Int. J. Heat Mass Transfer. Vol. 35, No. 11, pp. 2963–2968, 1992
Printed in Great Britain

0017–9310/92 $5.00+0.00

The use of gas absorption correlations for mass transfer coefficients in distillation processes

G. OVEJERO,† R. VAN GRIEKEN, L. RODRIGUEZ and J. L. VALVERDE
Department of Chemical Engineering, School of Chemistry, Complutense University,
Madrid 28040, Spain

(*Received* 28 *January* 1991 *and in final form* 6 *August* 1991)

Abstract—Individual mass transfer coefficients are measured under conditions where the involved overall resistance to the process is due mainly to one of the phases. The difficulty in reaching such conditions in distillation and the analogy of this unit operation with absorption suggest the use of individual coefficient correlations obtained in absorption experiments to design distillation columns. In this work a semi-empirical equation relating coefficients with physical properties of the two phases of distillation was obtained. The comparison among values of the mass transfer coefficients obtained by this equation and by the one determined from absorption experiments shows an acceptable agreement.

1. INTRODUCTION

NOWADAYS, a progressive tendency is observed towards the substitution of tray columns by packed bed columns in most of the mass transfer operations such as absorption and distillation. There are two important reasons for this change. The first one is that the new packing type allows a good distribution and flow of phases with a high interfacial contact area and low energy consumption; the second one is the decrease in several costs (column, support, etc.) due to the design of new, light, resistive packings.

To design this kind of column it is necessary to know the mass transfer resistance and the interfacial surface, with a complex geometry, between both phases. These phenomena may be described by the individual mass transfer coefficients (k_G and k_L) and by the specific interfacial surface, a, defined for the corresponding type, size packing.

Generally, the experimental determination of k_G, k_L and a-values is easy in absorption compared with distillation, where the experimental conditions make it difficult to assume that mass transfer resistance is due just to one of the phases.

For a binary system, according to Fick's law, the molar flux of component i, N_i, may be expressed as:

$$N_i = \frac{y_{i_0}-y_i}{\dfrac{1}{k_{G_i}p}} = \frac{x_i-x_{i_0}}{\dfrac{1}{k_{L_i}c_t}} = \frac{y_{i_e}-y_i}{\dfrac{1}{K_{G_i}p}} = \frac{x_i-x_{i_e}}{\dfrac{1}{K_{L_i}c_t}}. \tag{1}$$

Assuming equilibrium at the interfacial surface, the overall coefficients are calculated as a function of the individual coefficients by means of

$$\frac{1}{K_{G_i}p} = \frac{1}{k_{G_i}p} + \frac{m'}{k_{L_i}c_t} = \frac{m''}{K_{L_i}c_t} \tag{2}$$

in a differential element, dA, with the following definitions for m' and m'':

$$m' = \frac{y_i - y_{i_0}}{x_i - x_{i_0}}, \quad m'' = \frac{y_{i_e} - y_i}{x_i - x_{i_e}}. \tag{3}$$

Traditionally in mass transfer analysis of the distillation processes, the assumption of equimolar transfer ($N_t = \Sigma_{i=1}^{n} N_i = 0$) establishes one of the major differences with the absorption processes. Therefore, the use of absorption coefficient correlations to determine distillation coefficients implies the corresponding modifications of the equations.

Different authors using wetted-wall columns [1–8] and Raschig ring bed columns [9, 10] in the distillation process achieved results pointing out that most of the resistance was in the vapor phase. Other authors, however, using packed bed columns [4, 11–15] and wetted-wall columns [16] concluded that resistance in the liquid phase may reach appreciable values. Finally, another research group found that the overall resistance to mass transfer was a function of phase composition [17].

From these results and with a view to the design of industrial columns for distillation operations, different workers suggested the use of individual mass transfer coefficients obtained in analogous packed bed columns from absorption experiments.

The aim of the present work is to compare the use of correlations determined from absorption experiments to predict mass transfer coefficients for the distillation process.

2. EXPERIMENTAL: APPARATUS AND PROCEDURE

Figure 1 shows a schematic diagram of the experimental apparatus used. The contactor is basically

---

† Author to whom correspondence should be addressed.

