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Influence of scale on the hydrodynamics of bubble column reactors: an experimental study in columns of 0.1, 0.4 and 1 m diameters

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Abstract

Measurements of liquid velocities and liquid mixing have been performed in three different bubble columns of 0.15, 0.4 and 1 m diameter with water and air, at superficial gas velocity ranging from 0.05 to 0.20 m/s with high aspect ratios ($H_D/D > 4$). Liquid velocities are determined with a Pavlov tube calibrated up to 25% of gas holdup. Axial dispersion coefficient is determined using a new method which allows accounting for the upflow and downflow regions. The experimental results allow selecting reliable correlations of the literature. The Ueyama and Miyauchi model is successfully compared with the experiments and a new correlation for the kinematic viscosity is proposed.

Keywords: Hydrodynamics; Bubble column; Scale up; Pavlov tube; Axial dispersion coefficient; Modelling

1. Introduction

There is considerable interest, both within academia and industry, on the hydrodynamics of bubble column reactors. This interest stems from applications in emerging technologies for conversion of natural gas to liquid fuels (Krishna & Sie, 2000). The overall objective of our study is to investigate scaling up of bubble column reactors to commercial scale which could reach 10 m in diameter and 40 m in height.

Published studies on bubble column hydrodynamics have often been restricted to rather small columns ($D < 0.2$ m). We have therefore undertaken a comprehensive study of the hydrodynamics (measurements of gas holdup, liquid velocity profile, axial dispersion) using three different columns with diameters of 0.15, 0.4 and 1 m with air and water. The study focuses on the impact of the column diameter on hydrodynamics. Due to the complex nature of the flow, careful development and validation of the measurement techniques were undertaken to determine liquid velocities and axial dispersion coefficients in the three different columns, using identical experimental techniques.

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2. Experimental

2.1. Liquid velocity measurement: Pavlov tube

The liquid velocities were measured using a modified Pavlov tube based on the work of Hills (1974). This technique was chosen because the tube is easy to build and the method is easily adaptable to different liquids and to different column sizes. This Pavlov tube is made of a horizontal stainless tube divided by a wall into two independent compartments. An upward facing hole (0.5 mm) is drilled on one side of the wall, and a downward facing hole is drilled on the other side. Both compartments are connected to a differential pressure sensor. The differential pressure is sampled every 1/50 s and a mean axial liquid velocity is deduced from the kinetic energy balance:

$$V_L = \left(\sum_{i=1}^{N_1} \sqrt{\frac{2\,\Delta P_i}{\rho_L}} - \sum_{i=N_1+1}^{N} \sqrt{\frac{2(-\Delta P_i)}{\rho_L}} \right) \Big/ N, \qquad (1)$$

where N is the number of data points and N_1 the number for which ΔP_i is positive. In high gas velocity bubble columns, it is impossible to use conventional velocity measurement techniques such as LDV and Hot Film anemometry because of high gas holdups. Even with a Pavlov tube, preliminary testing is required in order to ensure the reliability of the

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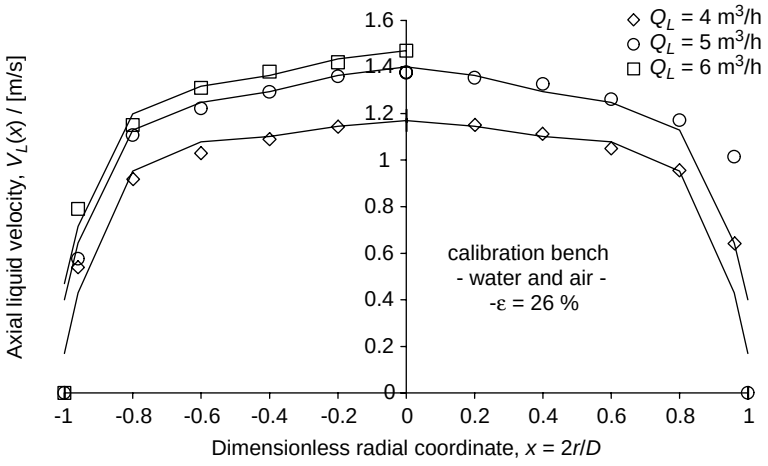

