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TECHNICAL NOTE

Optimum Plate Spacing for the Best Performance of the Enrichment of Heavy Water in Thermal Diffusion Columns of a Countercurrent-Flow Frazier Scheme

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ABSTRACT

The effect of plate spacing on the degree of separation and production rate for the enrichment of heavy water in a countercurrent-flow Frazier scheme with N flat-plate thermal diffusion columns of same size and with total expense fixed, has been investigated. The equations for estimating optimum plate spacing for maximum separation and maximum production rate have been developed. Considerable improvement in performance for the enrichment of heavy water is obtainable when the‘ thermal diffusion columns with optimum plate spacing are employed for operation. The fact that countercurrent-flow operation is more effective than concurrent-flow operation for the enrichment of heavy water in a Frazier scheme, is also confirmed.

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Key Words: Thermal diffusion; Heavy water; Plate spacing; Frazier scheme; Countercurrent-flow.

INTRODUCTION

It has been recognized that the thermal diffusion process is one of the feasible methods for separation of isotope mixtures. For separation of hydrogen isotopes, this method is particularly attractive because of the large ratio in molecular weights.[1−3] The first complete presentation of the separation theory of thermal diffusion was that of Furry, Jones, and Onsager.[4,5] The enrichment of heavy water by thermal diffusion was studied both theoretically and experimentally.[6,7] In industrial applications, thermal diffusion columns are connected in a series such as that shown in Fig. 1, called the Frazier scheme.[8] The separation theory of thermal diffusion in the Frazier scheme was given by Rabinovich and Suvorov.[9,10]

In addition to the desirable cascading effect, the convective currents arising due to the density difference in a thermogravitational thermal-diffusion column also produce an undesirable remixing effect.[1,2] Therefore, proper control of the convective strength might effectively suppress this undesirable remixing effect while still preserving the desirable cascading effect, and thereby lead to improved separation. One of the feasible ways for properly controlling the convective strength is suitably adjusting the plate spacing.[11] The effect of plate spacing on the degree of separation and production rate for the enrichment of heavy water in flat-plate thermal-diffusion columns of a concurrent-flow Frazier scheme, has been investigated.[12] It is the purpose of

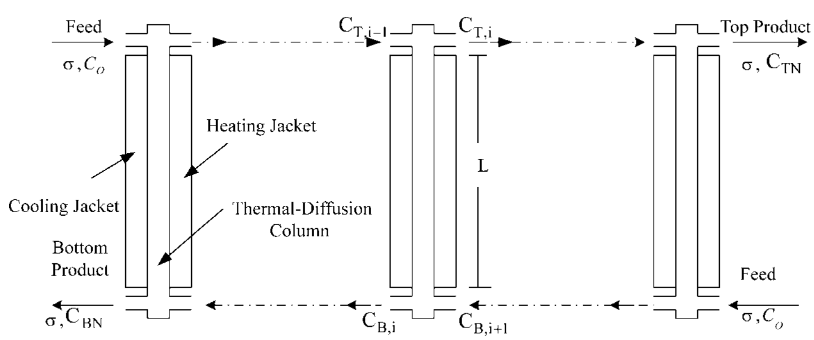

Figure 1. Countercurrent-flow Frazier scheme with N flat-plate thermal-diffusion columns of same size.

this work to investigate the improvement in performance for the enrichment of heavy water in a countercurrent-flow Frazier scheme with N flat-plate thermal-diffusion columns of the same size and with total expense fixed, and the performance efficiencies in a countercurrent-flow scheme employed in the present study will be compared with those in a concurrent-flow scheme employed in the previous work.

