EXPERIMENTAL APPROACH TO MASS TRANSFER IN BINARY PACKED COLUMN DISTILLATION

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Experimental studies of mass transfer in binary packed column distillation were made for the acetone-ethanol, acetone-methanol, methanol-ethanol and methanol-water systems under total reflux conditions for wide ranges of vapor flow rate and liquid concentration.

Measurements of the wetted area of the packings were made and the data were compared with Onda's correlation. Diffusion fluxes under low mass flux conditions with a short packed column were examined. The effect of condensation of mixed vapors or evaporation from liquids on the diffusion fluxes were studied.

A new method for predicting the separation performance of binary packed column distillation was developed with correlations for the diffusion fluxes under high mass flux conditions with a short column and for the wetted area of the packings. Predicted axial distributions of reflux flow rates and liquid concentrations in a long packed column by the present method were compared with the observed values.

Introduction

Mass transfer in binary packed column distillation is a very complicated phenomenon, where interactions between mass and heat fluxes take place through the boundary conditions at the vapor-liquid interface, and the vapor-liquid contacting area is affected by the liquid flow rates and concentrations.

In our previous work,[8−10,12] experimental studies were made of heat and mass transfer in binary and ternary distillation by wetted-wall columns. These showed that mass and heat transfer in the distillation column were vapor-phase controlled and that the effect of convective mass fluxes caused by partial condensation of mixed vapors on the diffusion fluxes could not be neglected. The purpose of the present work is to make an experimental approach to mass transfer in binary packed column distillation under total reflux conditions for wide ranges of vapor flow rate and liquid concentration, and to develop a new method for predicting the separation performance of binary packed column distillation with consideration of the effect of interaction between heat and mass fluxes.

1. Experimental Apparatus and Procedures

1.1 Experimental apparatus

Figure 1 shows a schematic diagram of the experimental apparatus. The test sections, packed with 3-mm Dixon packings, were 50 mm- and 600 mm-long,

22 mm-inner diameter glass packed columns with Swagelok joints at the top and bottom. For measurements of reflux temperatures, two 1 mm-outer diameter sheathed Chromel-Alumel thermocouples were attached at the top and bottom of the columns. On the wall of the 600 mm-long column, sampling taps were attached at 50 mm intervals from the bottom of the column for measurements of liquid concentrations.

1.2 Measurements

1) Wetted area of packings: Measurements of the wetted area of the packings were made at room temperatures by measuring the colored area of the
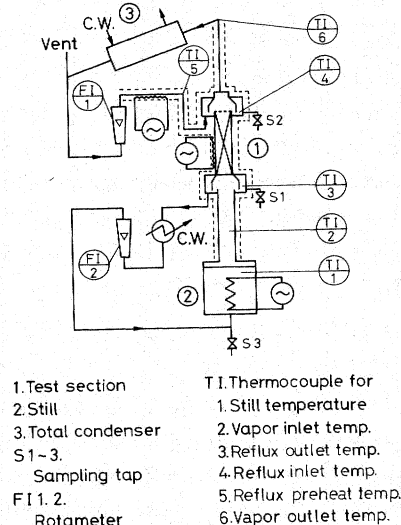

1.Test section	T I.Thermocouple for
2.Still	1. Still temperature
3.Total condenser	2.Vapor inlet temp.
S1~3.	3.Reflux outlet temp.
Sampling tap	4.Reflux inlet temp.
FI 1. 2.	5.Reflux preheat temp.
Rotameter	6.Vapor outlet temp.

Fig. 1. Schematic diagram of experimental apparatus

packings after running with binary solutions of various liquid concentrations mixed with a small amount of a dye, alcohol fluorescent green U.[2]

2) Interfacial area: The interfacial area for mass and heat transfer in packed column distillation, a_s, was taken equal to the wetted area of the packings, a_w, according to the suggestions by Onda et al.[18]

3) Reflux flow rates and concentrations: The bottom and top reflux flow rates, L_b, L_t, were measured by two precision rotameters within 1% error. The bottom and top liquid concentrations, ω_{LAb}, ω_{LAt}, were measured by a gas chromatograph with an accuracy better than 0.1 mol%.

