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Liquid Phase Mass Transfer with Concurrent Flow Through Packed Towers

Liquid phase mass transfer coefficients were measured for the absorption of carbon dioxide into water in concurrent flow. Liquid flow rates ranged up to 300 kg-mol/min. m² or about 60,000 lb./hr. ft.². In comparison with countercurrent flow, the coefficients were lower by as much as a factor of 3 with a significant gas rate effect. Correlations for concurrent coefficients with liquid and gas flow rates were obtained in the form $k_x a = CL^r G^s$ for 19.0-mm Berl saddles and 6.35-mm and 12.7-mm Raschig rings.

RAYMOND C. UFFORD
and
JOSEPH J. PERONA

Department of Chemical and Metallurgical Engineering
The University of Tennessee
Knoxville, Tennessee 37916

SCOPE

Recent interest and development in the field of gas-liquid reactions in packed beds stimulated this study. The literature contains much information about systems in which the gas and liquid streams flow countercurrently through the packed bed, but it contains relatively little about concurrent flow systems. The advantage of countercurrent operation in mass transfer is that it is more efficient in maintaining a relatively large concentration difference between the phases throughout the tower; however, countercurrent operation is limited by flooding, and greater pressure drops are required for given flow rates in countercurrent flow.

Correspondence concerning this paper should be addressed to J. J. Perona.

In gas-liquid reaction systems the absorbed gas is removed by the reaction, and the concentration of the absorbed gas is lowered from what it would be in purely physical absorption. The effect of the reaction is to maintain a larger concentration difference between phases and to lessen the advantage of countercurrent flow.

In accounting quantitatively for the effect of the reaction on the rate of absorption, it is convenient to express the rate as the product of the rate in physical absorption and an enhancement factor, which depends on the reaction kinetics and the physical mass transfer coefficient. Therefore, design calculations to find optimum conditions for packed tower gas-liquid reactors in concurrent flow require physical mass transfer coefficients in concurrent flow.

The literature contains a few references related to mass transfer in concurrent flow. The work of Dodds et al. (1960), Wen et al. (1963), McIlvirod as discussed by Wen et al. (1963), Reiss (1967), and Danckwerts and Gillham (1966) should be noted. While these studies have provided useful results for specific situations, they have not provided correlations or data which can be used to predict mass transfer coefficients in industrial packings over a wide range of flow rates.

The objective of this study was to produce mass transfer correlations for concurrent flow which would permit design studies of packed tower gas-liquid reactors to go forward. A secondary objective was to obtain a comparison of liquid phase coefficients for concurrent and countercurrent flow.

CONCLUSIONS AND SIGNIFICANCE

Liquid phase mass transfer coefficients were measured for gas rates ranging from 0.112 to 0.314 kg-mol/min. m^2 and for liquid rates up to 300 kg-mol/min. m^2. The maximum liquid rates which can be achieved in countercurrent operation for these gas rates are lower by factors of 2 to 8 for the various packings. The measured coefficients for each packing were well represented by the form $k_xa = CL^rG^s$ as shown in Figures 1 through 6. Best values for C, r and s are listed in Table 1.

In comparison with coefficients from the literature for countercurrent flow, concurrent coefficients differ in that they are affected significantly by the gas rate and are lower over most of the range of gas rates covered in this work.

EXPERIMENT

Carbon dioxide was absorbed from a carbon dioxide-air mixture into water. Carbon dioxide concentrations of 10 and 20% in air were used. The packed beds in which the absorption process was carried out were made from 10.16 cm I.D. Pyrex glass pipe. The column diameter is small compared with industrial packed beds but is typical of the size used in pilot plant process development and in experiments designed to test the theories of gas-liquid reactions. Three beds were used, 25.4 cm, 30.48 cm, and 86.4 cm in length. A closed top was designed and constructed to hold pressure, and the gas and liquid streams were fed into the top of the tower and flowed concurrently in downflow. The liquid was distributed through four holes 12.7 mm in diameter located symmetrically about the center of a stainless steel distribution plate. The gas tube released the gas below the plate and less than 6 mm above the packing.

