling of the product is carried out at the ends opposite to the supply entrance. Frazier (5) gave a theory of this process on the basis of the simplified model represented, while an analytical solution was given by Rabinovich and Sovorov (10, 11). The result is

$$\begin{aligned}\Delta_v &= C_{BN} - C_{TN} \\ &= \frac{-C(1-C)HL}{K}\left[1 - \left\{\frac{HL/K}{H(1/\sigma_T + 1/\sigma_B) + HL/K}\right\}^N\right]\end{aligned} \tag{1}$$

where

$$H = \frac{\alpha\bar{\rho}g\bar{\beta}_T(2\omega)^3B(\Delta T)^2}{6!\mu\bar{T}} \tag{2}$$

$$K = \frac{\bar{\rho}g^2\bar{\beta}_T^2(2\omega)^7B(\Delta T)^2}{9!\mu^2D} \tag{3}$$

In obtaining this solution, assumptions were made that the parameters of all the columns were the same in magnitude, that the ordinary diffusion in the transverse direction is generally negligible compared with the convection, and that the product form of concentration $C(1 - C)$ is constant.

For the enrichment of heavy water by thermal diffusion in the H_2O–HDO–D_2O system, Yeh and Yang (14) assumed that the concentrations were locally in equilibrium at every point in thermal diffusion columns and, thus, the quadratic form of concentration for D_2O becomes the following expression:

$$\begin{aligned}C(1-C) &= A \text{ (a constant)} \\ &= C_0[0.05263 - (0.05263 - 0.0135K_{eq})C_0 \\ &\quad - 0.027\{C_0K_{eq}(1-(1-0.25K_{eq})C_0)\}^{1/2}]\end{aligned} \tag{4}$$

THEORY OF INCLINED FRAZIER SCHEME

It was mentioned previously that separation in thermal diffusion columns can be improved by properly tilting the columns to reduce the strength of convective currents. The equation of separation for an inclined Frazier scheme with all columns inclined in the same angle θ can be readily obtained from Eq. (1), with the gravitational acceleration g replaced by $g \cos \theta$. The result is

$$\Delta = C_{BN} - C_{TN} \tag{5}$$
$$= \frac{-AHL}{K \cos \theta}\left[1 - \left\{\frac{HL/K \cos \theta}{H \cos \theta(1/\sigma_T + 1/\sigma_B) + HL/K \cos \theta}\right\}^N\right]$$

Making a material balance for the whole scheme yields

$$\sigma_T C_{TN} + \sigma_B C_{BN} = (\sigma_T + \sigma_B)C_0 \tag{6}$$

or

$$m = \sigma_B(C_{BN} - C_0) = \sigma_T(C_0 - C_{TN}) \tag{7}$$

in which m denotes the removal rate of heavy water. Thus, the concentrations of heavy water in the exiting streams are related with m as

$$C_{BN} = C_0 + m/\sigma_B \tag{8}$$

$$C_{TN} = C_0 - m/\sigma_T \tag{9}$$

Consequently, the removal rate is related to the concentration difference Δ defined in Eq. (5) as

$$\Delta = \left(\frac{\sigma_T + \sigma_B}{\sigma_T \sigma_B}\right)m \tag{10}$$

The following dimensionless variables are introduced:

$$\sigma' = \frac{L\sigma_B\sigma_T}{K(\sigma_B + \sigma_T)} \tag{11}$$

$$M = \frac{m}{A\sigma'(-H)} = \frac{\Delta K}{AL(-H)} \tag{12}$$

With the use of Eqs. (10)–(12), Eq. (5) becomes

$$M = \frac{1}{\cos \theta}\left[1 - \left(\frac{\sigma'}{\cos \theta^2 + \sigma'}\right)^N\right] \tag{13}$$

THE BEST ANGLE OF INCLINATION FOR MAXIMUM SEPARATION

The best angle of inclination θ^* for maximum removal rate M_{max} in the Frazier scheme is obtained by differentiating Eq. (13) with respect to θ and setting $\partial M/\partial\theta = 0$. After simplification, this gives

$$\theta^* = \cos^{-1}\left[\frac{\sigma'(1 - W)}{W}\right]^{1/2} \tag{14}$$

$$M_{max} = \left(\frac{W}{1 - W}\right)^{1/2}(1 - W^N)\sigma'^{-1/2} \tag{15}$$

The value of W in the above equations is determined from the following equation:

$$2NW^{N+1} - (2N + 1)W^N + 1 = 0 \tag{16}$$

Graphical representations of Eqs. (16), (14), and (15) are given in Figs. 3, 4, and 5, respectively. There is an important restriction on the existence of the best angle of inclination. Since $0 \leq \theta \leq \pi/2$, it follows that $0 \leq \cos\theta \leq 1$, and from Eq. (14)

$$\sigma' < W/(1 - W) \tag{17}$$

which should be satisfied if the maximum removal rate exists.

