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The Enrichment of Heavy Water in a Continuous-Type Inclined Thermal Diffusion Column

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Abstract

Equations of the best angle of inclination and the maximum separation for the enrichment of heavy water in a continuous-type inclined thermal diffusion column have been derived. Considerable improvement in separation was obtained by employing the inclined column instead of using the Clusius-Dickel column.

INTRODUCTION

The enrichment of heavy water from the H_2O–HDO–D_2O system in a thermogravitational thermal diffusion column has been studied, both theoretically and experimentally, by Yeh and Yang (*1–3*). The most important assumption in their work is that the concentrations are locally in equilibrium at every point in the column. A linear approximation to concentration product, $c\hat{c}$, was also made for simplicity.

Actually, the convective currents in a Clusius-Dickel column have two conflicting effects: a desirable cascading effect and an undesirable remixing effect. It therefore appears that proper control of the convective strength might effectively suppress the undesirable remixing effect while still preserving the desirable cascading effect, thereby leading to improved separation (*4–15*).

A simple and flexible way of adjusting the convective strength is to tilt a flat-plate column with a hot plate on top so as to reduce the effective gravitational force. The theory for an inclined flat-plate column for a binary mixture system has been studied by Powers and Wilke (*4*) and Chueh and

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Yeh (*9*). Theoretical considerations show that when the column is at the best inclination, maximum separation, maximum production, or minimum column length may be obtained. Experimental results for the system benzene–heptane are in excellent agreement with theory.

It is the purpose of this work to investigate the effect of inclination on the degree of separation for the H_2O–HDO–D_2O system.

CLUSIUS-DICKEL COLUMN

The transport equation of D_2O for the H_2O–HDO–D_2O system in a Clusius-Dickel column has been derived by Yeh and Yang (*1*):

$$\tau = H_0 c\hat{c} - K\frac{dc}{dz} \tag{1}$$

where

$$c\hat{c} = c\left\{0.05263 - (0.05263 - 0.0135K_{eq})c - 0.027\left\{cK_{eq}\left[1 - \left(1 - \frac{K_{eq}}{4}\right)c\right]\right\}^{1/2}\right\} \tag{2}$$

for a whole range of concentrations, and

$$c\hat{c} = c[0.05263 - (0.05263 - 0.0135K_{eq})c - 0.27K_{eq}^{1/2}c^{1/2}] \tag{3}$$

for low and short concentration ranges.

The most important assumption in their work is that the concentrations are locally in equilibrium at every point in the column. They also obtained the separation equation by a linear approximation to the concentration product, $c\hat{c}$:

$$c\hat{c} = a + bc \tag{4}$$

and the results quantitatively confirm the experimental data. The separation equation of D_2O for the whole range of concentrations under steady-state continuous operations thus derived is (*2*)

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$$\Delta_{0,L} = c_B - c_T$$

$$= \left(c_F + \frac{a}{b}\right)\left[\frac{1 - e^{bA(1-n/b)}}{e^{bA(1-n/b)} - n/b} - \frac{1 - e^{-bA(1+n/b)}}{e^{-bA(1+n/b)} + n/b}\right] \quad (5)$$

in which

$$H_0 = \alpha_0 \bar{\rho} g B \bar{\beta}_T (2\omega)^3 (\Delta T)^2 / 6! \mu \bar{T} \quad (6)$$

$$K = K_0 + K_1 \quad (7)$$

$$K_0 = (2\omega)^7 g^2 \bar{\beta}_T^2 \bar{\rho}\ \beta\ (\Delta T)^2 / 9! \mu^2 D \quad (8)$$

$$K_1 = 2\omega \bar{\rho} D B \quad (9)$$

$$n = \sigma / H_0 \quad (10)$$

$$A = H_0 L / 2K \quad (11)$$

$$b = \frac{d}{dc}(c\hat{c})|_{c=c_F} \quad (12)$$

$$a = c_F \hat{c}_F - b c_F \quad (13)$$

The term H_0 represents the effectiveness of separation by thermal diffusion and the term K represents the effect of remixing due to convection (K_0) and ordinary diffusion (K_1). K_1 is generally negligible compared with K_0.