SEPARATION THEORY

Equation of Separation

The scheme proposed by Frazier to connect several flate-plate thermo-gravitational thermal-diffusion columns with countercurrent-flow sampling streams is shown in Fig. 1, while Fig. 2 shows the flows and fluxes in the column. All flat-plate columns have the same dimensions of gap (2w), length L, and width B. The separation equation for the enrichment of heavy water from H_2O—HDO—D_2O system in a countercurrent-flow Frazier scheme was derived in a previous work.[13]

$$\Delta = C_{B,1} - C_{T,N} = \frac{2ALN(-H)}{(N+1)K + \sigma L} \tag{1}$$

where

$$\mathrm{H} = \frac{\alpha\rho\beta g(2w)^3 B(\Delta \mathrm{T})^2}{6!\mu \mathrm{T}_m} \tag{2}$$

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

$$\mathrm{A} = C_0\Big[0.05263 - (0.05263 - 0.0135\mathrm{K}_{eq}) - 0.027 \{C_0\mathrm{K}_{eq}[1 - (1 - 0.25\mathrm{K}_{eq})C_0]\}^{1/2}\Big] \tag{4}$$

In obtaining Eq. (1) the assumptions were made that the product form of concentration was approximately considered as a constant A with the value taken at its feed concentration C_0, and that the concentrations of water-isotope

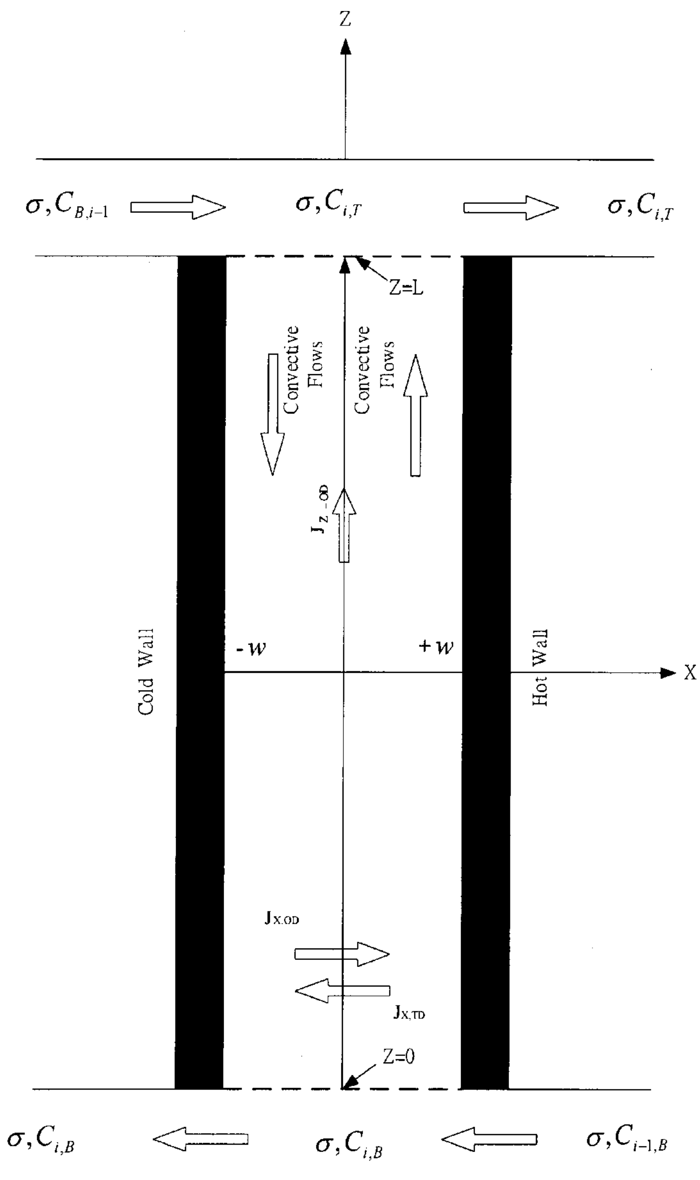

Figure 2. The flows and fluxes in ith column of a countercurrent-flow Frazier scheme.

mixture are locally in equilibrium with equilibrium constant K_{eq} at every point.[6,7]

$$H_2O + D_2O \leftrightarrow 2HDO \tag{5}$$

Separation Equation with the Consideration of Constant Total Expense

Eq. (1) may be rewritten as

$$\Delta = \frac{2\mathrm{A}LN(-a)(2w)^5}{(N+1)b(2w)^9 + \sigma L} \tag{6}$$

where

$$a = \frac{\alpha\rho\beta g B(\Delta\mathrm{T}/2w)^2}{6!\mu \mathrm{T}_m} = \mathrm{H}/(2w)^5 \tag{7}$$

$$b = \frac{\rho\beta^2 g^2 B(\Delta\mathrm{T}/2w)^2}{9!D\mu^2} = \mathrm{K}/(2w)^9 \tag{8}$$

