Simultaneous Measurement of Interfacial Area and Mass Transfer Coefficient in Three-Phase Fluidized Beds

The technique of simultaneous measurement of the mass transfer coefficient and the interfacial area (Robinson and Wilke, 1974) was used to measure the volumetric liquid-phase mass transfer coefficient ($k_L\underline{a}$), the interfacial area ($\underline{a}$) and the true liquid-phase mass transfer coefficient (k_L) in three-phase fluidized beds. The eddy-cell model of Lamont and Scott (1970) was found to give a reasonable fit to the k_L data, if the proportionality constant is modified. The $k_L\underline{a}$ and $\underline{a}$ data were correlated by empirical equations. Some information on bubble diameter was deduced from the interfacial area and holdup measurements.

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SCOPE

Much attention has been devoted recently to the phenomenon of three-phase fluidization with the interest arising mainly from the possible application to coal liquifaction and other hydrogenation processes. It has been reported that in most of these processes gas-liquid mass transfer is the rate-controlling factor and invariably this particular process is characterized by the volumetric mass transfer coefficient, $k_L\underline{a}$. However, it seems more appropriate to attempt the prediction of k_L and $\underline{a}$ (the constituent parameters of $k_L\underline{a}$) from separate correlations, because there is enough evidence in the literature to suggest that k_L and $\underline{a}$ depend on completely different parameters of the system (e.g., k_L depends mainly on diffusivity and viscosity, while $\underline{a}$ depends mainly on surface tension and ionic concentration of the solution).

This paper attempts to study systematically $k_L\underline{a}$, k_L, and $\underline{a}$ in three-phase fluidized beds. Separate measurements of k_L and $\underline{a}$ were previously reported by Lee and Worthington (1974), and Ostergaard (1978), in both cases doubts exist as to the correctness of the published values. Lee and Worthington (1974) measured interfacial areas not inside the bed but in the region above it; the two values may be quite different as was discovered in the study presented here. The values of $k_L\underline{a}$ reported by Ostergaard (1978) differ by a factor of about 5 from the earlier reported data (Ostergaard and coworkers; 1971, 1972). There is also no agreement between the interfacial areas obtained by Lee and Worthington (1974), and Ostergaard (1978).

The technique of Robinson and Wilke (1974) has been used here to study the effect of various process parameters such as particle diameter, gas and liquid velocity, gas-phase resistance, decrease in gas flow rates, etc. on the above quantities. Some information on bubble diameters has been deduced. The results obtained here are likely to further the understanding of the phenomenon of gas-liquid mass transfer in the three-phase fluidized beds and they may also provide guidance in the choice of the optimum size of fluidized particles.

CONCLUSIONS AND SIGNIFICANCE

On the basis of the technique developed by Robinson and Wilke (1974), a simple chemical system was proposed and used for the measurement of $k_L\underline{a}$, k_L, and $\underline{a}$ in three-phase fluidized beds; this system seems to be applicable to measurements of mass transfer parameters in almost any contactor, except in packed beds. Measurements of $k_L\underline{a}$, k_L, and $\underline{a}$ in three-phase fluidized beds of 1.98-, 4.08-, and 5.86-mm particles showed that the particle size has a profound effect on $\underline{a}$ and therefore on $k_L\underline{a}$, while there was little difference in k_L values among the beds of various particles.

Further, it was found that both $k_L\underline{a}$ and $\underline{a}$ increased with increasing superficial gas velocity (u_g) for all three-particle sizes. There was no effect of liquid velocity on $k_L\underline{a}$ and $\underline{a}$ in beds of the two larger particles, while both parameters increased with increasing liquid velocity (u_l) in the bed of the smallest particles. The true mass transfer coefficient k_L was found to be independent of the liquid velocity for all three particle sizes, while an increase in u_g was found to increase k_L for the smallest particle, decrease k_L for the 4.08-mm particles, and no effect was observed for the largest size of particles.

A comparison of the interfacial area and $k_L\underline{a}$ obtained in three-phase fluidized beds with the values published for other contactors revealed that three-phase beds with particles of the size of 4 mm and larger were probably the most efficient gas-liquid contactors, while beds of particle size of 2 mm are comparable in performance to packed beds. The difference between the values of $k_L\underline{a}$ in beds of the smallest and those for larger particles was by a factor of 5 or more.

The results indicate that a great improvement in the gas-liquid mass transfer rate in various hydrogenation and other processes can be achieved by using catalyst particles of a relatively large size. However, this improvement in gas-liquid mass transfer is achieved only at the expense of a reduction in the rate of the surface reaction due to a reduced catalyst surface area. The latter problem can be solved to some extent by employing a porous catalyst. Although pore diffusion may tend to slow down the surface reaction, it is believed that this would be more than compensated by the improvement in the gas-liquid mass transfer rate.

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The industrial importance of three-phase fluidized bed has been emphasized by many authors; Ostergaard (1968) published an excellent review of industrially important processes. It has been reported that in many of the processes described in literature, mass transfer across the gas-liquid interface is the rate-controlling step. Despite such importance of gas-liquid mass transfer in three-phase fluidized beds, very few studies are found in literature; in the majority of the reported investigations, only volumetric liquid-phase mass transfer coefficients were measured. However, it is generally recommended to attempt separate correlations for k_L and $\underline{a}$, as each parameter depends on a different set of variables. The present investigation attempts to study the effect of various process parameters on $k_L\underline{a}$, k_L, and $\underline{a}$ in three-phase fluidized bed of different particle sizes.

PREVIOUS WORK

Ostergaard and coworkers (1971, 1972) studied absorption of CO_2 and O_2 into water in beds of 1- and 6-mm glass ballotini and in a bubble column. They found that the volumetric absorption coefficient increased with increasing gas flow rate, but it remained unaffected by variations in the liquid flow rate for the 6-mm particles, while it increased with an increase in liquid flow rate in the bed of 1-mm particles. Absorption coefficients in beds of 6-mm particles were approximately ten times greater than in beds of 1-mm particles, while coefficients in the solid-free bubble column were of intermediate magnitude. The authors also observed that $k_L\underline{a}$ decreased with the bed height in beds of 1-mm particles and in bubble columns, but in beds of 6-mm particles the coefficients passed through a maximum. In a later study, Ostergaard (1978) reported values of $k_L\underline{a}$ obtained by chemical absorption of CO_2 into a carbonate-bicarbonate buffer for particles of the same sizes as used in the two previous studies. However, the $k_L\underline{a}$ values measured by the chemisorption technique were about five times lower than those obtained earlier by the physical adsorption method.

Dakshinamurty and coworkers (1973, 1974, 1976) also studied the process of physical absorption of CO_2 and O_2 into water in three-phase fluidized beds and confirmed the findings of Ostergaard and coworkers (1971, 1972). Finally, Lee and Worthington (1974) investigated absorption of CO_2 into water and a sodium sulphate solution in a bed of particles 6 mm in diameter. In addition to measurements of $k_L\underline{a}$, they also measured the interfacial area immediately above the bed by the light transmission technique and assumed that it was virtually the same as within the bed. The authors found a similar effect of u_g and u_l on $k_L\underline{a}$ as observed by Ostergaard and coworkers (1971, 1972); but, in contrast to the findings of the above authors, they reported that $k_L\underline{a}$ steadily decreased with the bed height.

EXPERIMENTAL TECHNIQUE

There are many techniques reported in literature for determining $k_L\underline{a}$ and $\underline{a}$. Sharma and Danckwerts (1970) compared the available techniques (physical absorption/desorption and chemical absorption) and systems for measuring $k_L\underline{a}$ and concluded that chemical methods were generally superior to physical absorption, as they did not require any knowledge of flow pattern or the use of very accurate analytical methods. A comparison of the available techniques for the measurement of interfacial area (viz., the light-scattering method, the chemical method, and the photographic method) was carried out by Landau et al. (1977) and the authors concluded that the light transmission technique gave the best results followed by the chemical method and the photographic method. However, optical techniques were developed for gas-liquid dispersions only and they cannot be applied to systems containing a solid phase.