Although the method of linear approximation can be employed to obtain the separation equations applicable to the whole range of concentrations, the application of optimization to these equations for optimal design and operation is very complicated and difficult. Therefore, a further approximation to $c\hat{c}$ may be made; for example,

$$c\hat{c} \doteqdot c_F \hat{c}_F = p \text{ (a constant)} \quad (14)$$

Then $a = P$, $b = 0$, and Eq. (5) reduces to

$$\Delta_0 = \frac{-2p}{n}[1 - e^{-nA}]$$

$$= \frac{2p(-H_0)}{\sigma}\left\{1 - \exp \frac{-\sigma L}{2(K_0 + K_1)}\right\} \quad (15)$$

Some values of $\Delta_{0,L}/\Delta_0$ for $K_{eq} = 3.793$ ($\bar{T} = 30.5°C$) calculated from Eqs. (5) and (15) are presented in Fig. 1. We found from Fig. 1 that $\Delta_0 \approx \Delta_{0,L}$ for most of the application range of $(-A)$ and $(-n)$. Although Eq. (5) is more precise than Eq. (15), for further application of the separation equation to the optimal column design, Eq. (15) is much simpler and more convenient to use.

INCLINED COLUMN

Jones, Furry, and Onsager (*16–19*) have presented a separation equation for continuous operation in a Clusius-Dickel column, with top and bottom products withdrawn at the same rates, for an equifraction binary system:

$$(\Delta_0)_{\text{binary}} = \frac{H_0}{2\sigma}\left[1 - \exp\frac{-\sigma L}{2(K_0 + K_1)}\right] \tag{16}$$

For an inclined column, g must be replaced by $g \cos\theta$, and the separation equation becomes (*4, 9*)

$$(\Delta_i)_{\text{binary}} = \frac{H_0 \cos\theta}{2\sigma}\left[1 - \exp\frac{-\sigma L}{2(K_0\cos^2\theta + K_1)}\right] \tag{17}$$

When the enrichment of D_2O from the H_2O–HDO–D_2O system for a whole range of concentrations in an inclined thermal diffusion column is considered, the separation equation may be obtained from Eq. (15) with g replaced by $g \cos\theta$:

$$\Delta_i = \frac{2p(-H_0)\cos\theta}{\sigma}\left[1 - \exp\frac{-\sigma L}{2(K_0\cos^2\theta + K_1)}\right]$$

The best angle of inclination for maximum separation is obtained by partially differentiating Eq. (18) with respect to θ and setting $\partial\Delta_i/\partial\theta = 0$. After calculation and neglecting K_1, we have (*9*)

$$\theta^* = \cos^{-1}\left(\frac{\sigma L}{2.42\,K_0}\right)^{1/2} \tag{19}$$

with the restriction

$$\sigma L/K_0 < 2.52 \tag{20}$$

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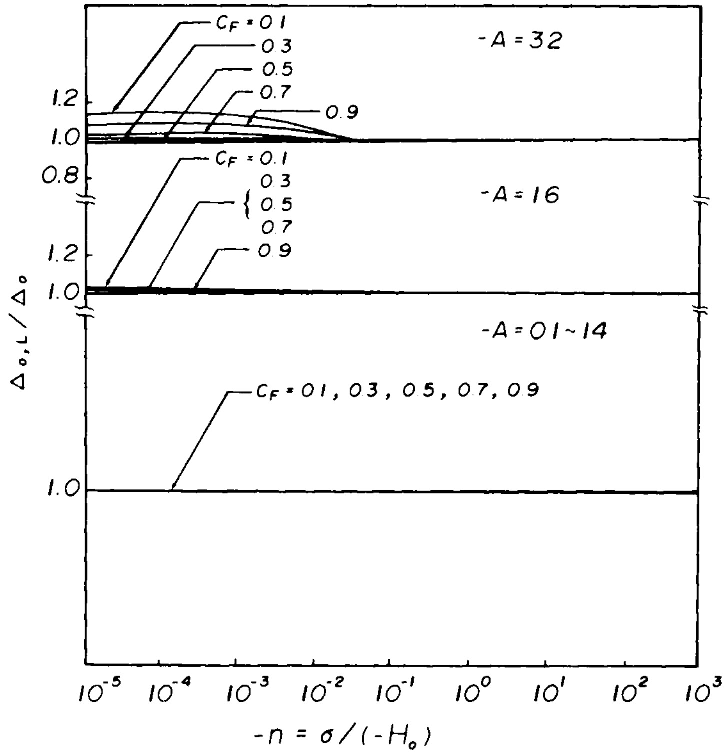

FIG. 1. Some values of $\Delta_{0,L}/\Delta_0$ for $K_{eq} = 3.793$ ($\bar{T} = 30.5°C$).