The plate spacing (2w) in a thermal diffusion column is generally so small that changing (2w) will not cause any additional fixed charge. The expenditure of making a separation by thermal diffusion essentially includes two parts: a fixed charge and an operating expense. The fixed charge is roughly proportional to the equipment cost, say mainly the material cost of a thermal diffusion column (BL), while the operating expense is chiefly heat. The heat transfer rate is obtainable from the expression, kBL (ΔT/2w). Based on these terms, we shall take into account the influence of plate-spacing change on the degree of separation and the output with the consideration of constant total expense [i.e., ΔT/2w as well as a and b in Eqs. (7) and (8) are constant].

Maximum Separation

The optimum plate spacing $(2w)_\Delta$ for a maximum separation Δ_{max} is obtained by partially differentiating Eq. (6) with respect to (2w) and setting $\partial\Delta/\partial(2w) = 0$. After differentiation and simplification one obtains

$$(2w)_\Delta = \left[\frac{5\sigma L}{4b(N+1)}\right]^{1/9} \tag{9}$$

Substitution of Eq. (9) into Eq. (6) yields

$$\frac{\Delta_{\max}}{\mathrm{A}} = \left[\frac{8N(-a)}{9\sigma}\right]\left[\frac{5\sigma L}{4b(N+1)}\right]^{5/9} = \frac{8N(-a)(2w)_{\Delta}^{5}}{9\sigma} \tag{10}$$

Maximum Output

The equation of output can be derived by rewriting Eq. (6) as

$$\sigma = \frac{2(-a)N\mathrm{A}(2w)^5}{\Delta} - \frac{b(N+1)(2w)^9}{L} \tag{11}$$

The optimum plate spacing $(2w)_\sigma$ required to obtain the maximum production rate $\sigma_{\max}$ for given scheme which is to give a specified degree of separation Δ, is ready to obtain by partially differentiating Eq. (11) with respect to (2w) and setting $\partial\sigma/\partial(2w) = 0$. The results are

$$(2w)_\sigma = \left[\frac{10(-a)\mathrm{A}NL}{9b(N+1)\Delta}\right]^{1/4} \tag{12}$$

$$\sigma_{\max} = \left[\frac{8(-a)\mathrm{A}N}{9\Delta}\right]\left[\frac{10(-a)\mathrm{A}NL}{9b(N+1)\Delta}\right]^{5/4} = \frac{8(-a)\mathrm{A}N(2w)_{\sigma}^{5}}{9\Delta} \tag{13}$$

The Best Performance for Concurrent-Flow Operation

The effect of plate spacing on the degree of separation and production rate for the enrichment of heavy water in a concurrent-flow Frazier scheme with N flat-plate thermal-diffusion columns of the same size and with total expense fixed, has been investigated in a previous work.[12] Considerable improvement in performance is obtainable when the thermal diffusion columns with optimum plate-spacing are employed for operation. The equation for estimating optimum plate spacing for maximum separation and maximum production rate developed, are

$$(2w)_\Delta = \left[\left(\frac{\sigma L}{2b}\right)(W_\Delta - 1)\right]^{1/9} \tag{14}$$

$$\frac{\dot{\Delta}_{\max}}{\mathrm{A}} = \left(\frac{-aL}{b}\right)\left[\left(\frac{\sigma L}{2b}\right)(W_\Delta - 1)\right]^{-4/9}(1 - W_\Delta^{-N}) \tag{15}$$

$$(2w)_\sigma = \left[\left(\frac{-aL\mathrm{A}}{b\Delta}\right)(1 - W_\sigma)\right]^{1/4} \tag{16}$$

$$\dot{\sigma}_{\max} = \frac{(2b/L)\left[(-aL\mathrm{A}/b\Delta)(1 - W\sigma)\right]^{9/4}}{W_\sigma^{-1/N} - 1} \tag{17}$$

where the parameter values, W_Δ and W_σ, are determined from the following equations:

$$4W_\Delta^{N+1} - (9N + 4)W_\Delta + 9N = 0 \tag{18}$$

$$9W_\sigma^{(N+1)/N} - \left(9 + \frac{4}{N}\right)W_\sigma + \frac{4}{N} = 0 \tag{19}$$

