Liquid Dispersion in Large Diameter Bubble Columns, with and without Internals

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There is considerable interest within both academia and industry in the hydrodynamics of bubble column reactors. This interest stems from applications in emerging technologies for conversion of natural gas to liquid fuels (Krishna and Sie, 2000). Because of their efficient liquid mixing, bubble columns are suitable reactors for carrying out highly exothermic reactions using vertical cooling tubes.

Published studies on bubble column hydrodynamics are often restricted to rather small columns (D < 0.2 m) without internals. In small columns without internals, the axial dispersion coefficient $D_{ax,1D}$ was found to be proportional to the product of the time-averaged centre-line liquid velocity $V_L(0)$ times the column diameter D (Krishna, 2000). There are no general guidelines available in the literature for determining the axial dispersion coefficient for large diameter columns with vertical cooling tubes as internals. We have therefore undertaken a comprehensive study of the hydrodynamics (liquid velocity profile, axial dispersion) in a 1 m diameter bubble column with air-water system, with and without internals. In an earlier publication, careful development and validation of the measurement techniques were undertaken to determine liquid velocities and axial dispersion coefficients (Forret et al., 2003). The present study focuses on interpretation and modeling of liquid mixing in bubble columns both with and without vertical internals.

Experimental

Experiments were performed in a 1 m diameter bubble column, with air-water system in batch mode (no liquid flowrate), with and without internals. The gas distributor is a perforated plate (313 of 2 mm diameter holes with a pitch of 50 mm). The internals used in this study have the following characteristics: 56 tubes, each 63 mm in diameter, arranged in a square pitch of 108 mm.

Liquid velocities and fluctuating velocities were determined with an improved Pitot tube, calibrated up to 25 % of gas holdup (Forret et al., 2003). Liquid backmixing was studied using a standard tracer method based on conductivity measurements. The tracer was injected into the batch liquid phase above the dispersion height. Since there is a radial tracer concentration profile in large columns, the axial dispersion coefficient has to be determined with a specific method to account for the upflow

Liquid mixing has been studied in a 1 m diameter bubble column, with and without internals (vertical cooling tubes). The presence of internals significantly affects both large scale recirculation and local dispersion. The most common approach to model liquid mixing is the one-dimensional axial dispersion model, validated many times in small bubble columns without internals. This paper shows that this model is still appropriate to large columns, but without internals. A two-dimensional model, taking into account a radially dependent axial velocity profile, and both axial and radial dispersion, is required to account for the internals on liquid mixing.

Le mélange du liquide dans une colonne à bulles de 1 m de diamètre a été étudié, avec et sans internes (tubes verticaux simulant des échangeurs de chaleur). La présence d'internes affecte de manière significative à la fois la recirculation globale du liquide ainsi que la dispersion locale. L'approche la plus couramment employée pour modéliser le mélange du liquide est le modèle de dispersion axiale mono dimensionnel, validé maintes fois pour les petites colonnes à bulles sans internes. Cet article montre que ce modèle reste valable pour les colonnes de grande taille, sans internes. Par contre, la prise en compte des effets des internes sur le mélange liquide passe par l'utilisation d'un modèle bidimensionnel, prenant en compte le profile radiale de la vitesse axiale ainsi que les dispersions axiale et radiale.

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and downflow regions (Forret et al., 2003). Two series of samples (dual samplings) are withdrawn simultaneously at two radial positions (x = 0.35 and 0.85) located respectively in the upflow and downflow zones. Then a cross-sectional averaged concentration is defined by :

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Figure 1. Comparison between cross-sectional average over samplings at two and four positions in the same axial plane (z = 0.41, $H_D = 4.3$ m).



Figure 2. Cross-sectional averaged tracer time response with and without internals.

$$\overline{C} = \frac{(1 - \varepsilon_1)C_1 + (1 - \varepsilon_2)C_2}{2(1 - \overline{\varepsilon}_g)}$$
(1)

where ε_1 and ε_2 (respectively C_1 and C_2) are the gas holdups (respectively concentrations) in the upflow and downflow regions. We have also verified that we obtained roughly the same average from simultaneous withdrawals at four radial positions (x = 0, 0.3, 0.6 and 0.95) in the same axial plane (see Figure 1). Thus we have access both to local and cross-sectional averaged tracer time responses.

Results and Discussion

Figure 2 shows the experimental tracer concentrations that have been cross-sectionally averaged as a function of time with and without internals for the very same conditions (superficial gas velocity $U_g = 0.15$ m/s; sampling height z = 0.35 with $H_D = 4.3$ m and z = 0.41 with $H_D = 3.7$ m). Clearly, the cross-sectional averaged responses with and without internals are similar at z = 0.35 (open squares and black squares). However, at a different height (z = 0.41 with $H_D = 3.7$ m, closer to the free surface where the tracer is injected corresponding to z = 1), the cross-sectional averaged responses with and without internals are significantly different (open dots and black dots). Clearly, internals significantly affect the liquid mixing pattern inside the bubble column.