Recently, Robinson and Wilke (1974) proposed a novel technique for a simultaneous determination of $k_L\underline{a}$ and $\underline{a}$. The technique involves a simultaneous physical desorption of one gas from and absorption with chemical reaction of another solute gas into the liquid phase. The rate of desorption of the first species is used to evaluate the overall mass transfer coefficient $k_L\underline{a}$ and the rate of absorption with chemical reaction is then used to calculate the interfacial area $\underline{a}$. The main advantage of the method is that it ensures that both $k_L\underline{a}$ and $\underline{a}$ (and therefore k_L) are determined under truly identical physico-chemical and hydrodynamic conditions. For this reason, it was decided to employ this method in the present investigation.

SIMULTANEOUS EVALUATION OF $k_L\underline{a}$, $\underline{a}$, AND k_L

Robinson and Wilke (1974) performed their measurements in an agitated vessel by simultaneously absorbing CO_2 from a CO_2-N_2 mixture into a weak (<0.1M) solution of KOH and desorbing oxygen from the same solution into the CO_2-N_2 mixture. The change in O_2 concentration in the KOH solution was measured by a dissolved oxygen probe of Johnson et al. (1964); the change in the carbonate concentration was determined by potentiometric titration. For a fast pseudo-first order reaction in the liquid phase, the rate of absorption is given by (Danckwerts, 1951):

$$R_A\underline{a} = \underline{a}C_A^* \, (k_{LA}{}^2 + D_A k_2 C_0)^{0.5} \tag{1}$$

The rate expression for the physical desorption of oxygen is given by:

$$R_B\underline{a} = k_{LB}\underline{a} \cdot \Delta C_{B,m} \tag{2}$$

where $\Delta C_{B,m}$ is the mean value of the difference in oxygen concentration between the interface and the bulk of the liquid phase. Since $k_L \propto \sqrt{D}$, it holds that:

$$k_{LB} = k_{LA}\sqrt{D_B/D_A} \tag{3}$$

and Eq. 2 becomes

$$R_B\underline{a} = k_{LA}\underline{a}\sqrt{D_B/D_A}\,\Delta C_{B,m} \tag{4}$$

To calculate the interfacial area, Eq. 1 is rearranged to give:

$$\underline{a} = \{[(R_A\underline{a}/C^*_A)^2 - (k_{LA}\underline{a})^2]/D_A k_2 C_0\}^{0.5} \tag{5}$$

The calculation technique derived above involves:

1. Experimental measurement of $R_A\underline{a}$ and $R_B\underline{a}$, with A referring to the absorption of CO_2 and B referring to the desorption of oxygen.
2. The use of $R_B\underline{a}$ to calculate $k_{LB}\underline{a}$ by Eq. 2 and $k_{LA}\underline{a}$ by Eq. 4.
3. The use of $R_A\underline{a}$ and $k_{LA}\underline{a}$ to calculate $\underline{a}$ by Eq. 5.
4. The calculation of k_{LB} by dividing $\underline{a}$ into $k_{LB}\underline{a}$.

The essential condition for the success of the method is that the volumetric mass transfer coefficients for chemical absorption and physical desorption be equal after correction for a different diffusivity. However, Joosten and Danckwerts (1973) reported differences in $k_L\underline{a}$ values for physical and chemical absorption in packed beds. These differences were attributed to pockets of stagnant liquid that might become completely saturated in physical absorption. Since it is highly unlikely that stagnant pockets of liquid can exist in three-phase fluidized beds (due to a very high degree of turbulence in the bed), there is no reason to believe that $k_L\underline{a}$ would be different during physical desorption and chemical absorption. This assumption was, nevertheless, put to an experimental test in a bed of 4.08-mm particles; it was found that the volumetric mass transfer coefficients determined by physical desorption agreed remarkably well with those obtained by chemisorption. (For details see Dhanuka, 1978.)

CHEMICAL SYSTEM

The system originally proposed by Robinson and Wilke (1974) could not be used in this study because of relatively low conversions in the liquid and large changes in the ionic strength of the solution during the process of absorption. The latter would affect the rate constant for the reaction between CO_2 and OH^- in a way that cannot yet be predicted with sufficient accuracy. A system free of these drawbacks was found in a 1.01 M sodium carbonate-sodium bicarbonate solution with a buffer ratio of 1 containing 0.01 M sodium hypochlorite to enhance the reaction rate. In this system, the change in the ionic strength of the solution resulting from the absorption of CO_2 was always below 5%.

The concentration of oxygen in the solution was measured by a dissolved oxygen probe, Model 8012, from Electronic Instruments Ltd. A calibration procedure for measuring the oxygen concentration in ionic solutions, in particular in a solution of Na_2CO_3-$NaHCO_3$, was developed and is briefly described in Appendix I. A more detailed description of the procedure can be found elsewhere (Dhanuka, 1978). Before a sample of solution could be subjected to the standard titration analysis for carbonate (Vogel, 1951), a stoichiometric amount of sodium arsenite was added to reduce the hypochlorite present in the solution which would otherwise bleach the indicator.

The kinetics of the reaction between dissolved carbon dioxide and sodium carbonate catalyzed by sodium hypochlorite has not yet been properly established. Measurements of the reaction rate constant were undertaken using a stirred cell. The results of these measurements are described in Appendix II.

EXPERIMENTAL

The three-phase fluidized bed was contained in a perspex tube 50 mm i.d., consisting of two sections, each 750 mm long. The bed was supported by a wire mesh through which the liquid was introduced after passing through a calming section. The gas was introduced through a ring distributor containing four equispaced 1-mm diameter holes. The ring was placed in such a way that the top portion of the ring just touched the wire screen at the bottom of the column. This arrangement ensured that both the gas and the liquid entered the column separately.

The liquid samples were withdrawn at a height of 620 mm from the bottom of the column, which formed the test section. The distributor was also part of the test section. The sampling device consisted of an upward-facing orifice; the flow of the liquid sample was controlled with a fine control needle valve. This arrangement prevented gas bubbles from being entrained into the sampling line. The liquid sample was first passed through an airtight tube containing the oxygen probe and then it was collected for chemical analysis. The pressure drop was also measured at the same height of 620 mm from the bottom. A detailed description of the experimental setup is available elsewhere (Dhanuka, 1978).

Three sizes of glass ballotini were used in this investigation with diameters 1.98 mm, 4.08 mm, and 5.86 mm.* The mass of the particles in the bed was approximately 1.5 kg. The superficial liquid velocity was varied from 6.1 to 13.8 cm/s and the superficial gas velocity from 2 to 8 cm/s.

The data for calculating $k_{LB}\underline{a}$, k_{LB} and $\underline{a}$ were collected for all three particle sizes by the technique described above. The $k_{LB}\underline{a}$ values for each particle size were also determined by physical desorption of oxygen from the same solution of carbonate-bicarbonate buffer into a gas phase consisting of pure nitrogen. This was carried out to investigate the effect of the reduction in gas flow rate resulting from the absorption of CO_2 in the liquid phase on the mean value of the volumetric mass transfer coefficient. This reduction in CO_2 systems was as high as 60-70% in some cases and might have a significant effect in many processes of industrial importance.

No investigation was undertaken of the effects of the increase of the gas flow rate caused by oxygen desorption and pressure drop; the former constituted only 1-2% of the total flow rate, while the latter represented at the most only 10% of the total pressure, both of which were neglected. The other effect that was subjected to test was that of the gas-phase resistance. For this purpose, a mixture of 75% CO_2 and 25% N_2 (at inlet) was passed through a bed of the 5.86-mm particles; the choice of the particle size was made on the grounds that these particles were expected to show the lowest resistance in the liquid, thus facilitating the detection of any contribution of the gas phase to the total resistance.

