ADVANCED DIAGNOSTIC TECHNIQUES FOR THREE-PHASE SLURRY BUBBLE COLUMN REACTORS (SBCR)

Annual Technical Progress Report No. 2 for the Period July 1, 2000 – June 30, 2001 DE-FG-26-99FT40594

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ABSTRACT

This report summarizes the accomplishment made during the second year of this cooperative research effort between Washington University, Ohio State University and Air Products and Chemicals. The technical difficulties that were encountered in implementing Computer Automated Radioactive Particle Tracking (CARPT) in high pressure SBCR have been successfully resolved. New strategies for data acquisition and calibration procedure have been implemented. These have been performed as a part of other projects supported by Industrial Consortium and DOE via contract #DE-2295PC95051 which are executed in parallel with this grant. CARPT and Computed Tomography (CT) experiments have been performed using airwater-glass beads in 6" high pressure stainless steel slurry bubble column reactor at selected conditions. Data processing of this work is in progress. The overall gas holdup and the hydrodynamic parameters are measured by Laser Doppler Anemometry (LDA) in 2" slurry bubble column using Norpar 15 that mimic at room temperature the Fischer Tropsch wax at FT reaction conditions of high pressure and temperature. To improve the design and scale-up of bubble column, new correlations have been developed to predict the radial gas holdup and the time averaged axial liquid recirculation velocity profiles in bubble columns.

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EXECUTIVE SUMMARY

The objective of this cooperative research effort between Washington University, Ohio State University and Air Products and Chemicals is to advance the understanding of the hydrodynamics of Fischer-Tropsch (FT) Slurry Bubble Column Reactors (SBCR) via advanced diagnostics techniques. The emphasis during this second year was: i) to implement the achieved solutions to the technical difficulties encountered during implementation of CARPT in high pressure SBCR and also to implement new achieved strategy, data acquisition and calibration procedure, ii) to perform CARPT and CT experiments using air-water-glass beads in 6" high pressure SBCR, iii) to perform LDA and overall gas holdup measurements in 2" slurry bubble column using Norpar 15 which mimics at room temperature FT wax at FT reaction conditions of high pressure and temperature, iv) to develop new correlations to predict the radial gas holdup and the time averaged axial liquid recirculation velocity profiles in bubble columns for improved design and scale up of bubble column reactors.

This report summarizes the accomplishments made during the second year of this project. The report is organized in individual sections. Each section represents a distinct task.

Section 1 provides an introduction and motivation for this collaborative project.

Section 2 provides a review of the objectives and tasks set for the project, list of accomplishments during the first and second year and plan for the third year.

Section 3 describes the experimental facilities and the advanced techniques like LDA, CARPT, CT used to study hydrodynamics of high pressure slurry bubble columns.

Section 4 discusses the results of the performed experiments at Ohio State University and Washington University. Also it outlines the technical difficulties encountered in implementing CARPT in high pressure stainless steel SBCR and their resolutions.

Section 5 and Appendix provides details for the development of the new correlations for radial gas holdup and axial liquid recirculation velocity profiles.

1. INTRODUCTION AND MOTIVATION

Synthesis gas (mixture of carbon monoxide and hydrogen) from coal is one of the most abundant and reliable sources of energy and chemicals. Fischer-Tropsch (FT) Chemistry is an acknowledged route for clean utilization of coal/natural gas-derived synthesis gas in production of fuels and chemicals. Based on reaction engineering considerations and economics (i.e., from the heat transfer and high volumetric productivity points of view), slurry bubble column reactors (SBCR) operated at high gas velocities in churn turbulent flow regime are the preferred reactors for commercialization of FT synthesis.

A slurry bubble column reactor (SBCR) is a cylindrical vessel in which gas (containing one or more reactants, e.g. synthesis gas for FT processes) is sparged through the liquid (containing liquid reactant(s) and products), and a finely dispersed catalyst. As long as the operating liquid superficial velocity (in the range of 0 to 2 cm/s) is an order of magnitude smaller than the superficial velocity of the gas (1 to 30 cm/s), and the catalyst particles are small (less than 50 μ m) and not excessively heavy, the gas dominates the hydrodynamics and, by buoyancy forces resulting from the non uniform cross-sectional gas holdup distribution induces liquid velocities order of magnitude larger than the liquid superficial velocity. The finely dispersed catalyst follows the motion of the liquid in SBCR.

It is well known that fluid dynamics (phase velocities and holdup distribution) affect the phase mixing and transport (gas-liquid interfacial area, transport coefficients) between the phases, and hence, to a large extent affect the reactor conversion and selectivity. If the reaction involves phase volume changes (e.g. gas is either consumed or produced), the phase mixing, and transport parameters are affected along the column. Thus numerous design and operating variables, listed in Figure 1.1 and physicochemical and thermodynamic properties of the fluid affect the many highly interactive phenomena in SBCR. All of these in turn affect reactor performance.

A slurry bubble column reactor for the FT synthesis and other syngas processes in order to be economically successful, must operate at high volumetric productivity which requires high activity catalyst, high catalyst loading of the slurry, large gas flow rate and high gas conversion. The ability to achieve complete catalyst suspension and the desired flow pattern (degree of backmixing) of the liquid phase are crucial to the targeted reactor performance. In order to accomplish these an improved understanding and quantification of the key hydrodynamic phenomena is required. However, reliable data and tested models or theories for design and scale-up of SBCR for FT synthesis are still scarce.

Therefore, the overall objective of this project is to properly describe the distribution of phases and liquid (slurry) circulation and turbulence in SBCR for Fischer-Tropsch synthesis by studying the microstructure of the gas-liquid-solid mixtures in a comparable fluid to FT waxes in 2 inch diameter column and develop a fundamental understanding as to how important the physical and fluid dynamic properties can be "finger-printed" via various diagnostic techniques such as laser doppler anemometry (LDA), optical probe and differential pressure fluctuation technique and by measuring large scale hydrodynamic parameters at high pressure and high gas velocity in a 6 inch diameter slurry bubble column using computed tomography (CT) and computer



Figure 1.1: Variables that affect SBCR performance

automated radioactive particle tracking (CARPT). CARPT and CT are the only non-invasive techniques that can provide information on slurry velocity and density profiles in 3D domain. Such data provides a firm scientific and engineering basis for scale-up and design of FT SBCR. In addition the obtained results can be utilized as a benchmark to validate the computational fluid dynamic codes.

This grant enables a unique integration of the expertise of the two universities (Washington University,WU and Ohio State University, OSU) and industry (Air Products and Chemicals, APCI) towards achieving the goals set for the project. This study complements well the work in progress at WU, OSU, Iowa State University (ISU) and Sandia National Laboratory, Contract No. DE-FC-22-95PC95051, related to the LaPorte Advanced Fuels Demonstration Unit (AFDU) operated by Air Products with the Department of Energy funding which focuses on advancing the state-of-art in understanding the fluid dynamics of slurry bubble columns and replacing empirical design methods with a more rational approach.