4) Mass and diffusion fluxes: The observed average vapor-phase mass fluxes of each component, N_A, N_B, were determined from the bottom and top reflow rates and concentrations by:

$$N_A = (L_t\omega_{LAt} - L_b\omega_{LAb})/a_s V \quad (1a)$$

$$N_B = (L_t(1 - \omega_{LAt}) - L_b(1 - \omega_{LAb}))/a_s V \quad (1b)$$

where a_s is the interfacial area per unit volume. The observed average vapor-phase diffusion fluxes of the more volatile component, J_{As}, were obtained by[8,9]:

$$J_{As} = N_A - \rho_{Gs}v_s\omega_{GAs} = N_A - (N_A + N_B)\omega_{GAs} \quad (2)$$

where ω_{GAs} is the average vapor concentration at the vapor-liquid interface. The average vapor-phase diffusion fluxes were made dimensionless by[8,9]:

$$Sh_{GA}(J_{As}/N_A) = J_{As}D_p/\rho_{Gs}\mathscr{D}_{GAB}\Delta\omega_{GA} \quad (3)$$

where $\Delta\omega_{GA}$ is the average concentration driving force, defined as:

$$\Delta\omega_{GA} = \frac{1}{\omega_{GAt} - \omega_{GAb}} \int_{\omega_{GAb}}^{\omega_{GAt}} (\omega_{GA}^* - \omega_{GA})d\omega_{GA} \quad (4)$$

1.3 Vapor-liquid equilibria and physical properties

Vapor-liquid equilibria of the acetone-ethanol, acetone-methanol, methanol-ethanol and methanol-water systems were estimated from the vapor pressures of pure components by Antoine's equation and from the liquid-phase activity coefficients by Wilson's equation, where Antoine's constants and Wilson's parameters for the acetone-ethanol system by Gmehling et al.[5] and for the other binary systems by Holmes et al.[6] were used. The surface tensions of nonaqueous solutions and of pure organic liquids were estimated by Macleod-Sugden's method[20] and those for the methanol-water solutions by Tamura's method.[20] The thermal conductivities and the viscosities of mixed vapors were estimated by Wilke's method.[20] The binary diffusion coefficients and the viscosities of pure vapors were estimated by Hirschfelder's method[20] and the thermal conductivities of pure vapors by Eucken's method.[20]

Table 1. Ranges of variables for measurements of wetted area of the packings

System	Z [m]	x_A [—]	σ_L [N/m]	Re_L [—]
Acetone-ethanol	0.05	0.20–0.81	0.024–0.026	0.07–0.78
Acetone-methanol	0.05	0.55	0.023	0.08–0.92
Methanol-ethanol	0.05	0.50	0.024	0.03–0.33
Methanol-water	0.05	0.10—0.77	0.024–0.051	0.04–0.61
	0.60	0.52	0.028	0.27

Table 2. Ranges of variables for binary packed column distillation

System	Z [m]	x_{Ab} [—]	Re_L [—]	Re_G [—]	Sc_{Gs} [—]
Acetone-ethanol	0.05	0.15–0.75	0.11–1.08	40–380	0.66–0.80
	0.60	0.02–0.11	0.19–0.46	72–175	0.69
Acetone-methanol	0.05	0.22–0.52	0.11–0.90	34–260	0.62–0.75
Methanol-ethanol	0.05	0.17–0.76	0.08–0.82	32–280	0.68–0.89
	0.60	0.16–0.34	0.09–0.73	33–260	0.79–0.84
Methanol-water	0.05	0.16–0.75	0.15–0.74	41–200	0.54–0.62
	0.60	0.13–0.20	0.09–0.61	83–220	0.58

1.4 Ranges of variables

Measurements of the wetted area of the packings were made for binary solutions of the acetone-ethanol, acetone-methanol, methanol-ethanol and methanol-water systems with the 50 mm- and 600 mm-long columns for wide ranges of liquid concentrations at room temperratures. The ranges of variables for the measurements of the wetted area are summarized in **Table 1**.

Binary distillation runs were made for the acetone-ethanol, acetone-methanol, methanol-ethanol and methanol-water systems under total reflux conditions with 50 mm- and 600 mm-long columns over wide ranges of vapor flow rates and liquid concentrations. **Table 2** shows the ranges of variables for the binary distillation runs.

