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Andrzej Górak^a; Alfons Vogelpohl^b

^a INSTITUTE OF CHEMICAL ENGINEERING ŁÓDŹ TECHNICAL UNIVERSITY, ŁÓDŹ, POLAND ^b

INSTITUT FÜR THERMISCHE VERFAHRENSTECHNIK DER TECHNISCHEN UNIVERSITÄT
CLAUSTHAL, CLAUSTHAL-ZELLERFELD, WEST GERMANY

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Experimental Study of Ternary Distillation in a Packed Column

ANDRZEJ GÓRAK*

INSTITUTE OF CHEMICAL ENGINEERING
ŁÓDŹ TECHNICAL UNIVERSITY
90-926 ŁÓDŹ, POLAND

ALFONS VOGELPOHL

INSTITUT FÜR THERMISCHE VERFAHRENSTECHNIK DER
TECHNISCHEN UNIVERSITÄT CLAUSTHAL
3392 CLAUSTHAL-ZELLERFELD, WEST GERMANY

Abstract

Ternary distillation of a methanol/isopropanol/water mixture was studied in a 0.1-m diameter column with SULZER-CY packing. The experimental data have been compared with results from the multicomponent mass transfer model of Krishna and Standart. The influence of model simplifications and the accuracy of the model parameters have been studied. Finally, the suitability of simple mass transfer models for predicting concentration profiles in packed columns was looked into.

INTRODUCTION

The number of theoretical plates (NTP) is the most often used designing method for multicomponent distillation. The number of real plates is then obtained using a calculated or assumed value of the plate efficiency or the HETP value for packed columns. An inaccurate plate efficiency (or HETP) can result in large errors of the calculated product compositions, even if the number of theoretical plates has been computed with sufficient accuracy.

Most methods for calculating plate efficiencies (or HETP values) don't

*To whom correspondence should be addressed.

take into consideration the kinetics of multicomponent mass transfer and are based on the assumption that the influence of diffusional interactions between components on the rate of mass transfer is negligible. The theoretical and experimental investigations of multicomponent diffusion, condensation, and distillation (Table 1) have shown, however, that "cross effects" should be taken into consideration in multicomponent mass transfer calculations, especially if the compounds involved have a different chemical structure or/and the diffusivities of the binary subsystems are significantly different. This demands a more complex mathematical description of the transfer phenomena in comparison to binary mixtures. These methods are not used, unfortunately, in design practice because of the complex calculation algorithms, long computation times needed, and their poor experimental verification. It is, therefore, necessary to investigate the kinetic models of multicomponent mass transfer, regarding their applicability in distillation design methods, the computational effort on one hand and the resulting accuracy on the other hand, as well as the agreement between calculation and experimental results.

Mathematical models of interphase mass transfer in multicomponent distillation can be divided into two groups.

Models where multicomponent mass transfer is described by equations analogous to those used in binary systems. The relation between the molar fluxes of each component, transferred through the interface and the mass transfer coefficients as well as the driving force, is then given by a simple equation

$$N_i = k_i \Delta y_i, \quad i = 1, 2, \dots, n \quad (1)$$

Models which take into account the diffusional interactions between components. They are based on a solution of the Stefan-Maxwell equation. Relationships from the last group of models are either:

Equations which are analogous to the analytical solutions of the Stefan-Maxwell equation. Applicability of these equations has been confirmed only for a few multicomponent systems (33).

Equations derived from approximate solutions of the Stefan-Maxwell equation. These equations contain the characteristic phenomena of multicomponent mass transfer like "reverse diffusion" or the diffusional barrier. They also allow the mass transfer coefficients for each component to be correctly defined and seem to be the most accurate with respect to other models.

TABLE 1. Literature Review of Experimental Investigations of Multicomponent Distillations in Tray and Packed Columns

No.	System	Author(s)	Ref.
1	Acetone/ethanol/water	Burghardt et al.	7
2	Acetone/methanol/water	Diener and Gerster	8
3	Acetone/methanol/water	Vogelpohl	29
4	Acetone/methanol/ethanol	Bevers	5
5	Acetone/methanol/ethanol	Free and Hutchinson	9
6	Benzene/methylcyclopentane/ <i>n</i> -hexane	Miskin et al.	23
7	Benzene/toluene/ <i>m</i> -xylene	Nord	25
8	Benzene/toluene/ <i>m</i> -xylene	Westphely	30
9	CH ₄ /C ₂ H ₆ /C ₃ H ₈ /+ C ₄ /+ C ₅	Rajko and Aleksandrov	32
10	CH ₃ Cl ₃ /benzene/ <i>n</i> -heptane	Aittaama	1
11	CCl ₄ /benzene/methyl ethyl ketone	Lutugina and Kovalitchev	16
12	Cyclohexane/benzene/CCl ₄	Lutugina et al.	17
13	Cyclohexane/methyl ethyl ketone/ isopropanol	Lutugina et al.	18
14	Ethanol/benzene/ <i>n</i> -heptane	Aittaama	2
15	Ethanol/ <i>n</i> -butanol/water	Aittaama	1
16	Ethanol/ <i>tert</i> -butanol/water	Martinez	21
17	Ethyl acetate/acetic acid/acrylic acid/ethyl acrylate	Kibol and Artamonov	15
18	Ethane-decane (11 <i>n</i> -alkanes)	Molokanov	24
19	<i>n</i> -Hexane/methylcyclopentane/ethanol	Young and Weber	31
20	<i>n</i> -Hexane/methylcyclopentane/ ethanol/benzene	Young and Weber	31
21	Methanol/ethanol/water	Stefanovskaia et al.	27
22	Methanol/ethanol/water	Bogoslavski and Planovski	6
23	Methanol/ethanol/ <i>n</i> -propanol/ isobutanol/water	Gelbin	11
24	Methanol/isopropanol/water	Cermak	4
25	Methanol/isopropanol/water	Goldberg et al.	12
26	Methanol/isopropanol/water	Burghardt et al.	7
27	Methanol/isopropanol/water	Vogelpohl and Ceretto	28
28	Methyl ethyl ketone/ <i>n</i> -heptane/toluene	Tchernykh et al.	10
29	<i>tert</i> -Butanol/ <i>n</i> -propanol/ <i>n</i> butanol/ <i>n</i> -amyl alcohol	Planovski et al.	26
30	Trimethylcarbinol/propanol/butanol/ pentanol	Katalov et al.	19
31	Trimethylcarbinol/propanol/butanol/ pentanol	Mazaev et al.	22
32	Oxygen/nitrogen/argon	Haselden and Thorogood	14
<i>Packed Columns</i>			
1	Acetone/methanol/benzene	Kowalska et al.	20
2	Acetone/methanol/ethanol	Bevers	5
3	Acetone/methyl acetate/methanol	Westphely	30
4	Benzene/toluene/xylene	Arikwar	3
5	Ethane-decane (11 <i>n</i> -alkanes)	Molokanov	24
6	Ethanol/benzene/ <i>n</i> -heptane	Gorak	13
7	Methanol/isopropanol/water	This work	

Toor (34) and Stewart and Prober (35) presented the first linearized solution of the multicomponent mass transfer equations. The solutions are equivalent (36) and have been developed under the assumption that the multicomponent diffusivity matrix is constant over the diffusion path. The same assumption is made by Burghardt and Krupiczka (37) in their model of interphase mass transfer. The linearized equations allow the molar fluxes of species to be determined from an explicit equation, but require an iterative computation of average compositions over the diffusion path.