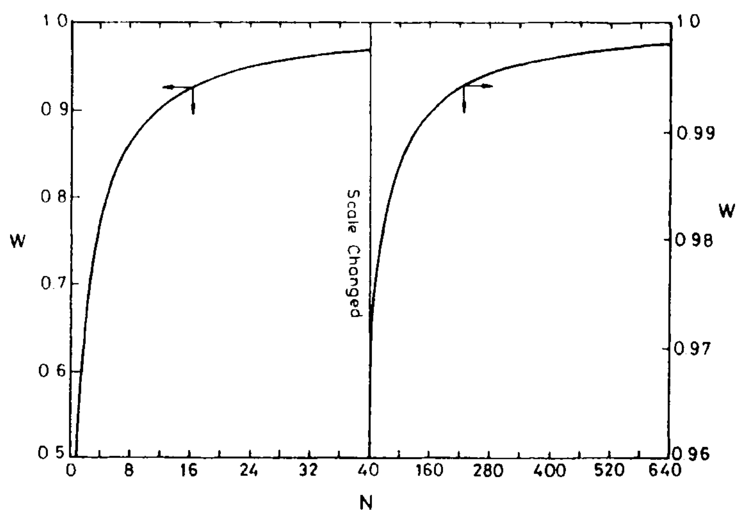

FIG. 3 Graphical solution of Eq. (16) for $0 < W < 1$.

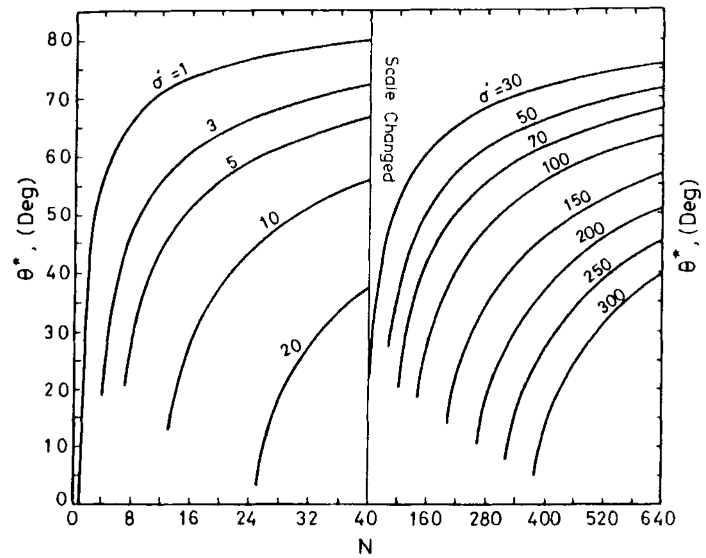

FIG. 4 Best column angle of inclination vs column number of scheme.

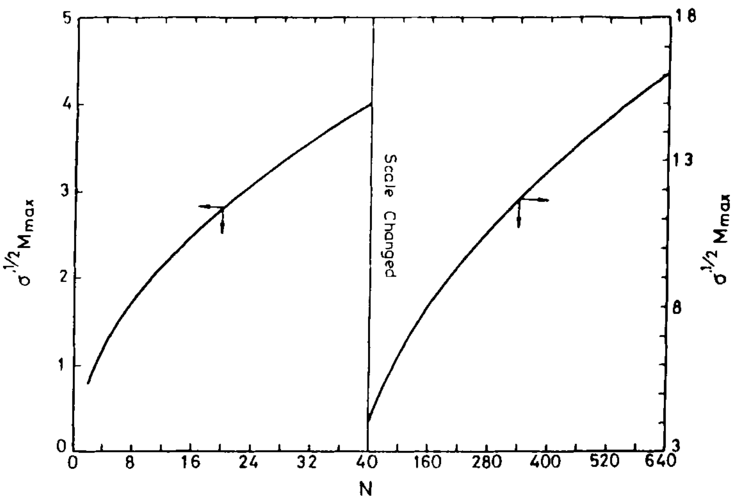

FIG. 5 Maximum separation vs column number of scheme.

