settling suspension) and the analogy between momentum and mass transfer:

$$\frac{\bar{k}_L}{k_w} = 3.48 \left\{ \frac{V^2}{gD_T} \sqrt{\frac{gD_p}{(\rho_p/\rho_w - 1)W^2}} \right\}^{-0.81} \quad (5)$$

Figure 4 shows a comparison of Eq. (5) with the observed values shown in Fig. 1. The weighted average settling velocity W in Eq. (5) was evaluated for a particle falling in a stagnant liquid. From the figure, the observed values deviate remarkably from Eq. (5) for the tube of small diameter and at high slurry velocities. Equation (5) is not always applicable to the experimental results obtained under flow conditions where no saltation was observed.

Acknowledgment

The authors would like to thank Mitsubishi Chemical Industries Ltd. and the Asahi Glass Foundation for Industrial Technology for their financial assistance. The assistance of student colleagues (Messrs. T. Yasumoto, T. Henmi, M. Isé, H. Funayama, K. Suzuki, H. Abé and H. Mabusé) is also appreciated.

Nomenclature

C_D	= drag coefficient of single particle moving at relative velocity U_r	[—]
$\mathcal{D}$	= diffusion coefficient	[m ² /s]
D_p	= particle diameter	[m]
D_T	= tube diameter	[m]
f	= friction factor for smooth tube ($=0.082Re_T^{-1/4}$)	[—]
g	= acceleration of gravity	[m/s ²]
$\bar{k}_L$	= average liquid-side mass transfer coefficient between particles and liquid	[m/s]
k_w	= mass transfer coefficient between tube wall and liquid	[m/s]

L_T	= tube length	[m]
m_d	= delivered concentration of particles [volume fraction]	
Re_T	= Reynolds number ($= (U_w)_{av} D_T / \nu$)	[—]
Sc	= Schmidt number ($= \nu / \mathcal{D}$)	[—]
Sh	= Sherwood number ($= \bar{k}_L D_p / \mathcal{D}$)	[—]
U_r	= relative velocity between particles and liquid	[m/s]
$(U_w)_{av}$	= average liquid velocity across cross-section of tube	[m/s]
V	= average velocity of suspension	[m/s]
W	= weighted average settling velocity	[m/s]
ϵ	= energy dissipation rate per unit mass of liquid defined by Eq. (1)	[m ² /s ³]
ϵ_{f0}	= energy dissipation rate per unit mass of liquid defined by Eq. (2)	[m ² /s ³]
ϵ_{s0}	= energy dissipation rate per unit mass of liquid defined by Eq. (3)	[m ² /s ³]
ν	= kinematic viscosity	[m ² /s]
ρ_w	= liquid density	[kg/m ³]
ρ_p	= particle density	[kg/m ³]

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(Presented in part at 2nd Pacific Chem. Eng. Congress, Denver, U.S.A., Aug., 1977.)

A METHOD FOR SOLVING DESIGN PROBLEM OF MULTICOMPONENT DISTILLATION

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A further rigorous method is proposed for determining the number of plates in enriching and stripping sections, n and s , required for a specified doublet separation in multicomponent distillation by use of an ordinary column. This method employs the same approach proposed by Murdoch and Holland³⁾ (M-H method) to assign the degrees of freedom as the

design problem, and to determine n and s by a condition of minimum total number of plates, $n+s \rightarrow \min$. However, the proposed method is characterized by an accurate determination of an extremely small amount of molar rates of nondistributed components existing in the distillate (heavier than the heavy key) and bottoms (lighter than the light key) in the plate-to-plate calculation procedure⁵⁾ within allowable significant figures of the computer, where the molar rates are

Received September 19, 1981. Correspondence concerning this article should be addressed to I. Yamada.

determined by the condition of "plate-matching".

1. Plate-to-Plate Calculation Procedure⁵⁾

Denoting that the plates are numbered toward the feed plate for both sections of the column, the equations used in the plate-to-plate calculation procedure are as follows:

Enriching section

$$y_{j+1,i} = ax_{j,i} + bx_{D,i} \quad (1)$$

$$x_{j,i} = \frac{y_{j,i} - bx_{D,i}(1 - E_{j,i}^{MV})}{K_{j,i}E_{j,i}^{MV} + a(1 - E_{j,i}^{MV})} \quad (2)$$

$$\sum_i \frac{y_{j,i} - bx_{D,i}(1 - E_{j,i}^{MV})}{K_{j,i}E_{j,i}^{MV} + a(1 - E_{j,i}^{MV})} = 1 \quad (3)$$

where $a \equiv R/(R+1)$, $b \equiv 1/(R+1)$, and $E_{j,i}^{MV}$ denotes the Murphree plate efficiency for the vapor phase. When an extremely small amount of $x_{D,i}$ for the non-distributed component is once assigned, $y_{1,i}$ is determined by the boundary condition at the condenser, and $x_{1,i}$ can be calculated from Eq. (2) with $K_{1,i}$ obtained by solving Eq. (3). Then $y_{2,i}$ is calculated by Eq. (1). These alternating calculations can be continued toward the feed plate as $j=2, 3 \dots f$.

