N_{Pe_m} = $(N_{Re_m})\ (N_{Pr})$

N_{Pr} = Prandtl number $\left(\dfrac{c_p\mu}{K}\right)$

N_{Re} = Reynolds number $\left(\dfrac{4r_o u_b}{\nu}\right)$

N_{Re_m} = $\dfrac{r_o u_m}{\nu}$

Greek Letters

α = thermal diffusivity $\left(\dfrac{K}{c_p\rho}\right)$

β = dimensionless axial distance $\left(\dfrac{x}{r_o}\right)$

ϵ_h = eddy diffusivity of heat (L^2/t)

ϵ_m = eddy diffusivity of momentum (L^2/t)

ϵ = dimensionless distance from center to wall $\left(\dfrac{r}{r_o}\right)$

θ = dimensionless temperature $\left(\dfrac{T-T_o}{T_i-T_o}\right)$

λ = eigenvalue

μ = viscosity (M/Lt)

ν = kinematic viscosity (L^2/t)

ρ = density (M/L^3)

σ = $\displaystyle\int_o^\epsilon \frac{d\epsilon}{f}$

Subscripts

b = mean value

i = inlet value

l = index

m = maximum value

n = mode of eigenvalue

o = wall value

∞ = asymptotic value

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Mass Transfer and Effective Interfacial Areas in Packed Columns

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Experimental studies on the mass transfer rates were performed for three kinds of operations: vaporization of water into air; absorption of methanol vapor by water; and distillation of three binary mixtures, trichloroethylene-toluene, ethanol-water, and acetone-water, by use of columns of the same diameter packed with 25- and 15-mm. Raschig rings. Correlations are presented for the individual phase height of a transfer unit and also for the effective interfacial areas. The effective areas for a given packing vary depending on the kind of operation as well as on the liquid rate and surface tension. The effective interfacial area for vaporization is significantly larger than that for absorption. The effective area for distillation seems to be roughly the same as that for absorption, if correction is made for the effect of surface tension.

Separation of volumetric mass transfer coefficients in packed columns, such as k_Ga and k_La, into the mass transfer coefficient, k_G or k_L, and the effective interfacial area a was attempted by Shulman and co-workers (*22*) and by the present authors (*30*). However there remains a question of whether or not the effective area varies with the kind of operation. This paper compares the effective areas for vaporization and absorption and presents the results of a study on a packed distillation column. Columns of the same diameter were used throughout the three kinds of experiments.

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Part I. Absorption and Vaporization

Data on the liquid phase mass transfer rates in packed absorption columns obtained by various investigators are in approximate agreement, whereas there exists considerable disagreement among data on the gas phase mass transfer rates in packed columns. One explanation for this could be that for vaporization or gas phase controlled absorption a relatively short packed section is required, and accordingly the fractional end effects, which depend on the mechanical design of the column, are relatively large. Another possibility is the difference in the effective interfacial areas between absorption and vaporization columns. In the present experiment a single packed column, which is of the same diameter as the one for a previous study (*30*) on the liquid phase mass transfer rates, was used for the absorption of methanol vapor contained in air by water and for the vaporization of water into air. It was confirmed by calculation that the rate of methanol absorption was almost entirely controlled by the resistance of the gas phase.

EXPERIMENTAL

A glass column 12 cm. in inside diameter was packed with 15- and 25-mm. ceramic Raschig rings to the heights of 4 and 8 in. The liquid distributor consisted of seven nozzles. The liquid level in the column bottom was controlled so that the height of the shower below the packing was kept constant. The liquid outlet pipe at the bottom center was tapered to avoid entrapping of gas by the outgoing liquid. For vaporization runs the column was insulated with felt 2 in. thick.

In vaporization runs air was electrically heated before it was supplied to the column bottom. Temperature of water entering the column was so controlled in a thermostated heating tank that the temperature of water at the top of the column was within 0.1°C. of the temperature at the bottom. By this method the conditions of adiabatic humidification were reached faster than by the usual method of recirculating water.

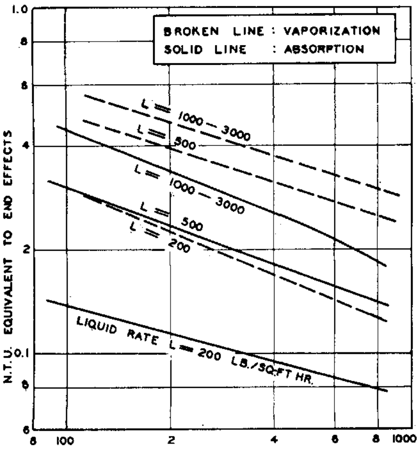

Fig. 1. End effects vary with liquid and gas rates.

In absorption runs the methanol vapor from an electrically heated evaporator was mixed with air in a separate unirrigated packed bed before the mixed gas was sent to the column bottom.

It took 2 to 4 hr. to attain steady conditions both in vaporization and absorption runs. In vaporization runs humidities of air at the inlet and exit were determined by a gravimetric method with phosphorus pentaoxide and silica gel. In absorption runs methanol vapor in a known amount of gas sample was dissolved by water, oxidized by oxalic acid into formaldehyde, and the aldehyde was analyzed by means of the Schiff's reagent and a photoelectric colorimeter. Experimental data for vaporization and absorption runs are given in Tables 1 to 4.*

CORRECTIONS FOR END EFFECTS

End effects are due to the mass transfer at the showers above and below the packed section and at the free liquid surface in the column bottom. It has been customary to express end effects in terms of an equivalent height of packing. However this method is sound only if the rate of the mass transfer due to end effects and that in the packed section proper vary exactly the same way with liquid and gas rates. The present data indicated that this was not the case.