More recent publications of Krishna and Standart (38) give an exact solution of the multicomponent mass transfer equations. The dependence between fluxes on one side and mass transfer coefficients and driving forces on the other side is implicit, however. The solution simplifies for low molar rates typical for distillation problems. All mathematical models of multicomponent diffusion published up to now have been compared by Smith and Taylor (39); they suggested using the Krishna/Standard model for distillation problems.

A comparison of the solutions of various diffusional mass transfer models cannot, of course, answer the question of how good these models are for predicting multicomponent distillation. A large number of experimental data have been published on multicomponent distillation whereas a lack of data is evident with respect to multicomponent distillation in packed columns. Unfortunately, a lot of experiments are uncompletely published, so they may not be used to verify theoretical predictions. Experimental results on distillation systems with more than three components are rarely published.

The aim of the present work was to check the Krishna/Standart method against experimental results from ternary distillations in a packed column. In addition, the influence of simplifying assumptions of the mass transfer model as well as experimental errors of the model parameters on the accuracy of predicted product compositions was studied.

EXPERIMENTAL

The ternary mixture methanol/isopropanol/water has been chosen as the test system. Calculated concentration profiles of this experiment system show a strong sensitivity on model parameters (40) and simplifications of the model (41). In addition, distillation experiments with the test system were run by different authors, resulting in opposing conclusions with respect to the suitability of matrix methods for design purposes. From a theoretical analysis of the role of cross effects on calculated distillation profiles of the test system, Toor and Burchard (42) reasoned that

diffusional interactions have to be taken into consideration. In their study the average column efficiency was 39% below the binary efficiencies. Experimental distillation results from a plate column, presented by Vogelpohl (43), revealed a different plate efficiency for every component. Cermak (4), in his study on ternary distillation in a plate column, compared the experimental concentration profiles with theoretical profiles calculated on the basis of the Toor (34) model and on the hypothesis of equal efficiencies, and he found the best agreement between experimental and calculated curves by assuming equal efficiencies for all components. Goldberg et al. (12) applied Eq. (1) to multicomponent distillation problems, and they claimed sufficient accuracy using pseudo-compositions. Their own experimental distillation results in a plate column confirmed their conclusions. Burghardt et al. (7) discussed the suitability of the matrix method in multicomponent distillation calculations on the basis of their own experimental investigations of the test system in a plate column. They state that "expressing the mass fluxes solely in terms of its own concentration gradients may lead to considerable errors because of the influence of cross effects on the mass fluxes."

These contradicting conclusions from distillation data of the methanol/isopropanol/water mixture motivated us to use the same system for distillation experiments with a packed column.

Figure 1 shows a schematic drawing of the distillation apparatus consisting of a packed column (3), a reboiler (2), and a condenser (5). The column had an inner diameter of 0.1 m and was packed with SULZER-CY packing, divided into two parts of 0.8 m height each. All experiments were performed under atmospheric pressure and at total reflux conditions. The vapor rate was determined from an enthalpy balance around the condenser. The composition of the samples was analyzed by gas chromatography. The vapor rate in the columns was varied between 40 and 80% of the rate at the flooding point. The compositions and rates from the experiments are shown in Table 2.

THEORETICAL

Multicomponent distillation in a packed column at total reflux conditions may be described by the following set of differential and matrix equations (45):

$$\frac{d(Gy)}{dA} = \frac{d(Lx)}{dA} = N \quad (2)$$

TABLE 2
Experimental Results

Experiment	Composition of vapor or liquid phase (mol%)												Molar flow rate of vapor (liquid) in the column (mol/m ² · s)
	Vapor below the packing			Liquid below the packing			Vapor in the middle of the packing			Liquid at the top of the packing			
	y ₁	y ₂	y ₃	x ₁	x ₂	x ₃	y ₁	y ₂	y ₃	x ₁	x ₂	x ₃	
1	—	—	—	1.14	71.26	27.59	14.00	57.93	28.05	86.54	9.74	3.70	28.81
2	—	—	—	0.99	71.16	27.83	12.15	59.23	28.61	84.94	10.60	4.44	20.93
3	—	—	—	3.91	68.69	27.38	73.98	18.84	7.16	99.56	0.24	0.19	37.57
4	—	—	—	1.30	70.63	28.06	47.03	36.32	16.63	99.15	0.63	0.20	31.56
5	—	—	—	0.43	70.70	28.86	16.17	56.04	27.78	97.77	0.04	2.17	19.32
6	—	—	—	0.28	59.87	39.84	8.31	61.62	30.06	82.29	12.99	4.70	40.72
7	—	—	—	0.22	59.52	40.25	9.98	60.47	29.54	83.87	11.84	4.28	28.65
8	—	—	—	1.01	62.56	36.42	12.79	58.48	28.72	84.10	11.70	4.18	40.31
9	0.39	62.83	36.76	0.29	62.49	37.21	14.87	56.67	28.44	87.02	9.35	3.61	31.75
10	0.62	67.80	31.56	0.42	68.20	31.37	9.09	60.98	29.91	79.75	14.71	5.52	48.04

11	0.39	67.95	31.64	0.21	68.35	31.42	10.69	59.84	29.45	79.89	14.11	5.99	29.89
12	1.73	63.15	35.10	1.14	63.13	35.71	9.37	60.75	29.87	80.39	14.26	5.34	41.28
13	0.37	63.41	36.21	0.30	62.98	36.71	6.73	62.54	30.72	67.07	23.13	9.78	49.26
14	1.74	63.33	34.91	1.05	63.51	35.43	6.70	62.48	30.80	66.67	23.20	10.12	42.13
15	1.39	63.78	34.82	0.89	63.68	35.42	9.64	60.65	29.70	67.32	22.50	10.17	28.91
16	0.46	58.42	41.10	0.28	57.31	42.39	6.10	59.58	34.31	69.14	21.16	9.68	44.46
17	0.23	60.00	39.76	0.12	58.41	41.45	6.12	59.92	33.95	68.41	21.44	10.13	33.36
18	0.85	59.35	39.78	0.50	57.98	41.51	3.95	64.25	31.78	48.77	35.13	16.09	41.69
19	0.63	59.85	39.50	0.22	58.06	41.71	4.01	64.19	31.78	50.98	33.19	15.81	32.02
20	0.58	59.81	39.59	0.30	58.21	41.48	4.04	64.30	31.64	53.57	32.01	14.40	41.72
21	0.32	59.30	40.37	0.14	57.62	42.23	4.31	64.28	31.40	54.88	30.78	14.33	31.72
22	0.84	58.69	40.46	0.85	58.09	41.05	4.37	64.07	31.55	53.81	31.78	14.39	42.52
23	0.61	60.09	39.29	0.63	58.43	40.93	5.20	62.80	31.99	55.98	29.68	14.33	33.69
24	0.63	58.60	40.75	0.39	58.08	41.52	4.21	64.31	31.46	57.10	29.65	13.23	41.55
25	0.39	60.27	39.33	0.22	58.60	41.16	4.77	63.91	31.31	57.97	28.78	13.23	33.30
26	0.73	59.62	39.63	0.50	58.53	40.96	4.43	64.09	31.47	59.76	27.85	12.38	39.54
27	0.54	60.00	39.45	0.34	58.94	40.71	4.57	64.13	31.29	60.65	27.00	12.33	30.80
28	0.94	58.94	40.10	0.53	57.62	41.84	5.40	63.52	31.06	61.47	26.72	11.80	42.71
29	0.67	59.93	39.38	0.40	59.09	40.50	6.36	62.67	30.96	61.78	26.26	11.95	33.06