RESULTS AND DISCUSSION

Volumetric Liquid-Phase Mass Transfer Coefficient

The values of $k_{LB}\underline{a}$ were calculated from the experimental data by Eq. 2, using the logarithmic mean value of the concentration difference in the liquid phase: this is equivalent to assuming that the liquid phase was in plug flow. Available literature on liquid mixing in three-phase fluidized beds (Ostergaard and Michelsen, 1968; Michelsen and Ostergaard, 1970; Kim et al., 1972; Ostergaard, 1978; El-Temtamy et al., 1979) suggest that liquid mixing can be ignored if the particle diameter is greater than 3 mm and the column diameter less than 15 cm, both conditions were satisfied in the beds of 4.08- and 5.86-mm particles. In the case of the smallest 1.98-mm particles, liquid mixing does take place. However, El-Temtamy et al. (1979) reported quite low axial dispersion coefficients in beds of 2-mm particles with values in the range 20-60 cm^2/s. Estimations of volumetric mass transfer coefficient with and without allowance for liquid mixing showed that no significant error was introduced when the effect of back mixing was ignored.

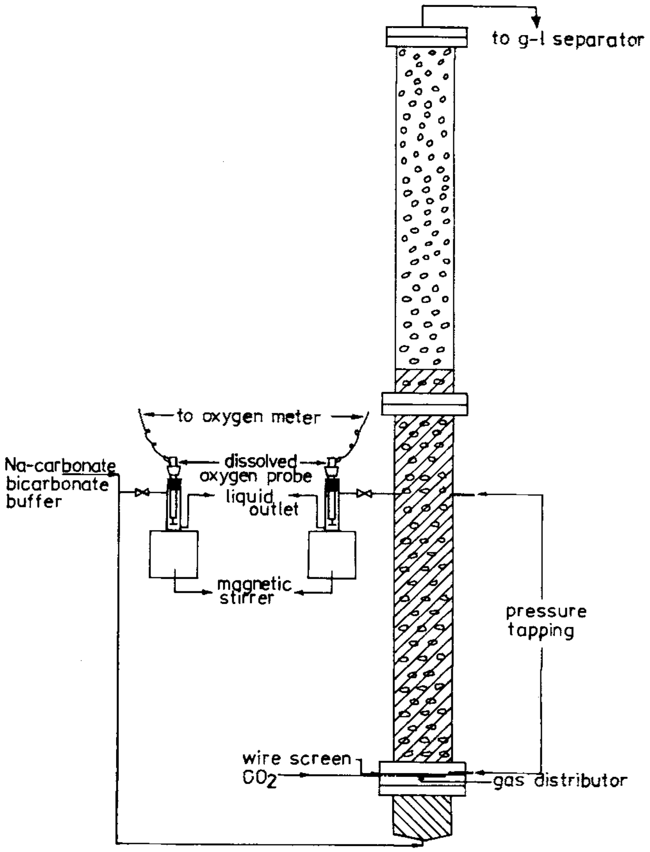

Figure 1. The experimental column.

The experimental results of $k_{LB}\underline{a}$ for oxygen desorption into the CO_2 and N_2 phases, plotted as a function of the superficial gas and liquid velocities, are shown in Figs. 2, 3 and 4 for the 1.98 mm, 4.08 mm and 5.86 mm particles, respectively. The superficial gas velocity in the case of CO_2 was calculated as the arithmatic mean of the superficial velocities at the inlet and outlet.

Figure 2 shows that the $k_{LB}\underline{a}$ value of the 1.98-mm particles increases linearly with the gas velocity and it also increases with the liquid velocity. The beneficial effect of the liquid velocity probably arises from the reported reduction of bubble coalescence and increases in the gas holdup with an increase in the liquid flow rate (Dhanuka and Stepanek, 1978), both of which combine to increase the interfacial area and thus $k_{LB}\underline{a}$.

The gas flow rate affects the $k_{LB}\underline{a}$ through an increase in the interfacial area resulting from increased gas holdup, as well as through an increase in the mass transfer coefficient due to the presence of larger size bubbles resulting from increased coalescence. The observed effect of liquid flow rate is in qualitative agreement with the results of Ostergaard and Fosbøl (1972) for a bed of 1-mm glass beads. However, it contradicts the findings of

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*The diameters represent statistical means of micrometer measurements of 50 particles.

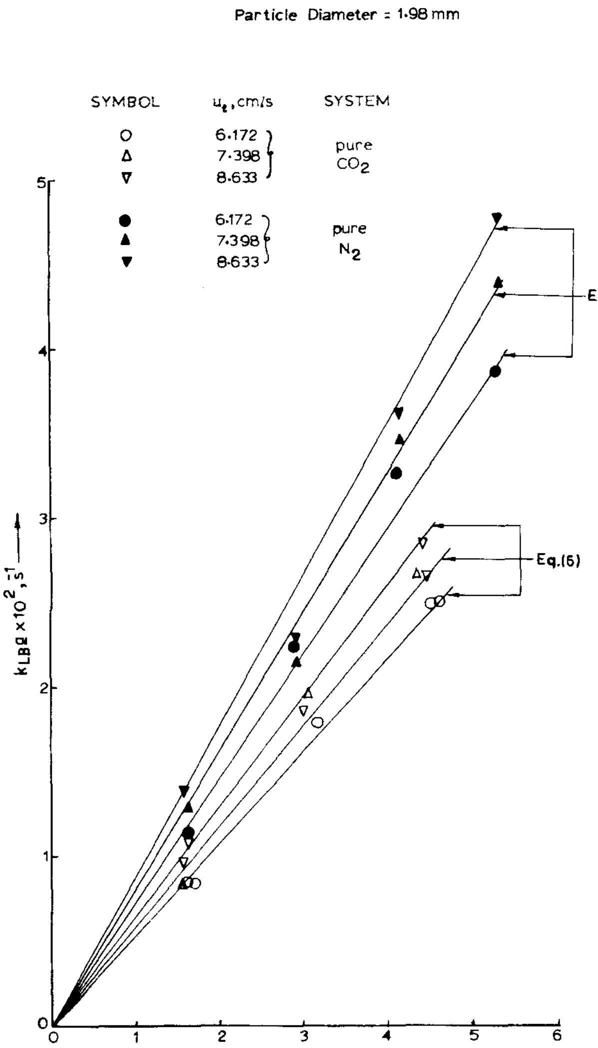

Figure 2. Effect of gas and liquid velocities on the volumetric mass transfer coefficient in a bed of 1.98-mm particles.

Dakshinamurty and coworkers (1973, 1974), who reported no effect of liquid velocity in beds of 1.06 mm sand.

As mentioned earlier, an attempt was made to study the effect of the decrease in gas flow rate along the bed on $k_{LB}\underline{a}$ by using the average gas velocity in the reactor as the basis of comparison. It was found that $k_{LB}\underline{a}$ values at the same average superficial gas and liquid velocities in the case of 1.98-mm particles were higher by about 30% with N_2 as gas phase (where no decrease in gas flow occurred), than with CO_2 (Fig. 2), while no such difference observed in the case of the two larger particles (Figures 3 and 4). This observation is of practical importance, as small particles are widely used in industrial three-phase fluidized beds.

For the observed difference, consider the two beds illustrated in Figure 5. The arithmatic mean of the inlet and outlet CO_2 flow rate in the second bed (bed 'b') is equal to the flow rate of nitrogen in the first bed (bed 'a'). A cross section BB can be defined in the bed 'b', where the local flow rate is exactly equal to the arithmatic mean flow rate and a corresponding section AA can be specified in the bed 'a'at exactly the same height. Then, below the cross section BB, the gas flow rate in the bed 'b' is always higher than in the bed 'a' and the opposite is true for the volume above the cross section. As mentioned earlier, bubble coalescence in beds of small particles increases with increased gas velocity. Thus, the average bubble diameter leaving the reference section in bed 'b' is larger than that leaving the bed 'a', although the local gas flow rates leaving these sections are exactly equal. Since in beds of small particles only bubble coalescence can take place, this means that the average bubble diameter in bed 'b' is larger than that leaving the bed 'a'. Besides, larger bubble diameter in bed 'b' may also arise from the fact that a higher inlet flow rate of CO_2 than of N_2 has to be maintained through the distributor (which influences the inlet bubble diameter) to ensure the same mean flow rate in the two beds under comparison. In both cases, larger bubbles mean lower interfacial area, which in turn leads to a reduction of the $k_{LB}\underline{a}$ values in bed 'b' against those in bed 'a'.

