Performance of Packed Columns: VI. Mass Transfer Rates for Dehumidification at High Solute Concentrations

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Air-carbon tetrachloride gas streams were dehumidified with cold liquid carbon tetrachloride in a short 4.0-in. diameter column packed with 0.5-in. Raschig rings in an attempt to explain reported unsuccessful attempts to correlate dehumidification data obtained at high solute concentration.

The data were in satisfactory agreement with a previously presented correlation if the Schmidt number was evaluated at average film conditions and interfacial conditions were employed with rigorous calculation methods to evaluate the mass transfer coefficients.

The previous paper (*10*) of this series presented a correlation for mass transfer in packed columns at low and high solute concentrations which was based on adiabatic vaporization data.

$$\left[\frac{k_G M_M P_T}{G}\right]\left[\frac{P_{BM}}{P_T}\right]^{2/3}\left[\frac{\mu}{\rho D_V}\right]^{2/3} = 1.195\left[\frac{D_P G}{\mu(1-\epsilon)}\right]^{-0.36} \quad (1)$$

Cairns and Roper (*1*) obtained a similar correlation for adiabatic vaporization data employing a wetted-wall column:

$$\left[\frac{k_G M_M P_T}{G}\right]\left[\frac{P_{BM}}{P_T}\right]^{0.83}\left[\frac{\mu}{\rho D_V}\right]^{2/3} = 0.025\left[\frac{DG}{\mu}\right]^{-0.2} \quad (2)$$

In a later attempt to test the usefulness of Equation (2) Cairns and Roper (*2*) obtained mass transfer data at high humidities for dehumidification runs in a wetted-wall column. These data however could not be correlated by Equation (2) or any other satisfactory method.

Yoshida and Tanaka (*11*) on the other hand, working with columns packed with Raschig rings, were able to obtain satisfactory correlations of both vaporization and dehumidification data obtained at low humidities. There are several possible explanations for the different conclusions reached by Cairns and Roper (*2*) and Yoshida and Tanaka (*11*). The first is that correlations obtained for low solute concentrations do not apply at high concentrations for dehumidification. The second is that packed-column data lead to correlations which are not similar in form to those obtained for wetted-wall columns. The third, and most likely explanation, is that Cairns and Roper (*2*) used an oversimplified method of calculation to evaluate their data. They assumed the bulk liquid temperature was equal to the interfacial temperature and employed a logarithmic mean driving force to calculate the mass transfer coefficients. These simplifications are suitable for adiabatic vaporization work when the liquid temperature is held constant at the adiabatic saturation temperature but are not justified for dehumidification work where heat transfer takes place in the liquid phase and the interfacial temperature may be substantially different from the bulk liquid temperature. Yoshida and Tanaka (*11*) working at low humidities were able to estimate interfacial temperatures and employ a rigorous trial-and-error, graphical method of calculation to evaluate the mass transfer coefficients.

In order to test the explanations offered above and the extension of the use of Equation (1) to dehumidification work an experimental investigation was undertaken to obtain suitable dehumidification data. A short 0.5-in. Raschig ring packed column was employed to dehumidify air-carbon tetrachloride gas streams at low and high solute concentrations with cold liquid carbon tetrachloride. This system has a number of advantages over a wetted-wall column employing the air-water system. A short packed column has fluid-flow characteristics representative of all irrigated packed columns and has been shown (*9, 10*) to yield correlations in which the effects of variables such as those contained in the Schmidt and Reynolds numbers are the same regardless of column height and diameter provided the column diameter to nominal packing-size ratio is greater than 8.0.

When the air-carbon tetrachloride system is used, the physical properties of the gas stream are found to vary considerably with concentration which is not the case for the air-water system. For example the Schmidt number varies more than fivefold for air-carbon tetrachloride in the 0 to 0.70 solute mole fraction range, whereas it can be assumed constant for the air-water sys-

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tem over the same concentration range. For this reason correlations developed with the air-carbon tetrachloride system are more likely to be of a sufficiently generalized nature to permit extension of their use to other systems. In addition, when physical properties vary rapidly with concentration, there is an opportunity to determine whether dimensionless groups evaluated at film or bulk conditions provide better correlations.

Short packed columns have a shortcoming however in that excellent gas and liquid distribution as well as even small end effects combine to yield much higher mass transfer coefficients than can be expected from the same packings in commercial installations. This factor limits the use of a short packed column to determining the form of a correlation and the effects of the variables. The actual prediction of mass transfer coefficients for design purposes should be based on methods which have been developed from data obtained with columns representative of those used in commercial installations. Such methods have been discussed in previous papers of this series (*6, 7, 8*).