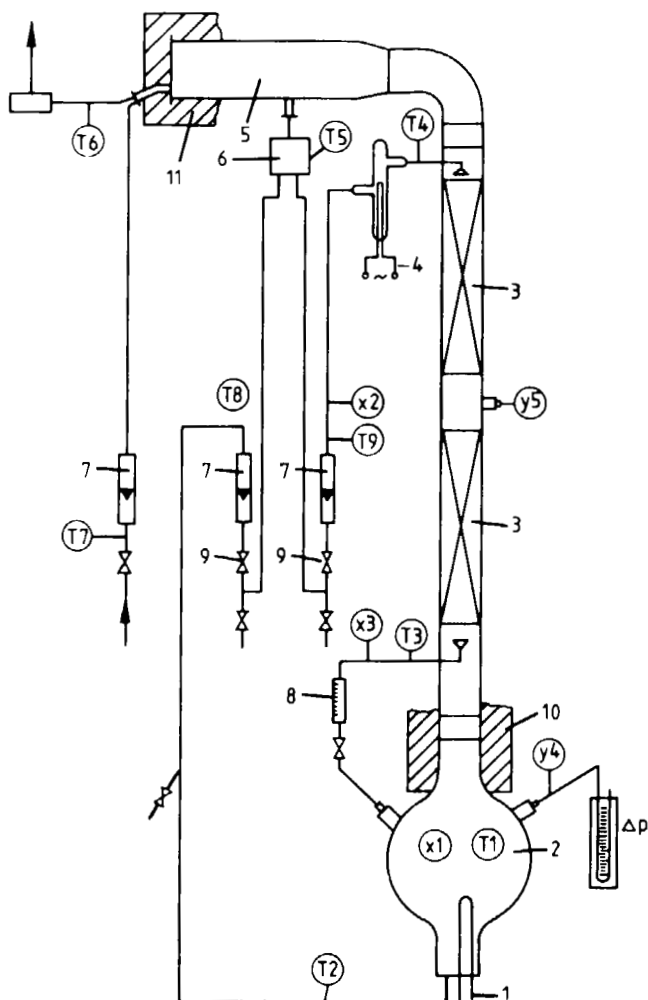


FIG. 1. Experimental apparatus: 1, electrical heater; 2, reboiler; 3, SULZER-CY packing; 4, reflux heater; 5, condenser; 6, reflux divider; 7, flow meter; 8, calibrated volume; 9, valves; 10, insulation; x1, x2, x3, liquid sampling ports; y4, y5, gas sampling ports; T1–T9, temperature measurement.

$$dL = dG = \sum_{i=1}^n N_i dA \quad (3)$$

$$\sum_{i=1}^n N_i r_i = 0 \quad (4)$$

$$\mathbf{x} = \mathbf{y} \quad (5)$$

This model is based on the assumption that thermal distillation and surface tension effects are negligible and only vaporization enthalpies need to be taken into account in the heat balances.

Integrating the set of Eqs. (2)–(5), we obtain the concentration profiles within the packing as well as the product compositions. The initial conditions are the mole fractions of the components of both phases at the lower end of the packing and the molar flow rates of vapor and liquid in the column. An analytical integration of the set of equations is impossible because the mass fluxes on the right sides of Eqs. (2) and (3) are an implicit function of the mole fractions on the left side of the equations.

The mass fluxes are obtained from the dependence

$$\begin{aligned} \mathbf{N} &= (\dot{\mathbf{k}}_G \bar{\beta}_G^{-1} \mathbf{M} \dot{\mathbf{k}}_L^{-1} \beta_L^{-1})^{-1} (\mathbf{y}^* - \mathbf{y}) \\ &= \dot{\mathbf{K}}_{OG} (\mathbf{y}^* - \mathbf{y}) \end{aligned} \quad (6)$$

where

$$\beta_{Gij} = \delta_{ij} - y_i \frac{r_i - r_n}{r_G}, \quad i, j = 1, 2, \dots, n-1 \quad (7)$$

and

$$r_G = \sum_{i=1}^n y_i r_i \quad (8)$$

The elements of matrix \mathbf{M} are defined as

$$M_{ij} = \frac{\partial y_i^*}{\partial x_j}, \quad i, j = 1, 2, \dots, n-1 \quad (9)$$

The matrices of the mass transfer coefficients in both phases are expressed as

$$\dot{\mathbf{k}}_G = \mathbf{k}_G \mathbf{E}_G = k_G \Phi \exp \Phi (\exp \Phi - 1)^{-1} \quad (10)$$

$$\dot{\mathbf{k}}_L = \mathbf{k}_L \mathbf{E}_L = \mathbf{k}_L \Theta (\exp \Theta - 1)^{-1} \quad (11)$$

where

$$\Theta = \Gamma^{-1} \Phi \quad (12)$$

The elements of matrix Γ and Φ are defined as

$$\Gamma_{ij} = \delta_{ij} + \frac{x_i}{x_j} \frac{\partial \ln \gamma_i}{\partial x_j}, \quad i, j = 1, 2, \dots, n-1 \quad (13)$$

$$\Phi_{ii} = \frac{N_i}{K_{Gin}} + \sum_{\substack{j=1 \\ j \neq i}}^n \frac{N_i}{k_{Gij}}, \quad i, j = 1, 2, \dots, n-1 \quad (14)$$

$$\Phi_{ij} = -N_i \left(\frac{1}{k_{Gij}} - \frac{1}{k_{Gin}} \right)$$

The matrix of the mass transfer coefficients \mathbf{k}_G in the vapor phase is defined by the inverse matrix \mathbf{B}_G :

$$B_{Gii} = \frac{y_i}{k_{Gin}} + \sum_{\substack{k=1 \\ k \neq i}}^n \frac{y_k}{k_{Gik}}, \quad i, j = 1, 2, \dots, n-1$$

$$B_{Gij} = -y_i \left(\frac{1}{k_{Gij}} - \frac{1}{k_{Gin}} \right) \quad (15)$$

The matrix of mass transfer coefficients in the liquid phase \mathbf{k}_L results from the product

$$\mathbf{k}_L = \mathbf{B}_L^{-1} \cdot \Gamma \quad (16)$$

The matrices Φ_L , \mathbf{B}_L , and β_L are obtained from the same equations as the matrices Φ_G , \mathbf{B}_G , and β_G by replacing of concentrations, binary diffusivities, and the mass transfer coefficients in the gas phase through the corresponding values in the liquid phase.