THE IMPROVEMENT IN SEPARATION

The improvement in separation by operating at the best angle of inclination is best illustrated by calculating the percentage increase in separation based on the vertical scheme:

$$I = \frac{m_{max} - m_v}{m_v} = \frac{\Delta_{max} - \Delta_v}{\Delta_v} \tag{18}$$

It is noted that Eq. (18) holds according to Eq. (10) or (12), and can be rewritten as

$$I = \frac{M_{max} - M_v}{M_v} = \frac{M_{max}}{M_v} - 1 \tag{19}$$

The dimensionless removal rate of heavy water in the vertical scheme is obtained from Eq. (13) by setting $\theta = 0$:

$$M_v = 1 - \left(\frac{\sigma'}{1 + \sigma'}\right)^N \tag{20}$$

Substitution of Eqs. (15) and (20) into Eq. (19) results in

$$I = \frac{[W/(1 - W)]^{1/2}(1 - W^N)\sigma'^{-1/2}}{1 - [\sigma'/(1 + \sigma')]^N} - 1 \tag{21}$$

The graphical representation of Eq. (21) is given in Fig. 6 by using Eq. (16) or Fig. 3. It is seen from Fig. 6 that

$$I > 0, \qquad \text{as } \sigma' < W/(1 - W) \tag{22}$$

Therefore, in order to obtain a better separation in flat-plate thermal diffusion columns of the Frazier scheme, operation at the best angle of inclination is recommended.

For the purpose of illustration, let us employ some numerical values obtained in previous work (14) for the enrichment of D_2O in the H_2O–HDO–D_2O system as follows:

$$H = -1.473 \times 10^{-4} \text{ (g/s)}$$

$$K = 1.549 \times 10^{-3} \text{ (g·cm/s)}$$

where $L = 177$ cm, $B = 10$ cm, $\Delta T = 30.5$°C. From these values, the best angles of inclination and the maximum removal rates of heavy water

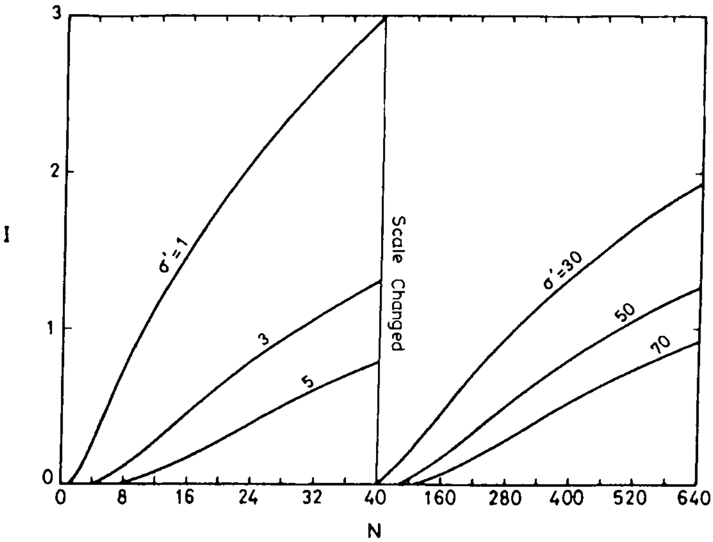

FIG. 6 Graphical representation of the improvement in separation for the inclined scheme based on the vertical scheme.

were calculated from the corresponding separation equations, and consequently the improvements in removal rate were obtained by substituting the appropriate values of removal rate into Eq. (19) or by directly calculating from Eq. (21). The results are presented in Tables 1–3, in which considerable improvement in thermodiffusion separation was obtained in the Frazier scheme by operating at the best angle of inclination.

Once the removal rate m is calculated, the concentrations of exiting streams may be estimated from Eqs. (8) and (9). Table 4 shows some values of these concentrations as well as the concentration differences between two exiting streams.