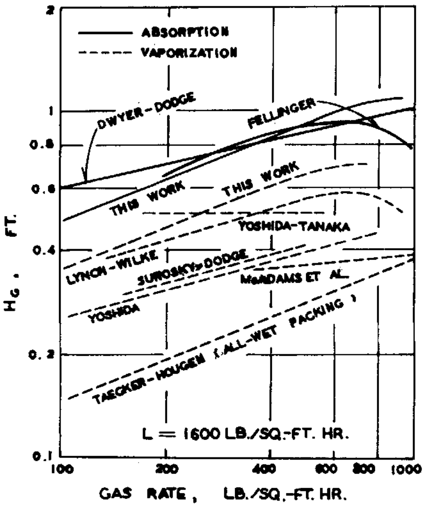

Fig. 2. Comparison of $(HTU)_G$ by various investigators. Dwyer, O. E., and B. F. Dodge, *Ind. Eng. Chem.*, 33, 485 (1941); Lynch, E. J., and C. R. Wilke, *A. I. Ch. E. Journal*, 1, 9 (1955); McAdams, W. H., J. B. Pohlenz, and R. C. St. John, *Chem. Eng. Progr.*, 45, 241 (1949); Surosky, A. E., and B. S. Dodge, *Ind. Eng. Chem.*, 42, 1112 (1950); Yoshida, F., and T. Tanaka, *ibid.*, 43, 1467 (1961); Yoshida, Fumitake, *Chem. Eng. Progr. Symposium Ser. No. 16*, 51, p. 59 (1955).

The magnitude of end effects was determined from the data for two packed heights, 4 and 8 in., on the assumption that the height of a transfer unit and the end effects are not affected by the packed height, which seems justifiable for such short packed sections. With the two packed heights no change in conditions of showers in the column bottom was observed visually. Thus for a given set of gas and liquid rates the following relationships were assumed to hold:

$$(N_t')_8 = 2(N_t)_4 + (N_t)_e \qquad (1)$$

$$(N_t')_4 = (N_t)_4 + (N_t)_e \qquad (2)$$

From Equations (1) and (2)

$$(N_t)_e = 2(N_t')_4 - (N_t')_8 \qquad (3)$$

Values of $(N_t)_e$ were calculated by the above equation from the smoothed values of $(N_t')_4$ and $(N_t')_8$ as functions of gas and liquid rates. In calculating values of $(N_t')_4$ and $(N_t')_8$ the logarithmic mean partial pressure difference was adopted both in vaporization and absorption. The equilibrium partial pressure of methanol over dilute aqueous solution was calculated by the equation presented by Zabban and Dodge (*31*).

Figure 1 shows the variation of the values of $(N_t)_e$ with liquid and gas rates. It is seen that the end effects cannot be expressed by an equivalent height of packing because the exponential effects of gas and liquid rates on the value of $(N_t)_e$ are different from those on the values of N_t for the

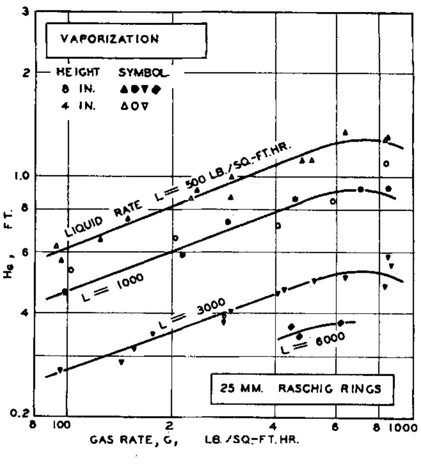

Fig. 3. Height of a transfer unit for vaporization with 25-mm. Raschig rings.

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* Tabular material has been deposited as document 7078 with the American Documentation Institute, Photoduplication Service, Library of Congress, Washington 25, D. C., and may be obtained for $2.50 for photoprints or $1.75 for 35-mm. microfilm.

packing proper. End effects are greater for vaporization than for absorption, and the difference between vaporization and absorption decreases with increasing liquid rate. At liquid rates over 1,000 lb./hr./sq. ft. the end effects are independent of liquid rates.

HEIGHT OF A TRANSFER UNIT AND EFFECTIVE INTERFACIAL AREA FOR GAS PHASE MASS TRANSFER

Figure 2 shows a comparison of the values of H_G obtained by various investigators for vaporization of water and absorption of ammonia both with air as carrier gas by use of 1-in. Raschig rings. Although there is considerable disagreement among the data, a trend is seen that the H_G values for absorption are in general higher than those for vaporization. The values of the gas phase Schmidt number for the air-ammonia and air-water vapor mixtures are nearly equal. The lowest broken line in the figure represents the data of Taecker and Hougen (*25*) on the constant-rate drying of unirrigated all-wet Raschig ring-packed beds. Their correlation is expressed by the dimensionless equation

$$(H_G)_o\, a_o = \frac{G_M}{k_G\, p_{BM}} = 0.935 \left(\frac{G\sqrt{A_p}}{\mu_G}\right)^{0.41} \left(\frac{\mu_G}{\rho_G\, D_G}\right)^{2/3} \qquad (4)$$

It is natural that H_G values for the all-wet packing are the lowest, since the whole surface area of the packing seems effective for mass transfer. The data of Shulman and co-workers (*22*) on the sublimation of dry naphthalene Raschig rings are in approximate agreement with the Taecker-Hougen correlation.

Figures 3 and 4 show the values of H_G obtained in the present work for the vaporization of water with 25- and 15-mm. Raschig rings, respectively, plotted against gas rates at various liquid rates.