2. OVERALL OBJECTIVES

The overall objectives of this cooperative university (WU and OSU) – industry (APCI) research is to advance the understanding of the hydrodynamics of FT SBCR via advanced diagnostics techniques. The goals set for these projects are as follows:

- **TASK 1:**Literature Review
 - Physicochemical properties and their effect on the hydrodynamics of bubble columns.
 - Models used to predict FT reactor performance.
- **TASK 2:** Based on Task 1, identify the range of intrinsic properties (density, viscosity and surface tension) of the fluids used for the FT synthesis.
 - Identify a solvent that, at room temperature and pressure up to 200 psig, will mimic the hydrodynamics of FT wax (at FT reaction conditions).
 - Identify the particle type and size to be used.
- **TASK 3:** Using the identified system (solvent-particle-air), perform the following investigation on the hydrodynamics in a 2" diameter column:
 - Using Laser Doppler Anemometer (LDA) investigate the effect of reactor pressure on the flow field and turbulent parameters.
 - Using Δp fluctuation measurements to identify the flow regime transition and investigate the effect of reactor pressure on the flow regime transition.
 - Using change in slurry height measure overall gas holdup.
 - Using Optical Probe measure bubble size and bubble rise velocity.
- **TASK 4:** Using the identified system in Task 2 or a system with similar properties, investigate the hydrodynamics in a 6" diameter column via CT and CARPT techniques. The following will be measured:
 - Phase distribution profiles using CT/CARPT/ Δp
 - Flow field and turbulent parameters using CARPT
 - Gas holdup using CT and differential pressure transducers.
- **TASK 5:** Evaluate scale-up procedure for slurry bubble column. Develop additional correlations, if needed.
- **TASK 6:** As a case study, examine the available CFD and mechanistic models against the newly obtained data.
- **TASK 7:**Final report

2.1 Accomplishments During the First Year

The first year was dedicated to the preparation of the technical review, experimental facilities and the advanced measurement techniques. A new correlation was developed to predict the liquid-solid mass transfer coefficient in high pressure bubble column based on the atmospheric pressure data. The accomplishments were as follows:

- 1. The technical review of the variables affecting SBCR performance, some aspects of bubble dynamics and hydrodynamic properties and the physical properties of FT waxes and catalyst has been performed.
- 2. The experimental facilities and the advanced measurement techniques have been prepared. The preparation includes the following units:
 - High pressure (up to 3000 psi) and high temperature (up to 250°C) 2-inch diameter slurry bubble column set-up.
 - High pressure (up to 200 psi) 6-inch diameter slurry bubble column set-up. Two facilities will be used; one for computer automated radioactive particle tracking (CARPT) and computed tomography (CT) techniques and another one for pressure drop measurements. The later facility consists of a 6-inch diameter column equipped with 6- windows and 15 ports along the column.
 - Laser Doppler Anemometry (LDA) for 2" slurry bubble column facility.
 - CARPT and CT for 6-inch slurry bubble column facility.
 - Techniques to measure in situ the intrinsic density, viscosity and surface tension of the liquid phase that will be used for hydrodynamics investigation which mimic the hydrodynamics of FT waxes.
- 3. The solvent that mimic FT waxes at FT operating conditions has been identified and the gas and solid phases to be used in the hydrodynamics investigation have been selected.
- 4. A new correlation to estimate the mass transfer coefficient at high pressure based on atmospheric pressure data has been developed.

2.2 Accomplishments During Second Year

- 1. Experimental investigation of hydrodynamics of Norpar 15- nitrogen-glass beads in 2" column using LDA/pressure drop/slurry height measurements has been executed.
- 2. The technical difficulties related to CARPT at high pressure stainless steel SBCR have been resolved.
- 3. Experimental investigation of the effect of reactor pressure and gas flow rate on the hydrodynamics of air-water-glass beads system in 6" column using CT/CARPT has been performed.
- 4. Correlations to predict radial gas holdup and axial liquid recirculation velocity profiles in bubble columns have been developed.

2.3 Plan for the Next Year

- 1. Complete the experimental investigation of hydrodynamics of SBCR using the selected fluid-air-glass beads (150µm) in 2" diameter column.
- 2. Complete the experimental investigation of hydrodynamics of SBCR using selected fluid-air-glass beads (150µm) in the 6" diameter column.
- 3. Evaluate the current scale-up procedure and CFD against the newly obtained data.

3. EXPERIMENTAL FACILITIES

The prepared experimental facilities were discussed in the technical report (no.1) of the first year. However, for clarity the facilities that are used in the investigations reported in this report are again outlined as follows.

3.1 High pressure and high temperature 2" diameter slurry bubble column

The schematic diagram of the 2" high pressure and high temperature slurry bubble column facility is shown in Figure 3.1. The column is made of stainless steel and is 95.9 cm in height and 5.1 cm in diameter. Three pairs of plane windows made of quartz are installed on the column; each window is 1.27 cm wide and 9.3 cm in height. These windows allow viewing throughout the entire test section of the column. The pressure is controlled by a back pressure regulator installed at the outlet of the column.

3.2 High pressure 6 inch diameter slurry bubble column

High pressure 6" slurry bubble column experimental setup (Figure 3.2) can be operated up to the maximum operating pressure of 200 psig at room temperature. Compressed air is used as gas phase and is supplied from the compressor with working pressure of 185 psi at the maximum flowrate of 310 SCFM. Air passes through an air filter, pressure regulator, flowmeter setup (4 rotameters of increasing range in parallel) and enters through a check valve (to prevent liquid/slurry phase back flow) into distributor chamber of the bubble column. Air exits the column through a demister, passes through the back pressure regulator to the atmosphere. Column design enables easy removal of the distributor chamber and sparger replacement. Two similar column designs are used to suit all the needed experiments. The first one, designated for CARPT/CT experiments, is a smooth 6" column equipped with just two probe ports (1") at each end of the column. The second one, designated for probe measurements, is also 6" column equipped with an array of additional 15 probe ports (1") and 6 (12"H x 1½2"W) view windows. View windows are mounted at three radially opposite sides in staggered positions to cover the middle and the top part of the column.







Figure 3.2: Gas flowsheet for the high pressure 6 inch diameter bubble column

3.3 Laser Doppler Anemometer (LDA)

The Laser Doppler Anemometer is capable of non-intrusive velocity measurements in fluid dynamics in both gas and liquid phases. It has up to three velocity components with high accuracy and high spatial resolution due to small measurement volume. The basic components of an LDA include a continuous wave laser, a traverse system, transmitting and receiving optics and a signal conditioner and a signal processor. Figure 3.3 shows the set-up of LDA system used in this work at OSU. A 300-mW air-cooled argon-ion laser and a beam separator are used to generate two pairs of beams of known wavelengths of 514.5 and 480 nm. The light is transmitted through a fiber optic cable and a probe with 3.40 and 3.22 μ m and measurement volumes of 0.164 X 0.164 X 2.162 mm and 0.156 X 0.156 X 2.05 mm for the 514.5 and 480-nm wavelengths respectively. The scattered light is collected with a detector and processed with a signal processor.

The principle of the operation follows that when a particle passes through the intersection volume formed by the two coherent laser beams, the scattered light received by a detector has components from both the beams. The components interfere on the surface of the detector. Due to changes in difference between optical path lengths of the two components this interference produces pulsating light intensity as the particle moves through the measurement volume.



Figure 3.3: Laser Doppler Anemometer Setup.

3.4 Computer Automated Radioactive Particle Tracking (CARPT)

CARPT is a technique for tracking of a single radioactive tracer particle by detecting the intensity distribution of emitted γ -rays (Figure 3.4). It consists of a number of scintillation detectors (16-32) to monitor the motion of a single small radioactive particle in multiphase systems. This technique has been used extensively at Washington University (Chemical Reaction Engineering Laboratory, CREL) to measure in a non-invasive manner the flow pattern and turbulent parameters of different multiphase flow reactors. A fully wettable, neutrally buoyant particle is used to simulate the motion of the liquid in gas-liquid system and particle of the same size and density of the solids used is employed for monitoring the motion of solids in two and three phase fluidized beds and other multiphase systems. Scandium 46 at activities of about $200 - 500 \mu ci$ is used in a composite made tracer particle. Collected data for the tracer particle location in time is used to compute the tracer particle velocity and "turbulent" parameters. Precise calibration and good radioactive particle tracking are essential in obtaining accurate and reproducible CARPT data. In this work the assessment of solids velocities and "turbulent" parameters in a FT slurry bubble column is sought. Thus, the radioactive tracer particle should, as closely as possible, track the solids present in the system. To accomplish this the tracer particle should be comparable in size and density to the solid phase particles. Scandium is a highly reactive rare earth metal whose reactivity increases with increase in surface area per unit volume (decrease of diameter). To resolve the issue of the reacting scandium tracer particle we have developed a new technique for coating and protecting the minute size tracking particles. A tracer scandium Sc45 particle of required diameter is protected with a thin coating of Parylene N, an extremely inert derivative of poly p-xylene with excellent thermal and mechanical properties. The coated Scandium particle is then irradiated in a nuclear reactor. The resulting radioactive scandium Sc46 particle (strength of up to 200 µCi and half-life of 83 days) with a total diameter within the solid phase particle size range is thus used as a tracer particle. Since the density of Parylene N is 1.11 g/cm³, application of different coating thickness lower the overall particle density from 2.99 g/cm³ (of pure scandium) to about 2 g/cm³.