The algorithm for the numerical integration of the set of differential equations (2)–(3) taking into account the heat balance (4) and the operating conditions (5) as well as the matrix equation (6)–(16) has been presented by Kowalska et al. (46). The size of the computer program and the computation time may be reduced by introducing some simplifying

assumptions. In the following analysis, the influence of such simplifications on the agreement between calculated and experimental data is examined.

The following simplifications of the mass transfer model have been tested:

Low mass fluxes

Equimolar mass transfer

Ideal liquid phase

Negligible cross effects

Mass transfer resistance only in the vapor phase

In addition, the influence of the following model parameters on the agreement between calculated and experimental product compositions has been investigated:

Accuracy of VLE data

Accuracy of the correlation equations for the physical properties and the binary mass transfer coefficients in both phases

Experimental error in the determination of the molar flow rate of both phases in the column as well as the concentration

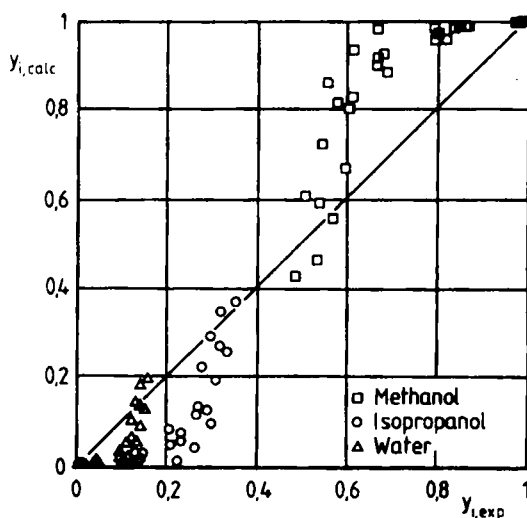


FIG. 2. Comparison of experimental and theoretical distillate compositions calculated from Krishna/Standart model.

RESULTS AND DISCUSSION

The above-described mathematical model of multicomponent distillation (Eqs. 2–16) was tested by comparing numerical solutions of the model with measured distillate compositions. The compositions and molar flow rates of both phases below the packing were given as initial values for the calculation of concentration profiles along the packing.

While evaluating the experimental samples, some difficulties arose. The mole fraction of methanol in the liquid below the packing was in general below 0.002, resulting in an excessively large error from the chromatography analysis. In these cases, therefore, the comparison between calculated and experimental data was performed only for the upper part of the packing (0.8 m).

The comparison between the theoretical and the experimental data is given in Fig. 2 and Table 3.

Influence of Simplifications of the Mass Transfer Model on the Agreement between Calculated and Experimental Data

Simplifications of the mass transfer model reduce numerical difficulties and save computation time. Therefore the distillate compositions determined experimentally have been compared with calculated compositions obtained with the model simplifications shown in Table 4.

The matrices E_G and E_L describe the influence of large mass fluxes on the transport process. The calculation algorithm simplifies if we assume that both are unit matrices (Variant 1) and allows determination of the mass flux vector N from Eq. (6) without iterations. The comparison of the distillate compositions calculated with or without the simplification

$$E_G = E_L = 1$$

with experimental data confirms the conclusion of other authors (39, 40, 46, 47) that this simplification gives a negligible error.

The simplification of equimolar mass transfer corresponds to the assumption

$$\beta_G = \beta_L = 1$$

As this simplification (Variant 2) did not significantly change the calculated distillate compositions, the distillation of the methanol/isopropanol/water mixture may be treated as equimolar.

TABLE 3
Comparison between Experimental and Theoretical Distillate Compositions Calculated from
Krishna/Standart Model

Experiment	Composition of distillate (mol%)						Absolute deviation		
	Experimental			Theoretical					
	x_1	x_2	x_3	x_1	x_2	x_3	Δx_1	Δx_2	Δx_3
1	86.5	9.7	3.7	99.0	0.7	0.3	12.5	- 9.0	-3.4
2	84.9	10.6	4.4	99.2	0.6	0.2	14.3	-10.0	-4.2
3	99.6	0.2	0.2	99.9	0.1	0.0	0.4	- 0.1	-0.1
4	99.2	0.6	0.2	99.8	0.2	0.1	0.7	- 0.4	-0.1
5	97.8	0.0	2.2	99.6	0.3	0.1	1.9	0.3	-2.0
6	82.3	13.0	4.7	95.3	3.4	1.3	13.1	- 9.5	-3.4
7	83.9	11.8	4.3	98.2	1.3	0.5	14.4	-10.5	-3.7
8	84.1	11.7	4.2	98.2	1.3	0.5	14.1	-10.4	-3.6
9	87.0	9.4	3.6	99.0	0.7	0.3	12.0	- 8.6	-3.3
10	79.8	14.7	5.5	95.2	3.5	1.3	15.5	-11.2	-4.2
11	79.9	14.1	6.0	98.4	1.2	0.4	18.6	-12.9	-5.5
12	80.1	14.3	5.3	96.4	2.6	1.0	16.1	-11.6	-4.3
13	67.1	23.1	9.8	88.0	8.6	3.4	21.0	-14.5	-6.3
14	66.7	23.2	10.1	90.5	6.8	2.7	23.9	-16.4	-7.4
15	67.3	22.5	10.2	98.0	1.4	0.5	30.7	-21.1	-9.6
16	69.1	21.2	9.7	86.6	9.5	3.9	17.5	-11.6	-5.7
17	68.4	21.4	10.1	91.5	6.0	2.4	23.1	-15.4	-7.7
18	48.8	35.1	16.1	36.5	41.0	22.6	-12.2	5.9	6.6
19	51.0	33.2	15.8	54.7	30.0	15.3	3.8	- 3.1	-0.6
20	53.6	32.0	14.4	40.4	38.7	20.9	-13.1	6.7	6.5
21	54.9	30.8	14.3	67.4	22.0	10.6	12.6	- 8.7	-3.7
22	53.8	31.8	14.4	53.3	31.0	15.7	- 0.5	- 0.7	1.4
23	56.0	29.7	14.3	83.7	11.4	4.9	27.8	-18.2	-9.4
24	57.1	29.7	13.2	50.0	33.0	17.0	- 7.1	0.4	3.8
25	58.0	28.8	13.2	78.1	15.1	6.8	20.2	-13.6	-6.4
26	59.8	27.9	12.4	61.8	25.7	12.5	2.1	- 2.1	0.2
27	60.7	27.0	12.3	76.8	15.9	7.3	16.2	-11.1	-5.0
28	61.5	26.7	11.8	79.9	14.0	6.1	18.5	-12.7	-5.7
29	61.8	26.3	12.0	92.5	5.3	2.1	30.8	-20.9	-9.8
Average absolute deviation							11.5	- 8.7	-3.4

In contrast to the linearized theory, the Krishna/Standart model allows for the nonideality of the liquid phase through the introduction of the matrix Γ (Eq. 16). The comparison between results calculated with and without the assumption $\Gamma = 1$ (Variant 3) shows that this simplification has no effect.