RESULTS AND DISCUSSION

Numerical Example

The improvement in performance resulting from operation at the optimum plate-spacing with total expense fixed, may be illustrated numerically by using the experimental data of the previous work for the enrichment of heavy water (D_2O) in the H_2O-HDO-D_2O system as follows[6]: $\Delta T = 47 - 14 = 33°C$; $T_m = 30.5°C$; $K_{eq} = 3.793$; $(2w) = 0.016 in = 0.0406\,cm$; $L = 177\,cm$; $B = 10\,cm$; $H = -1.473 \times 10^{-4}\,g/s = -0.53\,g/s$; $K = 1.549 \times 10^{-3}\,g\,cm/s = 5.576 gcm/h.$; $A \times 10^2 = 0.359$ $(C_0 = 0.1)$, $0.709(C_0 = 0.3)$, $0.761(C_0 = 0.5)$, $0.591(C_0 = 0.7)$, $0.237(C_0 = 0.9)$.

Accordingly, from Eqs. (7) and (8)

$$-a = \frac{0.53}{(0.0406)^5} = 4.8 \times 10^6\, g/h\, cm^5 \tag{20}$$

$$b = \frac{5.576}{(0.0406)^9} = 1.86 \times 10^{13}\, g/h\, cm^{12} \tag{21}$$

and for constant total expense

$$\frac{\Delta \mathrm{T}}{2w} = \frac{33}{0.0406} = 812.8°C/cm\,(\text{fixed value}) \tag{22}$$

From these values, the maximum separation Δ_{max}, the maximum production rate σ_{max} and their corresponding optimum plate spacings, $(2w)_\Delta$ and $(2w)_\sigma$, are calculated from the appropriate equations. The results are listed in Tables 1 and 2.

The improvement in performance by operating at the optimum plate spacing is best illustrated by calculating the percentage increase in performance based on that obtained at $(2w) = 0.0406\,cm$, i.e.

$$\mathrm{I}_\Delta = \frac{\Delta_{max} - \Delta}{\Delta} \tag{23}$$

$$\mathrm{I}_\sigma = \frac{\sigma_{max} - \sigma}{\sigma} \tag{24}$$

Comparison of Separation

It is seen in Table 1 that the optimum plate spacing, $(2w)\Delta$ for maximum separation, Δ_{max} increases when the flow rate increases, or as the column number decreases. The improvement in separation I_Δ based on the separation obtained at $(2w) = 0.0406$ cm, is achieved, especially for the case that the optimum plate spacing goes far from 0.0406 cm, or for the scheme of large column number N.

The comparisons of separation between concurrent-flow and counter-current- flow operations are also presented in Table 1. It is shown in this table that the separations obtained by countercurrent-flow operation are better than those obtained by concurrent-flow operation either at $(2w) = 0.0406$ cm or at $(2w)_\Delta$, and that $(\Delta - \dot{\Delta})/\dot{\Delta}$ increases when the flow rate σ decreases, or as column number N increases, while $(\Delta_{max} - \dot{\Delta}_{max})/\dot{\Delta}_{max}$ though also increases with N but nearly depends on σ. Further, in some cases, even $\Delta > \dot{\Delta}_{max}$; in other words, the effect of countercurrent flow on Δ operating

Table 1. Comparison of separations obtained at $(2w)_\Delta$ and at $(2w) = 0.0406$ cm; (a) N = 10; (b) N = 20; (c) N = 40.