DISCUSSION AND CONCLUSIONS

It has been shown that the undesirable remixing effect in thermal diffusion columns of the Frazier scheme with forward transverse flow can be effectively reduced and controlled by tilting the columns, resulting in substantial improvement in separation efficiency (see Fig. 6). The equa-

TABLE 1

Results of a Numerical Example with $N = 20$ ($W = 0.9415$)

						$m_v \times 10^2$ (g/h)					$m_{max} \times 10^2$ (g/h)				
σ_B/σ_T	σ_B (g/h)	σ_I (g/h)	σ'	θ^* (deg)	I (%)	$C_0 = 0.1$	$C_0 = 0.3$	$C_0 = 0.5$	$C_0 = 0.7$	$C_0 = 0.9$	$C_0 = 0.1$	$C_0 = 0.3$	$C_0 = 0.5$	$C_0 = 0.7$	$C_0 = 0.9$
0.5	0.5	1.0	10.58	35.8	3	1.70	3.44	3.90	3.42	2.20	1.75	3.55	4.03	3.53	2.27
0.5	1.0	2.0	21.16	0	0	2.45	4.97	5.63	4.94	3.17	2.45	4.97	5.63	4.94	3.17
0.5	1.5	3.0	31.74	0	0	2.82	5.72	6.48	5.68	3.65	2.82	5.72	6.48	5.68	3.65
1.0	0.5	0.5	7.94	45.4	10	1.38	2.80	3.18	2.79	1.79	1.52	3.09	3.50	3.07	1.97
1.0	1.0	1.0	15.87	6.8	0	2.15	4.36	4.94	4.34	2.78	2.15	4.36	4.94	4.34	2.78
1.0	1.5	1.5	23.81	0	0	2.56	5.20	5.89	5.17	3.32	2.56	5.20	5.89	5.17	3.32
2.0	0.5	0.25	5.29	55.0	26	0.99	2.00	2.26	1.99	1.27	1.24	2.52	2.85	2.50	1.61
2.0	1.0	0.5	10.58	35.8	3	1.70	3.44	3.90	3.42	2.20	1.75	3.55	4.03	3.53	2.27
2.0	1.5	0.75	15.87	6.8	0	2.15	4.36	4.94	4.34	2.78	2.15	4.36	4.94	4.34	2.78

TABLE 2

Results of a Numerical Example with $N = 40$ ($W = 0.9697$)

						$m_v \times 10^2$ (g/h)					$m_{max} \times 10^2$ (g/h)				
σ_B/σ_T	σ_B (g/h)	σ_I (g/h)	σ'	θ^* (deg)	I (%)	$C_0 = 0.1$	$C_0 = 0.3$	$C_0 = 0.5$	$C_0 = 0.7$	$C_0 = 0.9$	$C_0 = 0.1$	$C_0 = 0.3$	$C_0 = 0.5$	$C_0 = 0.7$	$C_0 = 0.9$
0.5	0.5	1.0	10.58	54.9	26	1.98	4.01	4.54	3.98	2.56	2.50	5.07	5.75	5.04	3.23
0.5	1.0	2.0	21.16	35.6	3	3.42	6.94	7.86	6.90	4.43	3.54	7.17	8.13	7.13	4.57
0.5	1.5	3.0	31.74	5.2	0	4.33	8.79	9.96	8.73	5.60	4.33	8.79	9.96	8.73	5.60
1.0	0.5	0.5	7.94	60.1	44	1.51	3.06	3.47	3.04	1.95	2.16	4.39	4.98	4.37	2.80
1.0	1.0	1.0	15.87	45.2	10	2.78	5.64	6.39	5.61	3.60	3.06	6.21	7.04	6.18	3.96
1.0	1.5	1.5	23.81	30.4	2	3.69	7.48	8.48	7.44	4.77	3.75	7.61	8.62	7.56	4.85
2.0	0.5	0.25	5.29	66.0	75	1.01	2.06	2.33	2.05	1.31	1.77	3.59	4.06	3.57	2.29
2.0	1.0	0.5	10.58	54.9	26	1.98	4.01	4.54	3.98	2.56	2.50	5.07	5.75	5.04	3.23
2.0	1.5	0.75	15.87	45.2	10	2.78	5.64	6.39	5.61	3.60	3.06	6.21	7.04	6.18	3.96