APPARATUS

A schematic diagram of the apparatus employed in this work is shown in Figure 1.

The packed column used consisted of a 4.0-in. I. D. pyrex tube packed to a depth of 1.5 in. with 0.5-in. diameter porcelain Raschig rings. The outside of the column was heated with nichrome heating wire and insulated to avoid condensation on the walls and maintain essentially adiabatic operation. Air flow to the column, provided by a blower, was metered by a calibrated set of rotameters. Coarse adjustments of air flow were made with a variable speed drive on the blower, and valves located below the rotameters were used for fine adjustments.

The air supplied by the blower was heated to approximately 700°F. by a 4.5-kw. heater with a thermostatic temperature controller. Carbon-tetrachloride vapor was supplied by a boiler heated with a 2-kw. immersion heater with power input regulated by a variable transformer. The carbon-tetrachloride vapor was superheated by passing through a length of pipe heated by two 400-w. flexible strip heaters. The hot carbon tetrachloride vapors were then mixed with the hot air from the blower and passed through a heated, insulated pipe into the bottom of the column. The inlet gas temperature was measured just below the column by an iron-constantan thermocouple. The hot gases then passed through a gas distributor consisting of a hollow cross of brass bar stock ⅝ in. wide and ⅜ in. deep. Two grooves were cut along the underside of each arm of the cross, and six ⅛ holes were drilled along each groove from the end to the center of the cross, making a total of forty-eight discharge holes.

The gas passed upward through the packing, and its temperature above the packing was measured by a mercury thermometer placed directly in the gas stream above the liquid distributor. From the column the exit gases passed through two double pipe heat exchangers that removed by condensation most of the carbon tetrachloride remaining in the gas stream. The condensed carbon tetrachloride was returned to the boiler and the air vented to the atmosphere.

Liquid was supplied to the column by a 5-gal. steel constant head tank equipped with an overflow to assure constant liquid level. This tank was located approximately 5 ft. above the top of the column. The liquid flow was metered by a calibrated rotameter and passed into the top of the column. It was discharged directly above the packing to minimize end effects. The liquid distributor consisted of ¼-in. copper tubing forming a continuous coil of two turns. Approximately fifty holes 1/32 in. in diameter were drilled along the underside of the distributor.

The temperature of the inlet liquid was measured by a glass enclosed iron-constantan thermocouple that extended directly into the liquid distributor through the liquid inlet line. A portion of the liquid leaving the packing was collected in a shallow trough and its temperature measured by an iron-constantan thermocouple always immersed in fresh liquid from the packing in this trough. The exit liquid passed into a 5-gal. steel receiving tank located below the column. A gear pump was used to circulate the liquid. A glass wool packed filter was placed in the line between the pump and the constant head tank. Also a heat exchanger consisting of 50 ft. of coiled 0.375-in. copper tubing in a galvanized iron jacket was placed in this line to cool the liquid to the proper inlet temperature.

Inlet gas samples were taken from the inlet gas line ½ in. below the distributor. Exit-gas samples were taken through a sampling tube located just above the liquid distributor. The samples were drawn through 0.125-in. copper tubing by a slight constant vacuum, and analyses were made with a Consolidated Electrodynamics Corporation Chromatographic Analyzer, type 26-201. The analysis was made for air, and the carbon tetrachloride concentration was determined by difference. It was found that air peaks were of constant base width, so the air concentration was determined by comparing the peak height for a given sample with the peak height for pure air. The carrier gas used was helium, and a constant flow rate was used that gave sharp air peaks reproducible to within 0.2%.

PROCEDURE

The chromatographic analyzer required approximately 8 hr. to reach equilibrium, and it was turned on and set the night before runs were to be made.

The air flow was turned on first and the air heater set at 700°F. Power was supplied to the vapor line heating coils to bring the piping and insulation up to operating temperature. Then the boiler and carbon tetrachloride vapor line strip heaters were turned on, along with the column heating coil and sample line heaters. As soon as the inlet gas came up to fairly constant temperature, the liquid flow was started. A very high liquid rate was maintained for approximately ½ hr.