Figure 3. Radial profile of the axial liquid fluctuating velocity u_{σ} (r) without and with internals

To complement those findings, measurements of liquid velocity were performed with an improved Pitot tube. In the presence of internals, large scale recirculation is enhanced, and the axial liquid velocity $V_1(\mathbf{r})$ increases in the core of the bed; the radial liquid velocity is negligible. Nevertheless, the normalized liquid velocity profile $V_1(\mathbf{r})/V_1(\mathbf{0})$ is not modified and remains constant : in the churn-turbulent regime (high gas velocity), the liquid flows upwards in the core region and downwards in the wall region. Also, even in the presence of internals, flow reversal still takes place at x = 0.7 (Hills, 1974). Moreover, the presence of internals leads to a decrease of the fluctuating velocity $u_{\sigma}(r)$. Figure 3 shows radial profiles of the axial fluctuating liquid velocity at a given superficial gas velocity of 0.15 m/s in the core region. We observe that the maximum of the fluctuating velocity u_{σ} occurs at the inversion flow (x = 0.7). Degaleesan (1997) has also shown that the maximum axial eddy diffusivity occurs at a point which is close to the inversion point of liquid recirculation velocity, where the maximum turbulent shear stress occurs. This gives support to the fluctuating velocity measured with the Pitot tube.

The decrease of the fluctuating velocity with internals can partially explain the increase of liquid recirculation intensity. This is also in agreement with the conclusion reached by Chen et al. (1999): turbulent stresses and eddy diffusivities are lower in the column with internals, due to the fact that they physically reduce the length scales of turbulence.

The concept of dispersion in bubble columns is rather empirical. It lumps together two distinct physical processes: 1) large scale recirculations, essentially axial, resulting from the upwards and downwards flow regions; 2) turbulence, or fluctuating velocity, that contributes both to radial and axial mixing. According to the operating conditions and the presence of internals, one of these processes may predominate the other. Our results show that internals increase the large scale recirculation contribution and decrease the fluctuations.

Modeling

Axial Dispersion Model (ADM)

The value of the axial dispersion coefficient obtained from experiments depends upon the details for the formulation of the axial dispersion model. The most common approach to model liquid mixing in bubble columns is the one-dimensional dispersed-plug-flow model, namely the axial dispersion model (ADM), defined by :

$$\frac{\partial C}{\partial t} = D_{ax,1D} \frac{1}{H_D^2} \frac{\partial^2 C}{\partial z^2}$$
(2)

The large scale recirculation combined with the dispersion is defined through one coefficient : the one-dimensional axial dispersion coefficient $D_{ax,1D}$. This model is a simplified representation of physical phenomena which occur in a bubble column. However, it is a reasonable way to characterize the mixing of a continuous phase with only one coefficient.

Cross-sectional averaged tracer time responses are compared to the one-dimensional ADM simulations. $D_{ax,1D}$ is determined by adjusting its value in order to simulated curves fit experimental tracer time responses. The ADM was shown to be reliable in large columns without internals (Forret et al., 2003), since a single axial dispersion coefficient $D_{ax,1D}$ can represent cross-sectional averaged tracer time responses at different heights *z*. Continuous lines in represent ADM simulations which are in agreement with the experiments (clear squares and dots) obtained without internals at two elevation heights.

The filled symbols in have been obtained in the presence of internals at the same operating conditions as that for the opened symbols, obtained without internals. Whatever the value of $D_{ax,1D}$, the ADM can not predict a maximum as a function of time (too far from the free surface where the tracer is injected). Nevertheless we observe experimentally a peak at z = 0.41 and H_D = 3.7 m: filled circles in Figure 2. The ADM is therefore not appropriate to model liquid mixing in large bubble columns with internals. In fact, liquid mixing is a resultant of global convective recirculation of the liquid phase (induced by the non-uniform gas radial holdup distribution), as well as the turbulent diffusion, due to the eddies generated by the rising bubbles (Degaleesan, 1997). Since the large scale recirculation contribution to liquid backmixing is enhanced in the presence of internals, the ratio convection-dispersion is modified, and the one-dimensional ADM, validated without internals, is not appropriate anymore to large bubble columns with internals.