Stripping section

$$x_{j+1,i} = a^*y_{j,i} + b^*x_{W,i} \quad (4)$$

$$y_{j,i} = y_{j-1,i} + E_{j,i}^{MV}(K_{j,i}x_{j,i} - y_{j-1,i}) \quad (5)$$

where $a^* \equiv R^*/(R^*-1)$, $b^* \equiv 1/(R^*-1)$. The plate-to-plate procedure in this section can be easily continued without any complicated procedure as mentioned for the enriching section, since the values of $x_{j,i}$ and $y_{j-1,i}$ which are required in the calculation of Eq. (5) have already been obtained by the previous step, where $y_{W,i} = K_{W,i}x_{W,i}$ in accordance with the boundary condition at the bottoms. That is, the bottoms is to be an ideal stage.

2. Assignment of Molar Rates of Nondistributed Component

The component i is numbered in order of reciprocal relative volatility, and h and l denote the heavy and light key components, respectively. Usually, the molar rates of nondistributed components $Dx_{D,i}$ ($i < h$) and $Wx_{W,i}$ ($i > l$), which cannot be specified previously as the design type of multicomponent distillation, are treated as zero^{1,2)}. However, if these are once set at zero, the values of $x_{j,i}$ for the nondistributed component, resulting from the plate-to-plate procedure are always zero throughout the column. This result obviously conflicts with the actual composition profile in the column. Therefore, one must consider a certain extremely small amount of unspecified $Dx_{D,i}$ ($i < h$) and $Wx_{W,i}$ ($i > l$) within a allowable significant figures of computer, and these rates must be determined by

Table 1 Specification for doublet separation of ternary and quaternary systems

(All runs are specified by $q=1.0$, $R=1.3R_m$)

Run	i	$\alpha(i)$	$F \cdot x_F(i)$	$D \cdot x_D(i)$	$W \cdot x_W(i)$	Key	Ref.
1)	1	1.0	30.0	—	30.0		
	2	2.0	40.0	1.0	39.0	h	
	3	3.0	30.0	24.0	6.0	l	
2)	1	1.0	20.0	0.1	19.9	h	
	2	2.0	5.0	4.9	0.1	l	
	3	3.0	75.0	75.0	—		$q=1.0$
3)	1	1.0	20.0	—	20.0		$R=R_m \times 1.3$
	2	1.4	30.0	—	30.0		
	3	2.4	30.0	0.2	29.8	h	
	4	3.5	20.0	19.8	0.2	l	
4)	1	1.0	20.0	0.2	19.8	h	
	2	1.4	30.0	29.8	0.2	l	
	3	2.4	30.0	30.0	—		
	4	3.5	20.0	20.0	—		
5)	1	1.0	20.0	—	20.0		
	2	1.4	30.0	0.5	29.5	h	
	3	2.4	30.0	29.5	0.5	l	
	4	3.5	20.0	20.0	—		
6)	1	1.0	50.0	—	50.0		
	2	1.4	10.0	1.0	9.0	h	
	3	2.4	20.0	19.0	1.0	l	
	4	3.5	20.0	20.0	—		
7)	1	1.0	20.0	0.25	19.75	h	
	2	1.4	10.0	9.75	0.25	l	
	3	2.4	20.0	20.0	—		
	4	3.5	50.0	50.0	—		
8)	1	1.0	50.0	—	50.0		
	2	1.4	25.0	1.0	24.0	h	
	3	2.4	15.0	14.0	1.0	l	
	4	3.5	10.0	10.0	—		

— denotes the nondistributed component. R_m was determined by Underwood's method⁴⁾.

a condition of "plate-matching" written by the following equations.

$$g_i \equiv [y_{f,i}]_D - [y_{f,i}]_W = 0 \quad (i \neq h, l) \quad (6)$$

$$g_h = \left[\frac{y_{f,h}}{y_{f,h}} \right]_D - \left[\frac{y_{f,h}}{y_{f,h}} \right]_W = 0 \quad (7)$$

where $[]_D$ and $[]_W$ denote the values obtained by the plate-to-plate procedure for enriching and stripping sections, respectively.