Figure 5 and 6 are similar plots for the absorption of methanol vapor by water with 25- and 15-mm. Raschig rings, respectively. The H_G values in Figure 3 approximate the correlation proposed by Sherwood and Pigford (*21*), and the H_G values in Figures 5 and 6 fairly agree with the data of Fellinger (*8*) on ammonia absorption by water, after correction is made for the difference in the Schmidt number. Comparison of Figures 3 and 4 with Figures 5 and 6, respectively, when one allows for the difference in the Schmidt number, reveals that the H_G values are higher for absorption than for vaporization at the same gas and liquid rates and in the same packed column. This can be explained by assuming different values of the effective interfacial area for the two types of operation, with the same value of k_G in both cases. It can be further assumed that the value of k_G is the same as in the all-wet packing studied by Taecker and Hougen at equal values of the Reynolds and the Schmidt numbers. Then

$$(H_G)_o\, a_o = H_G\, a_e \qquad (5)$$

Thus the fractional effective interfacial area a_e/a_o is given as the ratio of $(H_G)_o$ to H_G. This is the effective interfacial area based on all-wet packings. Such a procedure of estimating the effective interfacial area was originated by Weisman and Bonilla (*26*). However their results are questionable because of their scattered experimental data.

Figures 7 and 8 show the fractional effective interfacial areas thus calculated for the 25- and 15-mm. Raschig rings, respectively. The effective interfacial areas for vaporization represented by the broken lines are considerably larger than effective interfacial areas for absorption shown by the solid lines. The effective interfacial areas are mainly affected by the liquid rate and slightly by the gas rate.

Figure 9 shows a comparison of the fractional effective interfacial areas for various systems plotted against liquid rates at a gas rate of 200 lb./hr./sq. ft. The effective interfacial area for methanol absorption roughly agrees with the effective interfacial area obtained by Shulman and co-workers (*22*) on the basis of their data on the vaporization of naphthalene Raschig rings and the Fellinger (*8*) data on ammonia absorption. The thin broken lines represent the effective interfacial area previously obtained by the present authors (*30*) from liquid phase mass transfer data by the different method mentioned later.

The difference in the effective interfacial areas between vaporization and absorption could be explained if one considers that for vaporization nearly the whole interfacial area is effective, whereas for absorption semistagnant or slowly moving parts of the liquid surface are less effective than fast moving parts. Shulman (*22*) states that the ratio of the values of k_Ga for vaporization and absorption is equal to 0.85 times the ratio of the total to operating holdups. Calculation from the data for 25-mm. Raschig rings irrigated by water indicated that this is roughly true. It will be shown later that the effective interfacial area for vaporization is nearly equal to the wetted area.

HEIGHT OF A TRANSFER UNIT AND EFFECTIVE AREA FOR LIQUID PHASE MASS TRANSFER

The correlation of Sherwood and Holloway (*20*) for the liquid phase mass transfer rates in packed columns is well known. Shulman and co-workers employed the effective interfacial area,

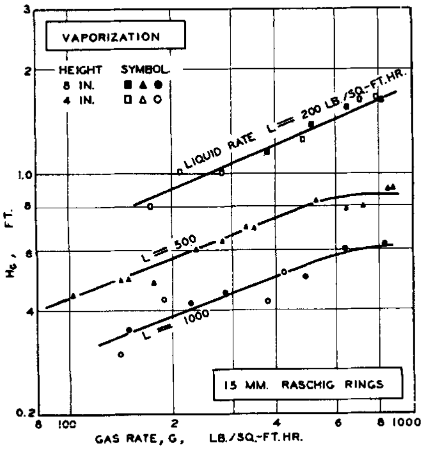

Fig. 4. Height of a transfer unit for vaporization with 15-mm. Raschig rings.

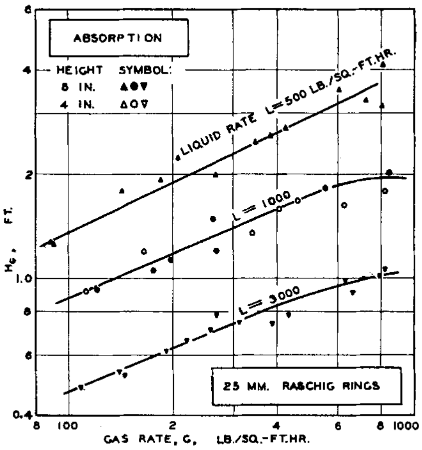

Fig. 5. Height of a transfer unit for absorption with 25-mm. Raschig rings.

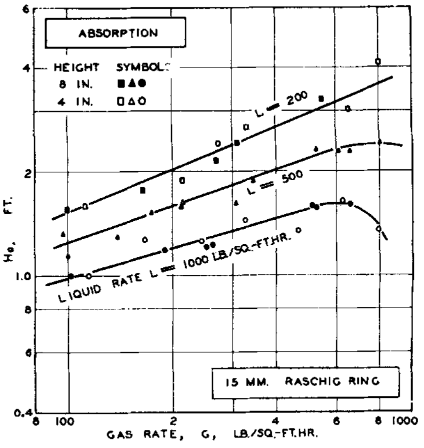

Fig. 6. Height of a transfer unit for absorption with 15-mm. Raschig rings.

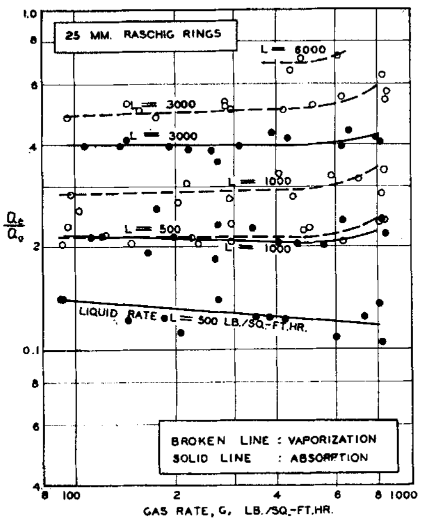

Fig. 7. Effective areas for 25-mm. Raschig rings.