Fig. 1. Radial profiles of axial liquid velocity.

measurement technique in these conditions. The Pavlov tube was thus calibrated in a 50 mm vertical column fed with water and air at known flowrates. The radial velocity profile was obtained by moving the tube along a column diameter. The Pavlov tube was first tested in a turbulent water flow with a known flow structure to determine the probe coefficient f_{corr}. It was then tested in a controlled gas liquid flow to ensure the consistency of measured liquid velocities at high gas holdups.

2.1.1. Calibration with water only

Under turbulent flow conditions in a pipe, the centre-line liquid velocity $V_L(0)$ is related to the cross-section average velocity $\bar{V}_L$ by

$$V_L(0) = \bar{V}_L(1 + 1.33\sqrt{f}) \quad \text{with } f = 0.316\,Re^{-1/4}. \tag{2}$$

Within the range of studied liquid velocity ($0.5 < \bar{V}_L < 1$ m/s), this leads to

$$1.19 < \frac{\bar{V}_L}{V_L(0)} < 1.21 \Rightarrow \bar{V}_L = \frac{Q_L}{\pi D^2/4} \approx \frac{V_L(0)}{1.2}. \tag{3}$$

This means that the centre-line liquid velocity $V_L(0)$ is sufficient to determine the mean velocity $\bar{V}_L$ within 2%. Knowing the volumetric flowrate Q_L, we obtained $\bar{V}_L = f_{\text{corr}}\frac{V_L(0)}{1.2}$ with $f_{\text{corr}} = 0.9$. This correction factor accounts mainly for the non-ideality of drilling the diametrically opposed holes in the Pavlov tube, rather than due to an incorrect alignment and orientation of the Pavlov tube. It was then assumed that the real velocity of the fluid was $f_{\text{corr}}V_L(r)$ at any location r.

2.1.2. Calibration in water and air

The calibration was conducted in controlled gas liquid flow conditions through a vertical pipe. A mass balance over the column cross-section yields

$$Q_L = \int_0^{D/2} 2\pi r[1 - \varepsilon(r)][f_{\text{corr}}V_L(r)]\,\mathrm{d}r. \tag{4}$$

The radial liquid velocity profile $V_L(r)$ was measured for different liquid and gas flowrates, and fitted by a polynomial (continuous lines, Fig. 1). The local gas holdup $\varepsilon(r)$ was either set to the average gas holdup $\bar{\varepsilon}$ (deduced from the pressure drop) or given by the empirical correlation proposed by Schweitzer, Bayle, and Gauthier (2001)

$$\varepsilon(x) = \bar{\varepsilon}[-1.638(x^6 - 1) + 1.228(x^4 - 1) - 0.939(x^2 - 1)], \quad x = 2r/D. \tag{5}$$

In both cases the average velocity $\bar{V}_L$ was in agreement with that deduced from the liquid flowrate within 10% up to $\bar{\varepsilon} = 0.25$.

2.2. Liquid backmixing measurement: tracer

Liquid backmixing was determined using a solution of potassium nitrate. The solution was injected into the batch liquid phase from above. Backmixing was studied by local conductivity measurements. To avoid problems of in-situ conductivity measurement due to air bubbles, liquid samples were withdrawn every second.

In the churn-turbulent regime (high gas velocity), the liquid flows upwards in the core region and downwards in the wall region. Fig. 2(a) shows typical local tracer concentration signals obtained from the upflow and downflow regions in a 1 m i.d. column. Since the curves are different, an appropriate average curve must be defined to obtain a significant axial dispersion coefficient. Therefore, at a given height, two series of samples are withdrawn simultaneously at two radial positions ($x = 0.35$ and 0.85) located respectively in the upflow and downflow zones. Then a cross-section average concentration is defined by:

$$\bar{C} = \frac{(1 - \varepsilon_1)C_1 + (1 - \varepsilon_2)C_2}{2(1 - \bar{\varepsilon})}, \tag{6}$$

where ε_1 and ε_2 (respectively C_1 and C_2) are the gas holdups (respectively concentrations) in the upflow and downflow regions. Flow reversal takes place at $x = 0.7$ (Hills, 1974 and Fig. 5 below). Then ε_1 and ε_2 are obtained upon integration of Eq. (5) from 0 to 0.7 and 0.7 to 1.