A detailed experimental setup and calculation procedure for CARPT experiments is given by Degaleesan (1997). In-situ calibration of detectors will be performed under the desired operating conditions using a calibration device that is operated under high pressure. CARPT data (tracer particle position in time) acquired over sufficiently long time, to insure enough particle occurrences in each column cell and good time/ensemble averaging, is used for calculation of the time averaged solids:

- a) velocities,
- b) "Reynolds" stresses,
- c) "turbulent" kinetic energy and
- d) eddy diffusivities.

This unique technique is essential for validation of hydrodynamic models used in design and scaleup and testing the effect of different design and operation variables (e.g. pressure, gas velocity, distributor design, internals, etc.) on the flow patterns in FT slurry bubble column reactors.



Figure 3.4: Configuration of the CARPT experimental setup.

3.5 Computed Tomography (CT)

CT is a technique for measurement of the cross-sectional phase holdup distribution in multiphase systems (Figure 3.5).

The CT technique has been extensively implemented at Washington University on various multiphase flow systems. It consists of an array of detectors and an opposing source rotate together around the object to be scanned. The scanner uses a cesium-137 encapsulated γ -ray source with activity of ~ 85 mCi. The array of detectors and the source are mounted on a gantry which can be rotated about the object to be scanned through a step motor. The entire system is completely automated to acquire the data required for the reconstruction of the phase distribution in a given cross-section. The Estimation-Maximization algorithm for image reconstruction has been implemented. It is based on maximum likelihood principles and takes into account the stochastic nature of the projection measurements.

Single source CT is used for phase holdup reconstruction in two phase (e.g., gas-liquid) systems. Theoretically, dual source CT is capable of resolving the holdups in three phase systems (e.g., gas-liquid-solid). However, because of numerical error accumulation dual source CT reconstruction of holdup profiles is yet not available (Daly et al., 1996). In this work we utilize a new combination of measurement techniques CT/CARPT/DP that has been developed in our laboratory (Chemical Reaction Engineering Laboratory, CREL) at WU as a part of the other projects which are currently under execution in parallel with this grant to evaluate holdup profiles of all three phases in a slurry system (Rados, 1999).



Figure 3.5: Configuration of the CT experimental setup (Kumar, 1994).

For clarity, a summary of such calculation methodology is again outlined below.

For a single γ radiation source, absorbance A over the path l is equal to:

$$A = -\ln\frac{I}{I_0} = \sum_{l} (\rho \mu)_{ij} l_{ij}$$
(3.1)

where I_0 is the intensity of radiation emitted by the source, I is the intensity of radiation received by the detector. Σ indicates the summation of the volumetric attenuation $(\rho\mu)_{ij}$ of each cell ijmultiplied by the path length in that cell l_{ij} along the path l, through which the radiation beam passes on its way from the source to the detector. If sufficiently large number of the scans of the operating column are taken from different directions (projections) then the volumetric attenuation in each cell $(\rho\mu)_{ij}$ can be calculated. To get the holdup distribution we have to measure the absorbance A_K for an empty column (K=G), for a column filled with liquid (K=L), for a column with solids and gas in voids between solid particles (K=GS) and for a column in operation with the gas-liquid-solid slurry (K=GLS). For each of these situations the detected intensity of radiation I_K and hence the measured absorbance A_K is different. Since the flow is time dependent larger number of acquired projections than cells (#equations >> #unknowns, over sampling) will yield more accurate time averaged attenuation coefficients (better statistics). In general I_0 is unknown and because of that the intensity of radiation I_K must be normalized with the intensity of radiation detected in the column containing only the gas phase I_G . In addition the intensity I_K must be corrected for the background (room) radiation intensity $I_{K,bck}$. This yields the following equation for A_K :

$$A_{K} = -\ln \frac{I_{K} - I_{K,bck}}{I_{G} - I_{G,bck}} = \sum_{l} \left[(\rho \mu)_{K,ij} - (\rho \mu)_{G,ij} \right] l_{ij}$$
(3.2a)

One defines relative volumetric attenuation as:

$$R_{K,ij} = (\rho\mu)_{K,ij} - (\rho\mu)_{G,ij}$$
(3.2b)

For the column containing packed bed of solids (uniform holdup of ε_s^0) and gas in voids between the solids particles the volumetric attenuation coefficient in cell *ij* is equal to:

$$(\rho\mu)_{GS,ij} = (\rho\mu)_{S,ij} \varepsilon_S^0 + (\rho\mu)_{G,ij} \left(1 - \varepsilon_S^0\right)$$
(3.3)

Substitution of Eq. (3.2b) into Eq. (3.3) (written for the gas-solid system, K = GS) after some manipulation yields the local solids volumetric attenuation coefficient:

$$(\rho\mu)_{S,ij} = \frac{R_{GS,ij} + (\rho\mu)_{G,ij} \varepsilon_S^0}{\varepsilon_S^0}$$
(3.4)

Similarly, for a slurry system:

$$(\rho\mu)_{GLS,ij} = (\rho\mu)_{G,ij} \varepsilon_{G,ij} + (\rho\mu)_{L,ij} \left(1 - \varepsilon_{G,ij} - \varepsilon_{S,ij}\right) + (\rho\mu)_{S,ij} \varepsilon_{S,ij}$$
(3.5)

Eq. (3.5) combined with Eq. (3.2b) (written for liquid, K=L and slurry, K=GLS) and Eq. (3.4) yields the expression for local gas holdup (cell *ij*):

$$\varepsilon_{G,ij} = \frac{R_{GS,ij} \frac{\varepsilon_{S,ij}}{\varepsilon_S^0} + R_{L,ij} \left(1 - \varepsilon_{S,ij}\right) - R_{GLS,ij}}{R_{L,ij}}$$
(3.6)

Clearly in order to close the system of equations we need one more equation for local solids holdup $\varepsilon_{S,ij}$. In dual source CT one more equation of the form (3.2) can be written for the other γ source. In this work the additional equation is generated from DP and CARPT measurements:

$$-\frac{1}{g}\frac{\Delta P}{\Delta z} = \rho_G \overline{\varepsilon_G} + \rho_L \left(1 - \overline{\varepsilon_G} - \overline{\varepsilon_S}\right) + \rho_S \overline{\varepsilon_S} \qquad DP \qquad (3.7)$$
$$\varepsilon_{S,ij} = n_{S,ij} \frac{\overline{\varepsilon_S}}{n_S} \qquad CARPT \qquad (3.8)$$

DP equation (3.7) assumes fully developed flow, no axial holdup profiles and negligible wall shear stress in the section Δz . Fully developed flow in slurry systems is usually reached at heights above two column diameters. Axial holdup profiles can be neglected over small Δz distances and the wall shear stress has been shown to be negligible compared to the pressure drop (Fan, 1989, less than 1%). CARPT equation (3.8) states that the volume averaged number of radioactive tracer particle occurances in the specific cell $n_{S,ij}$ is proportional to the solids holdup in that cell assuming that the radioactive tracer particle completely resembles solid phase particles and that all cells in considered cross plane are well perfused and readily accessible to the radioactive tracer particle (Moslemian et

al., 1992). This assumption is justified in churn turbulent regime. Finally, the combination of Eq. (3.7) and (3.8) yields the expression for the local solids holdup (cell *ij*):

$$\varepsilon_{S,ij} = \frac{-\frac{1}{g}\frac{\Delta P}{\Delta z} - \rho_G \overline{\varepsilon_G} - \rho_L \left(1 - \overline{\varepsilon_G}\right)}{\rho_S - \rho_L} \times \frac{n_{S,ij}}{\overline{n_S}}$$
(3.9)

Using the following iterative procedure the holdup profiles of all three phases can be calculated.