TABLE 4
Simplifications of Mass Transfer Models Used in This work

Simplification	Diffusion mass fluxes	Type of mass transfer process	Liquid phase	Cross effects have been neglected	Mass transfer resistance
1	Low	Nonequimolar	Real	No	In both phases
2	Low	Equimolar	Real	No	In both phases
3	Low	Nonequimolar	Ideal	No	In both phases
4	Low	Nonequimolar	Real	Yes	In both phases
5	Low	Nonequimolar	Real	No	In the vapor phase only

Neglecting of cross effects in both phases (the matrices K_L and K_G become diagonal—Variant 4) results in a large disagreement between calculated and observed compositions so that using the simple Eq. (1) for the prediction of concentration profiles may lead to large errors.

Even assuming transfer resistance only in the gas phase leads to smaller errors compared to neglecting cross effects. Mass transfer resistance in the liquid phase should be, however, considered in design procedures.

The concentration profiles, calculated using the above simplifications (Table 4), are shown in Fig. 3 for Experiment 18 as an example.

Influence of Experimental Errors of the Model Parameters on the Prediction of Product Compositions

The significance of experimentally determined multicomponent mass transfer model parameters in the design of distillation processes has been presented elsewhere (40) with the conclusion that the error of VLE data as well as the correlation equations for binary diffusivities and mass transfer coefficients have the biggest influence on the calculated results.

In this paper the effect of VLE data has been analyzed using four specially chosen correlations for predicting activity coefficients: the Wilson equation with parameters determined by Nagata (48) and Verhoye and de Schepper (49) and the polynomial equations of Toor (42) and Konstantinov (50). A comparison between measured and calculated distillate compositions using the above equations is given in Figs. 4a, 4b, and 4c. The largest disagreement between experimental and theoretical data is obtained using the Wilson equation and the smallest one from applying the Konstantinov equation.

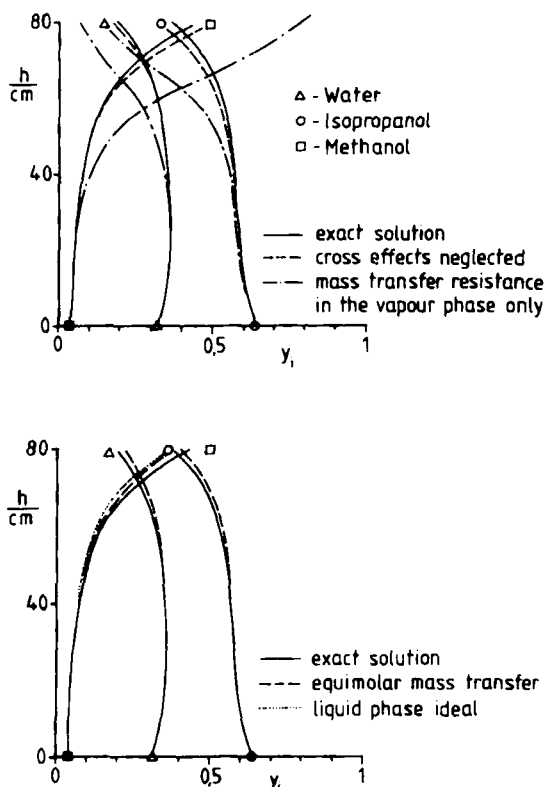


FIG. 3. Influence of model simplifications on calculated concentration profiles (based on Experiment 18).

The binary mass transfer coefficients necessary to calculate the elements of the matrices \mathbf{B}_G and \mathbf{B}_L (Eq. 15) are in general calculated from equations describing the mass transfer kinetics in the corresponding binary subsystems. Mass transfer coefficients are usually correlated as functions of the physical properties and the hydrodynamic conditions with an accuracy between 15 and 50%. For instance, the recently generalized correlation for mass transfer in packed distillation columns with randomly arranged rings and saddle types elaborated by Bravo and Fair (51) offers a correlating accuracy of $\pm 20\%$.

An elaborate study on hydrodynamics and mass transfer SULZER CY packing was carried out by Zogg (52). He proposes the following correlation equations for the mass transfer coefficients in the liquid phase:

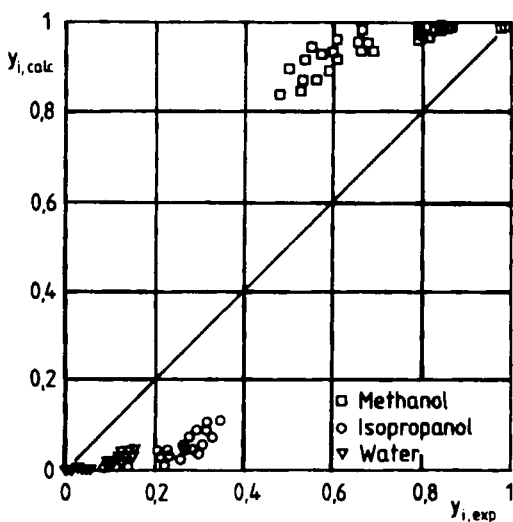


FIG. 4a. Comparison of experimental and theoretical distillate compositions calculated from VLE prediction equation of Toor (42).

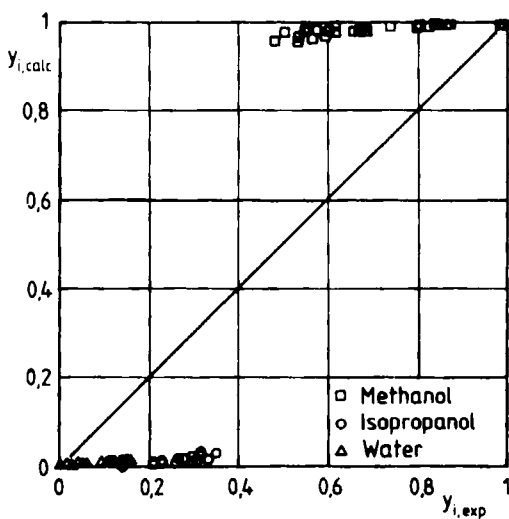


FIG. 4b. Comparison of experimental and theoretical distillate compositions calculated from VLE prediction equation of Nagata (48).

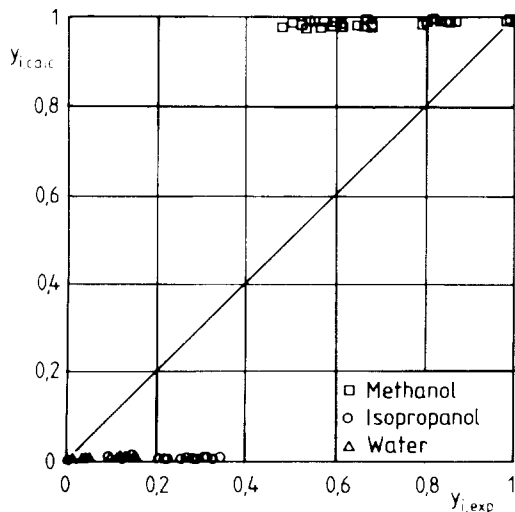


FIG. 4c. Comparison of experimental and theoretical distillate compositions calculated from VLE prediction equation of Verhoye and de Schepper (49).

$$\overline{Sh}_L = 3.415 + 1.2496 \sqrt{\frac{Re_L Sc_L}{\xi}} \exp \left(\frac{-0.72054 \xi^{0.6}}{(Re_L Sc_L)^{0.45801}} \right) \quad (17)$$

The correlation equation for the vapor phase mass transfer coefficient is

$$\overline{Sh}_G = 0.05144 Re_G^{0.8034} Sc_G^{1/3} \quad (18)$$

Zogg claims a maximum error of $\pm 2.2\%$ for Eq. (17) and a mean and maximum error of $\pm 6.9\%$ and $\pm 15\%$, respectively, for Eq. (18).