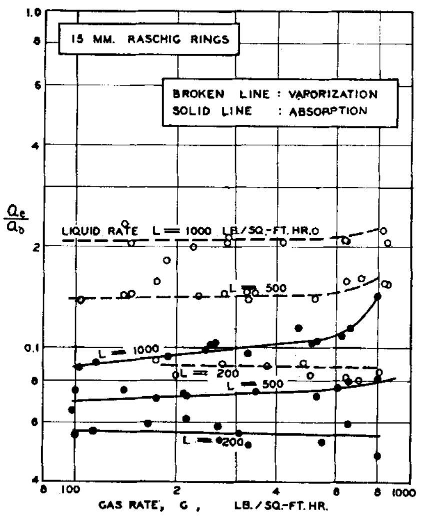

Fig. 8. Effective areas for 15-mm. Raschig rings.

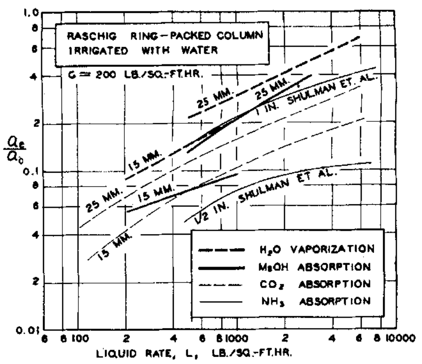

Fig. 9. Comparison of effective areas.

which they determined by the aforementioned procedure, in separating k_L from the $k_L a$ data of Sherwood and Holloway, obtaining the following dimensionless equation for k_L:

$$\frac{k_L d_p'}{D_L} = 25.1 \left(\frac{d_p' L}{\mu_L}\right)^{0.45} \left(\frac{\mu_L}{\rho_L D_L}\right)^{0.5} \tag{6}$$

In a previous study (*30*) the present authors determined the effective interfacial area for liquid phase mass transfer by comparing the rates of absorption of carbon dioxide by water and by methanol in a packed column with similar data with a bead column, that is a column of spheres connected in a vertical row. The effective interfacial area by Shulman is based on the gas phase mass transfer, whereas the present authors used the data on liquid phase mass transfer in evaluating the effective interfacial areas. As seen from Figure 9 the effective interfacial areas determined by two entirely different methods show approximate agreement. This may indicate soundness of both approaches.

It should be noted that the choice of the basis on which the values of k_G or k_L and accordingly the effective interfacial area in a packed column are determined is arbitrary, provided that the values of k_G or k_L in the reference apparatus, such as an unirrigated packed bed or a bead column, vary with gas or liquid rates just the same way as in the packed column. Furthermore it is possible to adopt different effective areas for the gas phase and liquid phase mass transfer. However it is more convenient to use the same effective area for the mass transfer in both phases. Thus in the present study the liquid phase mass transfer data of Sherwood and Holloway (*20*) and other sets of data were analyzed by use of the effective interfacial area on the basis of the Taecker-Hougen correlation for the gas phase mass transfer in all-wet packings and not on the basis of the bead column. Since the present data on methanol vapor absorption do not cover the necessary ranges of packing sizes and liquid rates, the Fellinger data (*8*) on ammonia absorption along with the Taecker-Hougen correlation were used for the determination of the effective interfacial area. The results are shown in Figure 10, in which the effective areas obtained from the present data on methanol absorption and the Taecker-Hougen correlation are also shown. On the basis of the Fellinger-Taecker-Hougen curves in Figure 10 and the liquid phase mass transfer data of Sherwood and Holloway, of Hikita et al. (*11*), and of the present authors (*30*) the following equation for the liquid phase height of a transfer unit was obtained:

$$\frac{H_L}{d_p} = 1.9 \left(\frac{L}{a_e \mu_L}\right)^{1/2} \left(\frac{\mu_L}{\rho_L D_L}\right)^{1/2} \left(\frac{d_p^3 g \rho_L^2}{\mu_L^2}\right)^{-1/6} \tag{7}$$

where the last term in the parentheses is the Galilei number. This type of correlation can be derived by dimensional analysis and was also proposed by Hikita, but he used the effective interfacial area based on the liquid phase mass transfer on a piece of Raschig ring. Davidson (*4*) derived theoretically an equation similar to Equation (7), but his exponent on the Reynolds number is 2/3. Figure 11 shows the comparison of the three correlations with the data points of four investigations on liquid phase mass transfer rates, that is data by Sherwood and Holloway (*20*), Jones (*15*), Hikita et al. (*11*), and the present authors (*30*). It is natural that the line representing Hikita's correlation is off the data points, because his effective area is different from the one defined here. The upper line of Davidson represents the case where the interface was assumed vertical and the lower line the case where the interface is inclined. Equation (6) is simpler than Equation (7) and gives a practically satisfactory correlation in most cases. Which of these equations has wider applicability could be known if data with wider variation in liquid viscosity were available, since the two equations give somewhat different net effect of liquid viscosity.

In the present correlation the effect of gas velocity on the effective interfacial area was assumed negligible, which might not be correct at low or high liquid rates, and especially near loading points as indicated by the data of Shulman (*22*). However in the absence of more extensive data effective interfacial areas given here may be used along with Equations (4) and (7) in most cases exclusive of extreme conditions. It is desirable to take an ample overdesign factor in designing a commercial column, because good liquid distribution as in laboratory columns cannot be expected in larger columns.