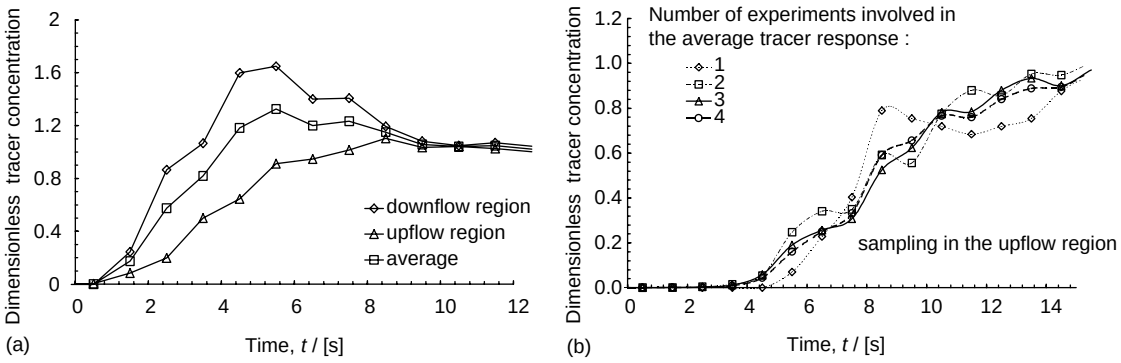

Fig. 2. Reliability of the dual sampling method.

Owing to the fast mixing process, particularly at large scale, three dual samplings are necessary to ensure accuracy and repeatability of measurements (Fig. 2(b)). Finally, it was checked that the average concentration signal was not dependent on the injection method (local or not) and several measurements were done at different heights in the column. The resulting average curves were then interpreted by the axial dispersion model.

3. Results and discussion

The average gas holdup $\bar{\varepsilon}$ is found to be independent of column diameter; see Table 1. This result checked in the 0.15, 0.40 and 1 m columns with air and water is in agreement with the conclusions reached by Joshi et al. (1998). However, the column diameter has a significant effect on liquid recirculations, as we shall demonstrate below.

Measurements of local liquid velocities were conducted at various scale with the Pavlov tube. If radial liquid velocity profiles are normalized by the centre-line liquid velocity, $V_L(r)/V_L(0)$, similar profiles are obtained independent of the column diameter up to 1 m (Fig. 3). Therefore, the knowledge of the centre-line liquid velocity $V_L(0)$ (maximum upward velocity measured along column axis) is enough to describe the whole liquid circulation hydrodynamics. This result is in agreement with Krishna (2000) data and Wu and Al-Dahhan (2001) correlation for $V_L(r)/V_L(0)$. Note however that the latter correlation does not agree well with the experiments close to the column wall (Fig. 3).

Fig. 4 illustrates the sensitivity of the centre-line liquid velocity $V_L(0)$ to column diameter D at $U_g = 0.15$ m/s. $V_L(0)$ increases strongly with D, whatever the gas velocities in the churn-turbulent regime. Comparing with published correlations shows that extrapolation is still risky.

Table 1
Effect of column diameter on average gas holdup ($U_g = 0.15$ m/s)

Column diameter D (m)	0.15	0.40	1
Average gas holdup $\bar{\varepsilon}$	0.240	0.241	0.252

Bernemann (1989) measured the liquid velocity by anemometry and assumed that the gas contribution was negligible. Although his result agrees with our measurements in the 0.4 m column at $U_g = 0.15$ m/s, his liquid velocities for larger flowrates are significantly higher than ours (results not shown). Since the calibration of the Pavlov tube at high gas holdup (high gas velocity) was satisfactory, we believe that the Pavlov tube is more appropriate than an anemometer which can be disturbed by the kinematic energy of the gas bubbles.