The mass transfer coefficients calculated from the correlations were increased or diminished by the mean or maximum error and used subsequently in the simulation method. The corresponding results were then compared with the experimental data. The comparison listed in Table 5 and Figs. 5a and 5b shows a significant effect which is proportional to the size of the error.

The same procedure was used to determine the effect of an error in the estimation of the binary diffusivities on the agreement between calculated and measured product compositions. The data presented in Table 6 and Figs. 6a and 6b give the same trend as observed with a change in the binary mass transfer coefficients.

TABLE 5
Compositions Computed Using Binary Mass Transfer Coefficients in Both Phases Reduced or Increased by Mean or Maximum Error of the Correlation Equations

Mean error of correlation										Maximum error of correlation			
Reduced					Increased					Reduced		Increased	
x_1	x_2	x_3	x_1	x_2	x_3	x_1	x_2	x_3	x_1	x_2	x_3	x_1	x_2
0.988	0.009	0.003	0.994	0.004	0.002	0.982	0.013	0.005	0.996	0.096	0.005	0.996	0.096
0.990	0.007	0.003	0.996	0.003	0.001	0.984	0.011	0.005	0.997	0.097	0.005	0.997	0.097
0.999	0.001	0.000	0.999	0.001	0.000	0.998	0.002	0.001	1.000	0.000	0.001	1.000	0.000
0.997	0.002	0.001	0.999	0.001	0.000	0.996	0.003	0.001	0.999	0.003	0.001	0.999	0.003
0.994	0.004	0.002	0.998	0.002	0.001	0.991	0.007	0.003	0.998	0.007	0.003	0.998	0.007
0.943	0.041	0.016	0.970	0.022	0.008	0.917	0.059	0.024	0.979	0.059	0.024	0.979	0.059
0.977	0.016	0.006	0.989	0.008	0.003	0.965	0.025	0.010	0.993	0.025	0.010	0.993	0.025
0.978	0.016	0.006	0.989	0.008	0.003	0.968	0.023	0.009	0.992	0.023	0.009	0.992	0.023
0.988	0.009	0.003	0.994	0.004	0.002	0.981	0.014	0.005	0.996	0.014	0.005	0.996	0.014
0.963	0.042	0.016	0.969	0.022	0.008	0.918	0.058	0.023	0.978	0.058	0.023	0.978	0.058

11	0.979	0.015	0.006	0.990	0.007	0.003	0.968	0.023	0.009	0.993	0.005	0.007
12	0.957	0.031	0.012	0.977	0.017	0.006	0.937	0.045	0.018	0.984	0.012	0.004
13	0.862	0.098	0.040	0.923	0.056	0.021	0.808	0.134	0.058	0.945	0.040	0.015
14	0.889	0.079	0.032	0.941	0.043	0.016	0.841	0.111	0.048	0.958	0.031	0.011
15	0.975	0.018	0.007	0.988	0.009	0.003	0.962	0.027	0.011	0.992	0.006	0.002
16	0.845	0.109	0.046	0.915	0.061	0.024	0.780	0.152	0.068	0.939	0.044	0.017
17	0.899	0.071	0.030	0.948	0.038	0.015	0.831	0.104	0.045	0.964	0.026	0.010
18	0.323	0.432	0.244	0.522	0.317	0.161	0.239	0.480	0.281	0.630	0.250	0.120
19	0.489	0.334	0.177	0.702	0.202	0.096	0.360	0.410	0.230	0.789	0.145	0.066
20	0.361	0.411	0.228	0.565	0.290	0.145	0.264	0.466	0.270	0.663	0.229	0.108
21	0.624	0.251	0.125	0.794	0.142	0.064	0.486	0.335	0.178	0.855	0.101	0.044
22	0.486	0.337	0.176	0.682	0.217	0.102	0.366	0.407	0.226	0.766	0.162	0.072
23	0.799	0.139	0.062	0.900	0.071	0.029	0.725	0.187	0.088	0.930	0.050	0.020
24	0.436	0.367	0.197	0.655	0.234	0.111	0.366	0.425	0.239	0.745	0.176	0.079
25	0.731	0.183	0.086	0.865	0.095	0.040	0.635	0.244	0.121	0.906	0.067	0.027
26	0.551	0.297	0.152	0.747	0.174	0.079	0.441	0.363	0.196	0.816	0.128	0.055
27	0.715	0.193	0.092	0.857	0.100	0.043	0.612	0.258	0.130	0.401	0.070	0.029
28	0.761	0.165	0.074	0.873	0.090	0.037	0.683	0.215	0.102	0.909	0.066	0.026
29	0.906	0.066	0.027	0.955	0.033	0.013	0.867	0.093	0.040	0.969	0.022	0.009

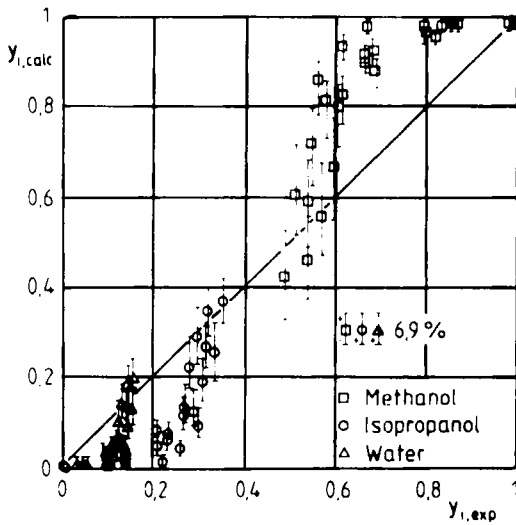


FIG. 5a. Comparison of experimental and calculated distillate compositions using mass transfer coefficients reduced or increased by 6.9% (mean error of correlation equation).

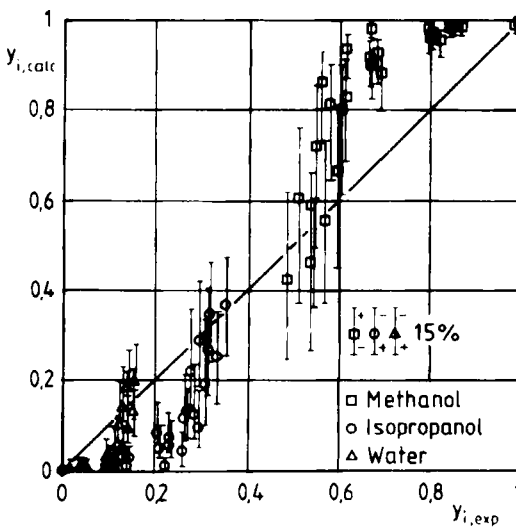


FIG. 5b. Comparison of experimental and calculated distillate compositions using mass transfer coefficients reduced or increased by 15% (maximum error of correlation equation).