2. Experimental Results

2.1 Measurements of wetted area

Figure 2 shows the observed wetted area of the packings for methanol-water solutions of various liquid concentrations ($x_A = 0.10, 0.25, 0.51, 0.77$) with the 50 mm-long column. The ordinate is the ratio of the wetted area to the total area, a_w/a_t, and the abscissa is the liquid-phase Reynolds number, Re_L. A systematic increase in the wetted area was observed with decrease of the surface tension, σ_L, and with increase of Re_L.

Figure 3 compares the observed wetted area for the acetone-ethanol, acetone-methanol, methanol-ethanol and methanol-water systems with Onda's correlation:[17]
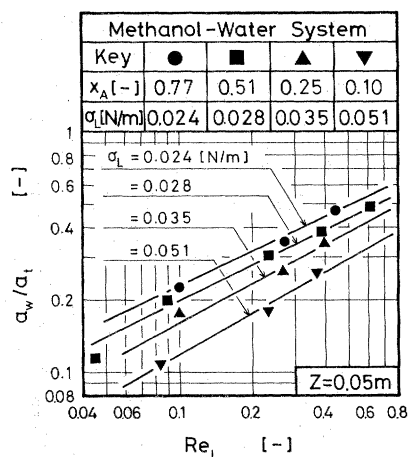
Methanol–Water System				
Key	●	■	▲	▼
x_A [-]	0.77	0.51	0.25	0.10
σ_L [N/m]	0.024	0.028	0.035	0.051

Fig. 2. Effect of surface tension and liquid-phase Reynolds number on wetted area of packings

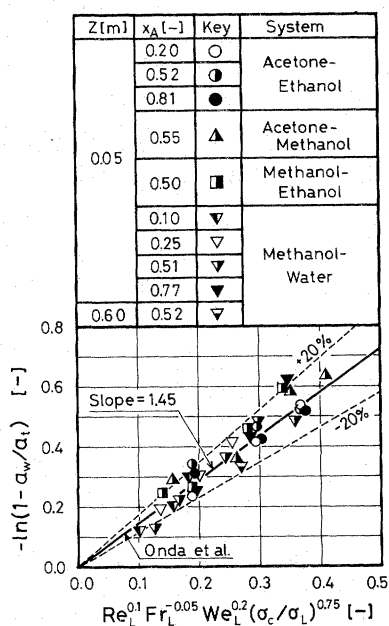| Z[m]	x_A[-]	Key	System
0.05	0.20	O	Acetone–Ethanol
	0.52	◑	
	0.81	●	
	0.55	▲	Acetone–Methanol
	0.50	◪	Methanol–Ethanol
	0.10	▽	Methanol–Water
	0.25	▽	
	0.51	▼	
	0.77	▼	
0.60	0.52	▽	

Fig. 3. Comparison of observed wetted area of packings with Onda's correlation

$$-\ln(1 - a_w/a_t) = 1.45 Re_L^{0.1} Fr_L^{-0.05}$$
$$We_L^{0.2}(\sigma_c/\sigma_L)^{0.75} \qquad (5)$$

The solid line in the figure represents Eq. (5). The data showed good agreement with Eq. (5). For this reason, the interfacial area for heat and mass transfer in packed column distillation was estimated from Onda's correlation in the latter part of the work.

2.2 Height of transfer unit

Mass transfer rates in packed column distillation have usually been described in terms of overall height of transfer unit (*HTU*) or number of transfer units (*NTU*) in previous works.[1,3,4,7,13–16,19] For this reason, the overall vapor-phase *HTU* was examined as for a first approximation. **Figure 4** shows the observed overall vapor-phase *HTU*, H_{OG}, for the
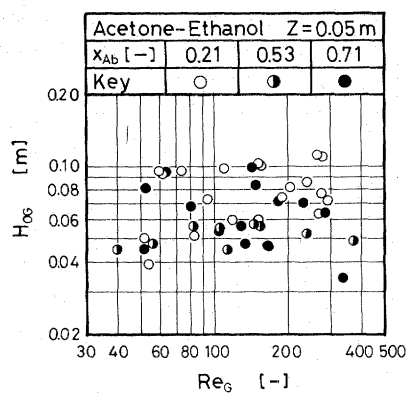
Acetone–Ethanol Z=0.05m			
x_{Ab} [-]	0.21	0.53	0.71
Key	O	◑	●