NOMENCLATURE

a	specific interfacial surface of the packing $[m^2 m^{-3}]$
a_S	specific total dry surface of the packing $[m^2 m^{-3}]$
c_t	total molar concentration $[kmol\ m^{-3}]$
D_L	solute diffusivity through liquid phase $[m^2 h^{-1}]$
Er	efficiency packing number $[1/a_S M]$
g	gravitational acceleration $[m\ s^{-2}]$
Ga	Galileo number $[g\rho_L^2/\mu_L^2 a_S^3]$
k_G	individual mass transfer coefficient for the vapor phase $[kmol\ h^{-1} m^{-2} bar^{-1}]$
k_L	individual mass transfer coefficient for liquid phase $[m\ h^{-1}]$
K_G	overall vapor phase mass transfer coefficient $[kmol\ h^{-1} m^{-2} bar^{-1}]$
K_L	overall liquid phase mass transfer coefficient $[m\ h^{-1}]$
L	molar flow of liquid phase $[kmol\ h^{-1}]$
L_m	specific wetting rate $[m^2 s^{-1}]$
M	height of the packing [m]
Mo	wetness capacity number $[\sigma_C/\sigma_L]$
N	molar flux $[kmol\ h^{-1} m^{-2}]$
p	total pressure [bar]
Re_G	Reynolds-number for vapor phase $[V_R \rho_G/a_S \mu_G]$
Re_L	Reynolds-number for liquid phase $[L_m \rho_L/\mu_L]$
S	transversal surface $[m^2]$
Sc	Schmidt-number $[\mu/\rho D]$
Sh_G	Sherwood-number for vapor phase $[k_G RT/V]$
Sh_L	Sherwood-number for liquid phase $[k_L/D_L a_S]$
Ts	surface tension number $[\mu_L^2 a_S/\rho_L \sigma_L]$
V	molar flow for vapor phase $[kmol\ h^{-1}]$
V_R	relative speed $[m\ s^{-1}]$
x_i	molar fraction of i component in the liquid phase
$(x_i)_0$	interfacial molar fraction of i component in the liquid phase
$(x_i)_e$	molar fraction of the i component in the liquid phase in equilibrium with a y_i molar fraction
y_i	molar fraction of i component in the vapor phase
$(y_i)_0$	interfacial molar fraction of i component in the vapor phase
$(y_i)_e$	molar fraction of the i component in the vapor phase in equilibrium with a x_i molar fraction
z	height of the column [m].

Greek symbols

$\Lambda_{12}, \Lambda_{21}$	Wilson parameters
μ_L	liquid viscosity $[kg\ m^{-1} s^{-1}]$
ρ_L	liquid density $[kg\ m^{-3}]$
σ_C	critical surface tension $[N\ m^{-1}]$
σ_L	liquid surface tension $[N\ m^{-1}]$.

constituted by a Pyrex glass column containing a string of graphite spheres and cylinders. The string was formed by 20 spheres of 18 mm diameter and 19 alternated cylinders of 6 mm diameter and height. The column was provided with seven special probes to take liquid samples from the liquid film. Reflux from the condenser, collected in the upper part of the column, was uniformly distributed at the top of the string to maintain a uniform film formation at the entrance of the column. At the bottom of the column, the liquid coming from the string was transferred to the boiler.

The boiler was heated by means of an electric resistance in order to maintain a constant boiling rate. Temperatures were measured in the boiler and at the entrance and the exit of the condenser.

Liquid samples were analyzed using a Perkin-Elmer SIGMA 3B gas chromatograph with a F.I.D. detector connected to an automatic integrator. PORAPACK Q 80/100 mesh size was used as the packing material (adsorbent) for the G.C. column.

