

TERNARY MASS TRANSFER IN PACKED DISTILLATION COLUMN

HITOSHI KOSUGE, JUNICHI MATSUDAIRA AND KOICHI ASANO

*Department of Chemical Engineering, Tokyo Institute of Technology,
Tokyo 152*

Key Words: Ternary Distillation, Packed Column, Mass Transfer, Diffusion Flux, Simulation

Experimental studies of ternary mass transfer in packed distillation columns were made for the acetone-methanol-ethanol system under total reflux conditions for wide ranges of vapor flow rates and liquid concentrations.

The observed diffusion fluxes of all the components in ternary distillation with a short packed column were proportional to their concentration driving forces, but the observed mass fluxes were not. The convective mass fluxes were affected by the heat fluxes through the wall and were in good agreement with the theoretical ones. The diffusion fluxes under high mass flux conditions were well correlated by taking into account the effect of partial condensation of mixed vapors.

A new method for predicting separation performance of ternary distillation in a packed column was developed by use of the correlation for the ternary diffusion fluxes. Predicted distributions of reflux flow rate and liquid concentrations in a long column showed good agreement with the observed ones.

Introduction

Ternary mass transfer in a packed distillation column is a very complicated phenomenon. The observed numbers of transfer units showed irregular scattering and were sometimes unbounded under certain conditions. This may be due to the effects of interaction between heat and mass fluxes and interaction between diffusion fluxes through the concentration driving forces as well as the effect of liquid flow rates and concentrations. Recently some theoretical studies were made on binary¹²⁾ and ternary¹⁾ mass transfer in packed distillation columns based on the diffusion theory, but there are few studies which take into account the effect of interaction between

heat and mass flux under high mass flux conditions.

In our previous paper,⁵⁾ experimental studies of binary mass transfer in packed distillation columns were made for several systems. The results showed that the observed wetted areas of the packings were in good agreement with Onda's correlation⁹⁾ and that the observed diffusion fluxes under high mass flux conditions were well correlated by taking into account the effect of partial condensation of mixed vapors. The purpose of the present work is to make an experimental study of ternary mass transfer in a packed distillation column under total reflux conditions for wide ranges of vapor flow rates and liquid concentrations, and to develop a new method for predicting the separation performance of ternary distillation in a packed column.

* Received December 5, 1990. Correspondence concerning this article should be addressed to H. Kosuge.

1. Experimental Apparatus and Procedures

1.1 Experimental apparatus

The distillation columns were 50 mm- and 600 mm-long, 22 mm-inner diameter glass packed columns, which were packed with 3-mm Dixon packings. On the wall of the 600 mm-long column, sampling taps were attached at 50-mm intervals for measurements of liquid concentrations. Details of the experimental apparatus were shown elsewhere.⁵⁾

1.2 Measurements

1) Reflux flow rates and concentrations: The bottom and top reflux flow rates were measured by two precision rotameters within 1% error. The liquid concentrations of each component along the columns were measured by a gas chromatograph with an accuracy better than 0.1 mol%.

2) Mass and diffusion fluxes: The observed average vapor-phase mass fluxes for the i -th component, N_i , were obtained from the bottom and top reflux flow rates, L_b , L_t , and concentrations, ω_{Lib} , ω_{Lit} , by:

$$N_i = (L_t \omega_{Lit} - L_b \omega_{Lib}) / a_s V \quad (i = A, B, C) \quad (1)$$

where the interfacial area for mass and heat transfer, a_s , was taken equal to the wetted area of the packings¹⁰⁾ and was estimated by Onda's correlation.⁹⁾ The observed average convective mass fluxes, $\rho_{Gs} v_s$, and the observed average vapor-phase diffusion fluxes for the i -th component, J_{is} , were calculated by:^{3,4,6)}

$$\rho_{Gs} v_s = N_A + N_B + N_C \quad (2)$$

$$J_{is} = N_i - (N_A + N_B + N_C) \omega_{Gis} \quad (i = A, B, C) \quad (3)$$

1.3 Vapor-liquid equilibria and physical properties

Vapor-liquid equilibria of the acetone-methanol-ethanol system 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 by Holmes *et al.*²⁾ were used. The surface tensions of ternary solutions were estimated by Macleod-Sugden's method.¹¹⁾ The viscosities and thermal conductivities of mixed vapors were estimated by Wilke's method.¹¹⁾ The binary diffusion coefficients were estimated by Hirschfelder's method.¹¹⁾ The viscosities and thermal conductivities of pure vapors were estimated by Hirschfelder's and Eucken's methods,¹¹⁾ respectively.