Three types of ceramic packing were used—19.0 mm Berl saddles, 12.7 mm Raschig rings, and 6.35 mm Raschig rings. The largest amount of data was taken with the larger Raschig rings. Sherwood and Pigford (1952) suggest data be taken with apparatus of pilot plant or semiplant size to avoid abnormal wall effects and that the ratio of the nominal packing size to the tower diameter be at maximum 1:10 to 1:8. The Berl saddle packing in the column used in this investigation is just under the 1:8 limit having a ratio of 1:6. The effect of too large a packing is to cause liquid channeling to the column wall. This wall effect causes deviation from the ideal homogeneous model used to derive the mass transfer coefficient relationship. Both Porter and Templeman (1968) and Danckwerts and Gillham (1966) have examined the effect with height, and more recently Danckwerts (1970) gives a general suggestion that the height of the packed column should not exceed a few column diameters in length in order to avoid significantly adverse effects. All of the correlated data in this investigation was taken with column lengths under 3 column diameters. Further details of the experimental equipment and procedure are given in the Supplement.*

* The Supplement has been deposited as Document No. 02250 with the National Auxiliary Publications Service (NAPS), c/o Microfiche Publications, 305 E. 46 St., N.Y., N.Y. 10017 and may be obtained for $1.50 for microfiche or $5.00 for photocopies.

RESULTS

Liquid phase mass transfer coefficients were measured for gas rates ranging from 0.112 to 0.314 kg-mol/min. m^2 and for liquid rates up to 300 kg-mol/min. m^2. In English units these correspond to gas rates of 1.38 to 3.86 lb-mol/hr. ft.^{2} and liquid rates to over 60,000 lb./hr. ft.^{2}. Values of k_xa for 19.0-mm Berl saddles and 12.7-mm Raschig rings are shown in Figures 1 through 3. Log-log plots of the coefficient against liquid rate and gas rate are represented well by straight lines. Data for 6.35-mm Raschig rings yielded similar plots.

The optimum values for both slopes which produced the minimum value of the root mean error between the experimentally measured k_xa values and those calculated by the correlation equation $k_xa = CL^rG^s$ were determined by a computerized search program in a range about initial values of the parameters determined from the plots. The optimization program, Pattern, was used (Moore et al., 1968). It searches for the optimum value of a specified function by varying the values of the parameters that affect the function within specified ranges of the parameters. In the analysis of this work the minimum value of the root mean error between the experimental and calculated values for k_xa from the correlation equation was found. The values of C, r, and s producing this minimum for the data of a specific packing were the optimum correlation parameters.

Final values for C, r, and s are presented in Table 1. The power for the liquid rate dependence is close to unity, particularly for Raschig rings. This agrees with the results of Danckwerts and Gillham (1966) for CO_2 absorption into sodium sulfate solutions in concurrent flow with 38.1-mm Raschig rings. The data and correlations are compared in Figures 4, 5, and 6.

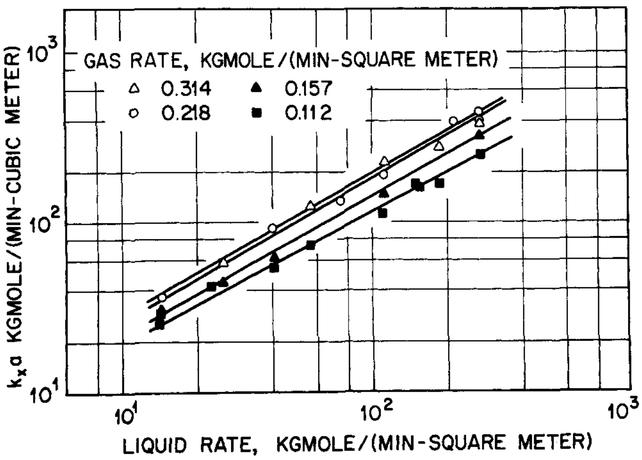

Fig. 1. Mass transfer coefficients for 19.0-mm Berl saddles.

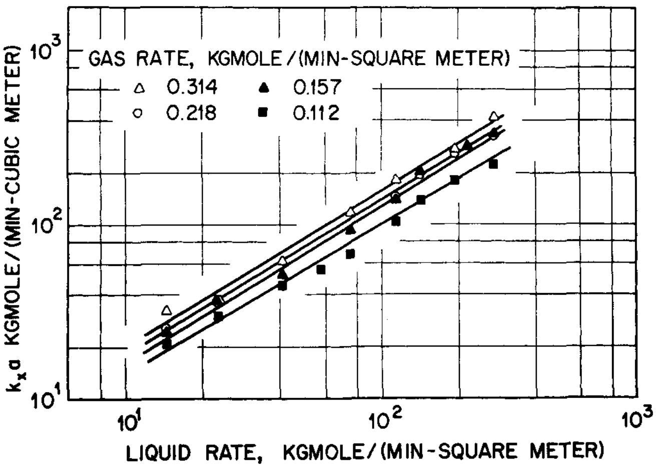

Fig. 2. Mass transfer coefficients for 12.7-mm Raschig rings.