3. Calculation Steps for Determining n and s

By the definition of component numbering, doublet separation means $l=h+1$; that is, no split key exists between h and l . Calculation steps are as follows:

- 1) Assume n
- 2) Assume $Dx_{D,i}$ ($i < h$) and $Wx_{W,i}$ ($i > l$)
- 3) Calculate $(y_{f,i})_D$ and $(y_{f,i}/y_{f,h})_D$
- 4) Repeat the plate-to-plate procedure in the stripping section until Eq. (7) is satisfied, and determine s and $(y_{f,i})_W$ ($i > l$) simultaneously.
- 5) Judge Eq. (6). If this equation is satisfied within a given accuracy for all components, the calculation to determine s , $Wx_{W,i}$ ($i > l$) and $Dx_{D,i}$ ($i < h$)

Table 2 Calculation results for number of plates in each section

Run	Murphree plate efficiency	n	L-M method s	$n+s$	$n/(n+s)$	n	Proposed method s	$n+s$	$n/(n+s)$
1	1.0	14.4	10.2	24.6	0.585	15.7	9.0	24.7	0.636
	0.5	29.0	18.8	47.8	0.607	31.0	17.0	48.0	0.646
2	1.0	20.1	7.8	27.9	0.720	17.0	9.8	26.8	0.634
	0.5	41.4	12.4	53.8	0.770	38.0	17.9	55.9	0.680
3	1.0	20.0	27.5	47.5	0.421	22.3	25.4	47.7	0.468
	0.5	41.2	52.0	93.2	0.442	44.7	48.8	93.5	0.478
4	1.0	32.3	21.7	54.0	0.598	29.4	24.1	53.5	0.550
	0.5	65.9	40.6	106.5	0.619	63.0	43.0	106.0	0.594
5	1.0	14.4	14.0	28.4	0.507	15.0	15.0	30.0	0.500
	0.5	29.6	25.4	55.0	0.538	30.0	28.5	58.5	0.513
6	1.0	6.4	10.5	16.9	0.379	8.2	11.4	19.6	0.418
	0.5	13.4	18.6	32.0	0.419	15.6	21.6	37.2	0.419
7	1.0	—	—	—	—	26.0	19.9	47.9	0.543
	0.5	—	—	—	—	50.0	39.0	89.0	0.562
8	1.0	8.3	12.2	20.5	0.405	10.3	12.1	22.4	0.460
	0.5	17.2	22.0	39.2	0.439	19.9	22.7	42.6	0.467

— not converged

has just converged; then go to the next step. Otherwise return to 4) after correcting $Dx_{D,i}$ ($i < h$) and $Wx_{W,i}$ ($i > l$) where the Regula-Falsis method or the conjugated gradient method is useful in finding the corrected values.

6) Repeat 1)–5) for various n

7) Determine n and s by $n+s \rightarrow \min$.

The calculation steps mentioned in 1)–6) are based on n assumed. When s is assumed the relationship between enriching and stripping sections is reversed. The selection of assumption of n or s depends on the number of nondistributed components in distillate and bottoms. If the number of nondistributed components in distillate is less than that in bottoms, n is assumed, otherwise s is assumed. Furthermore, when the ideal plate is assumed, that is, $E_{j,i}^M = 1$, the equations describing the plate-matching are given as follows:

$$g_i \equiv [x_{f,i}]_D - [x_{f,i}]_W = 0 \quad (i \neq h, l) \quad (8)$$

$$g_k \equiv \left[\frac{x_{f,l}}{x_{f,h}} \right]_D - \left[\frac{x_{f,l}}{x_{f,h}} \right]_W = 0 \quad (9)$$

4. Discussion and Conclusion

Some numerical examples are illustrated to demonstrate the superiority of the proposed method over the method of Lewis and Matheson²⁾ (L–M method), in which the number of plates in each section is determined by the following relationships.

$$\left[\frac{x_{f,l}}{x_{f,h}} \right]_D = \frac{x_{FL,l}}{x_{FL,h}} \quad \text{for } n \quad (10)$$

$$\left[\frac{x_{f,l}}{x_{f,h}} \right]_W = \frac{x_{FL,l}}{x_{FL,h}} \quad \text{for } s \quad (11)$$

For the doublet separation of ternary and quaternary systems as specified in Table 1, the values of n , s and $n+s$ calculated are listed in Table 2. As observed in Table 2, one will fail to obtain the converged solution by the L–M method for a specification of $D \ll F$ or $W \ll F$ (See Run 7), whereas the proposed method always gives the stable solution for any complicated specification.