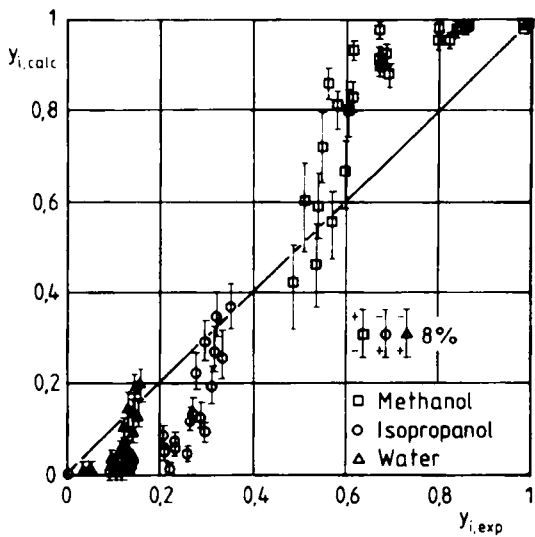


FIG. 6a. Comparison of experimental and theoretical distillate compositions calculated using binary diffusivities reduced or increased by 8% (mean error of correlation).

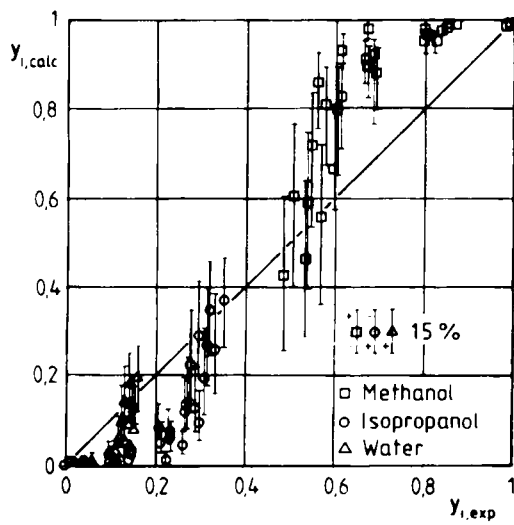


FIG. 6b. Comparison of experimental and theoretical distillate compositions calculated using binary diffusivities reduced or increased by 15% (maximum error of correlation).

TABLE 6
Distillate Compositions Computed Using Binary Diffusivities Reduced or Increased by Mean or Maximum Error of Correlation Equation

Experiment	Maximum error of correlation						Mean error of correlation					
	Increased			Reduced			Increased			Reduced		
	x ₁	x ₂	x ₃	x ₁	x ₂	x ₃	x ₁	x ₂	x ₃	x ₁	x ₂	x ₃
1	0.995	0.003	0.007	0.985	0.011	0.004	0.994	0.004	0.002	0.989	0.008	0.003
2	0.996	0.003	0.001	0.987	0.009	0.004	0.995	0.003	0.001	0.491	0.007	0.003
3	0.999	0.000	0.000	0.998	0.001	0.000	0.999	0.001	0.000	0.999	0.001	0.000
4	0.999	0.001	0.000	0.996	0.003	0.001	0.999	0.001	0.000	0.997	0.002	0.001
5	0.998	0.001	0.001	0.993	0.005	0.002	0.997	0.002	0.001	0.995	0.004	0.001
6	0.976	0.018	0.007	0.928	0.052	0.020	0.969	0.022	0.008	0.945	0.040	0.015
7	0.991	0.006	0.002	0.971	0.021	0.008	0.989	0.008	0.003	0.978	0.016	0.006
8	0.991	0.007	0.002	0.973	0.020	0.007	0.988	0.009	0.003	0.979	0.015	0.006
9	0.995	0.004	0.001	0.984	0.011	0.004	0.994	0.005	0.002	0.988	0.009	0.003
10	0.975	0.018	0.007	0.928	0.052	0.020	0.969	0.023	0.008	0.944	0.041	0.015
11	0.992	0.006	0.002	0.974	0.019	0.007	0.959	0.008	0.003	0.980	0.014	0.005

12	0.982	0.013	0.005	0.945	0.040	0.015	0.977	0.017	0.006	0.958	0.031	0.012
13	0.938	0.045	0.017	0.826	0.123	0.051	0.922	0.056	0.022	0.864	0.097	0.039
14	0.952	0.035	0.013	0.858	0.100	0.042	0.939	0.044	0.017	0.891	0.078	0.031
15	0.990	0.007	0.003	0.969	0.023	0.009	0.987	0.009	0.003	0.977	0.017	0.007
16	0.931	0.049	0.019	0.802	0.138	0.060	0.913	0.062	0.025	0.847	0.108	0.045
17	0.959	0.030	0.012	0.871	0.091	0.038	0.946	0.038	0.015	0.902	0.070	0.029
18	0.598	0.269	0.133	0.258	0.470	0.272	0.519	0.318	0.163	0.324	0.431	0.243
19	0.759	0.165	0.077	0.398	0.388	0.213	0.694	0.206	0.099	0.497	0.330	0.173
20	0.639	0.243	0.118	0.287	0.453	0.259	0.561	0.292	0.147	0.364	0.409	0.226
21	0.834	0.115	0.051	0.533	0.308	0.159	0.787	0.146	0.067	0.631	0.247	0.122
22	0.742	0.177	0.081	0.400	0.389	0.211	0.679	0.218	0.103	0.490	0.336	0.175
23	0.919	0.057	0.023	0.757	0.167	0.076	0.893	0.076	0.032	0.818	0.126	0.056
24	0.718	0.192	0.089	0.366	0.408	0.220	0.638	0.244	0.118	0.469	0.348	0.183
25	0.892	0.076	0.032	0.677	0.218	0.105	0.855	0.101	0.044	0.757	0.167	0.077
26	0.795	0.142	0.063	0.477	0.343	0.180	0.733	0.182	0.084	0.585	0.277	0.138
27	0.887	0.079	0.034	0.656	0.231	0.113	0.845	0.107	0.047	0.743	0.175	0.082
28	0.897	0.073	0.030	0.714	0.196	0.090	0.866	0.095	0.040	0.781	0.152	0.067
29	0.964	0.026	0.010	0.886	0.081	0.034	0.951	0.035	0.014	0.916	0.059	0.026

Results from Different Mass Transfer Models

Finally, the two following other design methods have been tested, based on:

Linearized Stefan Maxwell equation (37)

Simple HETP-NTP method

From a comparison of the results from the linearized theory in Fig. 7 with the results from the exact solution in Fig. 2, it is obvious that the linearized theory gives good agreement between calculated and experimental data and thus confirms previous conclusions (39, 46, 47). The exact solution of Krishna/Standart is recommended for distillation calculations at small mass fluxes, however (40).

Prediction of product compositions by the NTP-HTP method is significantly more simple than using the exact or linearized solution of the Stefan-Maxwell equation.