COMPARISON OF CONCURRENT AND COUNTERCURRENT COEFFICIENTS

The work of Sherwood and Holloway (1939) included data for carbon dioxide absorption from air into water as well as for other systems. They present general correlations for countercurrent flow for 38.1-, 25.4-, and 12.7-mm Berl saddles and 50.8-, 38.1-, and 25.4-mm Raschig rings developed from their data. Correlation parameters for 19.0-mm Berl saddles can be easily estimated since the variation with packing size observed was small. They extend their coverage for Raschig rings to include 12.7- and 9.5-mm sizes by correlating data obtained by Allen in previous work; however, it should be noted that the parameters obtained for these latter two sizes deviated greatly from the parameters for the other size rings which showed only a slight variation of their parameter values.

The data of Koch et al. (1949) on carbon dioxide absorption from air into water with 9.5-, 12.7-, 19.0-, and 31.7-mm Raschig rings in countercurrent flow was used in the comparison with the results of the present investigation. They found little variation of $k_L a$ for all sizes of rings.

Investigators in countercurrent flow mass transfer in packed columns have not observed a significant gas rate effect on the mass transfer coefficient. Most of the results reported by Sherwood and Holloway were determined with a gas rate of 0.64 kg-mol/min. m^2, [230. lb./(hr. ft.^{2})], or greater; although they did take data at lower gas rates to determine if there was a significant gas rate effect. The gas rates used by Koch et al. were between 0.05 and 0.22 kg-moles/min. m^2, and the scatter observed in their data was large.

In the concurrent studies of McIlvriod as discussed by Wen et al. (1963), a gas rate effect was observed, and Reiss (1967) in his $k_L a$ determinations in concurrent flow observed a gas rate effect. McIlvriod used gas rates from 1.12 to11.2 kg-mol/min. m^2 which are in the higher range. He observed a gas rate effect with the glass bead packing at the lower rates in his range and observed that the effect of $k_L a$ was less pronounced at the higher gas rates. He did not observe a significant gas rate effect with the 6.35-mm carbon ring packing at the rates he used.

TABLE 1. CORRELATION PARAMETERS

Packing type	C	r	s
¾-in. Berl saddles	7.78	0.82	0.46
½-in. Raschig rings	3.59	0.93	0.42
¼-in. Raschig rings	4.23	1.06	0.75

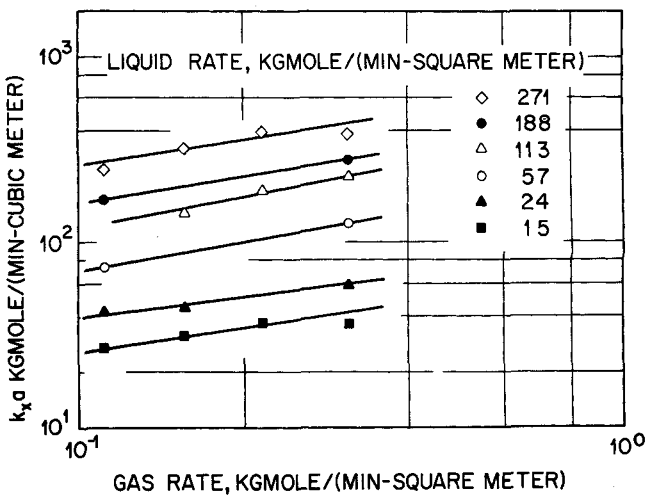

Fig. 3. Effect of gas rate (19.0-mm Berl saddles).

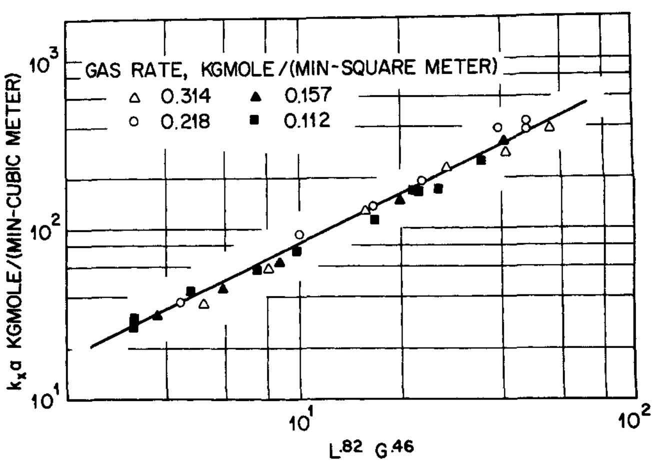

Fig 4. Correlation for 19.0-mm Berl saddles.