1.4 Ranges of variables

Distillation runs were made for the acetone-methanol-ethanol system under total reflux conditions with the 50 mm- and 600 mm-long columns for wide ranges of vapor flow rates and liquid concentrations. The ranges of variables for this study are shown in **Table 1**. A series of distillation runs denoted by AME-1

Table 1. Ranges of variables

	Component	Z [m]	x_{ib} [—]	Re_G [—]	Sc_{Gis} [—]
AME-1	Acetone		0.22–0.26		0.72–0.74
	Methanol	0.05	0.57–0.61	45–200	0.68–0.70
	Ethanol		0.15–0.20		0.75–0.76
Path-1	Acetone		0.03–0.68		0.59–0.92
	Methanol	0.05	0.30–0.61	103–130	0.59–0.77
	Ethanol		0.02–0.45		0.69–0.80
Path-2	Acetone		0.05		0.68
	Methanol	0.60	0.46	115	0.62
	Ethanol		0.49		0.72
Path-3	Acetone		0.05		0.69
	Methanol	0.60	0.15	143	0.57
	Ethanol		0.80		0.71

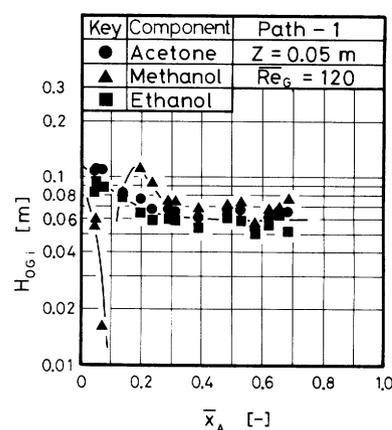


Fig. 1. Effect of liquid concentration of acetone on height of overall vapor-phase transfer unit

in **Table 1** were made with different vapor-phase Reynolds numbers at nearly constant liquid concentrations at the bottom of the 50 mm-long column and others denoted by distillation path-1 were made with the 50 mm-long column at nearly constant vapor-phase Reynolds number. The distillation runs denoted by distillation path-2 and path-3 were made with the 600 mm-long column.

2. Experimental Results

2.1 Height of transfer unit

Figure 1 shows the heights of overall vapor-phase transfer unit for all the components, H_{OGi} , along distillation path-1 at nearly constant vapor-phase Reynolds number with the 50 mm-long column. The abscissa represents the average liquid concentration of acetone, \bar{x}_A . The observed data for the intermediate component, methanol, showed a discontinuity near the concentration region of zero-driving force of methanol. Similar results have been also reported for ternary distillation by a sieve plate¹³⁾ and a wetted-wall column.⁴⁾ Such behavior of the height of transfer unit is peculiar to ternary or multicomponent distillation, but was not observed in binary distillation. This may indicate that the height of transfer unit is an unsuitable

parameter for expression of ternary mass transfer rates in a packed distillation column.

2.2 Mass and diffusion fluxes

Figure 2 shows the effect of concentration driving forces on the observed average vapor-phase mass and diffusion fluxes for all the components along distillation path-1 at nearly constant vapor-phase Reynolds number. The observed diffusion fluxes for all the components are proportional to their concentration driving forces. On the other hand, the observed mass fluxes for methanol and some for acetone take negative values in the concentration region of positive driving forces, and those for methanol and acetone are not proportional to their driving forces. Similar results were obtained for ternary distillation by wetted-wall columns.^{4,6)} These results indicate that the convective mass fluxes are affected not only by the concentration driving forces but also by other operating factors. To investigate this in more detail, the convective mass fluxes will be discussed in the following section.