TABLE 3
Results of a Numerical Example with $N = 80$ ($W = 0.9848$)

						$m_v \times 10^2$ (g/h)					$m_{max} \times 10^2$ (g/h)				
σ_B/σ_T	σ_B (g/h)	σ_T (g/h)	σ'	θ^* (deg)	I (%)	$C_0 = 0.1$	$C_0 = 0.3$	$C_0 = 0.5$	$C_0 = 0.7$	$C_0 = 0.9$	$C_0 = 0.1$	$C_0 = 0.3$	$C_0 = 0.5$	$C_0 = 0.7$	$C_0 = 0.9$
0.5	0.5	1.0	10.58	66.2	75	2.03	4.12	4.66	4.09	2.62	3.55	7.20	8.16	7.16	4.59
0.5	1.0	2.0	21.16	55.1	27	3.96	8.04	9.10	7.99	5.12	5.02	10.19	11.54	10.12	6.49
0.5	1.5	3.0	31.74	45.6	10	5.58	11.33	12.83	11.26	7.22	6.15	12.47	14.13	12.40	7.95
1.0	0.5	0.5	7.94	69.5	102	1.52	3.09	3.50	3.07	1.97	3.07	6.24	7.07	6.20	3.98
1.0	1.0	1.0	15.87	60.3	44	3.02	6.13	6.95	6.10	3.91	4.35	8.82	9.99	8.77	5.62
1.0	1.5	1.5	23.81	52.7	21	4.40	8.93	10.11	8.87	5.69	5.32	10.80	12.24	10.74	6.89
2.0	0.5	0.25	5.29	73.4	147	1.02	2.06	2.33	2.05	1.31	2.51	5.09	5.77	5.06	3.25
2.0	1.0	0.5	10.58	66.2	75	2.03	4.12	4.66	4.09	2.62	3.55	7.20	8.16	7.16	4.59
2.0	1.5	0.75	15.87	60.3	44	3.02	6.13	6.95	6.10	3.91	4.35	8.82	9.99	8.77	5.62

TABLE 4
Results of Numerical Examples: Concentrations of the Product Streams

						$C_0 = 0.3$								$C_0 = 0.5$							
						Vertical column				Inclined column				Vertical column				Inclined column			
N	σ_B (g/h)	σ_T (g/h)	σ'	θ^* (deg)	I (%)	$m_v \times 10^2$ (g/h)	C_{BN}	C_{IN}	Δ_v	$m_{max} \times 10^2$ (g/h)	C_{BN}	C_{IN}	Δ	$m_v \times 10^2$ (g/h)	C_{BN}	C_{IN}	Δ_v	$m_{max} \times 10^2$ (g/h)	C_{BN}	C_{IN}	Δ
20	1.0	0.5	10.58	35.8	3	3.44	0.334	0.231	0.103	3.55	0.336	0.229	0.107	3.90	0.539	0.422	0.117	4.03	0.540	0.419	0.121
20	1.0	1.0	15.87	6.8	0	4.36	0.344	0.256	0.088	4.36	0.344	0.256	0.088	4.94	0.549	0.451	0.098	4.94	0.549	0.451	0.098
20	1.0	2.0	21.16	0	0	4.97	0.350	0.275	0.075	4.97	0.350	0.275	0.075	5.63	0.556	0.472	0.084	5.63	0.556	0.472	0.084
40	1.0	0.5	10.58	54.9	26	4.01	0.340	0.220	0.120	5.07	0.351	0.199	0.152	4.54	0.545	0.409	0.136	5.75	0.557	0.385	0.172
40	1.0	1.0	15.87	45.2	10	5.64	0.356	0.244	0.112	6.21	0.362	0.238	0.124	6.39	0.564	0.436	0.128	7.04	0.570	0.430	0.140
40	1.0	2.0	21.16	35.6	3	6.94	0.369	0.265	0.104	7.17	0.372	0.264	0.108	7.86	0.579	0.461	0.118	8.13	0.581	0.460	0.121
80	1.0	0.5	10.58	66.2	75	4.12	0.341	0.218	0.213	7.20	0.372	0.156	0.216	4.66	0.547	0.407	0.140	8.16	0.582	0.337	0.245
80	1.0	1.0	15.87	60.3	44	6.13	0.361	0.239	0.122	8.82	0.388	0.212	0.176	6.95	0.570	0.431	0.139	9.99	0.600	0.400	0.200
80	1.0	2.0	21.16	55.1	27	8.04	0.380	0.260	0.120	10.19	0.402	0.249	0.153	9.10	0.591	0.455	0.136	11.54	0.615	0.442	0.173