Substitution of Eq. (19) into Eq. (18) gives

$$\Delta_{i,\max} = 0.903p\left(\frac{H_0^2L}{\sigma K_0}\right)^{1/2} \tag{21}$$

The solution of the conditions for best performance can be most conveniently represented graphically in dimensionless variables. We define the dimensionless flow rate σ' and reduced separation Δ' by

$$\sigma' = \sigma L/K_0 \tag{22}$$

$$\Delta' = \Delta\sigma/(-H_0) \tag{23}$$

Equations (19), (21), and (20) can then be rewritten, respectively, as

$$\theta^* = \cos^{-1}\left(\frac{\sigma'}{2.52}\right)^{1/2} \tag{24}$$

$$\Delta'_{i,\max} = 0.903p\sigma'^{1/2} \tag{25}$$

$$\sigma' < 2.52 \tag{26}$$

For $\sigma' > 2.52$, the best separation is obtained at the vertical position. In this case, Eq. (15) becomes

$$\Delta'_0 = 2p(1 - e^{-\sigma'/2}) \tag{27}$$

Equations (24) and (25) are plotted in Fig. 2 for $\sigma' < 2.52$, while Eq. (27) is plotted in Fig. 3 for $\sigma' > 2.52$.

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THE IMPROVEMENT OF SEPARATION

The improvement in separation by operating at the best angle of inclination is best illustrated by calculating the percentage increase in

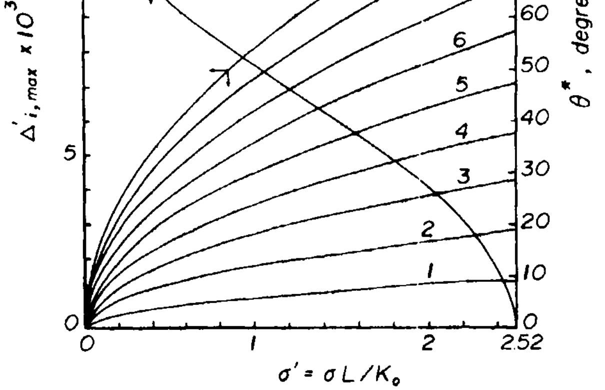

FIG. 2. Angle of inclination for best performance and reduced maximum separation vs reduced flow rate for $\sigma' < 2.52$.

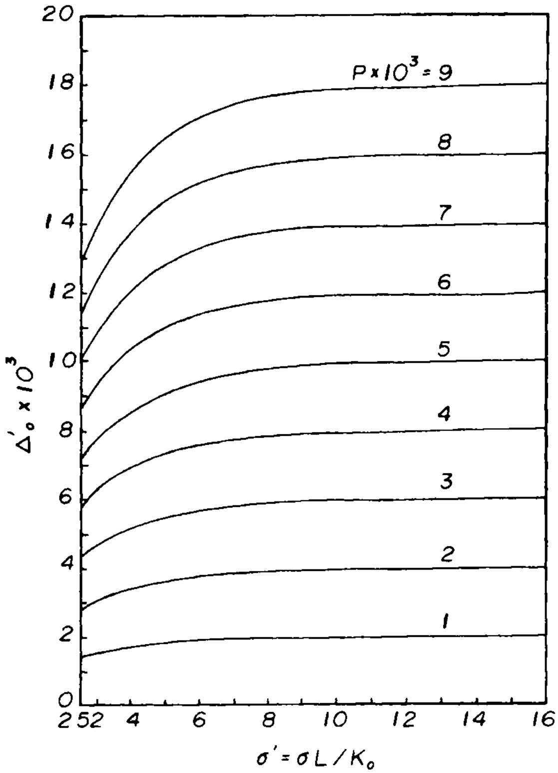

FIG. 3. Separation obtainable in vertical column vs reduced flow rate for $\sigma' > 2.52$.

separation based on the Clusius-Dickel column:

$$I = \frac{\Delta_{i,\max} - \Delta_0}{\Delta_0} \tag{28}$$

Substitution of Eqs. (15) and (21) into Eq. (28) yields

$$I = \frac{\Delta_{i,\max}}{\Delta_0} - 1 = \frac{[0.903p(-H_0)/\sigma]\sigma'^{1/2}}{[2p(-H_0)/\sigma](1 - e^{-\sigma'/2})} - 1$$

$$= \frac{0.451\sigma'^{1/2}}{1 - e^{-\sigma'/2}} - 1 \tag{29}$$

This result is presented graphically in Fig. 4. It is noted that $I > 0$ as $\sigma' < 2.52$.