2.3 Convective mass fluxes

Under nonadiabatic conditions, heat fluxes into or out of the column through the wall cause evaporation from liquids or partial condensation of mixed vapors, and thus may affect the convective mass fluxes. Figure 3 shows the effect of observed average heat fluxes through the wall, q_w , on the observed average convective mass fluxes with the 50 mm-long column obtained by Eq. (2). The heat fluxes through the wall were estimated from the enthalpy balance between the top and bottom of the column by

$$q_w = \{L_b(h_b - H_b) - L_t(h_t - H_t)\} / a_s V \quad (4)$$

where h_b and h_t are the vapor-phase enthalpies at the bottom and top, and H_b and H_t the liquid-phase ones. The convective mass fluxes increase with increase of heat flux through the wall, the positive value of which represents the heat flux out of the column to the surroundings.

In our previous paper,³⁾ an expression for convective mass fluxes was obtained from heat and mass balances for the falling liquid film for a ternary system in the distillation column, and was given as:

$$\rho_{Gs} v_s = \frac{J_{As}(\lambda_C - \lambda_A) + J_{Bs}(\lambda_C - \lambda_B) - q_G - q_w}{\lambda_A \omega_{GAs} + \lambda_B \omega_{GBs} + \lambda_C \omega_{GCs}} \quad (5)$$

Figure 4 shows a comparison of the observed convective mass fluxes with the theoretical ones by Eq. (5). Good agreement with the data and the theory was observed, as was the case for binary distillation in the packed column.⁵⁾

2.4 Dimensionless diffusion fluxes

As is clear from Fig. 2, the effect of interaction between diffusion fluxes of different components was

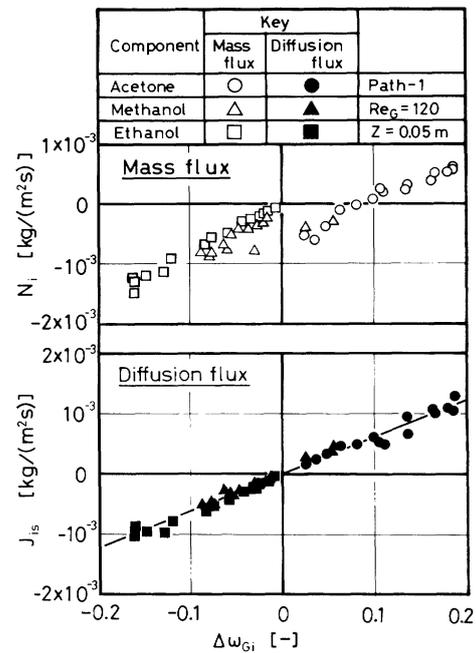


Fig. 2. Effect of concentration driving force on mass and diffusion flux

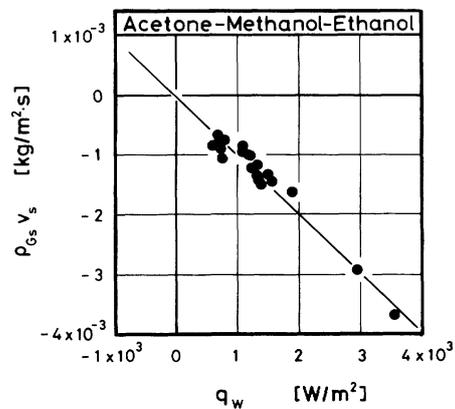


Fig. 3. Effect of heat flux on convective mass flux

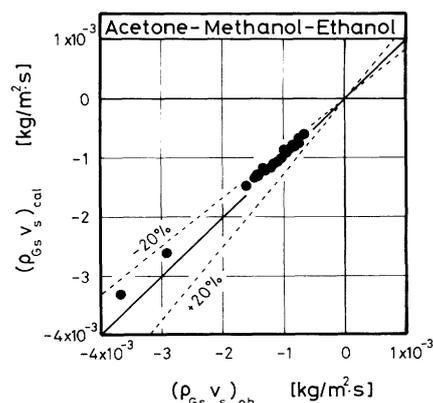


Fig. 4. Comparison of observed convective mass flux with theoretical ones by Eq. (5)