Fig. 4. Effect of vapor-phase Reynolds number and concentration on *HTU*

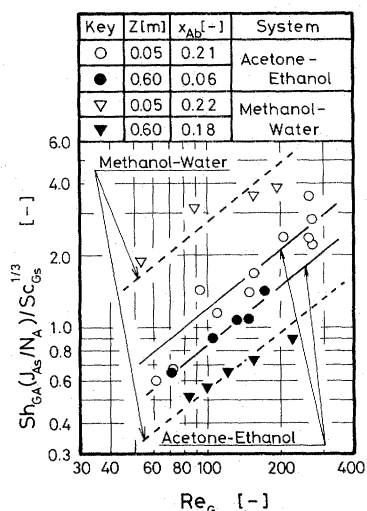| Key	Z[m]	x_{Ab}[-]	System
O	0.05	0.21	Acetone–Ethanol
●	0.60	0.06	
▽	0.05	0.22	Methanol–Water
▼	0.60	0.18	

Fig. 5. Effect of column length on diffusion flux for the acetone-ethanol and methanol-water systems

acetone-ethanol system with various liquid concentrations (x_{Ab}=0.21, 0.53, 0.71) with the 50 mm-long column. The abscissa is the vapor-phase Reynolds number, Re_G, given by:

$$Re_G = \rho_{G\infty} U_G D_p / \mu_{G\infty} \varepsilon \qquad (6)$$

where ε is the void fraction. An irregular scattering of the data with Re_G and x_{Ab} was observed. Similar results were observed for the acetone-methanol, methanol-ethanol and methanol-water systems. These facts indicate that *HTU* is an unsuitable parameter for expression of binary packed-column distillation.

2.3 Average diffusion fluxes and driving forces

Figure 5 shows the observed average vapor-phase diffusion fluxes, calculated by Eq. (3), for the acetone-ethanol and methanol-water systems under low mass flux conditions ($\rho_{Gs} v_s / \rho_{G\infty} U_G \approx 0$) with the 50 mm and 600 mm-long columns. The ordinate is the dimensionless diffusion flux, $Sh_{GA}(J_{As}/N_A)/Sc_{Gs}^{1/3}$. A decrease of the diffusion flux with increase of the packed height was observed. Similar results were observed for the methanol-ethanol system. A possible

explanation for this may be the effect of variations of the concentration driving forces in the columns.

To investigate this in more detail, axial distributions of the concentrations and of the concentration driving forces for the 600 mm-long column were plotted for the acetone-ethanol and methanol-water systems and are shown in **Fig. 6**. Higher concentration driving forces were observed only in the lower portion of the column, whereas lower and nearly constant driving forces were found in the middle and upper portions of the column. This may lead to the effect of packed height on the diffusion flux shown in Fig. 5.* The average diffusion flux with the 50 mm-long column is discussed in later sections.

2.4 Diffusion flux and effect of condensation of mixed vapors or evaporation from liquids

As was shown in our previous papers,[8-10,12] the vapor-phase diffusion flux in binary and ternary distillation by wetted-wall columns was affected by the convective mass fluxes caused by partial condensation of mixed vapors. **Figure 7** shows the observed average diffusion flux, calculated by Eq. (3), for the acetone-ethanol system with the 50 mm-long column under low mass flux conditions ($\rho_{Gs}v_s/\rho_{G\infty}U_G \approx 0$). The observed diffusion flux under low mass flux conditions was well correlated by the following equation:

$$[Sh_{GA}(J_{As}/N_A)]_0 = Sh_{GA}(J_{As}/N_A)\big|_{vs=0}$$
$$= 0.0306 Re_G^{0.805} Sc_{Gs}^{1/3} \qquad (7)$$

where subscript 0 denotes low mass flux conditions.