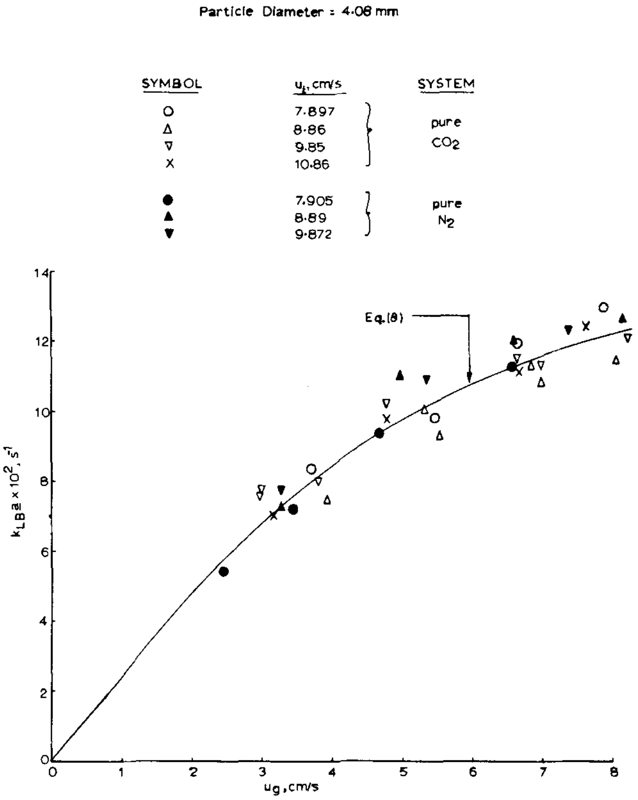

Figure 3. Effect of gas and liquid velocities on the volumetric mass transfer coefficient in a bed of 4.08-mm particles.

Figures 3 and 4 show that $k_{LB}\underline{a}$ in beds of 4.08- and 5.86-mm particles increases with the gas velocity but is independent of the superficial liquid velocity. The results are self-evident in view of the fact that the true mass transfer coefficient, as will be shown later, is virtually independent of the liquid velocity and that the interfacial area is proportional to the gas holdup. The latter varies only slightly with the liquid flow rate but increases with the gas velocity.

The volumetric mass transfer coefficients obtained here for the 5.86-mm particles agree within ± 30% with those reported by Ostergaard and coworkers (1971, 1972), Lee and Worthington (1974) and Dakshinamurty and coworkers (1973, 1974) for beds of particles of sizes around 6 mm, but differ by a factor of five and more from those recently reported by Ostergaard (1978). No data exist in the literature that could be compared with the results for the 4.08-mm particles. The $k_{LB}\underline{a}$ values obtained here were further compared with the corresponding results for bubble columns (Mashelkar, 1970), two-phase flow (Shilimkan and Stepanek, 1977), and most other gas-liquid contactors. The comparison showed that $k_{LB}\underline{a}$ values in beds of 1.98-mm particles are slightly lower than in bubble columns while the values in the beds of two larger particles were considerably higher than those reported for other contacting devices, with the bed of 5.86-mm particles being the most efficient contactor of them all.

The experimental data for oxygen desorption were correlated by nonlinear multiple regression and the following empirical equations were obtained:

For the 1.98-mm particle bed, with decreasing gas flow rate

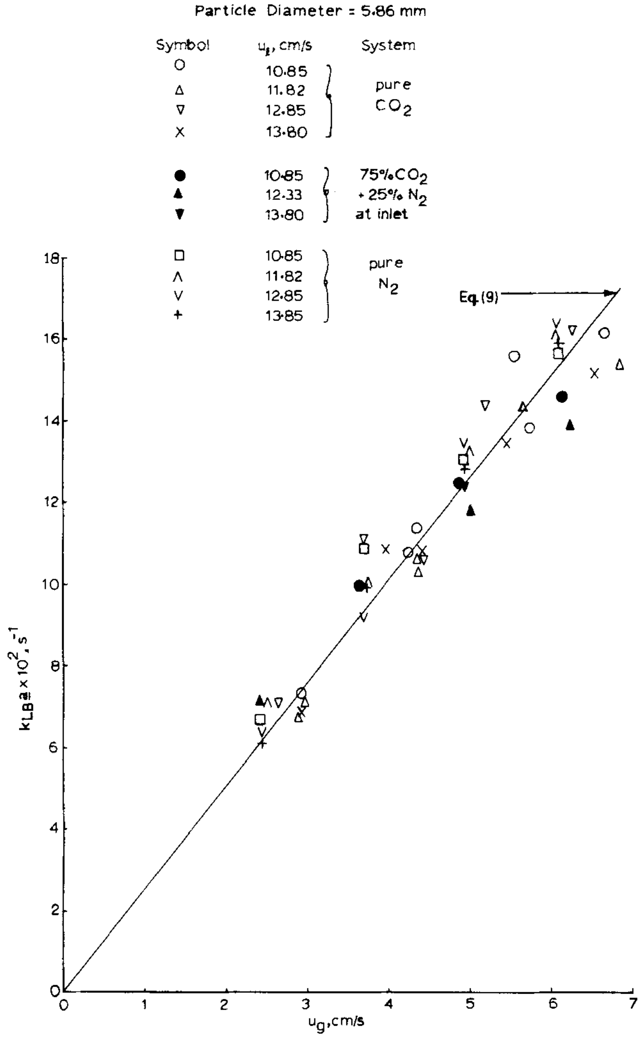

Figure 4. Effect of gas and liquid velocities on the volumetric mass transfer coefficient in a bed of 5.86-mm particles.

(i.e., pure CO_2 as the gas phase).

$$k_{LB}\underline{a} = (2.37 \pm 0.215) \times 10^{-3}\ u_g^{(1\pm0.063)}\ u_l^{(0.45\pm0.058)} \quad (6)$$

and with constant gas flow rate (i.e., pure N_2 as the gas phase).

$$k_{LB}\underline{a} = (3.25 \pm 0.304) \times 10^{-3}\ u_g^{(1\pm0.071)}\ u_l^{(0.55\pm0.84)} \quad (7)$$

For the 4.08-mm particle bed:

$$k_{LB}\underline{a} = (3.41 \pm 0.195) \times 10^{-2}\ u_g^{(0.555\pm0.075)} \quad (8)$$

and for the 5.86-mm particle bed:

$$k_{LB}\underline{a} = (2.52 \pm 0.041) \times 10^{-2} u_g \quad (9)$$

where $k_{LB}\underline{a}$ is expressed in s^{-1} and u_g and u_l in cm/s. The equations also include the standard deviation of the correlation coefficients.

Interfacial Area

Experimental values of the interfacial area are plotted in Figures 6, 7, and 8 for the 1.98-, 4.08-, and 5.86-mm particles respectively as functions of the arithmatic mean superficial gas velocity and the superficial liquid velocity.

Figure 6 shows that the interfacial area for the 1.98-mm particles increases with an increase in both the gas and the liquid superficial velocities which is a direct consequence of the similar effect of the two velocities on the gas holdup (Dhanuka and Stepanek, 1978). For the 4.08- and 5.86-mm particle beds (Figures 7 and 8), the interfacial area increases fairly linearly with increasing gas velocity but is independent of the liquid velocity. The observed effect of gas velocity on the interfacial area is due to a similar effect of gas velocity on gas holdup as reported by several authors (Dhanuka and Stepanek, 1978; Ostergaard, 1968).

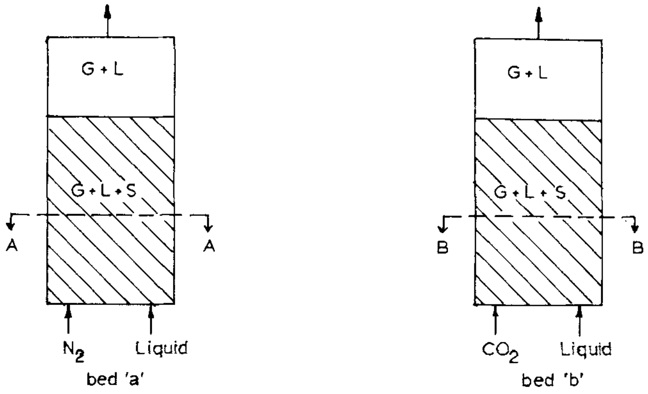

Figure 5. Comparison of beds of 1.98-mm particles with N_2 and CO_2 as the gas phase.