- 1) Guess the cross-sectional average solids holdup. The initial guess is based on the calculation of the cross-sectional average solids holdup from the overall gas holdup measurements and nominal solids loading (v_{s0}, volume of solids per volume of slurry suspension initially charged into the column) using the equation $\overline{\varepsilon_s} = v_{s0}(1 \overline{\varepsilon_G})$.
- 2) Using Eq. (3.8) calculate the solids holdup in each cell.
- 3) Using Eq. (3.6) calculate the gas holdup in each cell.
- 4) Calculate the cross-sectional average gas holdup.
- 5) Using Eq. (3.9) calculate the solids holdup in each cell.
- 6) Calculate the cross-sectional average solids holdup.
- 7) Compare the calculated and previous values (initial guess in the first iteration) of the crosssectional average solids holdup.

Using the solids holdups in each cell recalculated in step 5 repeat the steps 3 through 7 until specified tolerance is achieved (convergence).

4. **RESULTS AND DISCUSSION**

The following is the summary of the results for the investigations made during the second year.

4.1 Hydrodynamics measurements in 2" column using LDA and the change in the slurry height:

This work has been carried out at Ohio State University. The liquid and gas phase used for these experiments are Norpar15 (density =0.772 g/cc, viscosity =2.13 CPs, surface tension = 26.7 dynes/cm at ambient temperature and pressure) and nitrogen. The solid phase was glass beads of 150 μ m. All the experiments have been operated in batch liquid mode. The static liquid level is maintained at 50 cm above the distributor. Pliolite particles of 1.02 g/cm3 in density with a size range of 20-50 mm are used as the liquid tracer for LDA technique.

4.1.1 Overall gas holdup obtained by measuring the change in the slurry height

The overall gas holdup has been obtained by measuring the change in the slurry height. Figure 4.1 shows the pressure effect on the overall gas holdup. The overall gas holdup increases with increasing pressure. At superficial velocity of 35 cm/s, the overall gas holdup can reach 0.5 at pressure of 1.0 MPa while the overall gas holdup is only 0.3 at ambient pressure. Figures 4.2 and 4.3 gives the overall gas holdup at various pressures for slurry bubble column of 20 wt% and 40 wt% of solids loadings in slurry, respectively. The presence of particles reduces the gas holdup at all pressures, especially for velocities below 20 cm/s. The same conclusion was also reached by many researchers in literature viz. Koide et al. (1984), Kara et al. (1982), B.Gandhi et al. (1984), Krishna et al. (1997), etc. This decrease in gas holdup with increase in solids concentration is due to the fact that with increase in solids concentration, coalescence of small bubbles into larger ones occurs which results into increase in the mean bubble diameter. Elevated pressures also lead to higher gas holdups for all solids loadings. However, the effect is more profound at higher solids loadings.

4.1.2 Averaged liquid velocity and Reynolds stresses measured by LDA using Norpar 15nitrogen system

In order to let the laser beam to penetrate through the sight glass, only gas-liquid (Norpar 15nitrogen) system and superficial gas velocity up to 8 cm/s have been used in this investigation. Figure 4.4 shows the average liquid velocity at different superficial gas velocities and at ambient pressure. The average liquid velocity is higher in the center region of the column. A down flow is observed in the wall region due circulation. The inversion of velocity takes place at around $r/R\sim0.7$. The tangential and axial Reynolds normal stresses are illustrated in Figure 4.5 and 4.6, respectively. Both the tangential and axial Reynolds normal stresses are the lowest at the wall and the value levels off as the radial location moves toward the center. The center has slightly lower tangential and axial Reynolds normal stresses due to the swirling motion of the central bubble stream. In the center of the column, the flow is more frequently upward, whereas in the vortical region, the flow dynamically changes from upward to downward depending on the location of the central bubble stream. The flow in this region therefore experiences large fluctuations in the axial component of the liquid velocity, leading $\langle v'v' \rangle$ to peak closer to the center of vortical structures rather than in the center where the motion is primarily directed upward.



Figure 4.1: Pressure effect on overall gas hold up in 2" column using Norpar 15-nitrogen system



Figure 4.2: Effect of pressure on overall gas holdup in Norpar 15-nitrogen-glass beads system at 20 wt% solids loadings in 2" column



Figure 4.3: Effect of pressure on overall gas holdup in Norpar 15-nitrogen-glassbeads system at 40 wt% solids loadings in 2" column



Figure 4.4: Averaged liquid velocity measurement at various superficial gas velocities at ambient pressure



Figure 4.5: Tangential Reynolds normal stress at various superficial gas velocities at ambient pressure



Figure 4.6: Axial Reynolds normal stress at various superficial gas velocities at ambient pressure

4.2 Gas holdup profiles measured by Computed Tomography (CT)

In this work, the effects of pressure and superficial gas velocity on the gas holdup and its distribution have been studied first in air-water-glass beads system using CT. The reason behind using air-water-glass beads system is, we have extensive experience with this system and we have obtained a large data bank via CT and CARPT techniques in air-water system. In addition, the developed methodology of combination of CT/CARPT/DP discussed in section 3.5 will be evaluated and validated using air-water-glass beads as a system that is relatively easy to handle. The experiments and their operating conditions that have been performed using CT are listed in Table 4.1.

Run	Pressure Mpa (psi)	Superficial gas velocity, cm/s	Solids loadings Wt%, Vol%	Sparger
1*	0.1 (14.7)	8	20 (9.1)	PPL
2*	0.1 (14.7)	45	20 (9.1)	PPL
3	0.4 (58.0)	8	20 (9.1)	PPL
4*	0.4 (58.0)	45	20 (9.1)	PPL
5*	1.0 (145)	8	20 (9.1)	PPL

Table 4.1 : The Experiments performed using CT and CARPT Techniques in air-waterglass beads

*CARPT experiments were performed at these conditions.

where, PPL \rightarrow porous plate large hole sparger, 0.15% O.A.

All of the above experiments have been performed at three axial positions (2D, 5.5D, 9D) in 6" column.

Since at this stage, pressure drop measurements and full CARPT data processing were not performed, the developed methodology cannot be implemented to evaluate the distribution of phases across and along the column. However, for preliminary evaluation, we have assumed uniform distribution of solids across and along the column in order to process CT data for evaluating the gas holdup distribution. This may not represent the real situation which currently is refined using the developed methodology.

Figure 4.7 shows representative results of CT with assumption of uniform solids holdup (ε_s =0.078), at 8 and 45 cm/s using PPL. The radial gas holdup profile exhibits a characteristics shape obtained in gas-liquid system with greater gas holdup in the column center than by the walls. Also, one can observe that transition to churn-turbulent regime is characterized by the change in steepness of the radial gas holdup profile from relatively flat to almost parabolic.



Figure. 4.7: Gas holdup profile at 8 and 45 cm/s using PPL with assumption of uniform solid holdup at 9.1 vol% of solid loadings

where, 11: level 1 = 2D 12: level 2 = 5.5D 13: level 3 = 9D

The fact that, solids holdup is not uniform, will be further looked at once CARPT data processing is completed. The CARPT/CT/ ΔP algorithm explained in Section 3.5, will be used to calculate gas, liquid and solids holdup and their profiles in the column to account properly for the axial and the radial distribution of the phases holdups.

4.3 Computer Automated Radioactive Particle Tracking (CARPT)

During the process of implementing CARPT for the first time on high pressure steel column, several serious technical problems and issues have been encountered which need to be investigated and resolved carefully. Fortunately, all these problems have been successfully identified and resolved to implement CARPT technique at high pressure operation as a part of the other projects (Industrial Consortium and DF-22-95PC95051) which are executed in parallel

with this grant. In the following sections, we will briefly outline the problems and their implemented solutions.