Figure 8 shows the effect of condensation of mixed vapors or evaporation from liquids on the observed average diffusion flux by Eq. (3) for the acetone-ethanol system. The ordinate is the ratio of the observed diffusion flux values to those under low mass flux conditions (Eq. (7)), given by:
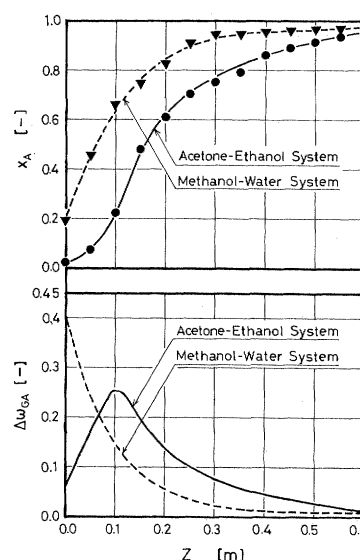
$$G_v = Sh_{GA}(J_{As}/N_A)/[Sh_{GA}(J_{As}/N_A)]_0 \qquad (8)$$

and the abscissa is a measure of the effect of condensation of mixed vapors or evaporation from liquids on the diffusion flux, $\rho_{Gs}v_s/\rho_{G\infty}U_G$. Although some scattering of data was observed, the effect of condensation of mixed vapors or evaporation from liquids on the diffusion flux was correlated by the following equation:

$$G_v = 1 - 83.3(\rho_{Gs}v_s/\rho_{G\infty}U_G) \qquad (9)$$

Figure 9 shows a final correlation for the average vapor-phase diffusion flux by Eq. (3) for the acetone-ethanol, acetone-methanol, methanol-ethanol and methanol-water systems with the 50 mm-long

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* A rapid decrease of the concentration driving forces in a smaller portion of the column may lead to a larger effect of packed height on the diffusion fluxes for the methanol-water ststem.

Fig. 6. Axial distributions of concentration driving force in the 600 mm-long column for the acetone-ethanol and methanol-water systems
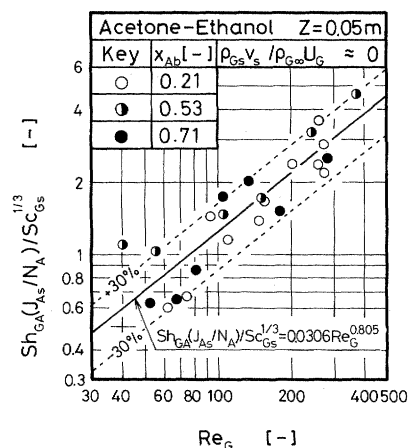
Fig. 7. Diffusion flux under low mass flux conditions for the acetone-ethanol system with the 50 mm-long column
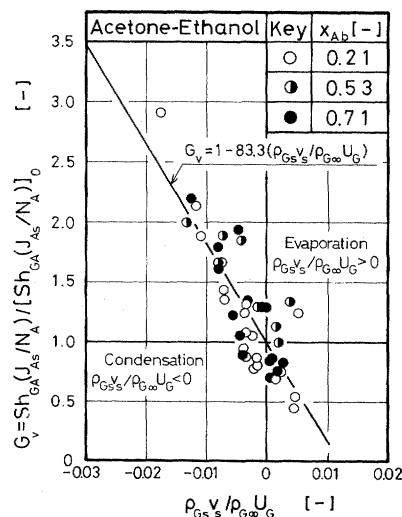
Fig. 8. Effect of condensation of mixed vapors or evaporation from liquids on diffusion flux for the acetone-ethanol system with 50 mm-long column
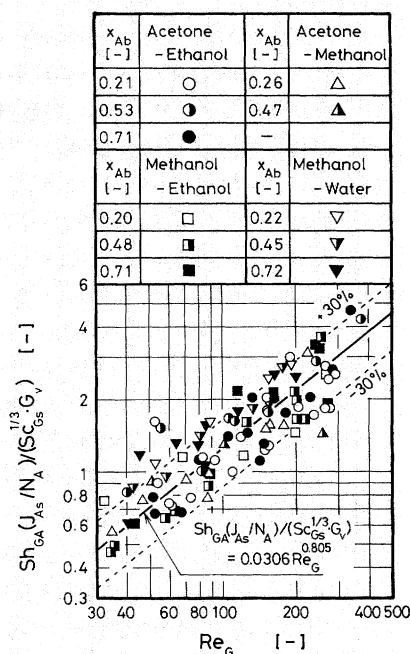
x_{Ab} [-]	Acetone -Ethanol	x_{Ab} [-]	Acetone -Methanol
0.21	O	0.26	△
0.53	◑	0.47	▲
0.71	●	-	

x_{Ab} [-]	Methanol -Ethanol	x_{Ab} [-]	Methanol -Water
0.20	□	0.22	▽
0.48	◪	0.45	▼
0.71	■	0.72	▼