Part II. Distillation

Since the pioneering work of Furnas and Taylor (*9*) several investigations have been reported on the mass transfer in packed distillation columns. Some of the papers claim that liquid-phase resistance was controlling, some state that vapor-phase resistance was predominating, and some conclude that resistances of both phases should not be neglected. The only work in which resistances of individual phases were determined is that of Edye (6). However the column used in his study was too small, 25-mm. in diameter and 60 mm. in packed height, to give any useful data, and the effects of properties of the system were not studied.

More difficulties are involved in experimental studies on packed distillation columns than in studies on absorption or vaporization columns. Very few reliable data are available on physical properties such as diffusivity and viscosity of liquid mixtures, especially at their boiling points. Careful examination of previous published data revealed that pinching at one end of the operating line is not uncommon, especially when the packed section is high. In such cases a small error in analysis would result in a serious error in values of mass transfer coefficients or in the height of a transfer unit.

In the present work no attempt was made to split the over-all mass transfer resistance into individual phase resistances, but rather it was planned to compare observed values of the over-all resistance with the values predicted from the correlations for individual resistances proposed in the first part of the present paper. Since the size of the column seems to be a factor, a column of the same diameter as in the vaporization and absorption studies was used.

Three binary systems, ethanol-water, trichloroethylene-toluene, and acetone-water, were chosen for various reasons. The first system was used because the slope of equilibrium curve varies widely, and physical properties of the liquid mixture are known as functions of composition. The second system was tested because it is a nearly ideal mixture, and its analysis is not difficult. The third system was employed because, at very low acetone concentrations, liquid-phase resistance to mass transfer is relatively large, because of the steep equilibrium curve, and physical properties are easily predictable.

EXPERIMENTAL

The apparatus shown in Figure 12 was an improved modification of the one used in a previous study (*29*). The column, made of copper, was 12 cm. in diameter and was packed with 15- or 25-mm. ceramic Raschig rings to a height of 50 cm. The column was covered with electrically heated air jackets to minimize heat losses. The vapor from the column was condensed in a total condenser at the top of the column, part of the condensate was withdrawn as distillate through a regulating valve, and the remainder was returned as reflux. No subcooling of reflux occurred, since it was not taken out of the column and was always in contact with the rising vapor. The auxiliary safety condenser was provided just for the case where controlling the rate of cooling water to the main condenser failed. The reflux withdrawn from the column bottom was returned to the electrically heated reboiler. Thus the column was operated as an enriching section either at partial or total reflux. The flow rates of the distillate and the reflux from the column were measured by means of orifices. Analyses were made of the liquid samples from the top and bottom of the column and the condensed vapor sample from the column bottom. The ethanol-water and trichloroethylene-toluene mixtures were analyzed by density measurement and the acetone-water mixture by an iodometric method. It was possible to determine the concentration of acetone in water as low as 10^{-8} mole fraction.

PHYSICAL PROPERTIES

For the ethanol-water system the vapor-liquid equilibrium data of Carey and Lewis (*2*) were used. Since the equilibrium data of Schoenborn et al. (*19*) for the trichloroethylene-toluene system do not agree with those of Stage (*23*), data for this system were taken by use of an equilibrium still similar to that of Hipkin and Myers (*12*). The data were thermodynamically checked by the method of Ibl and Dodge (*14*), showing approximate agreement with the data of Schoenborn. Table 5 gives the smoothed values of the equilibrium compositions and temperatures for the trichloroethylene-toluene system. Equilibrium data for the acetone-water system were also obtained by use of the same apparatus. The data can be expressed by the equation

$$y = 28.0\,x \qquad (8)$$

over the range of x from 0 to 0.015, within which the present experiment was performed. The data agree with the calculation by the Van Laar equation using the values of constants given by Perry (*17*).

Diffusivity and viscosity of the ethanol-water mixture at boiling points vary greatly with composition, but the data are available (*24*). Diffusivity of the liquid mixture of trichloroethylene-toluene was estimated by means of the Wilke correlation (*27*) and by assuming linear relationship between diffusivity and mole fraction, which seems justifiable for a nearly ideal solution (*1*). Diffusivity of the liquid mixture of acetone and water was estimated by the Wilke correlation, and viscosity was taken equal to that of water, since the acetone concentration in water was extremely low.

Diffusivities in the vapor phase were calculated by the Hirschfelder-Bird-Spotz correlation (*13*). Viscosities of vapor mixtures were estimated by the Wilke equation (*28*), and Lima's equation (*16*) was used for the estimation of the viscosities of the liquid mixtures, except the ethanol-water mixture for which data are available. Viscosity and density of the vapor mixture of acetone and water were taken equal to those of pure water vapor.

BASIS OF INTERPRETATION

The basic assumption in the present study is that the over-all resistance to mass transfer in distillation is the sum of the resistances of the vapor and liquid phases, there being no resistance at the interface. The fundamental difference in the mechanism between distillation and absorption or

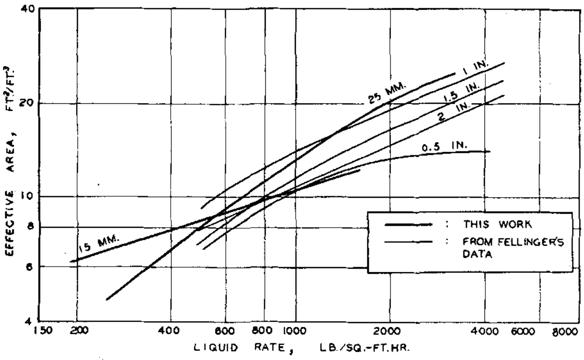

Fig. 10. Effective interfacial areas for absorption in Raschig ring. Packed column based on Taecker-Hougen correlation.