tions for the best angle of inclination and maximum separation for the enrichment of heavy water (Eqs. 14 and 15) have been derived, and their graphical representations are shown in Figs. 4 and 5, respectively. Further, the region within which inclination improves the separation has been delineated (Eq. 17).

Numerical examples for the enrichment of heavy water were given; considerable improvement in separation was obtained by operating at the best angle of inclination (see Tables 1–4). Tables 1–4 also show that when σ' or $\sigma_B\sigma_T/(\sigma_B + \sigma_T)$ increases, the removal rates of heavy water (m_v and m_{max}) and the concentrations of the exiting streams (C_{BN} and C_{TN}) increase whereas the best angle of inclination (θ^*) and the improvement of separation (I) as well as the concentration differences of the exiting streams (Δ_v and Δ_{max}) decrease. Besides, C_{BN}, C_{TN}, m_v, m_{max}, Δ_v, and Δ_{max} depend on the feed concentration (C_0) but θ^* and I do not.

It is seen in Table 1 that no improvement is obtained in the 20-columns Frazier scheme ($N = 20$) when $\sigma_B = 1.0$ and $\sigma_T = 2$ g/h ($\sigma' = 21.16$), or when $\sigma_B = 1.5$ and $\sigma_T = 1.5$ g/h ($\sigma' = 23.81$), or when $\sigma_B = 1.5$ and $\sigma_T = 3$ g/h ($\sigma' = 31.74$). In other words, when $\sigma' > W/(1 - W) = 16.09$, the best separation is obtained in vertical columns. This result confirms Eq. (17). Further, when $\sigma_B = 1.0$ and $\sigma_T = 1.0$ g/h ($\sigma' = 15.87$), or when $\sigma_B = 1.5$ and $\sigma_T = 0.75$ g/h ($\sigma' = 15.87$), the improvements are nearly zero because σ' approaches the critical value (16.09).

Tilting the columns probably offers the only effective and inexpensive way to improve the efficiency of an existing flat-plate thermal diffusion apparatus and is of great utility for feasibility studies in research or pilot-plant columns operating under widely different conditions.

SYMBOLS

A	$C(1 - C)$, a constant defined by Eq. (4)
B	column width (cm)
C	fractional mass concentration of heavy water (D_2O)
C_{BN}, C_{TN}	C in the product streams exiting from Nth column, for bottom and top ends, respectively.
D	ordinary diffusion coefficient (cm^2/s)
C_0	C in the feed streams
g	gravitational acceleration (cm/s^2)
H	system constant defined by Eq. (2) (g/s)
I	improvement in separation defined by Eq. (18)
K	system constant defined by Eq. (3) [(g·cm/s)]

K_{eq}	mass fractional equilibrium constant of H_2O–HDO–D_2O system
L	column length (cm)
M	dimensionless removal rate of heavy water defined by Eq. (12)
m	removal rate of heavy water defined by Eq. (7), (g/s)
p	pressure (Pa)
$\overline{T}$	mean absolute temperature (K)
ΔT	difference in temperature of hot and cold plates (K)
W	constant, evaluated from Eq. (16)

Greek Letters

α	thermal diffusion constant
$\overline{\beta}_T$	$-(\partial\rho/\partial T)_p$ evaluated at $\overline{T}$ [g/(cm^3·K)]
Δ	$C_{BN} - C_{TN}$, concentration difference of the product streams
θ	column angle of inclination (deg)
θ^*	best value of θ for maximum separation (deg)
$\overline{\rho}$	mass density evaluated at $\overline{T}$ (g/cm^3)
μ	absolute viscosity (cP)
σ_B, σ_T	mass flow rate at bottom, top side of the scheme (g/s)
σ'	dimensionless flow rate defined by Eq. (11) (g/s)
ω	half of plate spacing (cm)

Subscripts

max	maximum value obtained at the optimum condition
v	value obtained in vertical column

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