2.1. *Total reflux experiments*

Experiments were run at total reflux and under steady-state conditions, which considerably simplify the overall balances to give $x = y$ and $L = V$ at any point in the column, which, when combined with the operating advantages, makes total reflux experiments more practical and attractive. First, the boiler was charged with about 5 l of mixture, the column top pressure was adjusted with an inert stream (nitrogen) in order to obtain a constant pressure of 760 mmHg for all the experiments, and the heater of the boiler was adjusted to give the appropriate boiling rate conditions. Then, the system was operated for approximately 2 h to reach steady state conditions. Steady state was checked by successive measurements of the composition. After this period, samples were removed from the probes at several column heights. This procedure provided composition data minimizing liquid loss from the column without disturbing the continuity of the liquid film flow inside the column.

The sequence of data collection from different parts of the column was repeated twice consecutively, with about 15 min between reading or sampling at the same height of the column.

The composition ranges covered in the experiments were approximately a 0.3–0.95 mole fraction of the light component (methanol) for the methanol–water binary mixture; a 0.1–0.6 mole fraction for the iso-

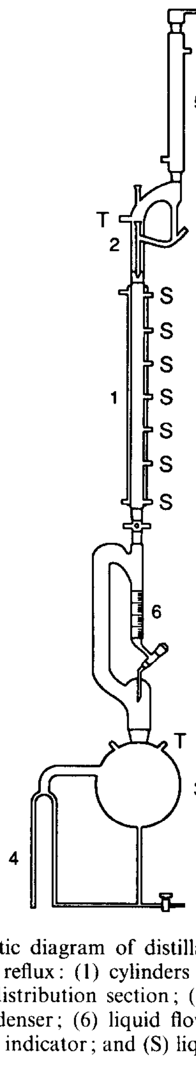

FIG. 1. Schematic diagram of distillation column operating under total reflux: (1) cylinders and spheres column; (2) reflux and distribution section; (3) boiler; (4) electric heater; (5) condenser; (6) liquid flow measure; (T) temperature indicator; and (S) liquid sampling.

Table 1. Coefficients for equation (6) proposed by Costa *et al.* [21]

Packing type	C	B	D
Pall rings	264.84	0.13	0.44
Raschig rings	414.08	0.15	0.65
Bundle rings	89.94	0.13	0.27
Hy-pack rings	25.23	0.13	0.00
Intalox saddles	43.76	0.13	0.13
Berl saddles	29.50	0.13	0.00

propanol–water binary mixture and 0.1–0.8 for the methanol–isopropanol binary mixture. The range of Reynolds-number was between 200 and 900 with Schmidt-numbers from 0.4 to 1.0. Reproducibility of the data was checked by carrying out an experimental run twice.

3. RESULTS AND DISCUSSION

3.1. *Temperature–composition relationship*

The state of the liquid phase is critical since it determines whether the resistance to mass transfer is mainly in the vapor phase or not. For wetted-wall column distillation of the methanol–water binary system, Kayihan *et al.* [8] found that the liquid phase was saturated, i.e. the measured liquid phase bulk temperature and bulk composition were found to lie on the bubble point curve. This suggested for their system that mass transfer resistance lay completely in the vapor phase. Other authors found the same experimental facts for other mixtures [18]. Bearing in mind that the mechanism for heat and mass transfer in the liquid phase for all binary mixtures in distillation should be the same, it is reasonable to expect the liquid phase of most other systems to be saturated as well. So it will be assumed that there is saturation of the liquid phase for all the mixtures used. In order to estimate vapor phase thermodynamics properties it also is a good approximation to assume that the vapor phase was at its dew temperature. The mass transfer for the distillation process in a column like the one used in this work may be described only in terms of overall vapor phase mass transfer coefficients.

3.2. *Evaluation of mass transfer coefficients*

The experiments were carried out using the 'differential height' technique described by Danckwerts *et al.* [19]. This technique, which was designed for absorption experiments, operates with a constant boiler concentration and removes samples at different heights of the column. Assuming liquid phase saturation and equality of the heat and mass transfer areas, the vapor overall mass transfer coefficients may be obtained in a straightforward manner: the rate of mass transfer in a differential element of volume within the column is given by

$$\mathrm{d}[Vy] = K_G aSp(y_e - y)\,\mathrm{d}z \tag{4}$$

where the transversal surface, S, was 5.2685×10^{-4} m^2.