4.3.1 Sc 46 CARPT Tracer Particle:

As mentioned earlier the radioactive particle should, as closely as possible, track the solids present in the system. To accomplish this, the tracer particle should be of comparable size and density to the solid phase particles. Scandium is a highly reactive rare earth metal whose reactivity increases with increase in surface area per unit volume i.e. decrease in diameter. In addition Sc is highly reactive with water and oxygen. To resolve the issue of reacting scandium tracer particle, we have developed a new technique for coating and protecting the minute size tracking particles. The Sc 46 particle is protected with inert material, parylene which is a derivative of poly p-xylene. The reasons for parylene as a choice for coating are,

- high melting point
- radiation resistance
- excellent mechanical properties –comparable to epoxy resins
- can be applied in layers below 0.1µm
- small density
- solvent resistance
- low permeability to moisture and gases
- no outgassing
- acid and base resistance
- uniform thickness on all surfaces

There are four types of parylenes, among which Parylene N was selected based on the specifications listed in Table 4.2.

	Tm(°C)	Stable in oxygen(°C)	Density(g/cm3)	Atoms other than C & H
Paralyne N	410	115	1.11	None
Paralyne C	290	125	1.29	1 Cl
Paralyne D	380	150	1.42	2 Cl
Paralyne HT	500	450	1.58	4 F

Table 4.2: Specifications of some Parylene family

The particle coating has been performed by Paratech Inc. with our close interaction and involvement. During coating, ionized air is blown over the particles to remove static electricity. On the walls of tumbler (core machine #1 with 7.5×4.5 " tumbler), static guard is sprayed to reduce static charge further. Then tumbler with particles is closed by masking tape. The required quantity of Parylene N dimmer is put in the system and is slowly vacuumed to 50 µm water and door & vaporizer temperature are set to 125°C. The rotational speed of tumbler was set to 4 rpm. Amongst the coated particle, several 'good' particles were selected for the irradiation on the basis of required size, shape and coating quality using microscope.

The irradiation of the selected particles was conducted at the flux of 4e14 n/cm².sec at ORNL (Oak Ridge National Laboratory) Nuclear reactor.

Figure 4.8 shows typical pictures of the coated tracer particle.



Figure 4.8: Typical pictures of tracer particles

4.3.2 CARPT calibration in high pressure slurry stainless steel column:

A reliable calibration technique is the one that assures accurate positioning of the particle which can be read with enough precision. In all CARPT experiments automated system was used. But due to the presence of the solids as the third phase, high reactor pressure and the flange design of high pressure calibration device, the calibration device failed to properly operate. Mechanical modifications have been implemented successfully to resolve these problems. In addition, several technical issues in the process of CARPT implementation have been encountered that affect considerably the accuracy of reconstructing the position of the radioactive particle. These problems as well have been successfully identified and resolved as outlined below.

i)Build-up problems:

Depending on whether the photon has encountered multiple collisions on its way before impinging on the detector, or has left the detector with residual energy, the energy deposited may be different from that of a genuine photon issued at the source (i.e. the photon that travels unhindered from the source to the detector crystal). There may also be small pulses recorded from electronic noise sources and may have nothing to do with the actual photon emission and detection process.

This problem was resolved by setting threshold energy value that captures only the photons with energy within the photo-peak as shown in Figure 4.9. Photons in the photo-peaks are the genuine photons that travel unhindered from the source to the detector and deposit all their energy in the detector crystal.



Figure 4.9: Typical differential pulse height spectrum

ii)Spectrum stability:

The problem of spectrum stability was resolved by properly controlling the laboratory temperature to avoid temperature rise of the electronic parts and electrical contacts, grounding and by frequently checking the Multi Channel Analyzer (MCA) and the detector stability.

iii) Pile-up problems:

Poor relative orientation of source and detector can cause detectors to be 'blinded' by excessive counts or receiving very low counts. When the tracer particle is very close to the detector, the gamma photons have a high probability of interaction with the detector crystal. Hence, many photons are counted while in the case when the particle radiation source moves away from the detector axis, a region of steep slope of counts versus distance is encountered which is characterized by large changes in counts with the small changes in distance. Also, there is asymptotic region in which large changes in distance make only small variations in measured counts. In this region, detectors are placed flush to the wall to avoid saturation of photons on crystal surface. As a result of which, placing detector in such region means using it to the full scale.

iv) Detector alignment:

The alignment of detectors plays very important role in CARPT calibration and experiments as it determines how the detectors "see" the tracer particle. The small change in detector alignment (angle) can cause considerable changes in particle position reconstruction. For proper alignment of detectors laser beam alignment procedure was developed.

Figures 4.10 & 4.11 shows the improvement obtained due to the resolution of the pile-up and alignment problems.



Figure 4.10: Calibration curve of single detector obtained with alignment and pile-up problems



Figure 4.11: Calibration curve of single detector after resolving detector alignment and pile up problems

v) Modification in existing reconstruction algorithm in slurry Stainless Steel column:

It was found that existing algorithm for particle position reconstruction which was primarily developed for gas-liquid systems by Degaleesan (1997) using plexiglas column was inadequate for slurry system in stainless steel column.

A new reconstruction algorithm was developed which takes into account the encountered problems such as detectors alignment and the loss in the strength of radiation due to the particle attrition. The newly developed algorithm results in a noticeable improvement in reconstruction algorithm as shown in Figures 4.12 and 4.13.



Degaleesan - 30 weighed det.

Figure 4.12: Calibration particle reconstruction using the algorithm developed by Degaleesan (1997) for gas-liquid system and plexiglas column



Figure 4.13: Calibration particle reconstruction using the newly developed algorithm in stainless steel column.

4.3.3 CARPT Results in high pressure SBCR:

The execution of CARPT experiments as indicated in, Table 4.1 with air-water-glass beads have been completed at Washington University in 6" column. The data processing which is time consuming is in progress. The results and findings will be reported upon completion of the data processing.

5. DEVELOPMENT OF CORRELATIONS FOR PREDICTION OF RADIAL GAS HOLDUP AND LIQUID RECIRCULATION VELOCITY PROFILES IN BUBBLE COLUMNS

In attempt to improve the design and scale-up of bubble columns, correlations have been developed to predict the radial gas hold up and axial liquid velocity profiles in bubble column reactors. The details of the developed correlations are described in the two full manuscripts published in Chemical Engineering Science Journal attached as Appendix A (Wu et al., 2001; Wu and Al-Dahhan, 2001)

6. NOMENCLATURE

- D Column diameter, m
- H Column height, m
- P Presure inside column, psi
- r/R Dimensionless radius
- u Axial liquid velocity, cm/s
- Ug Superficial gas velocity, cm/s
- u'u' Tangential Reynolds normal stress, (cm/s)²
- v'v' Axial Reynolds normal stress, (cm/s)²

Greek letters

- ε_{G} Cross-sectional average gas hold up
- μ_L Liquid viscosity, CPs
- ρ_L Liquid density, gm/cc
- σ_L Liquid surface tension, dynes/cm

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APPENDIX

DEVELOPMENT OF CORRELATION for PREDICTION OF RADIAL GAS HOLDUP & AXIAL LIQUID RECIRCULATION VELOCITY PROFILES

in BUBBLE COLUMNS

Predictions of Radial Gas Hold Up Profiles in Bubble Column Reactors

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Abstract

Gas holdup and its profile are important parameters to be characterized in bubble column reactors. Proper prediction of the radial gas holdup profiles is necessary for determining liquid mixing, flow regime transition, heat and mass transfer. In this study, the following gas holdup profile form, which can be fitted to the observed profiles, is proposed:

$$\varepsilon_G = \overline{\varepsilon}_G \left(\frac{n+2}{n+2-2c}\right) \left[1 - c(r/R)^n\right]$$

The parameters n and c needed to describe the gas holdup profile are correlated with appropriate dimensionless groups.

$$n = 2.188 \times 10^3 \text{ Re}_G^{-0.598} Fr_g^{0.146} Mo_L^{-0.004}$$

 $c = 4.32 \times 10^{-2} \text{ Re}_G^{-0.2492}$

However, the cross-sectional average gas holdup, ε_G , can be estimated using the available correlations for overall gas holdup. The agreement between the correlation predictions and experimental data is reasonable over wide range of operating conditions. \mathbb{C} 2001 Elsevier Science Ltd. All rights reserved.