Literature provides two sets of correlations predicting centre-line liquid velocities as a function of column diameter (Fig. 4). Our experimental data obtained in column diameters up to 1 m enable to discriminate one set. The lower curves (Miyauchi & Shyu, 1970; Nottenkämper, 1983; Zehner, 1986) seem the best which give a range of velocities at large scale of 1.5–2 m/s for a 5 m diameter column with air-water system. Scale-up trend is therefore assessed with reasonable accuracy using lower curves trends.

However, these curves reflect empirical correlations. It is therefore interesting to see a more physical approach. The model of Ueyama and Miyauchi (1979), based on a radial momentum balance along the column cross section is then used to describe liquid velocity profile $V_L(r)$ for column diameters up to 1 m; see Fig. 5. This model depends on an unknown turbulent kinematic viscosity ν_t. From our experiments in the 0.15, 0.40 and 1 m diameter columns, we obtained the following correlation:

$$\nu_t = 0.036 D^{1.6} U_g^{0.11}. \quad (7)$$

The strong influence of the column diameter D is apparent in the 1.6 power. With this approach, predictions of centre-line liquid velocities at large scale are close to predictions of the previously selected empirical correlations. Consistent tools are therefore available for liquid velocity scale-up.

The axial dispersion coefficient in the bubble column was obtained by analyzing the tracer time responses using the axial dispersion model. For a given superficial gas velocity, the dual-sampling method was used at three axial elevations. The same axial dispersion coefficient is able to reproduce average curves obtained at three height elevations as shown in

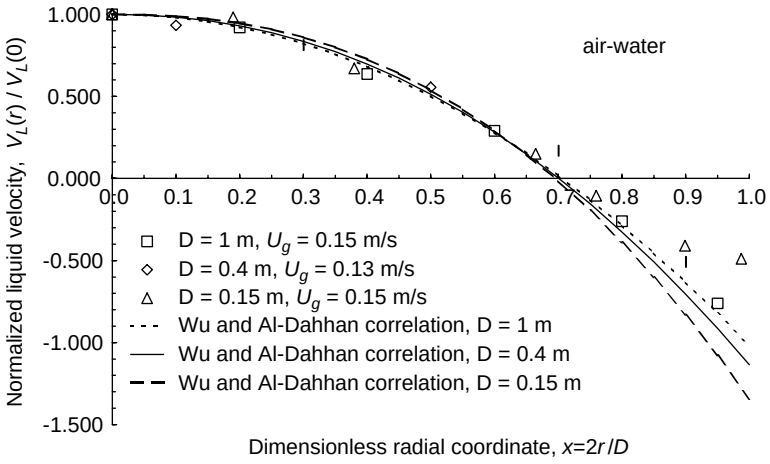

Fig. 3. Normalized liquid velocity profile $V_L(r)/V_L(0)$.

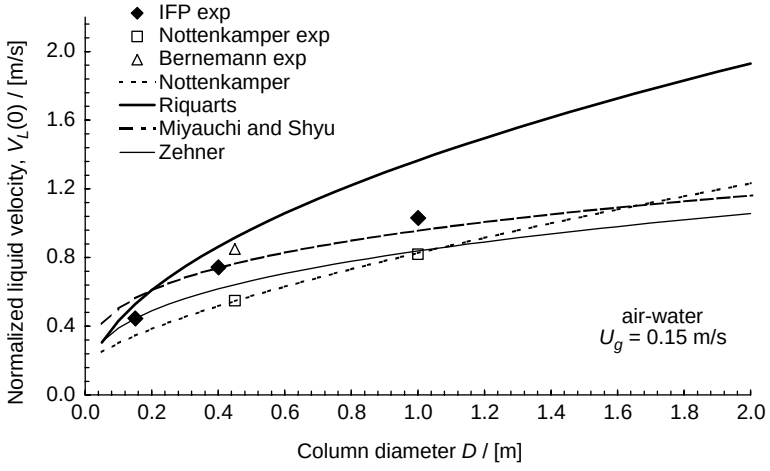

Fig. 4. Effect of column diameter on $V_L(0)$.