negligibly small. Therefore, the observed average diffusion fluxes were made dimensionless by the following equation:^{3,4,6)}

$$Sh_{Gi}(J_{is}/N_i) = J_{is} D_p / \rho_{Gs} D_{Gim} A \omega_{Gi} \quad (i = A, B, C) \quad (6)$$

where D_{Gim} is Wilke's effective diffusion coefficient for the i -th component by:⁷⁾

$$D_{Gim} = (1 - y_{is}) / (y_{js} / \mathcal{D}_{Gij} + y_{ks} / \mathcal{D}_{Gik}) \quad (i, j, k = A, B, C) \quad (7)$$

Figure 5 shows the observed dimensionless diffusion fluxes for all the components with the 50 mm-long column, where Sc_{Gis} in the ordinate is the multi-component Schmidt number. The solid line in the figure represents the binary correlation for diffusion fluxes under low mass flux conditions in the packed distillation column.⁵⁾ The observed data for the ternary system were in fairly good agreement with the binary correlation but some data were still larger than the correlation. This may be due to the effect of partial condensation of mixed vapors on the diffusion fluxes.

To confirm this, the observed diffusion fluxes corrected for the effect of partial condensation of mixed vapors are shown in **Fig. 6**. G_v in the ordinate represents the effect of partial condensation of mixed vapors given by:

$$G_v = 1 - 83.3(\rho_{Gs} v_s / \rho_{G\infty} U_G) \quad (8)$$

which was obtained in binary distillation for several systems.⁵⁾ The observed diffusion fluxes showed good agreement with the binary correlation for diffusion fluxes under low mass flux conditions.⁵⁾ From these results, the diffusion fluxes for ternary distillation could be correlated by the following equation:

$$Sh_{Gi}(J_{is}/N_i) = 0.0306 Re_G^{0.805} Sc_{Gis}^{1/3} G_v \quad (i = A, B, C) \quad (9)$$

3. Discussion

By use of the correlations for diffusion fluxes by Eq. (9) with Eq. (8), for convective mass fluxes by Eq. (5) and for wetted areas of the packings by Onda's correlation, the separation performance of ternary distillation in a packed column can be simulated. The following simplifying assumptions were made for the present calculations.

1) Mass and heat transfer processes in ternary distillation are vapor-phase controlled.^{3,4,6)}

2) Effect of interaction between diffusion fluxes is negligibly small; therefore the diffusion flux of each component is proportional only to its concentration driving forces.

3) Interfacial area for mass and heat transfer is equal to the wetted area of the packings.¹⁰⁾

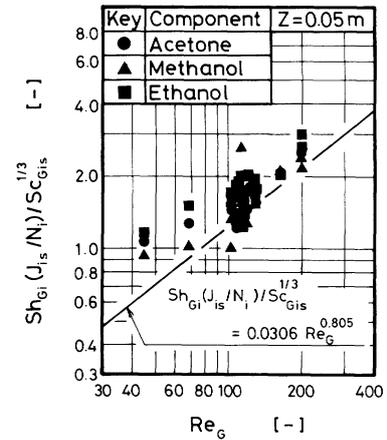


Fig. 5. Vapor-phase diffusion fluxes

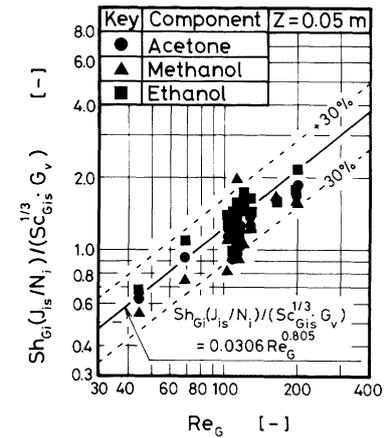


Fig. 6. Vapor-phase diffusion fluxes corrected for the effect of partial condensation of mixed vapors

