Gas Holdup and Volumetric Mass Transfer Coefficient in a Slurry Bubble Column

By *C. O. Vandu* and *R. Krishna**

The gas holdup, ε, and volumetric mass transfer coefficient, k_La, were measured in a 0.051 m diameter glass column with ethanol as the liquid phase and cobalt catalyst as the solid phase in concentrations of 1.0 and 3.8 vol.-%. The superficial gas velocity U was varied in the range from 0 to 0.11 m/s, spanning both the homogeneous and heterogeneous flow regimes. Experimental results show that increasing catalyst concentration decreases the gas holdup to a significant extent. The volumetric mass transfer coefficient, k_La, closely follows the trend in gas holdup. Above a superficial gas velocity of 0.04 m/s the value of k_La/ε was found to be practically independent of slurry concentration and the gas velocity U; the value of this parameter is found to be about 0.45 s^{-1}. Our studies provide a simple method for the estimation of k_La in industrial-size bubble column slurry reactors.

1 Introduction

Bubble columns are widely used in industry for carrying out a variety of chemical reactions, such as hydrogenations, chlorinations, oxidations and the Fischer Tropsch synthesis [1]. In bubble column slurry reactors catalyst particle sizes smaller than say 100 μm can be used, thus eliminating intraparticle diffusion resistances. These catalyst particles are held in suspension due to the liquid circulations caused by the rising gas bubbles. For successful scale-up of these processes it is necessary to estimate the hydrodynamics and mass transfer performance. There have been several experimental studies reporting the gas holdup, ε, and volumetric mass transfer coefficient, k_La, in slurry bubble columns. Most of these studies have reported experimental data on *either* ε *or* k_La in slurry bubble columns; for literature surveys see Inga and Morsi [2] and Behkish *et al.* [3]. In the present communication we have presented experimental data on *both* ε *and* k_La. Our main objective is to show that for high superficial gas velocities U the volumetric mass transfer per unit volume of dispersed gas, i.e. k_La/ε, has a nearly constant value, irrespective of the slurry concentration and the value of U. This should afford a simple procedure for the estimation of volumetric mass transfer coefficients in slurry bubble columns.

2 Experimental Setup and Procedure

The experiments were carried out in a bubble column of internal diameter of 0.051 m and constructed of glass. A sintered-glass plate, 3 mm thick with an average pore size of 70 μm, was used as the gas distributor. Two rotameters placed in parallel were used to control the rate of air flow into the column, while nitrogen flow was regulated through a manually operated control valve. In order to determine gas holdup, two pressure taps positioned at 0.03 m and 1.29 m from the base of the column were used. These taps were connected to pressure ports on a Validyne DP15 pressure transducer, which was in turn coupled to a PC via an analogue-to-digital converter consisting of the pressure transducer and a voltmeter. The experimental setup is shown in Fig. 1. A manometer tube placed between the lower pressure tap and the pressure transducer was used in calibrating the system to obtain the pressure reading which corresponded to the clear liquid height in the column.

Air was used as the gas phase (density, ρ_G = 1.3 kg m^{-3}; viscosity, $\mu_G = 1.7 \times 10^{-5}$ Pa s), ethanol as the liquid phase (density, ρ_L = 798 kg m^{-3}; viscosity, μ_L = 0.00127 Pa s; surface tension, σ = 0.023 N m^{-1}) into which passivated Raney cobalt catalyst particles (bulk density = 1177 kg m^{-3}; skeletal density = 4167 kg m^{-3}; mean diameter = 25.4 μm; particle size distribution: 10 % < 9 μm; 50 % < 28 μm; 90 % < 61μm) are suspended into the liquid phase in varying concentrations. Experiments were carried out with pure ethanol, volume fraction catalyst $\varepsilon_s = 0.01$ (= 3.6 wt.-% catalyst), and $\varepsilon_s = 0.038$ (= 13.4 wt.-%) catalyst. Note that the solids concentration is defined throughout this paper as the volume fraction, ε_s, of solids in the gas-free slurry. Also, the pore volume of the