Key words: bubble columns, gas holdup profiles, correlation

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1. Introduction

Gas holdup profile is one of the most important parameters in bubble column reactors. The spatial variation of gas hold up gives rise to pressure variation, which results in liquid recirculation in the bubble column. This liquid recirculation governs the rate of mixing, heat transfer and mass transfer. The ability to predict radial gas holdup profiles in bubble column reactors would help us in determining the flow regimes, liquid mixing, heat and mass transfer better. This should make bubble column scale-up more reliable.

The existence of a pronounced radial hold up profile is the characteristic of the heterogeneous regime of flow in the column which generates strong liquid recirculation. The magnitude of gas holdup radial gradients and the magnitude of liquid velocity driven by the gas depend on superficial gas velocity, column diameter, the nature of the gas-liquid system and the operating conditions (pressure and temperature of the reactor).

During the past three decades, a number of experimental measurements of gas holdup and gas hold up profile have been reported in the literature and have been summarized by Joshi *et at.*(1998). A variety of techniques, such as optical fiber probes, gamma ray densitometry, particle image velocimetry, and gamma ray and X-ray attenuation together with computer tomography have been employed for the local gas holdup measurements. Due to the complexity of the system, no fundamental equation is available at present for prediction of the gas hold up profiles in bubble columns, There are a number of empirical equations, similar in form, that can be fitted to the observed holdup profiles.

Nassos and Bankoff (1967) proposed the following equation for the radial holdup profile:

$$\varepsilon_G = \widetilde{\varepsilon}_G \left(\frac{n+2}{n}\right) \left[1 - \left(r/R\right)^n\right] \tag{1}$$

In eq(1), $\tilde{\varepsilon}_{G}$, which is the radial chordal average gas hold up along the column diameter, and the exponent n are parameters and r/R is the dimensionless radial position. The value of parameter n is indicative of the steepness of the holdup profile. When n is large the profile is flat, for small n the profile is steep. The steepness of holdup profile is reflected in the intensity of liquid circulation. Ueyama and Miyauchi (1979) reviewed the published literature and modified the above equation as follows to include the possibility of finite gas hold up close to the wall.

$$\varepsilon_G = \widetilde{\varepsilon}_G \left(\frac{n+2}{n}\right) \left[1 - c(r/R)^n\right]$$
(2)

In Eq.(2), c is an additional parameter which is indicative of the value of gas holdup near the wall. If c = 1 there is zero hold up close to wall, if c=0 hold up is constant with changing r/R.

Luo and Svendsen (1991) used Eq.(2) represented in terms of mean cross-sectional profile as given below:

$$\varepsilon_G = \overline{\varepsilon}_G \left(\frac{n+2}{n+2-2c}\right) \left[1 - c(r/R)^n\right]$$
(3)

By applying Eq. (3) to data, n was found from 1.4 to 11, and c from 0.5 to 1 according to the findings of different authors (Joshi *et al.*, 1998) and based on different systems investigated. In the absence of a firm theoretical prediction of the radial gas hold up profile correlations are needed for evaluating n and c based on the knowledge of the general operating variables and physical properties of the system in order to estimate the gas holdup profile by equation (3). In this work, such correlations have been developed as discussed below.

2. Correlation Development

Extensive gas holdup and gas holdup profile data have been acquired in the Chemical Reaction Engineering Laboratory (CREL) over the years under DOE contract DE2295PC95051 on the bubble column hydrodynamics, by employing gamma ray Computed Tomography (CT) over a wide range of superficial gas velocities (from 2 cm/s to 60 cm/s), at different pressures (0.1 MPa to 1.0 MPa) with 5 different gas distributors and in columns ranging in diameter from 0.19-0.44 m. The majority of the gas holdup profiles were measured in air-water system. However, airdrakeoil (light mineral oil) and air-propanol systems were also used at different operating conditions. The reproducibility of the measured gas holdup profiles was within $\pm 3\%$. By analyzing the experimental results carefully it was found that the shape of hold up profiles changes most significantly with superficial gas velocities. However pressure affects the shapes of hold up profiles but it has less effect compared to superficial gas velocity within the range of pressure and superficial gas velocity. Gas distributor does affect hold up in a certain range of gas velocities but it has a minor effect on gas holdup profiles particularly in the fully developed region and at high gas velocity. The shape of the gas holdup profile at different column heights seems to be unchanged at given gas velocity once the measurement has been taken at a certain distance from the distributor (2L/D or higher). Column diameter has been reported to have an effect on gas holdup profile (Kumar et al., 1997).

Based on the experimental observations and dimensional analysis, the following functional definition was proposed for parameters n and c:

$$n = af(\operatorname{Re}_G, Mo_L, Fr_G)$$
; $c = \beta \zeta(Re_G)$

The above dimensionless groups, Re_G , Mo_L , Fr_G , which are defined below reflect the effect of velocity and pressure, which change the density of the gas and has an effect on gas holdup profile, and the effect of gas and liquid physical properties. By fitting roughly two third of the available experimental data from the database consisting of our experiments and those in the literature (the remaining about one third of the experimental data are used to evaluate the developed correlations), the correlations listed below are generated for n and c:

$$n = 2.188 \times 10^3 \operatorname{Re}_{G}^{-0.598} Fr_g^{0.146} Mo_L^{-0.004}$$
(4)

$$c = 4.32 \times 10^{-2} \operatorname{Re}_{G}^{0.2492}$$
(5)

where

$$\operatorname{Re}_{G} = \frac{DU_{Sg}(\rho_{L} - \rho_{G})}{\mu_{L}}, \ Fr_{G} = \frac{U_{Sg}^{2}}{gD}, \ Mo_{L} = \frac{g\mu_{L}^{4}}{(\rho_{L} - \rho_{G})\sigma_{L}^{3}}$$

Equation (4) and (5) along with equation (3) are utilized for prediction of the gas holdup profiles for the whole set of the experiment data available. The cross sectional mean gas holdup values used in this study were evaluated from the experimental data. However, a favorite correlation for

the overall gas holdup can be used to estimate $\overline{\varepsilon}_{G}$ (Kemoun, Ong, Gupta, Al-Dahhan & Dudukovic, 2000).

As mentioned above, the majority of experimental data used were obtained by using an air-water system. Hence, based on the fitting performed in this study, n is almost independent of Mo_L (n $\alpha Mo_L^{-0.004}$). However, it was found that the liquid physical properties affect the overall gas holdup (Luo, Lee, Lau, Yang & Fan, 1999) and the holdup profile (Chen et al., 1998; Joshi et al., 1998). Therefore, at this stage, Mo_L is included in the correlation to be examined for any future necessary modification as gas holdup profiles become available for a wide enough range of liquid physical properties.

Due to the fact that most of the holdup profiles used were for air-water system, c was found to be only a function of Re_G . Liquid physical properties would affect the parameter c which needs to be examined further.

3. Comparison between the predicted and experimentally measured gas holdup profiles

3.1 Effect of superficial gas velocity

As mentioned earlier, gas holdup profiles vary significantly with gas velocity. The results are shown in Figure 1.

One can see from Figure 1 that gas holdup profiles become steeper with increased gas velocity (n changes from 3.73 at 8 cm/s to 2.02 at 60 cm/s). The steeper holdup profile is, the faster liquid recirculation rate is, hence liquid mixing, heat transfer and mass transfer rate will be improved accordingly. From Figure 1, one can observe that model predicts the experimental results reasonable well (mean relative error is within 15%).