		Countercurrent-flow operation							
	σ (g/h)	$\mathit{\Delta}/A$	$(2w)_\Delta \times 10^2$ (cm)	$(\Delta T)_\Delta$ (°C)	$\mathit{\Delta}_{max}/A$	I_Δ (%)	$\frac{\mathit{\Delta}-\dot{\mathit{\Delta}}}{\dot{\mathit{\Delta}}}$ (%)	$\frac{\mathit{\Delta}_{max}-\dot{\mathit{\Delta}}_{max}}{\dot{\mathit{\Delta}}_{max}}$ (%)	$\frac{\mathit{\Delta}-\dot{\mathit{\Delta}}_{max}}{\dot{\mathit{\Delta}}_{max}}$ (%)
(a)	0.1	23.72	3.63	29.50	26.68	12.48	42.12	11.35	−1.00
	0.2	19.38	3.92	31.86	19.63	1.29	23.36	11.53	10.11
	0.4	14.19	4.23	34.38	14.41	1.55	10.00	11.53	9.83
	0.8	9.24	4.57	37.14	10.60	14.72	3.36	11.57	−2.74
	3.2	2.99	5.32	43.24	5.72	91.30	0.34	11.50	−41.72
	12.8	0.81	6.20	50.39	3.09	281.48	0	11.55	−70.76
(b)	0.1	27.82	3.37	27.39	37.25	33.90	65.60	12.47	−16.00
	0.2	24.59	3.64	29.59	27.38	11.35	46.89	12.58	1.11
	0.4	19.95	3.93	31.94	20.12	0.85	25.39	12.53	11.58
	0.8	14.49	4.24	34.46	14.79	2.07	10.44	12.47	10.19
	3.2	5.49	4.95	40.23	7.99	45.54	1.10	12.54	−22.68
	12.8	1.57	5.77	46.90	4.31	174.52	0.64	12.24	−59.11
(c)	0.1	30.44	3.13	25.44	51.38	68.79	80.98	13.17	−32.95
	0.2	28.40	3.38	27.47	37.76	32.96	68.85	13.16	−14.89
	0.4	25.04	3.65	29.67	27.75	10.82	49.31	13.22	2.16
	0.8	20.25	3.94	32.02	20.41	0.79	26.64	13.26	12.38
	3.2	9.43	4.60	37.39	11.01	16.76	3.51	13.27	−2.98
	12.8	3.01	5.36	43.57	5.95	97.67	7.89	13.12	−42.78

Table 2. Comparison of production rates obtained at $(2w)_\sigma$ and at $(2w) = 0.0406$ cm; (a) N = 10; (b) N = 20; (c) N = 40.

		Countercurrent-flow operation							
	$\dot{\Delta}/A$ or Δ/A	σ (g/h)	$(2w)_\sigma \times 10^2$ (cm)	$(\Delta T)\sigma$ (°C)	σ_{max} (g/h)	I_σ (%)	$\frac{\sigma-\dot{\sigma}}{\dot{\sigma}}$ (%)	$\frac{\sigma_{max}-\dot{\sigma}_{max}}{\dot{\sigma}_{max}}$ (%)	$\frac{\sigma-\dot{\sigma}_{max}}{\dot{\sigma}_{max}}$ (%)
(a)	1	10.24	8.34	66.97	162.23	1494.04	0	27.77	−91.94
	2	4.95	6.93	56.33	34.11	589.09	0.20	27.80	−81.45
	4	2.30	5.83	47.39	7.18	212.17	0.44	27.99	−59.00
	8	0.98	4.90	39.83	1.51	54.08	4.26	27.95	−16.95
	16	0.32	4.12	33.49	0.32	0	77.78	28.00	80.00
(b)	1	20.52	8.34	67.79	343.90	1577.92	0.05	30.80	−92.20
	2	9.93	7.01	56.98	72.22	627.29	0.10	30.67	−82.03
	4	4.63	5.90	47.96	15.25	229.37	0.43	31.24	−60.15
	8	1.99	4.96	40.31	3.20	60.80	3.65	31.15	−18.44
	16	0.66	4.17	33.89	0.67	1.52	69.23	31.37	29.41
(c)	1	41.07	8.38	68.11	705.30	1617.31	0.02	31.77	−92.33
	2	19.89	7.05	57.30	148.62	647.21	0.15	32.08	−82.32
	4	9.30	5.93	48.20	31.29	236.45	0.65	32.30	−60.68
	8	4.00	4.99	40.56	6.60	65.00	3.36	32.80	−19.52
	16	1.36	4.19	34.06	1.38	1.47	70.00	31.43	29.52

at (2w) = 0.0406 cm may overcome the effect of concurrent flow even operating at its optimum plate-spacing.