It was also reported that the gas holdup decreases slightly with increasing liquid velocity. However no such effect on the interfacial area could be detected, probably due to the scatter in the measured values of the latter parameter.

A comparison among Figures 6, 7, and 8 reveals that the interfacial area in three-phase fluidized beds of the two larger particles is much larger than in beds of 1.98-mm particles. Fluidized beds of 4.08- and 5.86-mm particles also show a considerably higher interfacial area than all other contacting devices. Under comparable conditions, the interfacial areas in

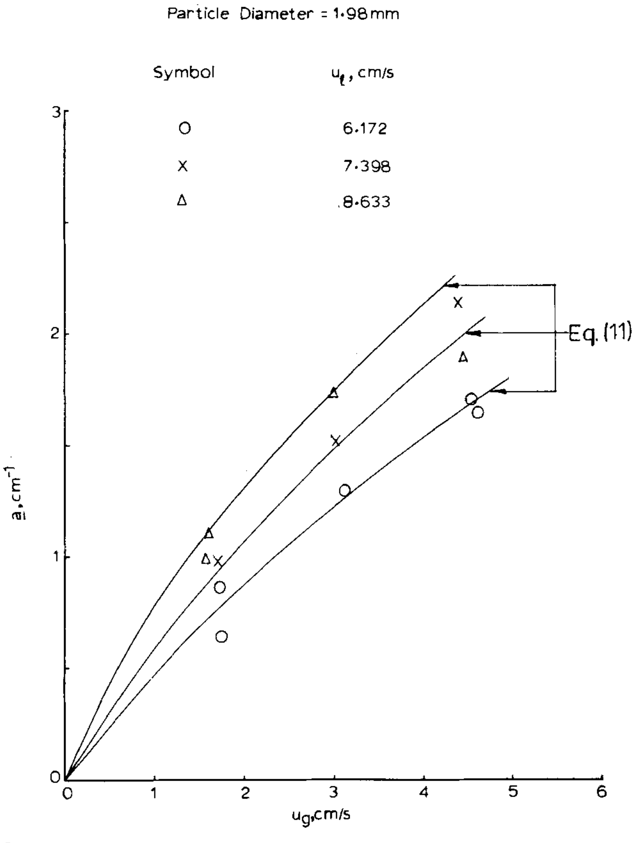

Figure 6. Effect of gas and liquid velocities on the interfacial area in a bed of 1.98-mm particles.

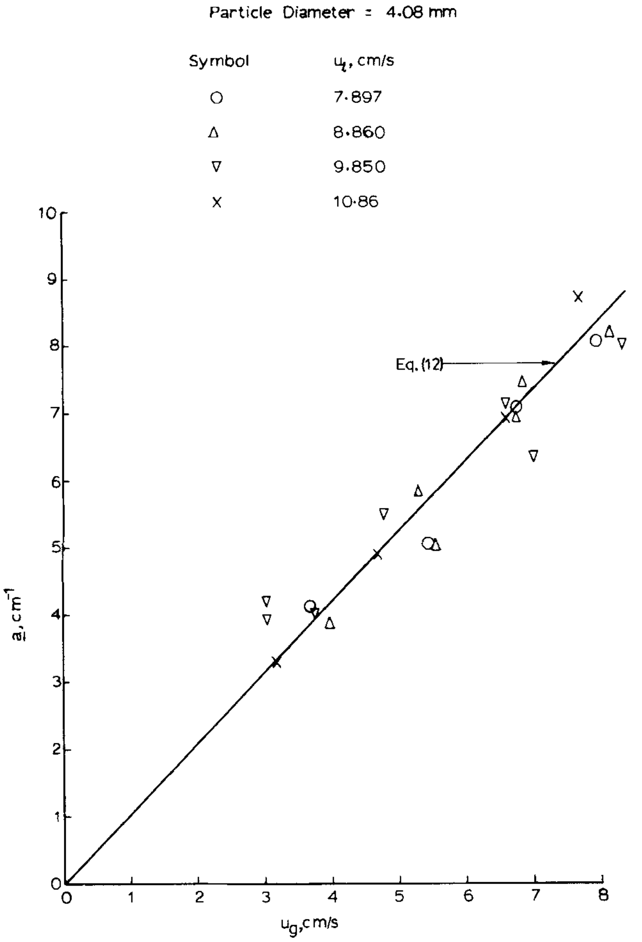

Figure 7. Effect of gas and liquid velocities on the interfacial area in a bed of 4.08-mm particles.

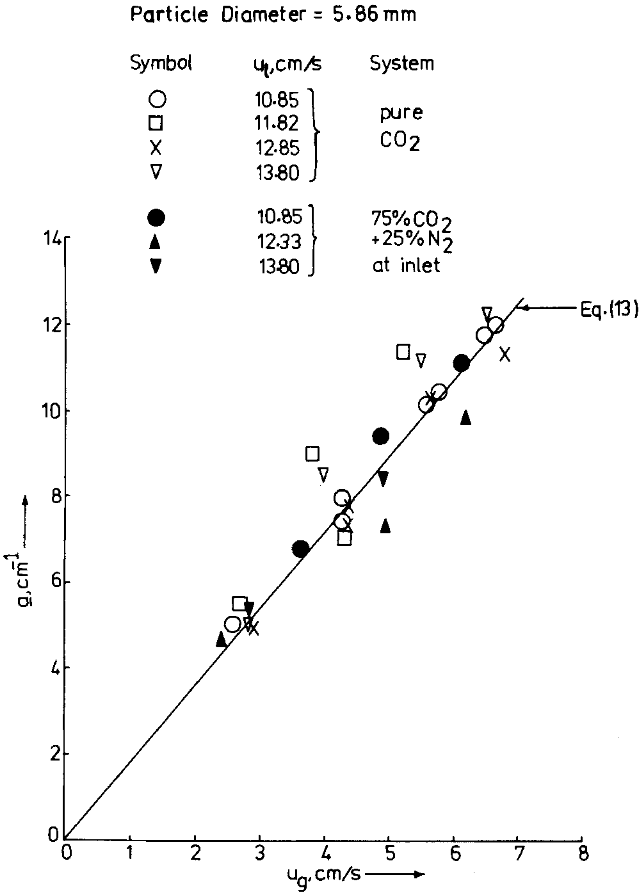

Figure 8. Effect of gas and liquid velocities on the interfacial area in a bed of 5.86-mm particles.

beds of 4.08- and 5.86-mm particles were found to be 4-5 cm^2/cm^3 and 7-9 cm^2/cm^3 respectively, while the corresponding values for bubble columns and for the fluidized bed of 1.98-mm particles were 1-2 cm^2/cm^3, for fixed beds around 1 cm^2/cm^3 and in agitated vessels around 3-4 cm^2/cm^3.

The interfacial areas in the bed of 5.86-mm particles are also about five times larger than those reported by Lee and Worthington (1974) for the region immediately above a fluidized bed of 6-mm particles, although, as mentioned earlier, the volumetric mass transfer coefficients $k_{LB}\underline{a}$ agreed within 30% in the two cases. This seems to suggest that rapid bubble coalescence occurs immediately above the bed.

A combination of the relationships for the interfacial area and for the gas holdup shows that the interfacial area is directly proportional to the gas holdup in beds of 4.08- and 5.86-mm particles, suggesting that the mean bubble diameter remains independent of the flow rates in such beds. The mean bubble diameter was, therefore, calculated from the following equation:

$$d_b = 6 \cdot \epsilon_g / \underline{a} \tag{10}$$

Since it was not possible to measure gas holdup simultaneously with mass transfer measurements, its values were calculated from the model of the three-phase fluidized bed proposed by Dhanuka and Stepanek (1978). The bubble diameters were found to be 1.8-2 mm in the beds of 4.08-mm particles and 1.2-1.4 mm in the beds of 5.86-mm particles, which are very small values indeed.