Under total reflux conditions

$$V = L = \text{constant}$$

so

$$K_G = \frac{V}{Sap}\frac{1}{(y_e - y)}(\mathrm{d}y/\mathrm{d}z) \tag{5}$$

where a-values were calculated by means of the equation

$$\frac{a}{a_S} = C\,Re_L^B Er^D Ts^{0.38} Mo^{0.18} \tag{6}$$

proposed by Costa *et al.* [21] (C, D and B values are shown in Table 1).

The pressure, p, was maintained at 760 mmHg and equilibrium data for the vapor phase (y_e) were cal-

Table 2. Wilson parameters for the three binary mixtures used in this work

Binary mixture	Wilson parameters Λ_{12}	Λ_{21}	Refs.
Methanol–water	0.4240	1.0140	[23] [24] [25] [26] [27]
Isopropanol–water	0.0891	0.6846	[28] [29] [30]
Methanol–isopropanol	2.3660	0.2890	[29] [31]

culated using the Wilson equation. The parameters of this equation are shown in Table 2.

Figure 2 shows the composition data plotted as a function of column height for the three different binary mixtures used at constant vapor flow.

Local values of K_G were calculated at several column heights where (dy/dz) values were measured from the slope of the y–z curve.

According to the heat and mass transfer analogy which was used for the vapor phase, *Sh* and *Sc* are a function of *Re*. Based on this expression, Costa *et al.* [20, 21] obtained a correlation for the individual mass transfer coefficients with packed columns by means of gas absorption data. In this correlation k_G and k_L were determined from experiments of previous work of the same authors. The equation [21] is given by

$$\frac{k_L}{D_L a_S} = 2.79\, Re_L^{2/3} Sc^{1/3} Ga^{0.38} Ts^{0.42} \tag{7}$$

$$\frac{k_G RT}{V} = 0.195\, Re_G^{-1/3} Sc^{-1/2} (p/p_B)^3. \tag{8}$$

Considering the similarities and differences with the absorption process, a similar correlation was assumed for distillation experiments:

$$\frac{k_G RT}{V} = a\left(\frac{V_R \rho_G}{a_S \mu_G}\right)^b \left(\frac{\mu_G}{\rho_G D_G}\right)^c \tag{9}$$

and constants a, b and c were estimated by means of the non-linear regression method of Marquardt [22]. Reynolds-, Schmidt- and Sherwood-numbers were calculated from experimental data.

Initial estimations of a, b and c were taken from the gas absorption results, equation (8). The best-fit correlation was

$$\frac{k_G RT}{V} = 0.0575\, Re_G^{-0.2} Sc_G^{-0.24}. \tag{10}$$

A statistical study of this expression showed a mean deviation below 20%.

However, distillation experiments are difficult to achieve and only under special conditions is it possible