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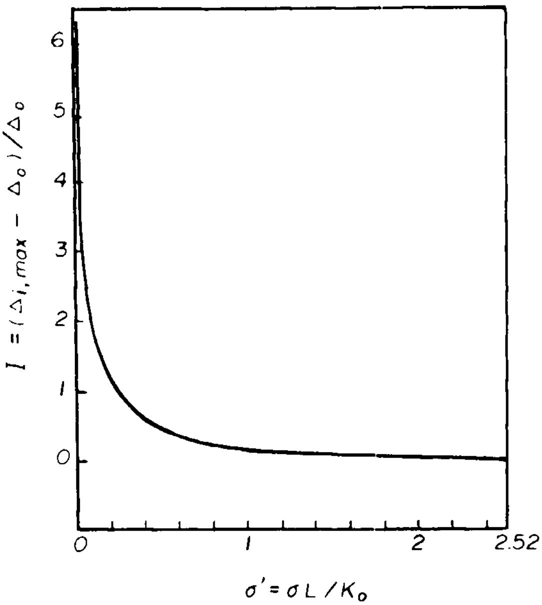

FIG. 4. Graphical representation of the improvement in separation for an inclined column based on a vertical one.

For the purpose of illustration, let us predict the improvement of separation by employing the inclined column under the design and operating conditions performed in previous works (*1–3*):

$$H_0 = -1.473 \times 10^{-4} \text{ g/s}$$

$$K_0 \approx K = 1.549 \times 10^{-3} \text{ g} \cdot \text{cm/s}$$

$$K_{eq} = 3.793 \ (\bar{T} = 30.5°\text{C})$$

$$L = 177 \text{ cm}$$

$$B = 10 \text{ cm}$$

From these values the separations were calculated from the corresponding separation equations, and consequently the improvements in separation were obtained by substituting the appropriate values of separation into Eq. (28). The results are presented in Table 1. Two more numerical examples are given by changing the column width to 30 and 100 cm, and the results are presented in Tables 2 and 3, respectively.

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TABLE 1
Results of Numerical Example for B = 10 cm

$H_0 = -1.473 \times 10^{-4}$ g/s L = 177 cm
$K_0 = 1.549 \times 10^{-3}$ g · cm/s $-A = H_0L/2K_0 = 8.42$
$K_{eq} = 3.793$ $1.89 \times 10^{-2} < -n < 7.54 \times 10^{-2}$

c_F	σ (g/h)	σ'	Δ_0 (%)	$\Delta_{i,max}$ (%)	θ^* (degree)	I (%)
0.1	0.01	0.317	5.59	9.69	69.2	73
0.1	0.02	0.635	5.18	6.85	59.9	32
0.1	0.04	1.270	4.47	4.84	44.8	8
0.3	0.01	0.317	11.03	19.12	69.2	73
0.3	0.02	0.635	10.22	13.52	59.9	32
0.3	0.04	1.270	8.83	9.56	44.8	8
0.5	0.01	0.317	11.84	20.53	69.2	73
0.5	0.02	0.635	10.97	14.52	59.9	32
0.5	0.04	1.270	9.48	10.26	44.8	8
0.7	0.01	0.317	9.20	15.94	69.2	73
0.7	0.02	0.635	8.52	11.27	59.9	32
0.7	0.04	1.270	7.36	7.97	44.8	8
0.9	0.01	0.317	3.69	6.40	69.2	73
0.9	0.02	0.635	3.42	4.53	59.9	32
0.9	0.04	1.270	2.96	3.20	44.8	8

CONCLUSIONS

On the basis of the results of this study, the following conclusions were reached.

(1) The separation equation for the enrichment of heavy water in a continuous-type inclined thermal diffusion column has been derived. Equation (18), thus obtained, is applicable to the whole range of concentrations.