4) Vapor-phase sensible heat flux, q_G , in Eq. (5) is given by⁵⁾

$$Nu_{Gs} = 0.0306 Re_G^{0.805} Pr_{Gs}^{1/3} G_v \quad (10)$$

Under these assumptions, calculations of separation performance of ternary distillation with the 600 mm-long packed column were made by using the step-by-step method.⁵⁾

Figure 7 shows a comparison of the theoretical profiles for the reflux flow rate and the liquid concentrations along the column with those observed in the 600 mm-long column under the conditions of $x_{Ab} = 0.05$, $x_{Bb} = 0.46$, $L_b = 1.53 \times 10^{-4}$ kg/s and $q_w = 461$ W/m². The solid lines in the figure represent the theoretical profiles and the solid circles are the observed data. Good agreement between the data and the theory was observed.

Figure 8 shows typical theoretical distillation paths. The solid lines in the figure represent the theoretical distillation paths by the present method. The arrows in the figure represent the observed liquid concentrations with the 50 mm-long column, where the tip and tail of the arrow represent the top and bottom liquid

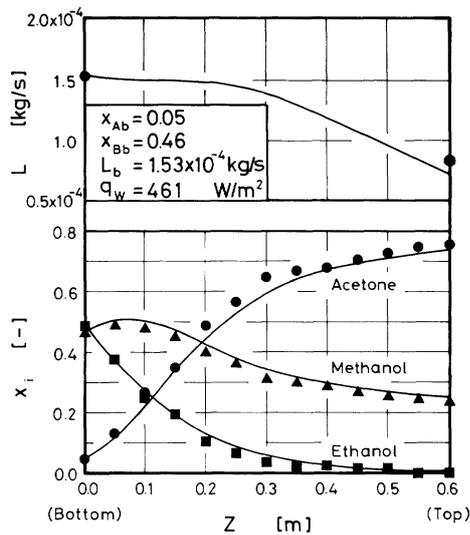


Fig. 7. Axial distributions of reflux flow rates and liquid concentrations for the acetone-methanol-ethanol system with 600 mm-long packed column

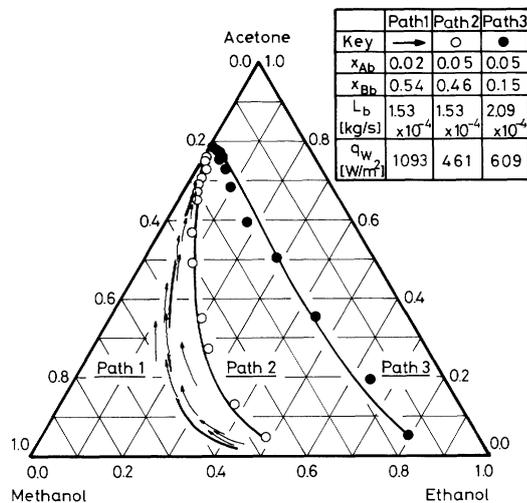


Fig. 8. Comparison of observed distillation paths with theoretical ones

concentrations, respectively, and the solid circles are the observed liquid concentrations with the 600 mm-long columns. Again, good agreement between the data and the theory was observed.

5. Conclusions

An experimental approach to ternary mass transfer in packed distillation columns for the acetone-methanol-ethanol system at total reflux conditions was taken and the following conclusions were reached.

1) The observed average vapor-phase diffusion fluxes for a ternary system under high mass flux conditions showed good agreement with the binary correlation for diffusion fluxes in the packed distillation column.

2) A new method was proposed for prediction of the separation performance of ternary distillation in

a packed column by using the present correlation for diffusion fluxes in a short column and Onda's correlation for wetted area of the packings. Predicted reflux flow rates and liquid concentrations in the 600 mm-long column showed good agreement with the experimental data.