Comparison of Production Rate

The production rates calculated at $(2w)_\sigma$ and at (2w) = 0.0406 cm for countercurrent-flow and concurrent-flow operations with various values of $\Delta/A(or\dot{\Delta}/A)$ are listed in Table 2. It is noted in this table that the optimum plate spacing, $(2w)_\sigma$, for maximum production rate, σ_{max} increases when the degree of separation decreases, but nearly unchanged with column number. The improvement in production rate, I_σ, based on the production rate obtained at (2w) = 0.0406 cm, is achieved, especially for the case that the optimum plate spacing goes far from 0.0406 cm, or for the scheme having larger column number.

The comparison of production rates between concurrent-flow and countercurrent-flow operations is also illustrated in Table 2. It is found in this table that the production rates obtained by countercurrent-flow operation are better than those obtained by concurrent-flow operation either at (2w) = 0.0406 cm or at $(2w)_\sigma$, and that $(\sigma - \dot{\sigma})/\dot{\sigma}$ increases when $\Delta(or\dot{\Delta})$ increases, while $(\sigma_{max} - \dot{\sigma}_{max})/\dot{\sigma}_{max}$ nearly does not depend on (Δ/A), or on ($\dot{\Delta}$/A). Further, in some cases, even $\sigma > \dot{\sigma}_{max}$; it means that the effect of countercurrent flow on σ operating at (2w) = 0.0406 cm may overcome the effect of concurrent flow even operating at its optimum plate spacing.

CONCLUSION

It has been shown that for the enrichment of heavy water in a thermal-diffusion column, proper control of convective strength by suitably adjusting the plate spacing may effectively suppress the undesirable remixing effect while still preserving the desirable cascading effect, resulting in substantial improvement in performance. Further, a Frazier scheme operating with countercurrent flow of feed is better than the one operating with concurrent flow.

The equations of the optimum plate spacing for maximum separation and maximum output for the enrichment of heavy water from H_2O-HDO-D_2O system by thermal diffusion in a countercurrent-flow Frazier scheme with total expense fixed, have been derived. A numerical example with the use of the experimental data obtained in a previous work[6] is given. Considerable improvement in performance is obtained by operating at the optimum plate-spacing as shown in Tables 1 and 2; the performance can be further improved in

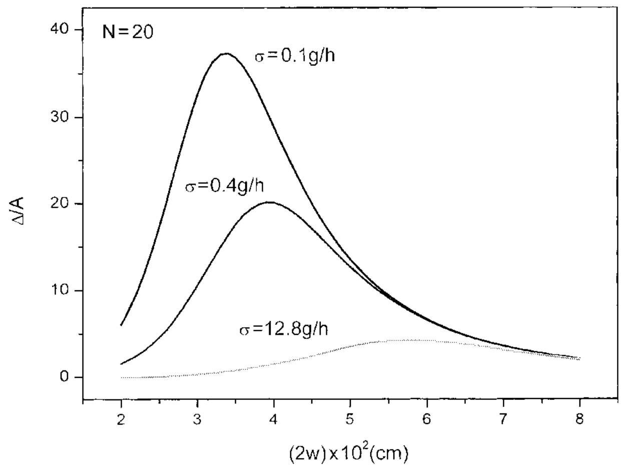

Figure 3. Δ/A vs. $(2w)$ for N = 20 with σ as parameter.

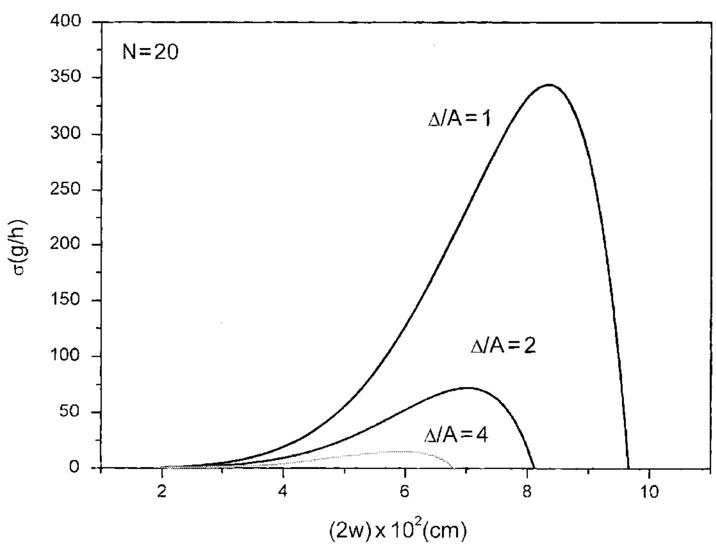

Figure 4. σ vs. $(2w)$ for N = 20 with Δ/A as parameter.

a countercurrent-flow scheme, rather than in a concurrent-flow one, as also shown in these tables. In some cases, the effect of countercurrent flow on performance operating at $(2w) = 0.0406\,\mathrm{cm}$ even overcomes the effect of concurrent flow operating at its optimum plate spacing.