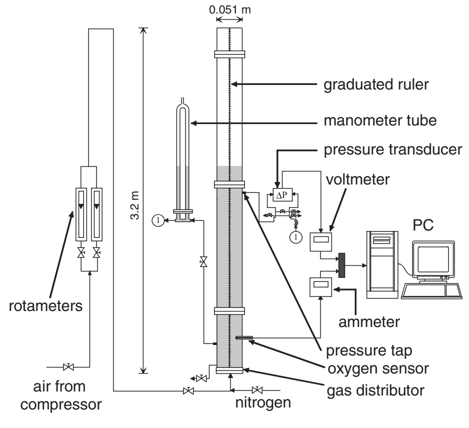

Figure 1. Typical experimental setup for the 0.051 m diameter column.

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particles, which is liquid filled during the experiments, is counted as being part of the solid phase.

In order to determine the gas holdup in each experiment, the gas flow rate into the column was adjusted using one of the two rotameters. Both rotameters were precalibrated to ensure that their float positions accurately corresponded to specified flow rates. With sufficient time given for steady state to be achieved, the increase in liquid pressure at the higher pressure tap was recorded. This increase occurred as a result of the upward displacement of the liquid due to the presence of gas bubbles in the column. The measured pressure signals obtained in the form of voltage readings were then interpreted to obtain information on the gas holdups.

For the determination of the volumetric mass transfer coefficient, k_La, an oxygen sensor probe (*Yellow Springs Incorporated* Model 5331) inserted into the column close to its base was used. Sensitivity of the probe to the presence of oxygen was ensured by the application of a 0.13 g/mL KCl solution between the tip of the probe and an outer membrane separating the probe from the contents of the column. Readings on the probe were fed to an analog-to-digital converter card located in the PC via an ammeter. The oxygen electrode is placed in the bubble column at a distance of 0.1 m above the gas distributor (see Fig. 1).

To determine the volumetric mass transfer coefficient, the liquid phase in the column was first stripped with nitrogen until its dissolved oxygen concentration became negligible. The nitrogen flow was then shut and after all bubbles had escaped, a step input of air was introduced into the column. When all the oxygen has been stripped out, air was sparged into the column and the oxygen uptake into the liquid phase was monitored continuously by means of the oxygen sensor. The total measurement time was chosen large enough in order to reach the oxygen saturation concentration, C^*.

Assuming the liquid phase to be perfectly mixed, the dissolved-oxygen concentration C is described by the relation (see our earlier work [4] for details of the derivation):

$$\frac{C}{C*} = 1 - \frac{1}{k_{sensor} - k_L a_L}\left[k_{sensor} e^{-k_L a_L t} - k_L a_L \, e^{-k_{sensor} t}\right] \quad (1)$$

where k_La_L is the volumetric mass transfer coefficient per unit volume of *liquid* in the bubble column and k_{sensor} is the time constant of the oxygen sensor.

In order to determine the oxygen sensor constant, k_{sensor}, experiments were performed in well-mixed ethanol/slurry systems in two separate glass beakers, one saturated with nitrogen (by continuously bubbling oxygen through it) and the other saturated with oxygen (by continuously bubbling air through it). In these experiments the sensor was first placed in the nitrogen-saturated system till it registered a negligible oxygen concentration. It was then quickly transferred to the oxygen-saturated system and the transient increase in oxygen concentration recorded. The sensor constant k_{sensor} was then determined by fitting the response to the relation:

$$\frac{C}{C*} = 1 - e^{-k_{sensor} t} \quad (2)$$

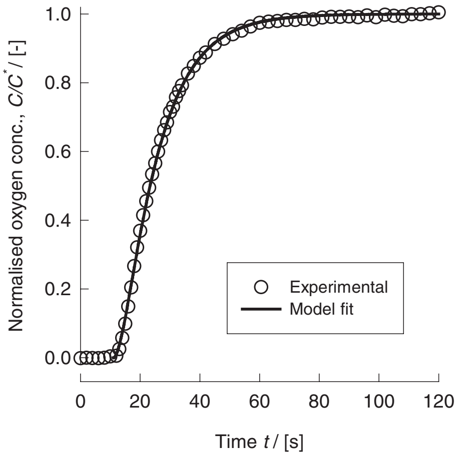

Figure 2. Typical oxygen sensor response, along with model fit (using Eq. (1)). Operation at $U = 0.054$ m/s. The slurry concentration $\varepsilon_s = 0.038$.