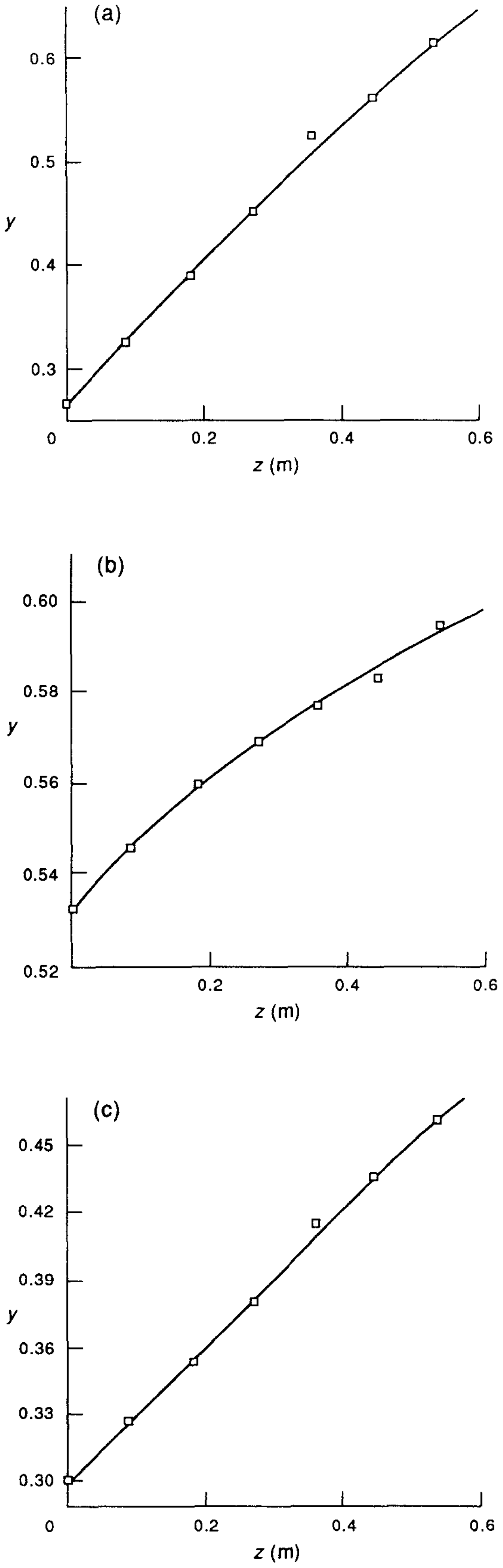

FIG. 2. Mole fraction vs column height for: (a) methanol–water system (methanol); (b) isopropanol–water system (isopropanol); and (c) methanol–isopropanol system (methanol).

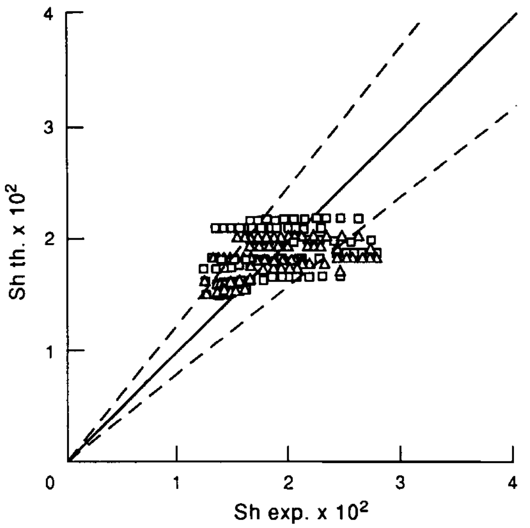

FIG. 3. Comparison of experimental and theoretical values of Sherwood numbers: (△) equation (8) and (□) equation (10).

to assume that most of the resistance to mass transfer lies in the vapor phase. All these problems do not appear when gas absorption experiments were carried out and it is easy to find correlations for mass transfer coefficients. Therefore, it should be interesting to use these correlations to obtain mass transfer coefficients for the distillation processes.

A comparison among the experimental results obtained by means of equation (5) and the predicted values calculated from equation (8) (gas absorption) and equation (10) (distillation) is shown for the three binary systems in Fig. 3. The values obtained with both equations ((8) and (10)) are very close and represent the experimental distillation data within ±20% error. Both expressions represent the distillation data with a reasonable accuracy.

Finally by means of equations (2), (3), (7) and (8), it could be verified that 85–100% of the overall resistance to mass transfer lay in the vapor phase for all the experiments carried out.