Fig. 9. Final correlation for diffusion flux with 50 mm-long column

Fig. 10. Axial distributions of concentration and reflux flow rate for the acetone-ethanol system
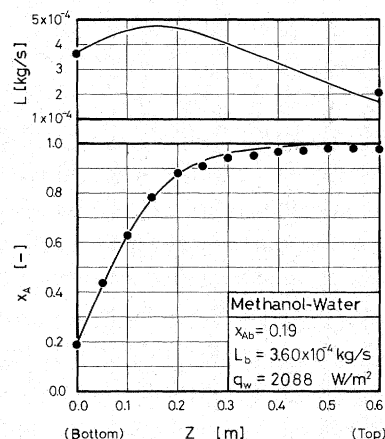
Fig. 11. Axial distributions of concentration and reflux flow rate for the methanol-water system

column. The ordinate is the diffusion flux corrected for the effect of condensation of mixed vapors or evaporation from liquids by Eq. (9). The solid line in the figure represents the low mass flux correlation for the diffusion flux. All the data showed good agreement with Eq. (7). This means that the diffusion fluxes in binary distillation with a column of short packed height for the acetone-ethanol, acetone-methanol, methanol-ethanol and methanol-water systems can be correlated by the following equation:

$$Sh_{GA}(J_{As}/N_A)=0.0306Re_G^{0.805}Sc_{Gs}^{1/3}G_v \qquad (10)$$

3. Discussion

By use of the correlations for diffusion flux by Eqs. (9) and (10) and for the wetted area of the packings by Eqs. (5), the separation performance of binary packed column distillation can be simulated. The following simplifying assumptions were made for the present calculations.

1) Mass and heat transfer processes in binary distillation are vapor-phase controlled.[8-9]

2) Interfacial area for mass and heat transfer is equal to the wetted area of the packings.[18]

3) Convective mass flux, $\rho_{Gs}v_s$, which is necessary for calculation of mass flux, is given by:[10]*

$$\rho_{Gs}v_s = \frac{J_{As}(\lambda_B - \lambda_A) - q_{Gs} - q_w}{\lambda_A \omega_{GAs} + \lambda_B \omega_{GBs}} \qquad (11)$$

where q_{Gs} is the vapor-phase sensible heat flux by:

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* The observed convective mass flux values with the 50 mm-long column were in good agreement with those calculated by Eq. (11).
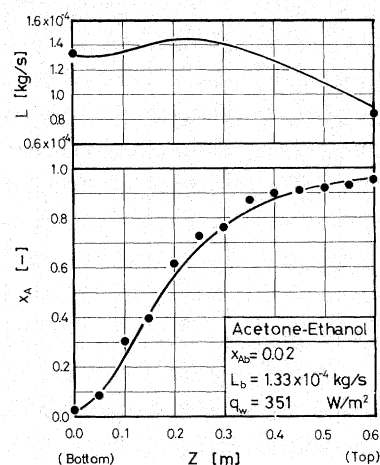
$$Nu_{Gs}=0.0306Re_G^{0.805}Pr_{Gs}^{1/3}G_v \qquad (12)$$

4) The surface temperatures of the liquids are at their bubble points and the vapor-free stream temperatures are at the their dew points.

Under these assumptions, calculations of separation performance of binary packed-column distillation with the 600 mm-long column were made by using the step-by-step method, the calculation steps of which were almost the same as those for ternary distillation by a wetted-wall column except for the basic equations and the number of components.[11]

Figure 10 compares the theoretical profiles for reflux flow rates and concentrations along the column with the observed ones with the 600 mm-long column at the conditions of $x_{Ab}=0.02$, $L_b=1.33 \times 10^{-4}$ kg/s and $q_w=351$ W/m² for the acetone-ethanol system, of which the surface tension is almost constant. The solid lines are the theoretical profiles and the solid circles are the observed data. Good agreement between the data and the theory was observed.