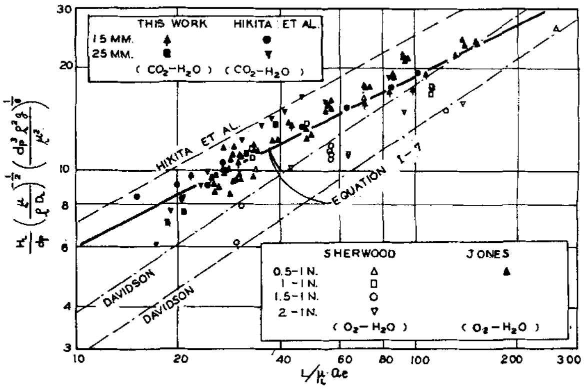

Fig. 11. Correlation for liquid phase mass transfer.

vaporization is that the former is equimolar counterdiffusion, while the latter is unidirectional diffusion. It is not likely that this difference alters the types of the correlations for the individual phase mass transfer coefficients or the values of constants and the exponents on dimensionless groups in the correlations. However it is conceivable that the effective interfacial area in a distillation column is different from that in an absorption or vaporization column irrigated by water or an aqueous solution owing to the difference in surface tension between water and organic liquids.

The well-known relationships between the over-all and the individual phase height of a transfer unit are

$$H_{OG} = H_G + (mG_M/L_M)\, H_L \quad (9)$$

and

$$H_{OL} = (L_M/mG_M)\, H_G + H_L \quad (10)$$

in which the average slope of equilibrium curve m is defined by Furnas (9) as

$$m = \frac{(N_t)_{OL}\, L_M}{(N_t)_{OG}\, G_M} \quad (11)$$

This definition of m is most convenient and seems sound, since it is derived directly from Equations (9) and (10).

If it is assumed that Equations (4) and (7) are valid for the mass transfer in the vapor and liquid phases, respectively, with proper values of the effective interfacial area a_e, then from Equations (4), (5), (7), and (9)

$$H_{OG} = (H_G)_o\, a_o/a_e + (m\, G_M/L_M)\, H_L$$

$$= 0.935 \frac{1}{a_e} \left(\frac{G\sqrt{A_p}}{\mu_G}\right)^{0.41} \left(\frac{\mu_G}{\rho_G\, D_G}\right)^{2/3}$$

$$+ m\,(G_M/L_M)\, 1.9\, d_p \left(\frac{L}{a_e\, \mu_L}\right)^{1/2}$$

$$\left(\frac{{d_p}^3\, g\, {\rho_L}^2}{{\mu_L}^2}\right)^{-1/6} \quad (12)$$

In the above equation all the quantities on the right-hand side except a_e are known or can be estimated for a given system at a given set of operating conditions. Therefore if the effective interfacial area a_e is known, the values of H_{OG} are calculable. Conversely, if the values of H_{OG} and all the quantities except a_e are known, it it possible to calculate the values of a_e.

DISCUSSION OF RESULTS

Experimental data on the three systems, trichloroethylene-toluene, ethanol-water, and acetone-water, are listed in Tables 6 to 11.* Data for those runs, for which the compositions of vapor at the bottom of the column calculated from the material balance differed more than 3% from the analyzed values, were considered inaccurate and were discarded. The number of such runs was about one tenth of the total number of runs. In calculating H_{OG} values the end effects were neglected, since they were known to be equivalent to a packed height of 2 to 5 cm. in this type of distillation column (29).

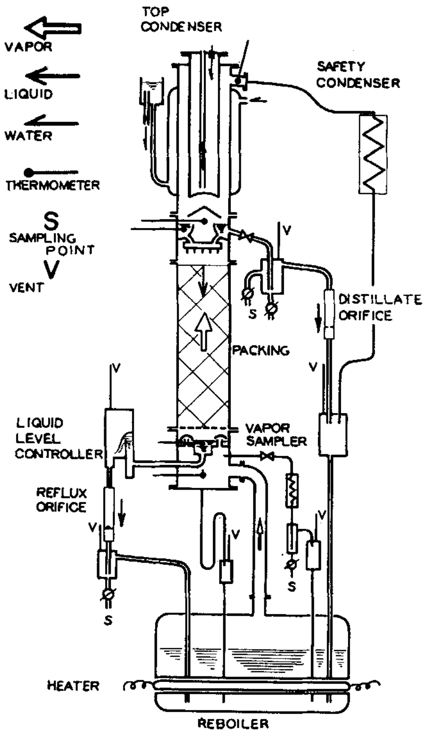

Fig. 12. Apparatus for distillation experiment.

TABLE 5. LIQUID-VAPOR EQUILIBRIUM DATA FOR TRICHLOROETHYLENE-TOLUENE MIXTURE

Composition, mole % trichloroethylene Liquid	Vapor	Temp., °C.
2.0	4.0	110.0
4.0	7.3	109.4
6.0	10.2	108.9
8.0	13.8	108.3
12.0	19.9	107.2
18.0	28.8	105.5
24.0	36.7	103.9
30.0	44.3	102.4
36.0	51.5	100.8
42.0	58.0	99.4
50.0	66.0	97.5
58.0	73.2	95.6
64.0	78.0	94.3
70.0	82.6	93.0
76.0	86.6	91.7
82.0	90.5	90.5
88.0	94.1	89.3
92.0	96.4	88.6
94.0	97.4	88.2
96.0	98.4	87.9
98.0	99.2	87.5

Since physical properties of vapor and liquid mixtures such as diffusivity and viscosity vary with composition, they should vary from one run to next for a given system, and even for a given run they should differ depending on the height in a packed bed. The values of the Reynolds, Schmidt, and Galilei numbers were calculated for each run, by use of the arithmetic mean values of the physical properties at the top and bottom of the packed bed.