In order to check the suitability of the NTP-HTP method, the number of theoretical plates (NTP) was first determined from dividing the height of the experimental packing (0.8 m) by the HETP value as given by Meier (44) and presented in Fig. 8. Product compositions were subsequently calculated using the above obtained NTP values and VLE data from the Konstantinov

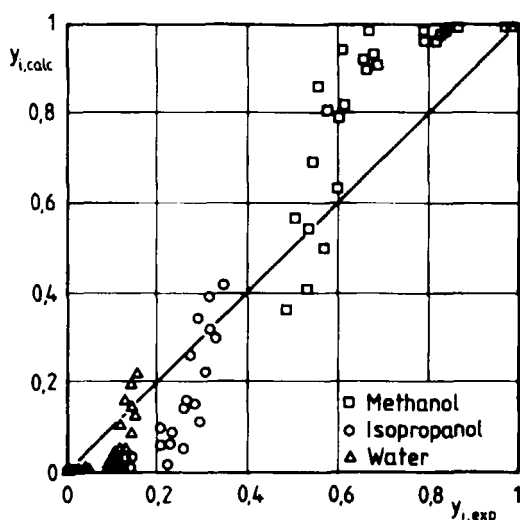


FIG. 7. Comparison of calculated and experimental distillate compositions calculated from the linearized theory (37).

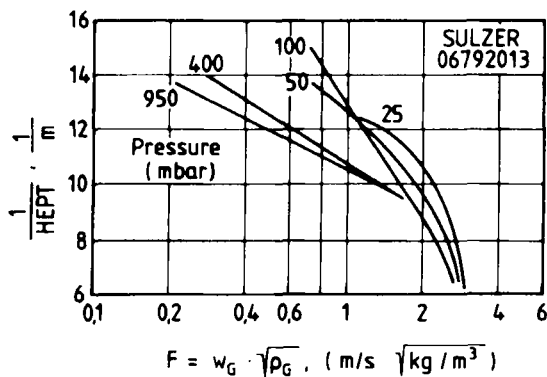


FIG. 8. Diagram used for HETP values.

equation. The agreement between the calculated and experimental data is significantly worse than in Fig. 2 where the kinetic model of mass transfer is used. The reason for the poor performance of the NTP-HETP method is seen in the very simplified kinetic dependence of Fig. 8.

CONCLUSIONS AND RECOMMENDATIONS

In a theoretical and experimental study the distillate concentrations for ternary distillation of methanol/isopropanol/water in a column of 0.1 m diameter packed with SULZER CY ordered packing have been compared. The theoretical predictions are based on a solution of the Maxwell-Stefan diffusion equation given by Krishna and Standart (38). The mean absolute deviation between calculated and experimental product compositions for all experiments and the components involved are

Methanol	$\Delta x_1 = .115$
Isopropanol	$\Delta x_2 = -.087$
Water	$\Delta x_3 = -.034$

The Krishna/Standart solution which neglects the effect of large fluxes gives the same results as the exact solution. The diffusional interactions between the transferred species as well as the mass transfer resistance in the liquid phase were found to affect the calculated concentration profiles significantly. Distillation of the test system may be treated as an equimolar process.

The VLE prediction method proved to have the largest effect on the calculated product compositions. For the system investigated, the best agreement was reached using the equation of Konstantinov (50). The error of the correlation equations for the binary mass transfer coefficients as well as for the binary diffusivities also plays the important role.

The agreement between experimental and calculated concentrations by the "plate to plate" method is very poor compared to the results from the multicomponent mass transfer model (Fig. 9). In general, the results obtained in this study are about 40% worse than those from other published experimental investigations (ethanol/benzene/*n*-heptane (53), acetone/methanol/benzene (46)). This is attributed to the strong sensitivity of the system investigated in VLE data and other model parameters (7, 40).

With respect to calculating multicomponent distillation, the conclusions from this investigation are:

Design methods taking into account mass transfer kinetics are significantly more accurate than the simple HETP-NTP method

The effect of large mass fluxes may be neglected in distillation, which significantly simplifies the calculation procedure

Distillation may be treated as an equimolar diffusion process

The most accurate VLE prediction method should be used

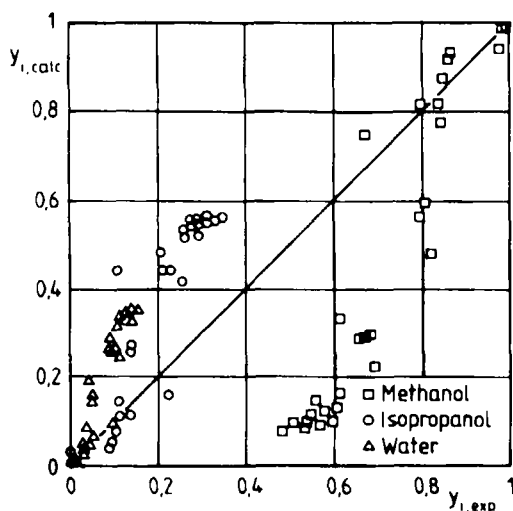


FIG. 9. Comparison of experimental and theoretical distillate compositions calculated from NTP-HETP model (VLE prediction equation of Konstantinov (50)).

The simulation method of multicomponent distillation in packed columns presented, verified by our experiments with the methanol/isopropanol/water system, can be recommended as sufficiently accurate for design purposes

Acknowledgment

A.G. thanks Mr I. Schmidt for his help with the experimental investigations and his numerical computations.

SYMBOLS

A	mass transfer surface (m^2)
D_{ij}	binary diffusivity (m^2/s)
G	vapor stream in the column (mol/s)
h	packing height (m)
k_{ij}	binary mass transfer coefficient in one phase ($\text{mol}/\text{m}^2\text{s}$)
L	liquid stream in the column (mol/s)
n	number of components in the mixture
N_i	molar flux of component i ($\text{mol}/\text{m}^2\text{s}$)
r_i	heat of vaporization (J/mol)
x_i	mole fraction of component i in liquid
y_i	mole fraction of component i in vapor

Quadratic Matrices

B	matrix defined by Eq. (15)
E	matrix which describes the influence of great mass fluxes on mass transfer coefficients
k	matrix of mass transfer coefficients in one phase in multicomponent mixtures
K_{OG}	matrix of mass transfer coefficients in multicomponent mixtures
M	matrix defined by Eq. (9)
β	matrix defined by Eq. (7)
Γ	matrix of activity coefficients
Φ	matrix defined in Eq. (14)
Θ	matrix defined in Eq. (12)
I	unit matrix

Vectors

\mathbf{N}	vector of molar fluxes
\mathbf{x}	vector of mole fractions in the liquid phase
\mathbf{y}	vector of mole fractions in the vapor phase
$\Delta\mathbf{y}$	vector of driving forces

Subscripts

i, j, k, n	number of component
G	vapor phase
L	liquid phase

Superscripts

*	equilibrium state
.	quantity modified by the effect of finite rates of transfer
-	average value

Greek

γ_i	activity coefficient of component i
δ_{ij}	Kronecker delta
ξ	dimensionless film length

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