The following empirical correlations were obtained by multiple nonlinear regression of the data. For beds of 1.98-mm particles with decreasing gas flow rate (CO$_2$ as gas phase),

$$\underline{a} = (0.232 \pm 0.066)\, u_g(0.677 \pm 0.06)\, u_l(0.534 \pm 0.13) \tag{11}$$

For beds of 4.08-mm particles,

$$\underline{a} = (1.05 \pm 0.045)\, u_g \tag{12}$$

and for beds of 5.86-mm particles,

$$\underline{a} = (1.8 \pm 0.076)\, u_g \tag{13}$$

In the above equations, $\underline{a}$ is in cm^2/cm^3 and both u_g and u_l in cm/s. As mentioned previously, the equations include the standard deviation of the correlation coefficients.

True Liquid-Phase Mass Transfer Coefficient

Figures 9, 10, and 11 show the experimental values of k_{LB} for oxygen plotted against the superficial gas velocity with u_l as parameter for the 1.98-, 4.08-, and 5.86-mm particles, respectively. These figures show that the k_{LB} values are, as expected, independent of u_l, but the effect of an increase in u_g is rather complicated: it increases k_{LB} in beds of 1.98-mm particles, decreases k_{LB} in beds of 4.08-mm particles, and no effect was observed in beds of 5.86-mm particles. A similar trend in k_{LB} values with increasing gas velocity in beds of 1-, 3-, and 6-mm particles was reported by Ostergaard (1978), although his absolute values differ from ours to a similar extent as did the volumetric mass transfer coefficient mentioned earlier.

The increase in k_{LB} with increasing gas velocity in the case of 1.98-mm particles can be explained by the fact that bubble coalescence increases with increasing gas velocity and this in turn leads to an increase in k_{LB} due to the freer motion of the gas-liquid interface of large bubbles. On the other hand, the bubble diameter in beds of the two larger particles remain fairly constant and the same should apply to k_{LB}. This is indeed true in the case of the 5.86-mm particle bed (Figure 11). However, the behavior of beds of 4.08-mm particles does not fit the pattern (Figure 10). The reduction in the mass transfer coefficient with u_g is probably caused by the changes in the mobility of the

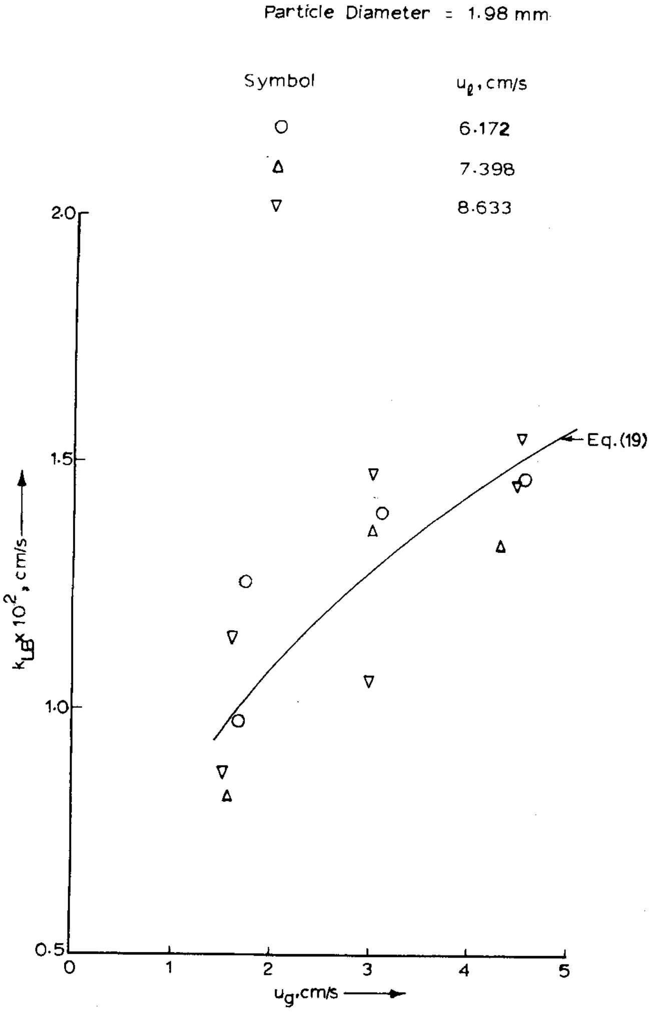

Figure 9. Dependence of the true mass transfer coefficient on the gas and liquid velocity in the bed of 1.98-mm particles.

gas-liquid interface with the slight reduction of the bubble diameter with the increasing gas flow rate.

Although this was not possible to detect from the bubble diameter calculations above, this trend could be noticed for the 4.08-mm particle bed from the drift-flux velocity calculations. However, in the case of 5.86-mm particle bed, the same calculations indicated constant bubble size. A more detailed description of the results of drift velocity calculations can be found elsewhere (Dhanuka, 1978). According to Calderbank (1967), bubbles with a diameter greater than 2.5 mm exhibit a free interface behavior, while bubbles with a diameter of 1 mm or less behave as rigid spheres. The data presented by Calderbank (1967) and most other correlations proposed in literature predict that the k_{LB} for bubbles 2.5 mm in diameter is larger than the corresponding value for 1-mm bubbles by a factor of about three with values falling off rapidly in the transition zone. This may explain the observed behavior of k_{LB} in beds of 4.08-mm particles, the bubble diameter in which lay in the transition region.

The experimental data were compared with published formulae for the prediction of the mass transfer coefficient. In the case of the Higbie's (1935) penetration theory, it was assumed that the exposure time can be approximated by the time taken by the bubble to rise to a distance equal to its diameter: it was found that the mass transfer coefficients predicted from this theory were larger by some 100-150% than the experimental values.

In another attempt, the data were compared with the equations proposed by Lochiel and Calderbank (1964) for mass transfer from a gas bubble into the continuous phase around axisym-

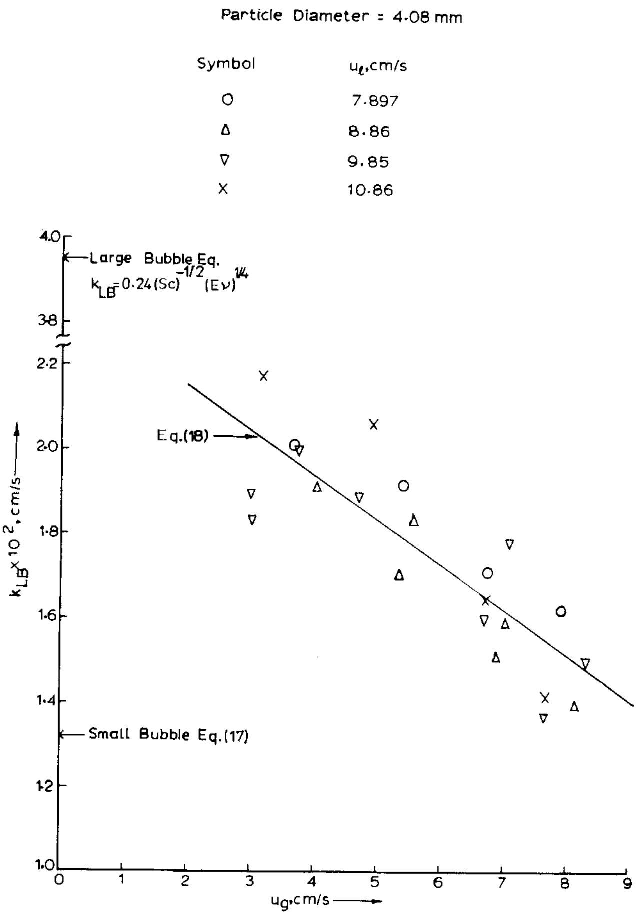

Figure 10. Dependence of the true mass transfer coefficient on the gas and liquid velocity in the bed of 4.08-mm particles.