2D Model

We therefore attempted a more detailed model to better describe experimental trends. A two-dimensional (2D) model was developed to account for the radial dependence of the axial velocity (large scale recirculation), axial dispersion ($D_{ax,2D}$) and radial dispersion ($D_{rad,2D}$); see Figure 4. One of the main differences between the 1D ADM and the 2D model is that the axial convective recirculation and the dispersion contributions to liquid mixing are separated in the 2D model. The 2D model equations are :

$$\frac{\partial C}{\partial t} = D_{ax,2D} \cdot \frac{1}{H_D^2} \cdot \frac{\partial^2 C}{\partial z^2} - V_L \cdot \frac{1}{H_D} \cdot \frac{\partial C}{\partial z} + \frac{D_{rad,2D}}{(1 - \varepsilon_g)}$$

$$\cdot \frac{\partial}{\partial r} \left(\left(1 - \varepsilon_g\right) \cdot \frac{\partial C}{\partial r} \right) + \frac{D_{rad,2D}}{r} \cdot \frac{\partial C}{\partial r}$$
(3)

with
$$V_L(x) = \frac{V_L(0)}{1.128} \left[2.976 \cdot \exp(-0.943 \cdot x^2) - 1.848 \right]$$
 (4)



CSTRA and CSTRB : extremely thin

Figure 4. Schematic of the two-dimensional model

and
$$\varepsilon_g(x) = \overline{\varepsilon}_g \begin{bmatrix} -1.638(x^6 - 1) + 1.228(x^4 - 1)) \\ -0.939(x^2 - 1) \end{bmatrix}$$
 (5)

The local liquid velocity correlation (4) was developed at IFP, validated in different bubble columns sizes (D = 0.15, 0.40 and 1 m). It is close to the correlation proposed by Wu and Al-Dahhan (2001). The local gas holdup $\varepsilon_g(x)$ is given by the empirical correlation (5) proposed by Schweitzer et al. (2001), and verified and validated in the 1 m column using an optical probe. The average gas holdup and centre-line liquid velocity values are referred to our own experiments.

The 2D model simulations were first compared to the measurements without internals: cross-sectional averaged tracer time responses are used here. From the experimental set of data at $U_g = 0.15$ m/s, the best model adjustment was obtained with $D_{ax,2D}/D_{rad,2D} = 20$. We can note that, at this given superficial gas velocity, the cross-sectional averaged tracer time responses simulated by the model 2D are equivalent to responses simulated by the ADM with $D_{ax,1D} = 0.5$ m²/s (see Figure 5(a): cross-sectional averaged tracer time responses). Moreover, Figure 5(b) shows that the corresponding local tracer time responses (in the upflow and in the downflow region) are in reasonably agreement with the 2D model local simulations.

Figure 6 represents snapshots simulated by the 2D model each second after the injection of the tracer in those conditions $(D = 1 \text{ m}, U_g = 0.15 \text{ m/s}; H_D = 3.6 \text{ m}; D_{ax,2D}/D_{rad,2D} = 20)$. As expected, the tracer flows first downwards along the column wall while it diffuses towards the centre-line; at about t = 7 s, it spreads over the whole cross-section because it enters the inner upward flow. Note that the color at the outer sampling point (x = 0.8) is at most yellow, which means that there is no concentration overshoot.



Figure 5. (a) Cross-sectional averaged and (b) local tracer time responses without internals.



+: sampling point

Figure 6. Snapshots of tracer concentration simulated by the 2D model for D = 1 m without internals ($U_g = 0.15$ m/s; $H_D = 3.6$ m; $D_{ax2,D}/D_{rad,2D} = 20$).

The adjustment of model prediction with experimental data obtained at $U_g = 0.15$ m/s with internals lead to the following model parameters (Figure 7(a) cross-sectional averaged; Figure 7(b) local - tracer time responses): $D_{ax,2D}/D_{rad,2D} = 400$. The best fit obtained with internals was found with a significantly

lower $D_{rad,2D}$ than without internals. This trend is consistent with the decrease of fluctuating velocity observed experimentally; see Figure 3. Contrary to what was observed with the ADM, the 2D model is able to reproduce the transient concentration overshoot both for the cross-sectional average and local



Figure 7. (a) Cross-sectional averaged and (b) local tracer time response with internals.



+ : sampling point

Figure 8. Snapshots of tracer concentration simulated by the 2D model for D = 1 m with internals ($U_g = 0.15$ m/s; $H_D = 3.7$ m; $D_{ax2,D}/D_{rad,2D} = 400$).

concentrations observed at z = 0.41 with $H_D = 3.7$ m. The model parameters $D_{ax,2D}$ and $D_{rad,2D}$ should not only be linked to large scale recirculation (time-averaged liquid velocity) but also to fluctuating velocity. However, the 2D model predicts also a maximum of tracer concentration at z = 0.35 with

 H_D = 4.3 m, maximum not observed experimentally (Figure 2). This shows the limits of the 2D model, partially explained by the difficulty to model the surface free region at the top of the column and the distributor region where flow pattern is quite complex. In the present model, as shown in Figure 4, extremely



with internals : enhancement of large recirculation scale, decrease of radial dispersion

Figure 9. Schematic representation of liquid recirculation in bubble columns without (a) and with internals (b).

thin, perfectly mixed reactors are modeled at the top and the bottom of the column. In other words, we consider in these regions infinite mixing (both axial and radial) which constitutes a strong assumption. Regardless, this model contributes to the understanding of effects generated by the presence of internals on liquid dispersion.