With reference to Equation (12) the values of H_{OG} were directly obtained from experimental data, and all the quantities on the right-hand side of the equation except a_e were known or estimated by the aforementioned methods. Thus by solving a quadratic equation the value of a_e was calculated for each of the trichloroethylene-toluene runs. In Figure 13 the effective interfacial area a_e thus calculated from the data on the toluene-trichloroethylene runs are plotted on logarithmic coordinates against the superficial volumetric liquid velocity per unit cross section of the column. It seems that the effective interfacial area is better correlated by use of the volumetric velocity than by use of the mass velocity when liquids of different densities are involved. In case only one kind of liquid is used, it does not make much difference which kind of velocity is adopted in the correlation. In Figure 13 the data points are correlated by two separate curves, one for 15- and the other for 25-mm. Raschig rings, irrespective of vapor rates. The upward trend of the curves at high liquid rates is probably due to loading, since in this experiment at total reflux high liquid rates were accompanied by high vapor rates.

Comparison of the effective areas for absorption and vaporization and the wetted area is shown in Figure 14 for the 25-mm. Raschig ring. The wetted area was calculated by the following equation of Hikita (*10*):

$$\frac{a_w}{a_o} = 0.079 \left[L \left(\frac{\sigma}{20}\right)^{3q}\right]^{1/3} \quad (13)$$

in which

$$q = -0.74\, {d_p}^{-0.70}$$

In Figure 14 the solid lines represent wetted and effective areas for water as the irrigating liquid, whereas the broken lines show the effective area for the distillation of the trichloroethylene-toluene mixture and the wetted area for a liquid having surface tension of 20 dynes/cm., which is a rough average value for most organic liquids at their boiling points. This figure reveals that the effective area

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* See footnote on page 310.

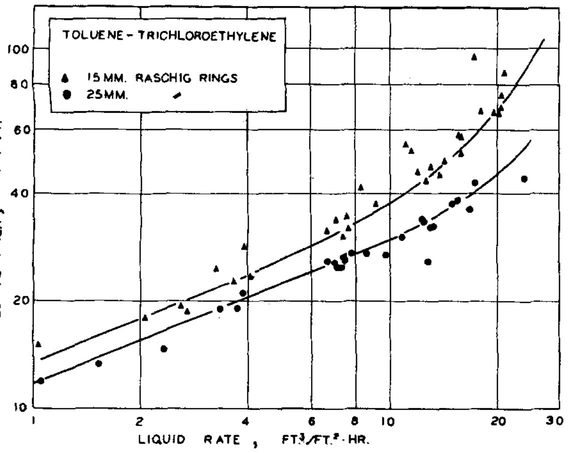

Fig. 13. Effective area for distillation.

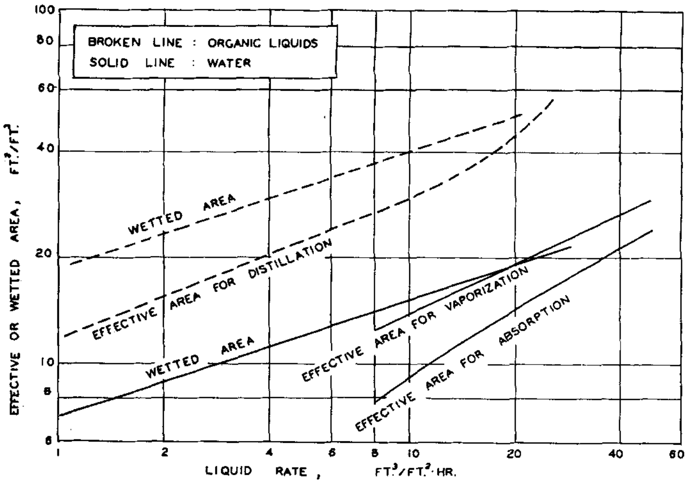

Fig. 14. Comparison of effective and wetted areas, 25-mm. Raschig rings.

for distillation is considerably larger than that for vaporization and absorption with water as the irrigating liquid. However it is substantially smaller than the wetted area for the distillation mixture. Thus the effective area for distillation seems to be roughly the same as that for absorption, if the effect of surface tension on the effective area is assumed to be similar to that on the wetted area. This conclusion hinges on the reliability of Equation (13), which is somewhat doubtful in the present case, because the equation was extrapolated down to a surface tension of 20 dynes/cm. The range of surface tension Hikita studied was 30 to 65 dynes/cm. Shulman (22) proposed that for liquids other than water the effective area for absorption be calculated as proportional to the operating holdup, which in turn was given as a function of the rate, surface tension, density, and viscosity of the liquid. Unfortunately his correlation is not usable here, because the liquid rate in the present work was below the range of liquid rate he studied. It is noted that, according to Shulman, the lower the liquid rate the greater the effect of surface tension on the operating holdup.

Observed height of transfer unit values for packed distillation columns are in general considerably lower than those for absorption columns irrigated by water or aqueous solutions. This could be attributable to larger effective area due to lower surface tension of organic liquids usually encountered in distillation.

To test the general applicability of Equation (12) the values of H_{OG} observed on systems other than trichloroethylene-toluene were compared with the values calculated by means of Equation (12) and Figure 13. The data used were those on the ethanol-water and acetone-water runs in the present work as well as the data on benzene-toluene systems which were published previously (*29*). The use of Figure 13 may be justified, since the surface tensions of these systems, or organic systems in general, are approximately equal to those of the trichloroethylene-toluene mixture, except for the dilute ethanol and acetone ranges of the ethanol-water and acetone-water mixtures. In Figure 15 the calculated and observed values of H_{OG} for the three systems are shown to be in rough agreement. The very large values of H_{OG} for the acetone-water system are evidently due to the very large values of m in the dilute acetone range within which the present data are limited. The fact that the observed values of H_{OG} for the acetone-water mixture are generally higher than the calculated values may be attributable to higher surface tension of the mixture at low concentration of acetone.