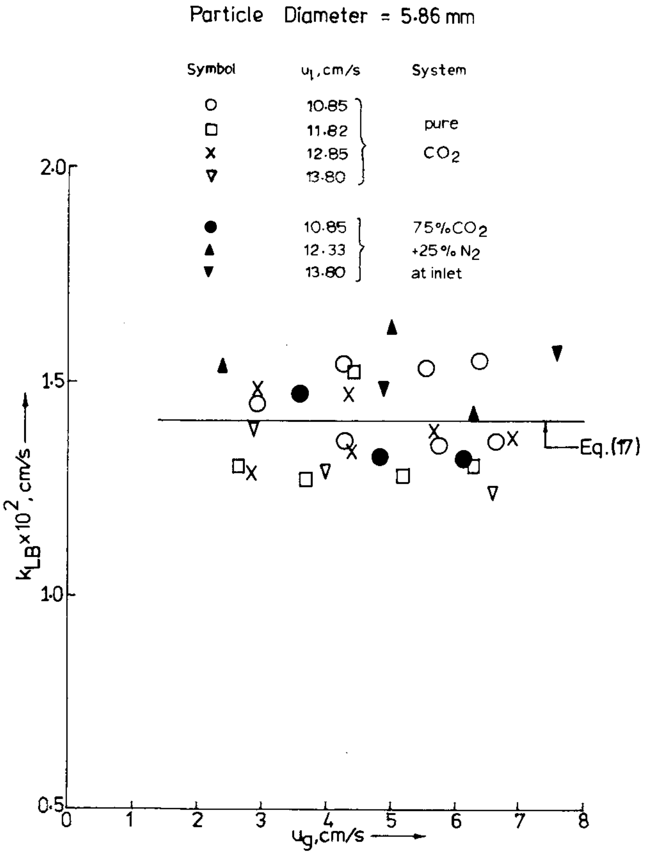

Figure 11. Dependence of the true mass transfer coefficient on the gas and liquid velocity in the bed of 5.86-mm particles.

metric bodies of revolutions. In this case, the predicted k_{LB} values were lower by almost an order of magnitude; the same difference was also obtained when comparing with the equations proposed by Calderbank and Moo Young (1961).

Finally, the data were compared with the eddy cell model of Lamont and Scott (1970). The authors derived the following formulae for k_{LB} for a fixed interface:

$$k_{LB} = 0.4\ Sc^{-2/3}(E\nu)^{1/4} \tag{14}$$

and for a free interface,

$$k_{LB} = 0.4\ Sc^{-1/2}(E\nu)^{1/4} \tag{15}$$

The rate of energy dissipation, E, in the liquid can be written in terms of pressure drop as:

$$E = \left(\frac{\Delta P}{h}\right)\left(\frac{1}{\rho_l}\right)\left(\frac{u_l}{\epsilon_l}\right) \tag{16}$$

The liquid holdup ϵ_l was calculated from the hydrodynamic model published earlier (Dhanuka and Stepanek, 1978).

The data for the bed of 5.86-mm particles were compared with Eq. 14, and those for the 1.98-mm particles with Eq. 15, since the *bubble interface* was rigid in the former and free in the latter case. Neither equation was expected to be strictly applicable to the data for the 4.08-mm particles which exhibit transition behavior.

For the 5.86-mm particle bed, it was found that Eq. 14 overestimated the k_{LB} values by about 40% which suggested that the value of the constant in Eq. 15 may not be correct. Indeed, the data were satisfactorily correlated (Figure 11) by:

$$k_{LB} = 0.24\ Sc^{-2/3}/(E\nu)^{1/4} \tag{17}$$

with an average deviation of ± 12%. The difference between the theoretical and the empirical value of the porportionality constant may be due to the fact that the assumption that all the energy dissipated in the liquid is converted into small scale eddies oversimplifies the real situation. The value of the constant lies among values reported by various investigators for widely different systems. Thus, Calderbank and Moo-Young (1961) found a value of 0.13 for solid dispersion in agitated vessels. Prasher (1973) reported a value of 0.196 for turbulent absorption in an open channel; Prasher and Wills (1973) obtained a value of 0.592 for gas absorption in agitated vessels.

The applicability of the model to the bed of 4.08-mm particles, which lies in the transition region between free and fixed interface, can be tested by calculating the k_{LB} values from both equations above: the results should provide limits for the experimental data. For this purpose, the earlier obtained empirical value of the proportionality constant of 0.24 rather than the theoretical value was used in both equations. The data lie close to the lower limit for the fixed interface indicating that the mobility of the bubble interface was very restricted. Also the bubble diameter slightly decreases with increasing gas flow rate, and this is reflected by the gradual approach of the mass transfer coefficient towards the lower limit as the gas flow rate increases. Since neither Eq. 14 nor Eq. 15 can be used for the 4.08-mm particles, the data were correlated by the following empirical equation:

$$k_{LB} \times 10^2 = (2.355 \pm 0.095) - (0.1045 \pm 0.016)u_g \tag{18}$$

In Eq. 18, both k_{LB} and u_g are expressed in cm/s; it also includes the standard deviation of the regression coefficients.

In the case of the 1.98 mm-particles, it was observed that the bubbles were much greater than 2.5 mm. Therefore, Eq. 15 for free interface was used for comparison, using the empirical value of 0.24 as the proportionality constant. The mass transfer coefficients so predicted were found to be greater than the experimental values by a factor of almost three. This discrepancy is probably due to two causes. First, the assumption that all the energy dissipated in the liquid is converted into small-scale eddies that govern the process of mass transfer may not be exactly true. This indeed seems to be the case, because the values of $(E\nu)^{1/4}$ calculated by means of Eq. 17 were found to be independent of u_g and u_l; therefore, if all the energy dissipated in the liquid contributed to mass transfer, k_{LB} should be independent of u_g. This is contrary to the observed behavior, and it seems that with an increase in u_g, more of the energy dissipated in the liquid is converted into small-scale eddies resulting in an increase in k_{LB}. Secondly, it is known that large bubbles are followed by a liquid wake and it has been reported by several authors (Valentine, 1967; Calderbank and Lochiel, 1964) that the existence of a liquid wake behind a large bubble reduces the mass transfer coefficient due to a reduction in the surface renewal rate.

In the absence of a model that would account for the distribution of the energy dissipating eddies and for the effect of liquid wakes on the mass transfer coefficient in the bed of 1.98-mm particles, an empirical correlation was obtained as:

$$k_{LB} = (0.8118 \pm 0.187) \times 10^{-3}\ u_g^{(0.4077\pm0.059)} \tag{19}$$

Here again, k_{LB} and u_g are in cm/s; the correlation also includes the standard deviation for the regression coefficient.

Effect of Gas-Phase Resistance

From a comparison of k_{LB} values obtained with pure CO_2 as gas phase with those obtained with a mixture of 25% N_2 and 75% CO_2 (at inlet), the effect of gas-phase resistance could be investigated. The gas-phase resistance cannot significantly affect the values of $k_{LB}\underline{a}$ for oxygen desorption, but it can affect the rate of CO_2 absorption. Then, Eqs. 1 and 4 could not be combined without making allowance for the finite gas-side resistance in Eq. 1.

Since the k_{LB} values obtained with the two different compositions of gas phase for the case of 5.86-mm particles are identical (Figure 11) and as the k_{LB} values in the other two beds are of same order of magnitude, it can be concluded that the process of gas-liquid mass transfer in three-phase fluidized beds is a liquid film controlled operation and that the gas-phase resistance is unimportant.

ACKNOWLEDGMENTS

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APPENDIX I: CALIBRATION OF THE DISSOLVED OXYGEN PROBE

A brief procedure of probe calibration, developed for the purpose of this study, is described below. A detailed proof of the calibration procedure can be found elsewhere (Dhanuka, 1978).