Figure 8 represents snapshots simulated by the 2D model each second after the injection of the tracer in those conditions $(D = 1 \text{ m}, U_q = 0.15 \text{ m/s}; H_D = 3.7 \text{ m}; D_{ax,2D}/D_{rad,2D} = 400).$ Compared to Figure 6, we observe that with internals the tracer flows down more rapidly, and remains stuck on the wall (lower radial dispersion). Then, it enters the upward flow region earlier than without internals. At t = 8 s, a significant radial concentration gradient is still present contrarily to Figure 6. The color at both the outer and inner sampling points reaches the red level before leveling off at the yellow level. This is reflected by the concentration overshoot observed in Figure 7(b). Finally, the delay between the concentration maxima reflect the large recirculation time. The recirculation of tracer is more pronounced with internals, due to enhancement of the convective contribution and decrease of the dispersion effect (decrease of fluctuating velocities). Figure 9 gives a schematic of liquid recirculation in bubble columns without and with internals. The main effect of internals is to reduce radial dispersion and to increase large scale recirculation.

Conclusion

- The large scale recirculation contribution to liquid backmixing is significantly enhanced in the presence of internals (increase of liquid recirculation intensity and decrease of fluctuating velocity).
- The time-averaged liquid axial velocity profile and the fluctuating axial velocity depend on the presence of internals. The axial velocity profile normalized by the centre-line velocity $V_L(0)$ seems to be independent of the internals. $V_L(0)$ is significantly increased by the internals.
- The standard one-dimensional axial dispersion model is still appropriate to describe mixing in large bubble columns without internals, even though there are radial tracer concentration gradients (agreement of the experimental cross-sectional averaged tracer time responses with ADM simulations).
- Since large scale recirculations dominate in the presence of internals, the ADM validated without internals is inappropriate to bubble columns with internals.

- A two-dimensional model is required for large bubble columns with internals. The effect of internals on time-averaged liquid velocity, as well as on liquid fluctuating velocity should be taken into account to simulate liquid mixing in the presence of internals.
- This paper shows that a two-dimensional model, taking into account a radially dependent axial velocity profile, an axial dispersion coefficient $D_{ax,2D}$, and a radial dispersion coefficient $D_{rad,2D}$, is able to reproduce effects of internals on liquid mixing. By reducing $D_{rad,2D}$, trends observed on liquid mixing with internals are simulated by the 2D model: the tracer flows more rapidly and less dispersed in loopings that become more pronounced in the presence of internals. The presence of internals has a strong influence on the ratio of the axial to radial dispersion $D_{ax,2D}/D_{rad,2D}$; see Figures 6 and 8. For a superficial gas velocity $U_g = 0.15$ m/s, this ratio is 20 without internals and increases 20-fold to 400 in the presence of internals.

Nomenclature

- ADM axial dispersion model
- C local tracer concentration, (mol/m³)
- C₁ tracer concentration in the upflow liquid
- flow region, (mol/m³)
- C_2 tracer concentration in the downflow liquid flow region, (mol/m³)
- C cross-sectional averaged tracer
- concentration, (mol/m^3)
- D column diameter, (m)
- $D_{ax,1D}$ axial dispersion coefficient determined by the onedimensional ADM, (m²/s)
- $D_{ax,2D}$ axial dispersion coefficient determined by the twodimensional model, (m²/s)
- $D_{rad,2D}$ radial dispersion coefficient determined by the two-dimensional model, (m²/s)
- *H* height of measurement or sampling, (m)
- H_D dispersion height or aerated height, (m)
- N number of acquisition data points
- r radial coordinate, (m)
- *u_i* instantaneous liquid velocity, (m/s)

$$u_{\sigma}(r) = \sqrt{\frac{\sum_{i=1}^{N} (u_i(r) - V_L(r))^2}{N}}$$
 fluctuating velocity, (m/s)

U_a superficial gas velocity, (m/s)

$$V_L(r) = \frac{\sum_{i=1}^{N} u_i(r)}{N}$$
 local axial liquid velocity, (m/s)

V_L(0) centre-line liquid velocity or maximum upward velocity measured along column axis, (m/s)

- x dimensionless radial coordinate x = 2r/D
- z dimensionless axial coordinate $z = H/H_D$

Greek Symbols

- ϵ_g local gas holdup
- $\overline{\varepsilon}_{g}$ average gas holdup
- ϵ_1^{j} gas holdup of the upflow liquid flow region
- ϵ_2 gas holdup of the downflow liquid flow region

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