(2) Solutions of the best angle of inclination and the maximum separation have been obtained as shown in Eqs. (19) and (21), respectively. Further, the region within which inclination improves the separation has been delineated by Eq. (20). Some graphical representations are given in Fig. 2 in terms of reduced flow rate and reduced separation.

(3) Tables 1, 2, and 3 showed some results of the numerical examples.

TABLE 2
Results of Numerical Example for B = 30 cm

$H_0 = -4.419 \times 10^{-4}$ g/s $L = 177$ cm
$K_0 = 4.647 \times 10^{-3}$ g · cm/s $-A = H_0L/2K_0 = 8.42$
$K_{eq} = 3.793$ $6.3 \times 10^{-3} < -n < 0.101$

c_F	σ (g/h)	σ'	Δ_0 (%)	$\Delta_{i,max}$ (%)	θ^* (degree)	I (%)
0.1	0.01	0.106	5.89	16.78	78.2	185
0.1	0.02	0.212	5.73	11.86	73.2	107
0.1	0.04	0.423	5.45	8.39	65.8	54
0.1	0.08	0.846	4.93	5.93	54.6	20
0.1	0.16	1.693	4.08	4.19	35.0	3
0.3	0.01	0.106	11.62	33.12	78.2	185
0.3	0.02	0.212	11.32	23.42	73.2	107
0.3	0.04	0.423	10.75	16.56	65.8	54
0.3	0.08	0.846	9.73	11.71	54.6	20
0.3	0.16	1.693	8.05	8.28	35.0	3
0.5	0.01	0.106	12.48	35.56	78.2	185
0.5	0.02	0.212	12.15	25.14	73.2	107
0.5	0.04	0.423	11.54	17.78	65.8	54
0.5	0.08	0.846	10.44	12.57	54.6	20
0.5	0.16	1.693	8.64	8.89	35.0	3
0.7	0.01	0.106	9.69	27.61	78.2	185
0.7	0.02	0.212	9.44	19.52	73.2	107
0.7	0.04	0.423	8.96	13.81	65.8	54
0.7	0.08	0.846	8.11	9.76	54.6	20
0.7	0.16	1.693	6.71	6.90	35.0	3
0.9	0.01	0.106	3.89	11.09	78.2	185
0.9	0.02	0.212	3.79	7.84	73.2	107
0.9	0.04	0.423	3.60	5.55	65.8	54
0.9	0.08	0.846	3.26	3.92	54.6	20
0.9	0.16	1.693	2.70	2.77	35.0	3

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TABLE 3
Results of Numerical Example for B = 100 cm

$H_0 = -1.473 \times 10^{-3}$ g/s L = 177 cm
$K_0 = 1.549 \times 10^{-2}$ g · cm/s $-A = H_0L/2K_0 = 8.42$
$K_{eq} = 3.793$ $1.89 \times 10^{-3} < -n < 0.121$

c_F	σ (g/h)	σ'	Δ_0 (%)	$\Delta_{i,max}$ (%)	θ^* (degree)	I (%)
0.1	0.01	0.0317	6.00	30.63	83.6	411
0.1	0.02	0.0635	5.95	21.66	80.9	264
0.1	0.04	0.127	5.86	15.31	77.0	162
0.1	0.08	0.254	5.67	10.83	71.5	91
0.1	0.16	0.508	5.34	7.66	63.3	43
0.1	0.32	1.016	4.74	5.41	50.6	14
0.1	0.64	2.031	3.79	3.83	26.1	0.9
0.3	0.01	0.0317	11.84	60.47	83.6	411
0.3	0.02	0.0635	11.74	42.76	80.9	264
0.3	0.04	0.127	11.56	30.23	77.0	162
0.3	0.08	0.254	11.20	21.38	71.5	91
0.3	0.16	0.508	10.54	15.12	63.3	43
0.3	0.32	1.016	9.35	10.69	50.6	14
0.3	0.64	2.031	7.49	7.56	26.1	0.9
0.5	0.01	0.0317	12.71	64.92	83.6	411
0.5	0.02	0.0635	12.61	45.90	80.9	264
0.5	0.04	0.127	12.41	32.46	77.0	162
0.5	0.08	0.254	12.03	22.95	71.5	91
0.5	0.16	0.508	11.31	16.23	63.3	43
0.5	0.32	1.016	10.04	11.48	50.6	14
0.5	0.64	2.031	8.04	8.11	26.1	0.9
0.7	0.01	0.0317	9.87	50.41	83.6	411
0.7	0.02	0.0635	9.79	35.65	80.9	264
0.7	0.04	0.127	9.64	25.21	77.0	162
0.7	0.08	0.254	9.34	17.82	71.5	91
0.7	0.16	0.508	8.78	12.60	63.3	43
0.7	0.32	1.016	7.80	8.91	50.6	14
0.7	0.64	2.031	6.25	6.30	26.1	0.9
0.9	0.01	0.0317	3.96	20.25	83.6	411
0.9	0.02	0.0635	3.93	14.32	80.9	264
0.9	0.04	0.127	3.87	10.13	77.0	162
0.9	0.08	0.254	3.75	7.16	71.5	91
0.9	0.16	0.508	3.53	5.06	63.3	43
0.9	0.32	1.016	3.13	3.58	50.6	14
0.9	0.64	2.031	2.51	2.53	26.1	0.9