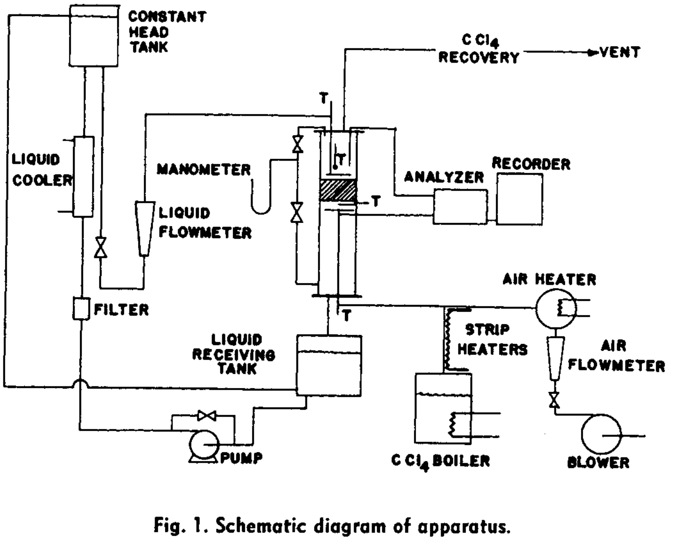

Fig. 1. Schematic diagram of apparatus.

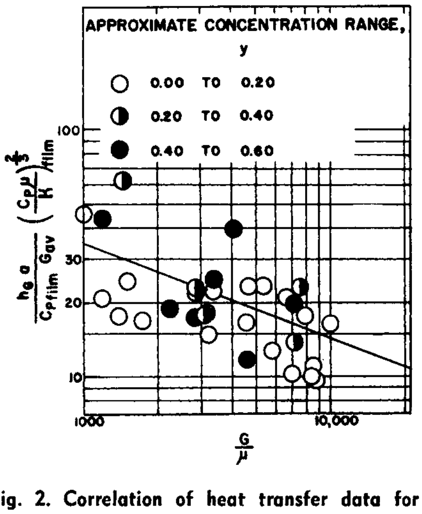

Fig. 2. Correlation of heat transfer data for vaporization runs.

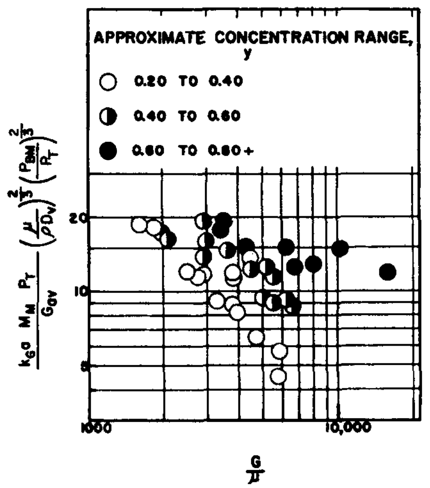

Fig. 3. Dehumidification data evaluated with bulk liquid conditions and logarithmic mean driving forces.

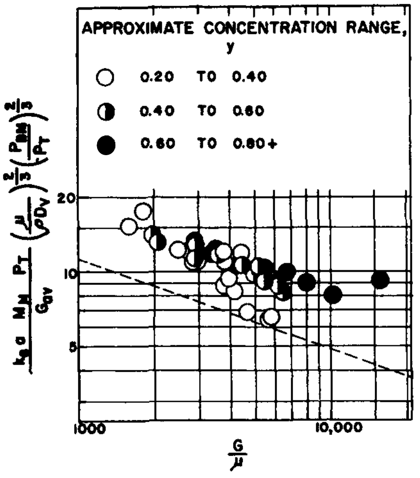

Fig. 4. Dehumidification data evaluated with interfacial conditions and the rigorous method of calculation.

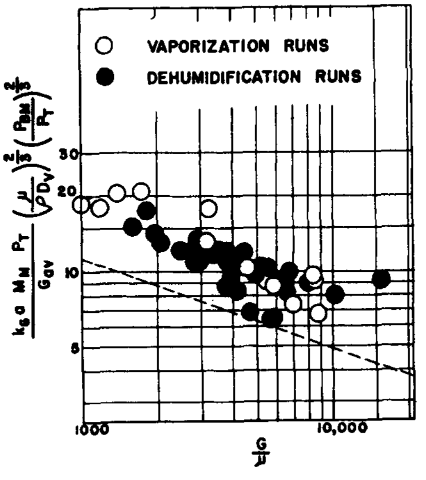

Fig. 5. Comparison of vaporization and dehumidification data obtained with the same packed column.

to thoroughly wet the packing, after which the desired flow rate was set. While the equipment was coming to equilibrium, a series of pure air peaks was taken on the chromatograph to be compared with sample peak heights.

Equilibrium conditions were determined by recording inlet and outlet gas and liquid temperatures. When all four temperatures became constant, equilibrium was attained. This condition required approximately 4 hr. from initial start-up to be fulfilled. After equilibrium had been reached, an outlet gas sample was taken for analysis. The analyzer required about 7 min. per sample, and readings of temperature, air flow, and column pressures were taken while the analysis was going on. Immediately after the outlet gas sample was taken, the inlet gas-sample line was connected to the analyzer, allowing the inlet gas about 7 min. to flush out the sample valve. The inlet gas composition remained constant for long periods and was analyzed as soon as the outlet gas analysis was complete.