Figure 11 shows a similar comparison for the

methanol-water system, of which the surface tension changes widely, at the conditions of $x_{Ab} = 0.19$, $L_b = 3.60 \times 10^{-4}$ kg/s and $q_w = 2088$ W/m^2. Again, good agreement between the data and the theory was observed. Similar results were obtained for the other runs with the 600 mm-long column as well as the 50 mm-long column.

5. Conclusions

An experimental approach to mass transfer in binary packed column distillation for the acetone-ethanol, acetone-methanol, methanol-ethanol and methanol-water systems at total reflux conditions gives the following conclusions.

1) A new correlation for the vapor-phase diffusion flux with consideration of the effect of condensation of mixed vapors or evaporation from liquids is proposed.

2) A new method for prediction of the separation performance of binary packed-column distillation is proposed by using the present correlation for diffusion fluxes with the 50 mm-long column and Onda's correlation for the wetted area of the packings. Predicted distributions of reflux flow rates and of liquid concentrations with the 600 mm-long column showed good agreement with the experimental data.

3) Overall vapor-phase height of transfer unit in packed column distillation showed irregular scattering in terms of liquid concentrations and vapor flow rates.

Nomenclature

a_s	= interfacial area per unit volume	[m^2/m^3]
a_t	= total surface area of packing per unit volume	[m^2/m^3]
a_w	= wetted area of packing per unit volume	[m^2/m^3]
c_p	= specific heat at constant pressure	[J/(kg·K)]
$\mathscr{D}_{AB}$	= binary diffusion coefficient	[m^2/s]
D_p	= nominal size of packing	[m]
Fr_L	= Froude number $(= a_t U_L^2/g)$	[—]
g	= gravitational acceleration	[m/s^2]
G_v	= function defined by Eq. (8)	[—]
H_{OG}	= overall vapor-phase height of transfer unit $\left(= Z \middle/ \int_{y_b}^{y_t} dy/(y^* - y)\right)$	[m]
J_A	= diffusion flux for more volatile component	[kg/(m^2·s)]
L	= mass flow rate of liquid	[kg/s]
N_A	= mass flux for more volatile component	[kg/(m^2·s)]
N_B	= mass flux for less volatile component	[kg/(m^2·s)]
Nu_G	= vapor-phase Nusselt number $(= q_G D_p/\kappa_G (T_s - T_{G\infty}))$	[—]
Pr_G	= vapor-phase Prandtl number $(= c_{pG}\mu_G/\kappa_G)$	[—]
q_G	= vapor-phase sensible heat flux	[W/m^2]
q_w	= wall heat flux	[W/m^2]
Re_G	= vapor-phase Reynolds number $(= \rho_{G\infty} U_G D_p/\mu_{G\infty}\varepsilon)$	[—]

Re_L	= liquid-phase Reynolds number $(= \rho_L U_L/\mu_L a_t)$	[—]
Sc_G	= Schmidt number $(= \mu_G/\rho_G \mathscr{D}_{GAB})$	[—]
Sh_G	= Sherwood number $(= N_A D_p/\rho_G \mathscr{D}_{GAB}\Delta\omega_{GA})$	[—]
U_G	= vapor-phase superficial velocity	[m/s]
U_L	= liquid-phase superficial velocity	[m/s]
V	= volume of test section	[m^3]
v	= normal component of interfacial velocity	[m/s]
We_L	= Weber number $(= \rho_L U_L^2/\sigma_L a_t)$	[—]
x	= liquid-phase mole fraction	[—]
y	= vapor-phase mole fraction	[—]
Z	= height of packed column	[m]
z	= distance from bottom	[m]
ε	= void fraction	[—]
$\Delta\omega$	= concentration driving force	[—]
κ	= thermal conductivity	[W/(m·K)]
λ	= latent heat of vaporization	[J/kg]
μ	= viscosity	[Pa·s]
ω	= mass fraction	[—]
ω_{GA}^*	= vapor-phase concentration in equilibrium with liquid concentration	[—]
ρ	= density	[kg/m^3]
σ_c	= critical surface tension	[N/m]
σ_L	= surface tension	[N/m]

⟨Subscript⟩

A	= more-volatile component	
B	= less-volatile component	
b	= bottom	
G	= vapor-phase	
L	= liquid-phase	
s	= vapor-liquid interface	
t	= top	
∞	= vapor-free stream or liquid main stream	

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