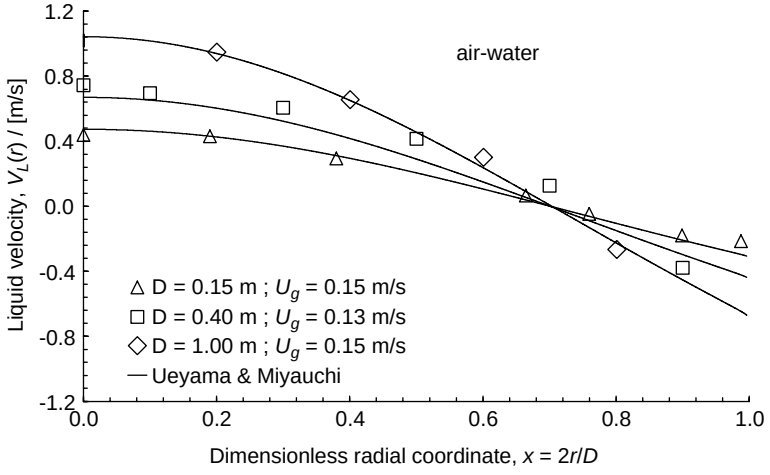

Fig. 5. Validation of the Ueyama and Miyauchi's model.

Fig. 6. This gives support both to the model and to the measurement technique. However, since our technique involves cross-section averaging, it could be hazardous to compare with previously published results. The axial dispersion coefficient is found to depend strongly on column diameter at a given superficial gas velocity (see Fig. 7). This is related to the recirculation and the dependence of the centre-line liquid velocity upon the diameter. Fig. 7 shows that our results

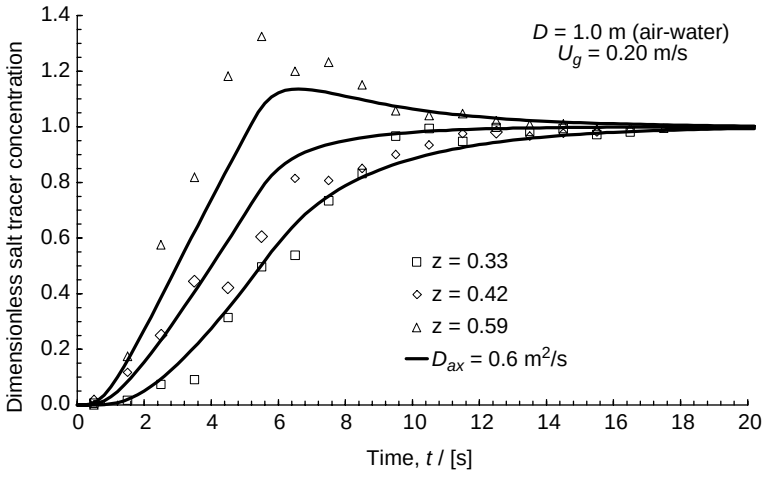

Fig. 6. RTD curves at different heights and validation of the ADM.

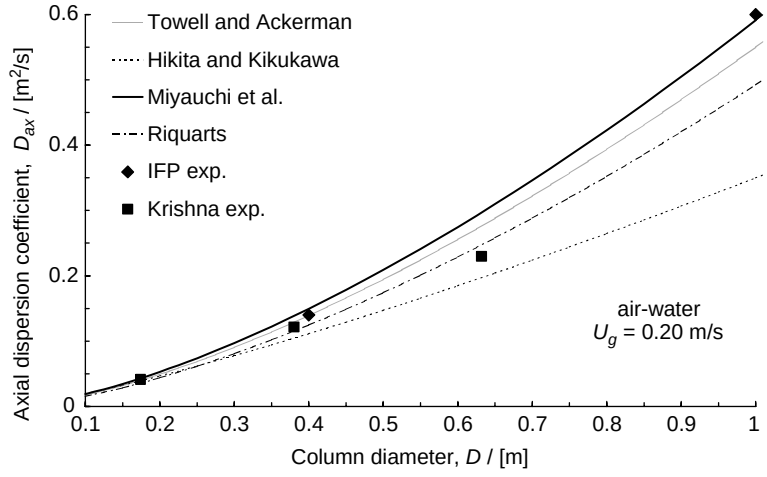

Fig. 7. Effect of column diameter on D_{ax}. Data from Towell and Ackerman (1972), Hikita and Kikukawa (1974) and Miyauchi et al. (1981).