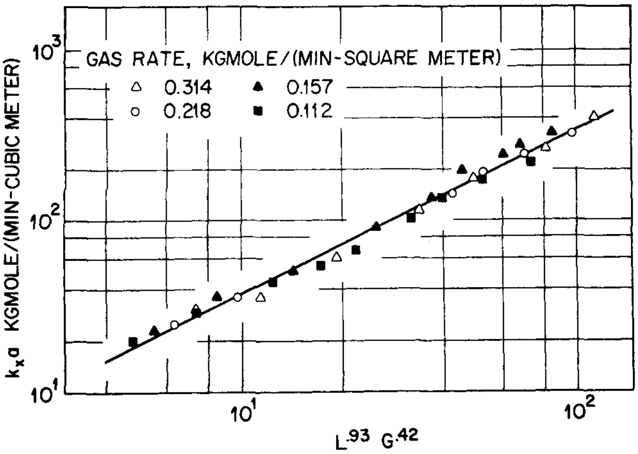

Fig. 5. Correlation for 12.7-mm Raschig rings.

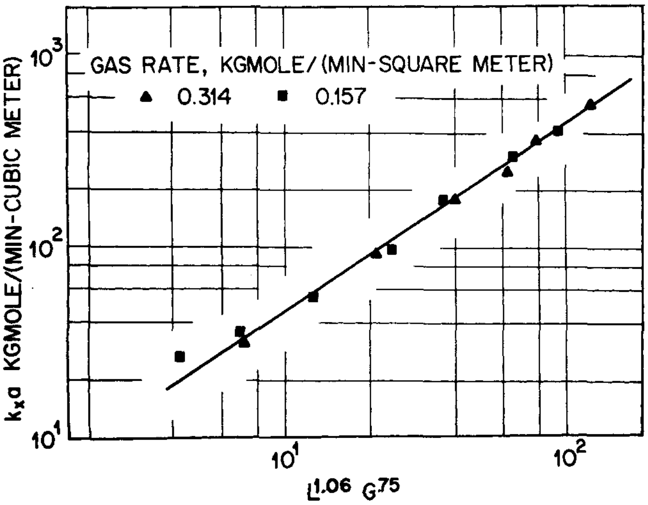

Fig. 6. Correlation for 6.35-mm Raschig rings.

The gas rate effect observed by Reiss is not immediately evident in the energy dissipation function correlation presented; however, the liquid phase energy dissipation function to which $k_L a$ is related is a function of the gas rate as well as the liquid rate.

Even though no gas rate effect has been observed in countercurrent operation, a comparison with the results of the present investigation in concurrent flow may be of interest. For 12.7-mm Raschig rings, as an example, the work of Sherwood and Holloway (1939) indicates values for $k_x a$ of about 60 and 110 for liquid rates of 20 and 40 kg-mol/min. m^2. The Koch correlation indicates values of about 45 and 90 at these liquid rates. The data points for concurrent flow in Figure 2 fall below these values.

Countercurrent flow is limited by loading and flooding at high liquid rates. The data of Koch et al. was taken at liquid rates between 2.77 and 18.7 kg-mol/min. m^2; whereas, the data of Sherwood and Holloway went to the loading point, which for 25.4-mm Raschig rings occurred at a liquid rate of approximately 77.0 kg-mol/min. m^2 for a gas rate of 0.27 kg-mol/min. m^2. For 12.7- and 6.35-mm Raschig rings the flooding rates are about 60 and 40 kg-mol/min. m^2 respectively.

EFFECT OF PACKING HEIGHT

Coefficients were smaller in deeper packed beds, in agreement with the results of previous investigators. Danckwerts and Gillham (1966) suggest that the effect is not significant when the height is less than seven column diameters. Wen et al. (1963) in their investigation of $k_G a$ in concurrent flow found a significant decrease above a packing height of 2 column diameters. The usual explanation for the effect is channeling of the liquid to the wall which becomes significant if the height is more than a few diameters.

The results of the present work show this effect to be of major importance. Coefficients measured with a height of 8.5 column diameters compared with those measured with a height of 2.5 diameters were much lower by factors of 3 to 4 at the lowest liquid rates. At the highest liquid rates the factor was about 1.5, and these factors hold for all three packings.

Porter and Templeman (1968) examined the chaneling of water in packed columns in the absence of a moving gas phase. They verified the general observation that the proportion of the liquid flowing down the wall decreased as the ratio of the column diameter to the packing particle diameter increased and as the liquid rate increased. If their results are applied to the packings and column diameter in the present work, the portion of wall flow for the Berl saddles is larger by factors of 2 to 3 than for the small Raschig rings. Yet the effect of packing height was just as severe for the rings. Hence wall flow alone does not appear to explain this effect, and further work is required in this area.

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