The sensor constant k_{sensor} did not have a fixed value but varied depending on the composition of the liquid medium into which the sensor was placed (this was set to the same composition as the contents of the bubble column for each experiment) as well as the amount of KCl present between the tip of the sensor and the membrane. Three values of the sensor constant were obtained for each set of experiments that were performed. The mean values of k_{sensor} were 0.25, 0.39 and 0.27 s^{-1} for pure ethanol, volume fraction catalyst $\varepsilon_s = 0.01$, and $\varepsilon_s = 0.038$, respectively.

The commonly used volumetric mass transfer coefficient per unit volume of (gas + liquid) dispersion, k_La, is then calculated from

$$k_L a = k_L a_L \varepsilon_L \quad (3)$$

where ε_L is the liquid holdup.

A sample experimental oxygen response is given in Fig. 2 along with the fit obtained by the use of Eq. (1). This response is for the experiment performed with 3.8 vol.-% slurry at a superficial gas velocity $U = 0.054$ m/s. Steady state was reached after about 90 seconds, and the predicted response by the use of Eq. (1) correlates very well with the experimental results. Similar responses and fits at other gas velocities were used in obtaining values of the volumetric mass transfer coefficients.

3 Experimental Results and Discussion

Fig. 3 shows the effect of slurry composition on gas holdup with varying superficial gas velocities U. These velocities cover both the homogeneous and heterogeneous flow regimes. It is observed that an increase in the volume fraction of solids in the slurry, ε_s, decreases the gas holdup. These data are in line with our previous measurements with paraffin oil/silica catalyst experiments [5]. Also noteworthy is the fact that the sharp maximum in the holdup curve for pure ethanol vanishes with increasing slurry concentrations. This sharp maximum denotes a shift in the regime from homogeneous bubbly flow to churn-turbulent flow. With the addition of catalyst particles, the coalescence of small bubbles is promoted and the dispersion consists predominantly of larger sized bubbles.

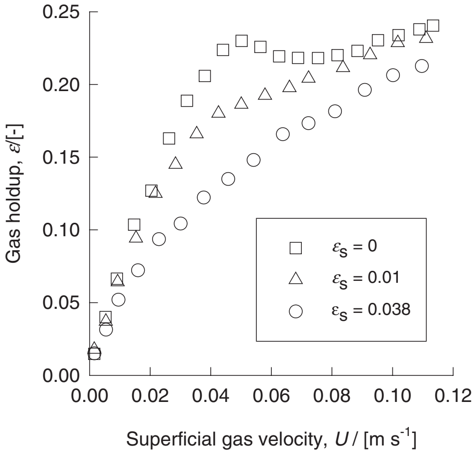

Figure 3. Gas holdup ε as a function of superficial gas velocity U for pure ethanol (ε_s = 0) and slurry concentrations ε_s = 0.01 and 0.038.

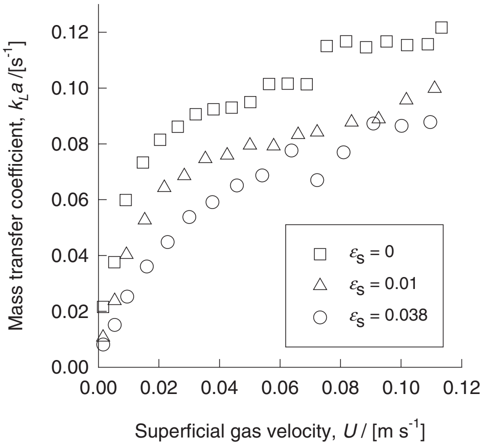

Figure 4. Variation of volumetric mass transfer coefficient k_La with superficial gas velocity U for pure ethanol (ε_s = 0) and slurry concentrations ε_s = 0.01 and 0.038.