When both gas samples had been analyzed, the boiler setting and air rate were changed to the next desired conditions and the column allowed to reach equilibrium once more. An average of one hour was required for the equipment to return to equilibrium under new conditions, at which time the above procedure was repeated. Pure air samples were taken before each run to assure accurate gas analysis.

In addition to the dehumidification runs a series of vaporization runs was made for comparison purposes. The procedure employed for these runs was described previously (*10*).

METHODS OF CALCULATION

Two methods of calculation were employed to evaluate the volumetric mass transfer coefficients k_Ga from the experimental data. The first is a simplified method which was employed for both the vaporization and dehumidification data, although its use can be justified only for the former. It assumes that the interfacial temperature is equal to the bulk liquid temperature for determining the driving forces, and a logarithmic mean driving force based on terminal conditions is employed as follows:

$$k_Ga = \frac{(N_Aa)}{P_T(\Delta y)_{lm}} \tag{3}$$

where

$$(\Delta y)_{lm} = \frac{(y_i - y)_1 - (y_i - y)_2}{\ln \dfrac{(y_i - y)_1}{(y_i - y)_2}} \tag{4}$$

and N_Aa is the pound moles of carbon tetrachloride transferred per hour per cubic foot of packing as calculated from the measured air rate and the analyses of the inlet and outlet gas streams.

The second method of calculation represents an attempt to evaluate k_Ga for the dehumidification runs by a rigorous procedure. It involves estimating the actual interfacial temperatures at several points in the column for each run. The true driving forces are then employed in a graphical integration procedure to evaluate k_Ga. An outline of the method follows; a complete calculation is available elsewhere (*4*).

The interfacial temperature can be determined by a trial-and-error procedure for any point in the column from the following transfer equation:

$$k_Ga\,P_T(y - y_i)\lambda_i + h_Ga(T_G - T_i) = h_La(T_i - T_L) \tag{5}$$

To use this equation it is necessary to know the appropriate values of the transfer coefficients k_Ga, h_Ga, and h_La. The mass transfer coefficient k_Ga was taken from the correlation for vaporization data obtained in a similar column by Shulman and Delaney (*10*). The gas-phase heat transfer coefficient h_Ga was estimated from the line on Figure 2 which combines the vaporization data of this work with that of Shulman and Delaney (*10*). The liquid-phase heat transfer coefficient h_La for the liquid rate employed was estimated to be 13,900 B.t.u./(hr.) (cu. ft.) (°F.) from the data of Yoshida and Tanaka (*11*) corrected for differences in interfacial areas by taking the ratio of h_Ga values and for differences in liquid physical properties by taking the ratio of heat transfer coefficients in tubes under similar conditions.

To evaluate a gas composition and temperature corresponding to an intermediate bulk liquid temperature the bulk gas and liquid temperatures are taken as varying linearly with gas-phase enthalpy. Since the gas-phase enthalpy varies rapidly with composition and relatively slowly with gas temperature, it can be used to calculate a gas-phase composition corresponding to a bulk liquid temperature at an intermediate point in the column.

The calculated values are used to plot an operating line y vs. T_i and an equilibrium line y_i vs. T_i on a y–T_i diagram. Vertical differences between the two lines give the true gas-phase driving force $y - y_i$ which is employed with the following rigorous equation developed from an equation given by Sherwood and Pigford (*5*):

$$\frac{k_Ga\ P_TZ}{G'} = \int_{y_2}^{y_1} \frac{dy}{(1-y)^2(y-y_i)} \tag{6}$$

The integral is evaluated graphically for each run.

Viscosities and diffusivities were calculated by the methods outlined by Hirschfelder, Curtis, and Bird (*3*). The reported gas rates are an average of the inlet and outlet rates. Molecular weights and physical properties are determined from an average of inlet and outlet compositions and temperatures except as noted. The Schmidt number, which varies rapidly with changing composition but is almost independent of temperature changes, is always evaluated at film conditions, that is an average of inlet and outlet bulk and interfacial conditions. The reported values of P_{BM} are the arithmetic average of the P_{BM} at the inlet and outlet ends of the column.

The experimental results are presented as plots of $[k_Ga\, M_M\, P_T/G]\ [\mu/\rho\, D_V]^{2/3}\ [P_{BM}/P_T]^{2/3}$ vs. G/μ in place of the terms shown in Equation (1). Shulman and Delaney (*10*) showed that data could be correlated equally well by either method and conclusions drawn from one applied to the other.