Figure 1: Effect of velocity on gas hold up profile (Diameter of column: 0.15m, Distributor D2: Perforated plate, Hole diameter: 0.5 mm, Number of holes: 163, Open area: 0.15%; P=0.1 MPa; Water-air system)

3.2 Effect of reactor pressure

It is shown that pressure not only changes the gas hold up but also changes the gas holdup profile as well (Joshi *et al.*, 1998).



Figure 2: Effect of pressure on gas hold up profile in 0.15m reactor diameter (Ug = 14 cm/s, Distributor D2: Perforated plate, Hole diameter: 0.5 mm, Number of holes: 163, Open area: 0.15%, Water-air system)

At higher pressure smaller bubbles are formed by breakage due to high gas density. Small bubble size increases the overall gas holdup, and as pressure increases, gas bubble size distribution becomes narrow which results in a slightly flatter hold up profile due to the uniform distribution of small bubbles. As shown in Figure 2, at Ug = 14 cm/s and pressure varies from 0.4 MPa to 1 MPa there is no major difference in the shape of gas hold up profile (n values varies from 3.146 to 3.168). Unfortunately, currently there is no gas hold up profile available at higher pressure and higher gas velocity. For the current operation condition, the model predicts the experimental results well (mean relative error is within 14%).

3.3 Effect of physical properties

As mentioned earlier, both holdup distribution and liquid recirculation depend on liquid physical properties. A noncoalescing system and a coalescing systems have different overall gas holdup and gas holdup profiles as well. Figure 3 illustrates that the proposed correlations predict the experimental results reasonably well for liquids of different physical properties.



Figure 3: Effect of liquid physical properties on hold up profile (Ug = 4.8 cm/s, D = 0.6m, air-0.22% propanol in water: Menzel *et al.*, 1990; Ug = 10cm/s, D = 0.44m, air-drakeoil: Chen *et al.*, 1998)

It is noteworthy that two sets of the data presented in Figure 3 are taken from the literature (Menzel *et al.*, 1990; Chen *et al.*, 1998), and are predicted well by the developed correlations (mean relative error within 17%). However, larger errors in the correlations predictions are obtained in the region near the wall for the data obtained from Menzel et al. (1990). This would be due to, as mentioned above, the majority of the measured holdup profiles used for the developed correlations (Eqs. (4) and (5)) were obtained for air-water system which affect the dependency of the parameters n and c on the liquid physical properties.

3.4 Effect of column diameter

As reported in the literature (Joshi *et al.*, 1998) with increase in column diameter D, the liquid recirculation velocity increases as $V_c \alpha D^{0.3to0.4}$. Hence, one would expect a steeper holdup profile in larger column diameter. This is not obvious from Figure 4 due to different superficial gas velocities used in columns of different diameter and this is the only available data at the moment.



Figure 4: Effect of Column Diameter on the Holdup Profile (Ug = 7.2 cm/s, D=0.6m: Menzel *et al*, 1990; Ug = 10 cm/s, D=0.44m: Chen *et al.*, 1998; Ug = 12 cm/s, D=0.19m: Kumar, 1996)

Figure 4 shows the comparison of the correlations prediction and the experimental data at different column diameters. One can see that the prediction agrees with our experimental results and literature experimental data (mean relative error is within 15%).

4. Summary

It should be pointed out that in all the data presented here the cross-sectional mean holdup was known. In design situation, that would not be the case. Then a favorite correlation for the overall gas holdup can be used (Kemoun et al., 2000) to determine the mean holdup $\overline{\epsilon}_G$. A correlation is proposed for prediction of radial gas hold up profiles, which are important in driving liquid recirculation in bubble column. As Figure 1-4 illustrate the agreement between the correlations predictions and the experimental data is reasonable over variable a range of operating conditions (mean relative error is less than 17%). Further work considering gas-liquid-solid slurry system is still in progress.

Notation

- c Parameter in Eq (2)
- D Column diameter, m
- Fr_g Gas Froude number, dimensionless
- g Acceleration due to gravity, m/s^2
- Mo_L Liquid Morton number, dimensionless
- n Parameter in Eq (1)
- r, R Column radius, m
- Re_G Reynolds number, dimensionless
- U_{Sg} Superficial gas velocity, m/s
- V_c Liquid circulation velocity, m/s

Greek letters

- $\varepsilon(r)$ Radial gas hold up profile
- $\overline{\varepsilon}_{G}$ Cross-sectional average gas hold up
- $\widetilde{\varepsilon}_{G}$ Radial chordal average gas holdup
- μ_L Liquid viscosity, Pa.s
- ρ_G Gas density, kg/m³
- ρ_L Liquid density, kg/m³
- σ_L Liquid surface tension, N/m

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Predictions of Axial Liquid Velocity Profile in Bubble Columns

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Abstract

The liquid flow and mixing behavior in bubble columns is partially described by means of global liquid recirculation velocity profile. Due to the complex character of the flow in bubble columns, prediction of the axial liquid circulation is still a difficult task. In this work, the following correlation is proposed for liquid recirculation profile:

$$\frac{V_L(r)}{V_{LO}} = 1 - 2.65 * n^{0.44} * c \left[\frac{r}{R}\right]^{2.65 * n^{0.44} * c}$$

where n and c are the gas radial holdup profile parameters evaluated by the correlations proposed by Wu, Ong and Al-Dahhan (Chemical Engineering Science, 56 (2001) 1207-1210).

$$n = 2.188 \times 10^{3} \text{ Re}_{G}^{-0.598} Fr_{g}^{0.146} Mo_{L}^{-0.004}$$
$$c = 4.32 \times 10^{-2} \text{ Re}_{G}^{0.2492}$$

The predictions of the developed liquid circulation correlation agree well with the experimental data obtained in our laboratory and reported in literature. The model is simple and is easy to use as an engineering tool to assess the liquid recirculation in bubble columns. © 2001 Elsevier Science Ltd. All rights reserved.

Key words: bubble columns, axial liquid velocity, correlation

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1. Introduction

Bubble column reactors are widely used as gas-liquid and gas-liquid-solid contactors in much industrial aerobic fermentations, hydrogenations and other chemical operations because of their simple construction and ease of maintenance. Bubble columns combine efficient oxygen transfer and mixing with low shear forces. The behavior of these reactors is determined by their hydrodynamic properties. The complex flow and mixing behavior found in bubble columns is often described by means of global parameters such as gas holdup and liquid circulation velocity. Due to the complex character of the flow in bubble columns, their design and scale up are still a difficult task.

Many models have been proposed to analyze and predict liquid circulation. Miyauchi and Shyu (1970) and Joshi and Sharma (1979) have predicted liquid velocity in relation to the local gas hold up. However, the local gas hold up must be obtained from experimental data for both models. Zehner (1986) introduced a friction factor for liquid velocity prediction, but the value assigned by him to this parameter is not easy to justify. Kumar (1994) developed a onedimensional momentum balance -based model which requires hold up profile and eddy viscosity or mixing length profile to which the model is found to be very sensitive. Various attempts have been made at developing functional forms for the eddy viscosity (Ueyama and Miyauchi, 1979) and mixing length (Luo and Svendsen, 1991) which are required for solving the one dimensional model. However, Kumar (1994) showed that there is truly no universal expression for the mixing length or the eddy viscosity that can be successfully used under wide range of operating conditions, to predict the recirculating liquid velocity profile. In his mixing length correlation there are five parameters which are fitted to experimental data. Recently Krishna, Urseanu, van Baten and Ellenberger (1999) proposed a computational fluid dynamics based model for prediction of hold up profile and axial velocity profile. CFD based model could be a powerful design and scale-up tool after it has been fully verified. This is not yet the case.