The practical plate spacing in a thermal-diffusion column actually is very small and its change is sensitive to the performance. For a designer, who may be also concerned about the sensitivity study for the plate spacing, more parameter studies on plate spacing were made and the results are shown in Figs. 3 and 4. It is seen from these figures that both the degree of separation and the production rate are very sensitive to the distance between two adjacent plates for low σ and high Δ/A, respectively, and that the optimizations are confirmed with those in Tables 1 and 2.

NOMENCLATURE

A	product form of concentration defined by Eq. (4)
a	a constant defined by Eq. (7) ($\mathrm{g\,h^{-1}\,cm^{-5}}$)
B	column width (cm)
b	a constant defined by Eq. (8) ($\mathrm{g\,h^{-1}\,cm^{-12}}$)
C	fractional mass concentration of heavy water (D_2O)
C_i	C in ith column
$C_{i,B}$,$C_{i,T}$	C_i in the product streams exiting from bottom and top ends, respectively
C_0	C in the feed stream
D	ordinary diffusion coefficient ($\mathrm{cm^2\,s^{-1}}$)
g	gravitational acceleration ($\mathrm{cm^2\,s^{-1}}$)
H	system const defined by Eq. (2) ($\mathrm{g\,s^{-1}}$)
I_Δ,I_σ	improvement in performance defined by Eqs. (23) and (24), respectively
$J_{X,OD}$,$J_{X,TD}$	mass flux of heavy water in x direction due to ordinary diffusion, due to thermal diffusion ($\mathrm{g\,cm^{-2}\,s^{-1}}$)
K	system constant defined by Eq. (3) ($\mathrm{g\,cm\,s^{-1}}$)
K_{eq}	mass equilibrium constant of H_2O-HDO-D_2O system
L	column length (cm)
$J_{Z,OD}$	mass flux of heavy water in z direction due to ordinary diffusion ($\mathrm{g\,cm^{-2}\,s^{-1}}$)
N	column number of a Frazier scheme
T_m	mean absolute temperature (K)
ΔT	difference in temperature between hot and cold plate (K)

$(\Delta T)_\Delta,(\Delta T)_\sigma$	ΔT for maximum separation, for maximum production rate (K)
W_Δ,W_σ	value determined from Eq. (18), from Eq. (19)
Z	axis of transport direction (cm)
α	thermal diffusion constant of water-isotopes mixture, <0
β	$-(\partial\rho/\partial T)_P$ evaluated at T_m ($g\,cm^{-3}\,K^{-1}$)
Δ	$C_{B,1} - C_{T,N}$, concentration difference of the product streams in a countercurrent-flow Frazier scheme
$\dot{\Delta}$	$C_{B,N} - C_{T,N}$, concentration difference of the product streams in a concurrent-flow Frazier scheme
$\Delta_{max},\dot{\Delta}_{max}$	maximum value of Δ or, $\dot{\Delta}$, obtained at $(2w)_\Delta$
2w	plate spacing (cm)
$(2w)_\Delta,(2w)_\sigma$	optimum value of (2w) for maximum separation, for maximum production rate (cm)
ρ	mass density evaluated at T_m ($g\,cm^{-3}$)
μ	absolute viscosity evaluated at T_m ($g\,cm^{-1}\,s^{-1}$)
$\sigma,\dot{\sigma}$	mass flow rate ($g\,s^{-1}$) in a countercurrent-flow scheme, in a concurrent-flow scheme
$\sigma_{max},\dot{\sigma}_{max}$	maximum value of σ, of $\dot{\sigma}$ obtained at $(2w)_\sigma$ ($g\,s^{-1}$)

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