Difficulties are encountered in an attempt to test the applicability of the present correlations to the data on packed distillation columns in the literature. As stated before, few published data, especially those at partial reflux, seem reliable. Some of the partial reflux data are based on vapor compositions at the column bottom obtained by unreliable material balance calculations. Physical properties such as liquid diffusivity can hardly be estimated for most nonideal mixtures. Column size and liquid distributor design, which seem to be important factors, used in some previous works are not comparable with those adopted in the present work. Thus only the total reflux data of Duncan et al. (*5*) on the trichloroethylene-toluene and carbon tetrachloride-benzene mixtures and of Schoenborn et al. (*19*) on the trichloroethylene-toluene mixture both obtained with ½-in. Raschig rings were compared with the H_{OG} values predicted by the present correlations. The results showed rough agreement.

These calculations indicated that in the general case where the value of mG_M/L_M is in the range of 0.3 to 3, the mass transfer resistance of vapor phase is predominating in distillation, for instance, about one fifth to one sixth of the total resistance being the resistance of liquid phase. This comes from the fact that H_L in a packed distillation column is considerably smaller than H_G in most cases because of higher temperatures usually encountered in distillation. The exceptionally high values of H_{OG} obtained for the acetone-water mixture in the present work are due to the purposely selected very large values of mG_M/L_M, around 28, in the dilute acetone range. In such special cases the liquid-phase resistance becomes significant and H_{OL} becomes very small as seen from Equation (10).

There are contradictory data and opinions (*3, 7, 11, 18*) on the effects of the column diameter and the packed height on the height of a transfer unit. This practically important problem deserves further investigation. In the present correlations for H_L and H_G the effects of packed height and column diameter were not studied.

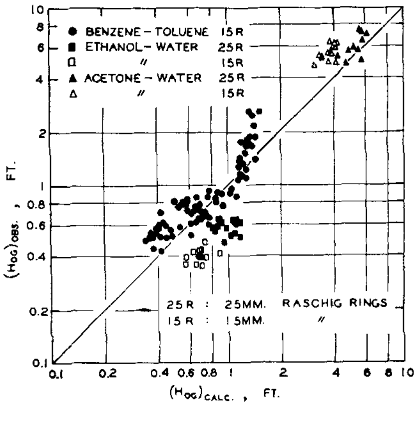

Fig. 15. Calculated and observed values of H_{OG} in packed distillation columns.

CONCLUSIONS

The correlations proposed in the present work for the individual phase height of a transfer unit along with the effective interfacial areas for absorption, vaporization, and distillation in packed columns might be imperfect in that the effects of packed height and column diameter, if any, were not studied. However the present correlations may indicate a rational approach in interpreting the effects of physical properties of systems and of operating conditions. The correlations here proposed are based on the data taken with carefully designed experimental columns. The values of height of a transfer unit for commercial columns might be considerably higher than those for experimental columns owing to inevitably poor liquid distribution in larger columns. It is hoped that the effective interfacial areas for various kinds of operations be determined with larger columns and with larger packings. It is also necessary to extend one's knowledge on diffusivity in nonideal liquid mixtures in order to have a sound basis for the design of distillation columns.

NOTATION

A_p = surface area of a piece of packing, sq. ft.

a, a_e = effective interfacial area of packing, sq. ft./cu. ft.

a_o = total surface area of packing, sq. ft./cu. ft.

a_w = wetted area of packing, sq. ft./cu. ft.

D = diffusivity, sq. ft./hr.

d_p = size of packing piece, ft., in Equation (13) in.

d_p' = diameter of sphere possessing the same area as a packing piece, ft.

G = superficial mass velocity of gas, lb./(hr.) (sq. ft.)

G_M = superficial molal mass velocity of gas, lb.-moles/(hr.) (sq. ft.)

g = gravitational constant, ft./(hr.)^2

H_{OG},H_{OL} = height of an over-all transfer unit based on gas phase and liquid phase, respectively, ft.

H_G,H_L = height of a transfer unit for gas phase and liquid phase, respectively, ft.

$(H_G)_o$ = height of a gas phase transfer unit for all-wet packings, ft.

k_G = gas phase mass transfer coefficient, lb.-moles/(hr.) (sq. ft.) (atm.)

k_L = liquid phase mass transfer coefficient, ft./hr.

L = superficial mass velocity of liquid, lb./(hr.) (sq. ft.)

L_M = superficial molal mass velocity of liquid, lb.-moles/(hr.) (sq. ft.)

m = average slope of equilibrium curve defined by Equation (11), dimensionless

N_t,N_t' = number of gas phase transfer units excluding and including end effects, respectively, dimensionless; suffixes 4 and 8 denote packed heights in in.

$(N_t)_e$ = number of transfer units due to end effects, dimensionless

$(N_t)_{OG},(N_t)_{OL}$ = number of over-all transfer units based on gas phase and liquid phase, respectively, dimensionless

p_{BM} = logarithmic mean partial pressure of inert gas in the gas film, atm.

x = vapor-phase composition, mole fraction

y = liquid-phase composition, mole fraction

μ = viscosity, lb./(ft.) (hr.)

ρ = density, lb./cu. ft.

σ = surface tension, dynes/cm.

Subscripts

G and L = gas and liquid phases, respectively

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