REFERENCES

1. H. F. Johnstone and R. L. Pigford, Distillation in a wetted-wall column, *Trans. Am. Inst. Chem. Engng* **38**, 25–51 (1942).
2. A. L. Surowiec and C. C. Furnas, Distillation in a wetted-wall tower, *Trans. Am. Inst. Chem. Engng* **38**, 53–89 (1942).
3. R. E. Peck and E. F. Wagner, Film resistances in rectification, *Trans. Am. Inst. Chem. Engng* **41**, 737–753 (1945).
4. A. Paris and A. Delzenne, Ratio of the individual heights of a transfer unit in the two phases of a binary mixture subjected to extraction, *Genie Chimie* **86**, 83–100 (1961).
5. R. Hoffmann, Mass-transfer resistances of liquid and vapor phases in binary systems, *Ver. Deut. Ing.* **536**, 36 pp. (1969).
6. H. Michalski and M. Serwinski, Mass transfer in a wetted-wall column, *Zesz. Nauk. Politech. Lodz., Chem.* **26**, 109–128 (1972).
7. M. Serwinski and R. Zarzycki, Mass transfer in a wetted-wall column, *Zesz. Nauk. Politech. Lodz., Chem. Spozyw.* **24**, 7–34 (1973).
8. F. Kayihan, O. C. Sandall and D. A. Mellichamp, Simultaneous heat and mass transfer in binary distillation, II, *Chem. Engng Sci.* **32**, 747–754 (1977).
9. M. Serwinski, C. Strumillo and R. Zarzycki, Mass transfer in packed fractioning columns (I) effect of G_m/L_m ratio on the mass transfer resistance. *Chem. Stosowana, Ser. B*, **1**, 39–52 (1966).
10. K. J. Arwikar and O. C. Sandall, Liquid phase mass transfer resistance in a small scale packed distillation column, *Chem. Engng Sci.* **35**, 2337–2343 (1980).
11. C. C. Furnas and M. L. Taylor, Distillation in packed columns, *Trans. Am. Inst. Chem. Engng* **36**, 135–160 (1940).
12. D. W. Duncan, J. H. Kolffolt and J. R. Withrow, The effect of operating variables on the performance of a packed column still, *Trans. Am. Inst. Chem. Engng* **38**, 259–281 (1942).
13. G. W. Minard, J. M. Kolffolt and J. R. Withrow, Fibrous glass as a packing material for packed column distillation, *Trans. Am. Inst. Chem. Engng* **39**, 813–831 (1943).
14. D. W. Deed, P. W. Schutz and T. B. Drew, Comparison of rectification and desorption in packed column, *Ind. Engng Chem.* **39**, 766–774 (1947).
15. C. H. G. Hands and F. R. Whitt, Design of packed distillation columns–III, *J. Appl. Chem.* **1**, 67–73 (1951).
16. J. A. Storrow, The fractioning of binary mixtures in a wetted-wall column, *Trans. Am. Inst. Chem. Engng* **23**, 32–42 (1945).
17. J. Leffler and H. T. Cullinan, Variation of liquid diffusion coefficients with composition, *Ind. Engng Chem. Fund.* **9**, 84–93 (1970).
18. M. M. Dribika and O. C. Sandall, Simultaneous heat and mass transfer for multicomponent distillation in a wetted-wall column, *Chem. Engng Sci.* **34**, 733–739 (1979).
19. P. V. Danckwerts, W. Smith and H. Sawistowski, *International Symposium on Distillation* (Edited by P. A. Rotenburg), pp. 7–12. Institute of Chemical Engineers, London (1960).
20. E. Costa, A. Escardino, P. Fito and J. Costa, Transferencia de Materia en los Procesos de Rectificación, *An. Quim.* **62B**, 1191–1202 (1966).
21. E. Costa, G. Ovejero, M. A. Uguina and C. Lopez, Transferencia de materia en líquidos. Determinación de áreas interfaciales específicas efectivas de relleno, *An. Quim.* **85A**, 135–143 (1989); Mass transfer in liquids. Determination of effective specific interfacial areas for packing, *Int. Chem. Engng* **32**, 292–301 (1992).
22. D. W. Marquardt, An algorithm for least squares estimation of non-linear parameters, *J. Soc. Ind. Appl. Math.* **11**, 413–422 (1963).
23. J. Ocon and F. Rebolleda, Vapor–liquid equilibrium (VI), binary system MeOH–H$_2$O, *Anal. Fis. Quim.* **54B**, 525–530 (1958).
24. H. E. Hughes and J. O. Maloney, Application of radioactive tracers to diffusional operations—binary and ternary equilibrium data, *Chem. Engng Prog.* **48**(4), 192–200 (1952).
25. R. S. Ramalho, F. M. Tiller, W. J. James and D. W. Bunch, A rapid method for obtaining vapor–liquid equilibrium data, *Ind. Engng Chem.* **53**(11), 895–899 (1961).
26. S. J. Green and R. E. Vener, Vapor–liquid equilibria of formaldehyde–methanol–water, *Ind. Engng Chem.* **47**(1), 103–109 (1955).
27. E. Costa and J. Moragues, Distillation of binary liquid mixtures (I), determination of vapor–liquid equilibrium, *Anal. Fis. Quim.* **48B**, 397–408 (1952).
28. B. Choffe and L. Asselineau, Liquid–vapor equilibrium

between acetone, isoPrOH and water at 760 mm, *Revue Inst. Fr. Pétrole* **XI**(7), 948–959 (1956).