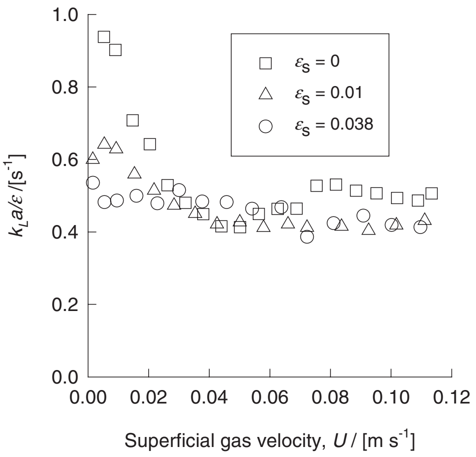

Figure 5. Variation of k_La /ε with superficial gas velocity U for pure ethanol (ε_s = 0) and slurry concentrations ε_s = 0.01 and 0.038.

The effect of solids concentration on k_La is shown in Fig. 4. The trend of k_La vs. U is similar to the trend observed for the gas holdup; increased solids concentration leads to a lowering in the value of k_La. In Fig. 5 we plot k_La /ε as a function of U. We note that above a superficial gas velocity U = 0.04 m/s the value of k_La /ε is practically constant and has a value of around 0.45. The physical significance of the parameter k_La /ε is that it represents the volumetric mass transfer coefficient per unit volume of gas bubbles; the constancy of this parameter implies that the effective bubble diameter d_b is independent of the gas velocity. Vermeer and Krishna [6] were the first to point out the constancy of k_La /ε in the heterogeneous flow regime. For the homogeneous regime of operation, this conclusion is not unexpected because there is negligible coalescence and breakup of bubbles. However, the constancy of the effective bubble diameter d_b in the heterogeneous flow regime can be rationalized only if we take account of the fact that frequent coalescence and breakup of the "large" bubbles causes the effective bubble diameter to be maintained at a low, constant, value; this conclusion was reached earlier by De Swart *et al.* [7] on the basis of video imaging experiments in a 2-D slurry bubble column.

4 Conclusions

We have measured the gas holdup, ε, and volumetric mass transfer coefficient, k_La, for the air-ethanol slurry systems with three different slurry concentrations, ε_s = 0, 0.01 and 0.038. Both ε and k_La show similar trends with varying U and ε_s. Increasing solids concentration tends to decrease both ε and k_La. This decrease is due to the increased coalescence of small bubbles to form larger bubbles. An interesting, and important, finding of this work is that the value of k_La/ε was found to be practically independent of column slurry concentration *and* superficial gas velocity U for operation above 0.04 m/s. The findings of this work provide a useful scale-up rule for estimating the mass transfer coefficients at high gas velocities and slurry concentrations.

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Symbols used

a_L	[m^{-1}]	gas-liquid interfacial area per unit liquid volume
a	[m^{-1}]	gas-liquid interfacial area per unit (gas + liquid) dispersion volume
d_b	[m]	diameter of bubble
C	[–]	oxygen concentration in the liquid phase, arbitrary units
C^*	[–]	saturation concentration of oxygen in liquid, arbitrary units
D_T	[m]	column diameter

k_L	[m s^{-1}]	liquid-side mass transfer coefficient
k_{sensor}	[s^{-1}]	sensor response constant
t	[s]	time
U	[m s^{-1}]	superficial gas velocity

Greek symbols

ε	[–]	gas holdup, dimensionless
ε_L	[–]	liquid holdup, dimensionless
ε_s	[–]	volume fraction of catalyst in slurry, dimensionless
μ	[Pa s]	viscosity of fluid phase
ρ	[kg m^{-3}]	density of phase
σ	[N m^{-1}]	surface tension of liquid phase

Subscripts

s	referring to solid catalyst
L	referring to liquid
T	tower or column

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