The calibration was based on the finding that the solution of sodium carbonate, sodium bicarbonate, and sodium hypochlorite can be replaced for calibration purposes by a solution of sodium chloride of equal ionic strength. The advantage of using a sodium chloride solution is that its dissolved oxygen can also be determined analytically by the Winkler method.

On the basis of the above conclusion, the probe was first calibrated with a 2M NaCl solution saturated with air in which the concentration of dissolved oxygen was determined by the Winkler method. Next, the dissolved oxygen readings were obtained by the probe and the Winkler method for four air saturated solution of NaCl of the strengths 1.9M, 1.95M, 2.05M, and 2.1M.

It was found that the ionic strength of the solution affected the probe reading systematically as can be seen from Figure A1, which gives the ratio of the actual oxygen content (determined by Winkler method) to that obtained by the probe against the ionic strength of the solution. The concentration of dissolved oxygen in the Na_2CO_3—$NaHCO_3$—$NaOCl$ solution could now be easily calculated from the reading of the dissolved oxygen probe after applying a correction obtained from Figure A1.

The linearity of the probe response over the oxygen concentration range encountered in the study was confirmed by saturating the 2M NaCl solution with artificial mixtures of 5%-, 10%-, and 15%-oxygen in nitrogen and measuring their oxygen concentration with the probe.

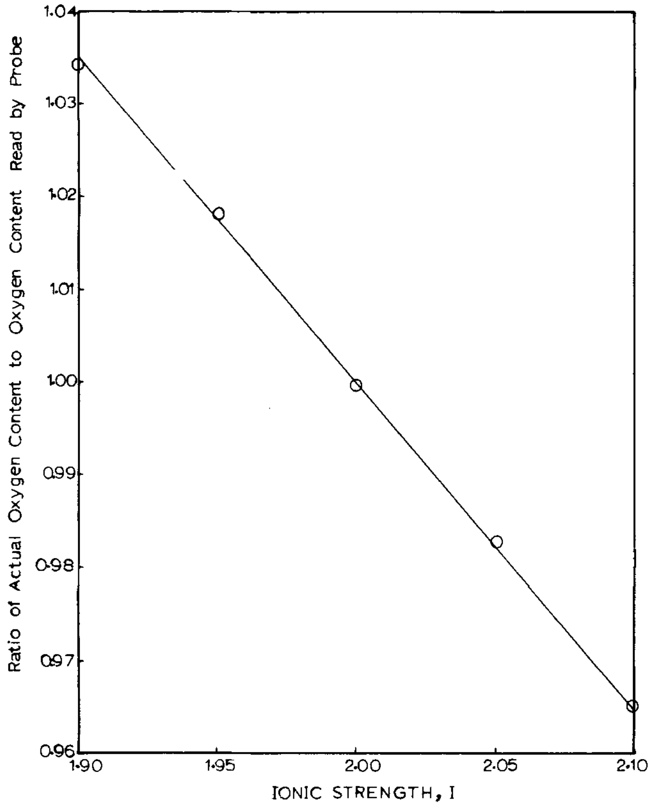

Figure A1. Dependence of the oxygen probe reading on the ionic strength of the solution.

APPENDIX II: KINETICS OF THE FAST REACTION OF CO_2 IN A Na_2CO_3-$NaHCO_3$ BUFFER WITH HYPOCHLORITE AS CATALYST

In a carbonate-bicarbonate buffer solution, dissolved CO_2 reacts with OH^- ions at a local rate of $k_2 C_{CO_2} C_{OH^-}$; the value of $k_2 C_{OH^-}$ has been reported by Roberts and Danckwerts (1962) and is $0.8 s^{-1}$ at 20°C in the buffer solution used in this study. The carbon dioxide also reacts with water at a rate $k_{H_2O} C_{CO_2}$, the value of k_{H_2O} being $0.02 s^{-1}$ at the same temperature. Further, in the presence of hypochlorite ions, there is also a catalysed reaction with water, the rate of which is $k_{OCl^-} C_{CO_2} C_{OCl^-}$. The rates of the three reactions add together to give the total rate of reaction of carbon dioxide:

$$R_{CO_2} = (k_2 C_{OH^-} + k_{H_2O} + k_{OCl^-} C_{OCl^-}) C_{CO_2} \qquad \text{(A1)}$$

The values of k_{OCl^-} varies greatly with the concentration and nature of the various ions in the solution (Sharma, 1964).

The above reaction can be considered to be pseudo-first order if the value of the quantity in parenthesis is the same at all times and in all the regions of the liquid; i.e., there should be no depletion of OCl^- ions near the interface due to formation of hypochlorous acid. A condition for this has been given by Danckwerts and Sharma (1966) and was verified for the present system.

The values of the reaction rate constant k_{OCl^-} were reported by Sharma and Danckwerts (1963), but for a different concentration of hypochlorite than was used in this study. There are also certain doubts regarding the accuracy of the reported results as they do not fit the Arrhenius equation. The values of k_{OCl^-} needed in the present investigation for a 0.01M solution of NaOCl were, therefore, determined in a 100-mm i.d. glass stirred cell operated semi-batchwise for periods of 8-10 minutes depending on the temperature of the solution, which was controlled to within ± 0.1°C. Further details of the system and experimental procedure are available elsewhere (Dhanuka, 1978). The following Arrhenius-type rate expression was fitted to the data:

$$k_{OCl^-} = (1.334 \times 10^{12} \pm 3.25 \times 10^{8}) e^{(50.8 \pm 0.17)/RT} \qquad \text{(A2)}$$

where R is universal gas constant expressed in kJ/mol · K and k_{OCl^-} in l/gmol · s. The correlation also includes the standard deviation of the regression coefficients.

NOTATION

$\underline{a}$ = interfacial area
C = concentration
C^* = saturation concentration
C_0 = reactant concentration in the bulk liquid
ΔC = concentration driving force
D = diffusivity
E = rate of energy dissipation
h = height of bed
k = rate constant
k_1 = pseudofirst order rate constant
k_2 = second order rate constant
k_L = true liquid-phase mass transfer coefficient
$k_L\underline{a}$ = volumetric liquid-phase mass transfer coefficient
ΔP = pressure drop
R = specific rate of absorption
Sc = Schmidt number
T = temperature
u = superficial velocity

Greek Letters

ρ = density
ϵ = bed porosity
ν = kinematic viscosity

Subscripts

A = absorbed species
B = desorbed species
g = gas
l = liquid

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R & D NOTES

The Axial Velocity of a Submerged Axially Symmetrical Fluid Jet

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To mix liquids in very large tanks it is often convenient to use a submerged jet of liquid formed by discharge through a pipe or nozzle. Problems frequently arise in the filling of tanks to mix the inflow with the tank contents. Tanks holding up to 159 000 m^3 (one million barrels) can be blended with inflow if the inflow is properly positioned and of sufficient velocity so that a jet is formed which will entrain surrounding fluid and mix in the turbulent core of the jet.

The axial velocity of such a submerged jet will determine the power required and the rate of blending or entrainment; thus, a knowledge of the velocity of submerged jets as they progress is necessary for the design of such systems. The data herein presented have been used successfully in the design of the filling-mixing systems for tanks ranging in size from 37.85 to 159 000 m^3 (Sarsten, 1972). See Rushton (1954), Fossett and Prosser (1949), Folsom and Ferguson (1949).

A fluid jet whose cross section is circular will entrain surrounding fluid as it flows from and along the axis of the nozzle. The axial velocities of the jet will increase as the jet flows from the nozzle and as its cross section increases owing to entrainment of surrounding fluid. The momentum of the jet will remain constant as it expands by entrainment.

The distance from the nozzle to a position along the axis of flow is designated as x, and the diameter of the nozzle is D_0. The velocity at the jet center line, or axis, is u_x at point x, and u_0 is the average velocity at the nozzle.

A large number of investigations have been made and reported in the literature on the behavior of air and gas jets in air under both isothermal and nonisothermal conditions. Those which report original data for isothermal conditions and for jet lengths between 20 and 100 jet diameters (x/D_0), are listed in the references. Some papers have been published on the flow behavior of liquid jets in liquids (Donald and Singer, 1959; others in the references). Of a number of books on the mechanics of

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