Nomenclature

D	= molar rate of distillate	[mol/hr]
$E_{j,i}^M$	= Murphree plate efficiency of component i at plate j	[—]
F	= molar rate of feed	[mol/hr]
$K_{j,i}$	= K -value for component i at temperature of plate j	[—]
n	= number of plates in enriching section	[—]
R	= reflux ratio	[—]
s	= number of plates in stripping section	[—]
W	= molar rate of bottoms	[mol/hr]
$x_{j,i}$	= mole fraction of component i in liquid leaving plate j	[—]
$y_{j,i}$	= mole fraction of component i in vapor leaving plate j	[—]
$y_{j,i}^*$	= mole fraction of component i in vapor phase in equilibrium with liquid phase leaving plate j	[—]

<Subscripts>

D	= distillate
F	= feed
FL	= liquid portion of feed
f	= feed plate
h	= heavy key component
i	= component number
j	= plate number
l	= light key component
W	= bottom product

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APPLICATION OF VACANCY SOLUTION THEORY TO ADSORPTION FROM DILUTE AQUEOUS SOLUTIONS

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In the design of purification facilities for removing organic pollutants from wastewaters by using activated carbon, equilibrium adsorption information is required. Since wastewaters contain more than one component, the desired equilibria are for multiple solutes rather than for a single solute. However, most of the published papers in this field deal with adsorption for removing a single solute from water. In the present study, the vacancy solution theory which has been proposed to predict gas mixture adsorption equilibria is applied to the adsorption equilibria of aqueous solutions in order to develop a new correlation method.

The vacancy solution theory, proposed by Suwanayuen and Danner^{3,4}, is found to be very useful for correlating single gas adsorption isotherms and gas mixture adsorption equilibria. By replacing the vacuum space with water molecule as a definition of "vacancy" in the theory, one can readily derive an adsorption isotherm equation for dilute aqueous solutions. In the derivation, the Gibbs adsorption equation for dilute solutions, $RT \ln c_i^\circ = (A/n_i^\circ) d\pi$, was used and the Wilson equation was adopted to describe the non-ideality in the adsorbed phase. The adsorption isotherm equation obtained for a dilute aqueous solution containing a single solute i is as follows.

$$c_i^\circ = \left[\frac{n_i^{\circ,\infty}}{H_i} \frac{\theta_i}{1-\theta_i} \right] \left[A_{i3} \frac{1-(1-A_{3i})\theta_i}{A_{i3}+(1-A_{i3})\theta_i} \right] \times \exp \left[-\frac{A_{3i}(1-A_{3i})\theta_i}{1-(1-A_{3i})\theta_i} - \frac{(1-A_{i3})\theta_i}{A_{i3}+(1-A_{i3})\theta_i} \right] \quad (1)$$

where Henry's Law constant, H_i , is defined by

$$H_i = \lim_{c_i^\circ \rightarrow 0} \left(\frac{n_i^\circ}{c_i^\circ} \right) \quad (2)$$

and

$$\theta_i = \frac{n_i^\circ}{n_i^{\circ,\infty}} \quad (3)$$

Four independent parameters, $n_i^{\circ,\infty}$, H_i , A_{i3} , and A_{3i} , in Eq. (1) should be determined empirically by fitting the experimental data. A least-squares fitting procedure was used to obtain these isotherm parameters from the single-solute adsorption data reported by Radke and Prausnitz¹. The values of the four parameters are listed in Table 1. The results are sensitive to selection of $n_i^{\circ,\infty}$ and A_{3i} among these parameters. If the adsorbed phase is assumed to be ideal (i.e., $A_{i3}=A_{3i}=1$), Eq. (1) is the same as the Langmuir adsorption isotherm. Every set of Wilson's parameters shown in Table 1 shows a negative deviation from the ideal-solution law (i.e., $\gamma_i, \gamma_3 < 1$). The deviation becomes large, with increasing values of A_{i3} and A_{3i} .

The vacancy solution theory can be extended to multicomponent systems. The adsorption equilibrium equation for a dilute aqueous multi-solute system is represented by

$$c_i = \gamma_i z_i n_T \frac{n_i^{\circ,\infty} A_{i3}}{n_T^\infty H_i} \exp(A_{3i}-1) \exp\left(\frac{\pi \bar{a}_i}{RT}\right) \quad (4)$$

where

$$\frac{\pi \bar{a}_i}{RT} = - \left[1 + \frac{n_T^\infty - n_i^{\circ,\infty}}{n_T} \right] \ln \gamma_3 x_3 \quad (5)$$

and

$$\ln \gamma_i = 1 - \ln \left(\sum_j x_j A_{ij} \right) - \sum_k \left(\frac{x_k A_{ki}}{\sum_j x_j A_{kj}} \right) \quad (6)$$

with

$$n_T^\infty = \sum_i z_i n_i^{\circ,\infty} \quad \text{and} \quad n_T = \sum_i n_i \quad (i=1, 2) \quad (7)$$

Received October 23, 1981. Correspondence concerning this article should be addressed to Y. Arai.