EXPERIMENTAL RESULTS

Dehumidification data were taken with inlet concentrations as high as 0.843 mole fraction carbon tetrachloride. To detect effects at different concentration levels an attempt was made to classify the data approximately by concentration level in some of the figures. Figure 3 shows the first attempt at correlation with the dehumidification data evaluated by the use of bulk liquid temperatures and logarithmic mean driving forces. This method was found to correlate vaporization data by both Shulman and Delaney (*10*) and Cairns and Roper (*1*). This method is no more successful in this case than it was with the dehumidification data of Cairns and Roper (*2*). The data are seen to scatter, although data at the same concentration level fall together. A simple correction for concentration level does not lead to a successful correlation because the effect of gas rate is not the one expected from Equation (1).

The second method of correlation is shown in Figure 4 with the data evaluated by means of interfacial conditions and the rigorous method of calculation. The broken line is the line of Shulman and Delaney (*10*) for vaporization data in a similar column. The data have been brought together with almost all of the separation at different concentration levels eliminated. In addition the best line through the data would show the effect of gas rate predicted by Equation (1). To explain why the data did not fall on the line given by Shulman and Delaney (*10*) for vaporization data a series of vaporization runs was made in the same column used for the dehumidification runs. Figure 5 shows both sets of data. It can be seen that both vaporization and dehumidification data come together satisfactorily when the data are obtained in the same column. The fact that the earlier data of Shulman and Delaney (*10*) lie on a parallel line below that of this work can probably be explained by the differences introduced in repacking a short column which contains as little as 1.5 in. of packing. Differences in the packing arrangement or some slight modification of end effects could easily produce the observed differences.

Of the three alternative explanations offered to explain the Cairns and Roper (2) results the present data indicate the use of incorrect interfacial temperatures, and the use of the logarithmic mean driving force accounts for the inability to obtain a satisfactory correlation.

SUMMARY AND CONCLUSIONS

Vaporization and dehumidification data from this and previous work at low and high solute concentrations indicate mass transfer coefficients for packings can be correlated by

$$\left[\frac{k_G\, M_M\, P_T}{G}\right]\left[\frac{P_{BM}}{P_T}\right]^{2/3}\left[\frac{\mu}{\rho D_V}\right]^{2/3} = 1.195\left[\frac{D_P G}{\mu(1-\epsilon)}\right]^{-0.36} \qquad (1)$$

when the Schmidt number is evaluated at average film conditions and interfacial conditions are employed with rigorous calculations to evaluate the mass transfer coefficients.

The constant 1.195 in Equation (1) is based on the work of previous investigators who employed columns and systems for which end effects could be eliminated or evaluated rather than on the data of the present work.

The present work makes it possible to extend the use of Equation (1) and the effective interfacial areas reported previously (*6, 7, 8, 9*) to predict k_Ga for a wide variety of gas-liquid contacting operations when the physical properties of the system are known.

ACKNOWLEDGMENT

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NOTATION

a = effective interfacial area, sq. ft./cu. ft.
C_P = specific heat, B.t.u./(lb.) (°F.)
D = equivalent diameter or characteristic dimension, ft.
D_P = diameter of sphere possessing the same surface area as a piece of packing, ft.
D_V = diffusivity of solute in gas, sq.ft./hr.
G = superficial gas rate, lb./(hr.) (sq. ft.)
G' = superficial molar inert gas rate, lb. moles/(hr.) (sq.ft.)
h_G = gas-phase heat transfer coefficient, B.t.u./(hr.) (sq.ft.) (°F.)
h_L = liquid-phase heat transfer coefficient, B.t.u./(hr.) (sq.ft.) (°F.)
k = thermal conductivity, B.t.u./(hr.) (sq.ft.) (°F./ft.)
k_G = gas-phase mass transfer coefficient, lb. moles/(hr.) (sq. ft.) (atm.)
M_M = mean molecular weight of gas. lb./lb. mole
N_A = rate of mass transfer, lb. moles/(hr.) (sq.ft.)
P_{BM} = mean partial pressure of inert gas in the gas phase, atm.
P_T = total pressure, atm.
T = temperature, °F.
y = solute mole fraction
$(\Delta y)_{lm}$ = logarithmic mean driving force, mole fraction
Z = height of packing, ft.
ϵ = void fraction, cu.ft./cu.ft.
λ = latent heat of vaporization, B.t.u./lb. mole
μ = gas viscosity, lb./(hr.) (ft.)
ρ = gas density, lb./cu.ft.

Subscripts

1 = bottom of packing
2 = top of packing
i = at the interface
G = gas phase
L = liquid phase

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