The objective of this work is to develop a simple model based on which axial velocity profile can be predicted in relation to the gas-input rate and the column dimensions without requiring an input of the local gas hold up profiles.

2. Correlation Development

A power law liquid velocity profile is widely accepted in the literature (Montserrat, T. et al., 1996, Garcia-Calvo, E. et al., 1994). The liquid rises with the bubbles in the central portion of the column and flows downward in the outer annular section. Hence, the liquid velocity distribution in a bubble column may be expressed as

$$\frac{V_L(r)}{V_{Lo}} = 1 - 2^{\frac{N}{2}} \left[\frac{r}{R}\right]^N \tag{1}$$

where N is the exponent of the liquid velocity profile and V_{Lo} is the liquid center line velocity. N varies from 2- 2.3 or higher based on different investigators (Kawase and Moo-Young 1986,1987 and Montserrat & Garcia-Calvo, 1996). In fact, liquid circulation is due to the existence of the gas holdup radial profile, and the radial gas hold up profile and liquid circulation are internally tied together. Both depend on superficial gas velocity, column diameter and the

physical properties of the gas-liquid system investigated. The liquid according to the velocity profile of Eq. (1) is in central core region of the bubble column and flows downward in the wall zone.

A correlation of a similar form was proposed for prediction of the radial gas holdup profile (Luo & Svendsen, 1991):

$$\frac{\varepsilon_G}{\overline{\varepsilon}_G} = \left(\frac{n+2}{n+2-2C}\right) \left[1 - C\left(\frac{r}{R}\right)^n\right]$$
(2)

In Eq (2), n is indicative of the steepness of the gas hold up profile, and c determines the value of hold up near the wall. It provides for the possibility for both zero and non-zero gas fraction values at the wall which may affect the circulation of liquid as well. Possibly exponent N in Eq. (1) also depends on the liquid properties and on the gas flow rates (Wu, Ong & Al-Dahhan, 2000). Hence, it may be necessary to include both n and c in Eq. (1) to predict the axial liquid velocity profile. To establish the needed relationship between the gas and liquid velocity profile, Eq. (1) is modified as

$$\frac{V_L(r)}{V_{Lo}} = 1 - f(n,c) [\frac{r}{R}]^{\xi(n,c)}$$
(3)

Correlations have been developed (Wu et al., 2001) for calculation of parameters n and c as follows:

$$n = 2.188 \times 10^3 \operatorname{Re}_{G}^{-0.598} Fr_g^{0.146} Mo_L^{-0.004}$$
(4)

$$c = 4.32 \times 10^{-2} \operatorname{Re}_{G}^{0.2492}$$
(5)

where,

$$\operatorname{Re}_{G} = \frac{DU_{Sg}(\rho_{L} - \rho_{G})}{\mu_{L}}, \ Fr_{G} = \frac{U_{Sg}^{2}}{gD}, \ Mo_{L} = \frac{g\mu_{L}^{4}}{(\rho_{L} - \rho_{G})\sigma_{L}^{3}}$$

By fitting our computer automated radioactive particle tracking (CARPT) data, it was found that $f(n,c) = \xi(n,c) = 2.65 * n^{0.44} * c$. Therefore, Eq. (3) becomes

$$\frac{V_L(r)}{V_{LO}} = 1 - 2.65 * n^{0.44} * c \left[\frac{r}{R}\right]^{2.65 * n^{0.44} * c}$$
(6)

As mentioned earlier, V_{Lo} is the axial liquid velocity in the center of the bubble column and can be obtained from either experiments or the models reported by Zehner (1986) and Riquart (1981).

$$V_{LQ} = 0.737 (U_G D)^{1/3}$$
 Zehaner(1986) (7)

$$V_{LQ} = 0.21(gD)^{1/2} (U_G^3 \rho_L / g\mu_L)^{1/8} \quad \text{Riquart}(1981)$$
(8)

From Eq. (4) and (5), one can see that when superficial gas velocity increases, c increases and n decreases. However n decreases with power 0.44 and c increases with power one, so that the overall effect is to render the axial velocity profile steeper with increased superficial gas velocity which is experimentally observed. The value of the term $f(n,c)=2.65*n^{0.44}*c$ is between 1.8-2.4 with column diameter 0.1-0.63m and for superficial gas velocity 0.02-0.6m/s and different gas-and liquid physical properties.

A predicted liquid velocity using Eq. (6) with Eq. (4) and (5) is shown in Figure 1 and compared with experimental data. From Figure 1, it can be seen that model matches experimental data well at different superficial velocities.





3. Evaluation of correlation predictions

As mentioned above, Eq. (6) was developed by only using part of our CARPT database, and it is necessary to determine whether Eq. (6) can predict the experimental results from the literature to evaluate the capability of the modified correlation. We have compared the correlation predictions to the experimental data from very different sources reported in the literature and this comparison is illustrated below.

Figure 2 shows the comparison of correlation predictions and experimental data for small column diameter.





It can been seen that for the column diameter equal to 0.172 m, the model can predict the axial velocity profile at different superficial gas velocity with reasonable accuracy. One can clearly see that the axial liquid velocity becomes steeper with the increase in superficial gas velocity, and the model predicts the point of zero velocity well. For the column diameter is as big as 0.6m, comparison of correlation predictions and experimental data is plotted in Figure 3, from which it is evident that the similar predictions as that represented in Figure 2 are observed.



Figure 3: Comparison of model prediction with the data of Menzel et al. (1990), Non-coalescence



Figure 4: Comparison of model with the data from HPA(Heat Pulse Anemometry), Degaleesan, 1997

Figure 4 shows the correlation predictions of the data observed by Heat Pulse Anemometry techniques, and the comparison is good.

From Figure 2-4, it is obvious that the correlation can predict the axial velocity profile of the experimental data within a range of conditions. This establishes that the proposed correlation could be used to predict axial velocity profile.

In order to compare the developed correlation with the one dimensional model (Kumar, 1994, Luo, 1991), both predictions of the proposed model and the 1D model are plotted in Figure 5 and Figure 6 for comparison with experimental data.





One can see that the proposed model is in reasonable agreement with the one dimensional model prediction. However, it predicts the time-averaged velocity profile at superficial gas velocity of 0.17 m/s better than the 1D model prediction of Luo and Svendsen (1991).

It should be mentioned that the data in these figures are not used in developing the correlation. In addition, with n and c developed under pressurized conditions, it may be possible for the model to predict the axial velocity profile in bubble columns operating at pressurized conditions. But this needs to be confirmed.





4. Summary

An existing correlation for prediction of the axial velocity profile is modified by using the correlations for the gas holdup n and c developed by Wu et al. (2001). The modified correlation can predict the experimental data reported in the literature well within a range of conditions. The correlation is simple and is easy to use as it requires an input only the superficial gas velocity, physical properties and column dimensions. It can be readily used for prediction of axial velocity profile over a range of conditions, which should help the process engineers assess convective liquid mixing in bubble column rapidly.

Nomenclature

c	Parameter in $Eq(2)$
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- D Column diameter, m
- Fr_g Gas Froude number, dimensionless
- g Acceleration due to gravity, m/s^2
- Mo_L Liquid Morton number, dimensionless
- n Parameter in Eq(2)
- N Parameter in Eq(1)
- r, R Column radius, m
- Re_G Reynolds number, dimensionless

- U_{Sg} Superficial gas velocity, m/s
- V_c Liquid circulation velocity, m/s
- $V_L(r)$ Axial liquid velocity profile, m/s
- V_{Lo} Axial liquid velocity in the center of the column, m/s

Greek letters

- $\varepsilon(r)$ Radial gas hold up profile
- $\overline{\varepsilon}_{G}$ Cross-sectional average gas hold up
- $\tilde{\varepsilon}_{G}$ Radial average gas holdup
- μ_L Liquid viscosity, Pa.s
- ρ_G Gas density, kg/m³
- ρ_L Liquid density, kg/m³
- σ_L Liquid surface tension, N/m

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