are consistent with literature data (Krishna, Urseanu, Van Baten, & Ellenberger, 1999). Nevertheless, comparing with correlations (Towell & Ackerman, 1972; Miyauchi et al., 1981; Riquarts, 1981) shows that most of them tend to underestimate D_{ax} at large scale (except Miyauchi et al., 1981). This can be explained by the local measurement made by many authors. We consider that local measurements are not appropriate in large diameter columns because radial mixing becomes less efficient when the diameter increases. Finally, to know whether D_{ax} is a relevant parameter, it is necessary to check that a single value represents the tracer curves at different elevations. Otherwise the axial dispersion model is questionable.

4. Conclusion

- A modified Pavlov tube (with two opposite holes) was calibrated in water and air. Up to a gas holdup of 25%, the measured liquid velocity is reliable within 10%.
- A rigorous experimental protocol is needed to determine the axial dispersion coefficient, especially in large columns. The liquid residence time distribution has been measured using the dual sampling method. The latter involves two simultaneous local measurements in the upflow and downflow regions and then cross-sectional averaging of these signals. Up to 1 m in diameter, the axial dispersion model is reliable since a single axial dispersion coefficient may represent three tracer time responses at different elevations.
- Liquid recirculation and mixing have then been measured with confidence in bubble columns up to 1 meter in diameter. Both liquid velocity and axial dispersion coefficient increase strongly with scale. However gas holdup is independent of scale ($D > 0.15$ m). Our results provide reasonable trend for scaling up $V_L(0)$ and D_{ax} in air–water systems.
- Several approaches can be used to scale up successfully liquid recirculation: carefully selected empirical correlations or more physical models.

- The radial profile of axial liquid velocity can be well represented by the model of Ueyama and Miyauchi (1979) provided that the turbulent viscosity is correlated by equation (7). We believe that this type of "more fundamental" model could be better for scale-up provided that the turbulent viscosity is well characterized.

Notation

C_1	tracer concentration in the upflow liquid flow region, mol/m^3
C_2	tracer concentration in the downflow liquid flow region, mol/m^3
$\bar{C}$	cross-section average tracer concentration, mol/m^3
D	column diameter, m
D_{ax}	axial dispersion coefficient, m^2/s
f_{corr}	correction factor, dimensionless
H	height of measurement or sampling, m
H_D	dispersion height or aerated height, m
N, N_1	number of data points
Q_L	volumetric liquid flowrate, m^3/s
r	radial coordinate, m
U_g	superficial gas velocity, m/s
$V_L(0)$	centre-line liquid velocity or maximum upward velocity measured along column axis, m/s
V_L	local liquid velocity, m/s
$\bar{V}_L$	cross-section average liquid velocity, m/s
x	dimensionless radial coordinate $x = 2r/D$, dimensionless
z	dimensionless axial coordinate $z = H/H_D$, dimensionless

Greek letters

ΔP_i	instantaneous differential pressure measured by the modified Pavlov tube, Pa
ε	gas holdup, dimensionless
ε_1	gas holdup of the upflow liquid flow region, dimensionless
ε_2	gas holdup of the downflow liquid flow region, dimensionless
$\bar{\varepsilon}$	average gas holdup, dimensionless
$\mathrm{e}\mu_L$	dynamic viscosity of the liquid, Pa s
v_t	turbulent kinematic viscosity, m^2/s
ρ_L	liquid phase density, kg/m^3

Dimensionless number

Re	Reynolds number ($\rho_L \bar{V}_L D/\mu_L$)

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