Nomenclature

a_s	= interfacial area per unit volume	[m ² /m ³]
c_p	= specific heat at constant pressure	[J/(kg·K)]
\mathcal{D}_{AB}	= binary diffusion coefficient	[m ² /s]
D_{Gim}	= Wilke's effective diffusion coefficient of i -th component defined by Eq. (7)	[m ² /s]
D_p	= nominal size of packing	[m]
G_v	= function defined by Eq. (8)	[—]
H	= liquid enthalpy	[J/kg]
h	= vapor-phase enthalpy	[J/kg]
H_{Ogi}	= height of overall vapor-phase transfer unit of i -th component	[m]
J_i	= diffusion flux for the i -th component	[kg/(m ² ·s)]
L	= mass flow rate of liquid	[kg/s]
N_i	= mass flux of i -th 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 ($= c_p \mu_G / \kappa_G$)	[—]
q_G	= vapor-phase sensible heat flux	[W/m ²]
q_w	= heat flux into or out of column through the wall	[W/m ²]
Re_G	= vapor-phase Reynolds number ($= \rho_{G\infty} U_G D_p / \mu_{G\infty}$)	[—]
Sc_{Gim}	= multicomponent Schmidt number for i -th component ($= \mu_G / \rho_G D_{Gim}$)	[—]
Sh_{Gi}	= Sherwood number for i -th component ($= N_i D_p / \rho_G D_{Gim} \Delta \omega_{Gi}$)	[—]
U_G	= vapor-phase superficial velocity	[m/s]
V	= volume of test section	[m ³]
v	= normal component of the interfacial velocity	[m/s]
x	= liquid-phase mole fraction	[—]
y	= vapor-phase mole fraction	[—]
Z	= height of packed column	[m]
z	= distance from the bottom	[m]

ε	= void fraction	[—]
$\Delta \omega_i$	= concentration driving force for i -th component	[—]
κ	= thermal conductivity	[W/(m·K)]
λ	= latent heat of vaporization	[J/kg]
μ	= viscosity	[Pa·s]
ω_i	= mass fraction	[—]
ρ	= density	[kg/m ³]

<Subscripts>

A	= most volatile component (= acetone)
b	= bottom
G	= vapor-phase
i	= i -th component
L	= liquid-phase
s	= vapor-liquid interface
t	= top
∞	= vapor free stream or liquid main stream

Literature Cited

- 1) Gorak, A. and A. Vogelpohl: *Separation Sci. Tech.*, **20**, 33 (1985).
- 2) Holmes, M. J. and M. van Winkle: *Ind. Eng. Chem.*, **62**, 21 (1970).
- 3) Kosuge, H. and K. Asano: *J. Chem. Eng. Japan*, **15**, 268 (1982).
- 4) Kosuge, H. T. Johkoh and K. Asano: *Chem. Eng. Commun.*, **34**, 111 (1985).
- 5) Kosuge, H., J. Matsudaira, K. Aoki and K. Asano: *J. Chem. Eng. Japan*, **23**, 593 (1990).
- 6) Kosuge, H., Y. Mochizuki and K. Asano: *Kagaku Kogaku Ronbunshu*, **10**, 219 (1984).
- 7) Krishna, R. and G. L. Standart: *Chem. Eng. Commun.*, **3**, 201 (1979).
- 8) Miyakawa, H., K. Honda and T. Munakata: *Kagaku Kogaku Ronbunshu*, **15**, 134 (1989).
- 9) Onda, K., H. Takeuchi and Y. Koyama: *Kagaku Kogaku*, **31**, 126 (1967).
- 10) Onda, K., H. Takeuchi and Y. Okumoto: *J. Chem. Eng. Japan*, **1**, 56 (1968).
- 11) Reid, R. C., J. M. Prausnitz and T. K. Sherwood: "The Properties of Gases and Liquids", 3rd ed., pp. 394, 473, 549, 604, McGraw-Hill, New York (1977).
- 12) Takamatsu, T., V. Lueprasitsakul and M. Nakaiwa: *J. Chem. Eng. Japan*, **21**, 595 (1988).
- 13) Vogelpohl, A: Proceedings of the Third International Symposium on DISTILLATION, Vol. 1, p. 2.1/25 (1979).