29. J. Kohoutova, J. Suska, P. J. Novak and J. Pick, Liquid–vapor equilibrium XLV: system methanol–2-propanol–water, *Collection Czechoslov. Chem. Commun.* **35**, 3210–3222 (1970).
30. A. Wilson and E. L. Simons, Vapor–liquid equilibria: 2-propanol–water system, *Ind. Engng Chem.* **44**(9), 2214–2219 (1952).
31. L. Ballard and M. Van Winkle, Vapor–liquid equilibria at 760 mm pressure, *Ind. Engng Chem.* **44**(12), 2450–2453 (1952).

UTILISATION DES FORMULES DE L'ABSORPTION DE GAZ POUR LES COEFFICIENTS DE TRANSFERT DE MASSE DANS LES MECANISMES DE DISTILLATION

Résumé—Des coefficients individuels de transfert de masse sont mesurés dans des conditions où la résistance globale au processus est due principalement à l'une des phases. La difficulté d'obtention de telles conditions en distillation et l'analogie de cette opération unitaire avec l'absorption suggèrent l'utilisation des formules des coefficients individuels obtenues dans les expériences d'absorption pour calculer les colonnes de distillation. La comparaison des valeurs des coefficients de transfert de masse obtenus avec cette équation et une determinée à partir des expériences d'absorption montre un accord acceptable.

DIE ANWENDUNG VON GASABSORPTIONS-KORRELATIONEN FÜR STOFFÜBERGANGSKOEFFIZIENTEN BEIM DESTILLATIONSPROZESS

Zusammenfassung—Individuelle Stofftransport-Koeffizienten werden unter Bedingungen gemessen, bei denen der komplexe Gesamtwiderstand des Prozesses hauptsächlich einer der Phasen zugeordnet werden kann. Die Schwierigkeit, solche Bedingungen bei der Destillation zu erreichen und die Analogie dieses Prozesses zur Absorption legen die Verwendung von individuellen Korrelationen für die Koeffizienten (aufgrund von Absorptionsexperimenten) bei der Auslegung von Destillationssäulen nahe. In der vorliegenden Arbeit wird eine halbempirische Gleichung ermittelt, welche die Koeffizienten mit den physikalischen Eigenschaften der beiden Phasen bei der Destillation in Beziehung setzt. Ein Vergleich der Stoffübergangskoeffizienten aufgrund dieser Gleichung und aufgrund von Absorptionsexperimenten zeigt eine annehmbare Übereinstimmung.

ИСПОЛЬЗОВАНИЕ СООТНОШЕНИЙ ДЛЯ ПРОЦЕССА ПОГЛОЩЕНИЯ ГАЗА ПРИ ОПРЕДЕЛЕНИИ КОЭФФИЦИЕНТОВ МАССОПРЕНОСА В ПРОЦЕССАХ ДИСЦИЛЛЯЦИИ

Аннотация—Экспериментально определяются коэффициенты массопереноса в условиях, когда суммарное сопротивление обусловлено преимущественно одной из фаз. Сложность достижения указанных условий при дисцилляции и аналогия с процессом поглощения обусловливают использование при разработке дисцилляционных колонн соотношений для индивидуальных коэффициентов, полученных в экспериментах по поглощению. В настоящем исследовании выводится полуэмпирическое уравнение, связывающее коэффициенты с физическими характеристиками обеих фаз при дисцилляции. Значения коэффициентов массопереноса, полученные из этого уравнения, удовлетворительно согласуются с результатами, найденными на основе экспериментов по поглощению.