Considerable improvement in separation was obtained in the given examples with the column tilted at the optimal angle of inclination.

(4) The most important assumption in this work is that the concentrations are locally in equilibrium at every point in the column. Unlike the linear approximation, further approximation to the concentration product, $c\hat{c}$, was made for whole range of concentrations in this study. With the values of $(-A)$ and $(-n)$ given in Tables 1, 2, and 3, we found from Fig. 1 that the further approximation to concentration product is doubtlessly applicable to the whole range of concentrations for practical use.

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SYMBOLS

A	$= H_0L/2K$
a	constant defined by Eq. (13)
B	column width (cm)
b	constant defined by Eq. (12)
c	fractional mass concentration of D_2O in H_2O–HDO–D_2O system
$c\hat{c}$	D_2O pseudoproduct form of concentration defined by Eqs. (2) and (3)
c_F	fractional mass concentration of D_2O in feed stream
c_B, c_T	c obtained at the bottom and top of column, respectively
D	mass diffusivity (cm²/s)
g	gravitational acceleration (cm/s²)
H_0	transport equation defined by Eq. (6) (g/s)
I	improvement in separation defined by Eq. (28)
K	transport coefficient defined by Eq. (7) (g · cm/s)
K_0	transport coefficient defined by Eq. (8) (g · cm/s)
K_1	transport coefficient defined by Eq. (9) (g · cm/s)
K_{eq}	mass-fractional equilibrium constant of H_2O–HDO–D_2O system
L	column length (cm)
n	system constant defined by Eq. (10)
P	system constant defined by Eq. (14)
$\bar{T}$	mean temperature (K)
ΔT	difference in temperature of hot and cold surfaces (°C)
z	axis parallel to the transport direction (cm)

Greek Letters

α_0	reduced thermal diffusion constant
$\bar{\beta}_T$	$= (\partial \rho / \partial T)$ evaluated at $\bar{T}$ (g/cm³ · K)

Δ	$= c_B - c_T$
Δ_0	Δ obtained in Clusius-Dickel column by the method of further approximation
$\Delta_{0,L}$	Δ obtained in Clusius-Dickel column by the method of linear approximation
Δ_i	Δ obtained in inclined column by the method of further approximation
Δ_i'	reduced separation of Δ_i defined by Eq. (23)
Δ_0'	reduced separation of Δ_0 defined by Eq. (27)
$\Delta_{i,\max}$	Δ_i obtained for maximum separation defined by Eq. (21)
$\Delta_{\max}'$	reduced separation of $\Delta_{\max}$ defined by Eq. (25)
$(\Delta_0)_{\text{binary}}$	Δ_0 for binary system defined by Eq. (16)
$(\Delta_i)_{\text{binary}}$	Δ_i for binary system defined by Eq. (17)
θ	angle of inclination of column plate from vertical (degree)
θ^*	angle of inclination for best performance (degree)
$\bar{\rho}$	mass density evaluated at $\bar{T}$ (g/cm^3)
μ	absolute viscosity (g · cm/s)
σ	mass flow rate (g/s)
τ	transport of D_2O along Z-direction (g/s)
ω	half of plate spacing (cm)
σ'	reduced flow rate defined by Eq. (22)

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