ENGINEERING DEVELOPMENT OF SLURRY BUBBLE COLUMN REACTOR (SBCR) TECHNOLOGY

Quarterly Technical Progress Report No. 13

For the Period 1 April – 30 June 1998

FINAL

Contractor
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Prepared for the United States Department of Energy
Under Cooperative Agreement No. DE-FC22-95PC95051
Contract Period 3 April 1995 – 2 April 2000

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Contract Objectives

The major technical objectives of this program are threefold: 1) to develop the design tools and a fundamental understanding of the fluid dynamics of a slurry bubble column rector to maximize reactor productivity, 2) to develop the mathematical reactor design models and gain an understanding of the hydrodynamic fundamentals under industrially relevant process conditions, and 3) to develop an understanding of the hydrodynamics and their interaction with the chemistries occurring in the bubble column reactor. Successful completion of these objectives will permit more efficient usage of the reactor column and tighter design criteria, increase overall reactor efficiency, and ensure a design that leads to stable reactor behavior when scaling up to large diameter reactors.

Summary of Progress

Task 2: Component Diagnostics Development

Bubble Size Measurement

An optical fiber probe was developed to measure the initial bubble size from a single nozzle and bubbling-jetting transition in liquid-solid suspensions. This work uses the previously developed bubble size probe to provide information on bubble formation. It will be extended to study liquid-solid systems, as well as gas-liquid systems.

(The Ohio State University)

Task 3: Model Selection and Development

Estimation of Eddy Diffusivities

A method for estimating turbulent eddy diffusivities was developed using the existing database for scaleup and design of bubble columns. This was implemented by using the cross-sectional averaged axial and radial turbulent eddy diffusivities calculated from the Computer Automated Radioactive Particle Tracking (CARPT) measurements in air-water in three column sizes (14, 19, and 44 cm diameters). The correlations were then developed by generalizing these methods. These correlations represent a preliminary attempt to describe the effects of scale and superficial gas velocities on the turbulent diffusivities because of the limited amount of data.

Column Design

The scaleup procedures for the gas holdup and liquid recirculating velocity which were developed previously (see the January – March 1998 quarterly report), along with the newly developed eddy diffusivities estimation procedure can be useful as design tools. A methodology has been proposed to estimate the mean liquid recirculation velocity and the turbulent eddy diffusivities for systems of industrial interest from more easily obtained laboratory data taken in air-water systems. This is a preliminary effort, and more data are needed. As discussed below (Task 6), this methodology has been used successfully in interpreting tracer data from the Alternative Fuels Development Unit (AFDU). This application provides at least indirect verification of the methodology.

(Washington University in St. Louis)

Task 4: SBCR Experimental Program

Initial Bubble Formation

Bubbles drive the flow in bubble columns. It is quite likely that flow conditions are determined, in part, by the way in which bubbles are formed. This effect is strong in the region of the column before flow is fully developed and at moderate gas flow rates. Although there is much work describing bubble formation, there is little work at high pressures and less involving the presence of solids. The newly developed optical probe was used to study the effects of particles and pressure on bubble formation in liquid-solid suspensions.

1. The particles have a significant effect on the initial bubble size and motion of bubbles. At identical operating conditions, the bubbles formed in the liquid-solid suspensions are larger than those in the liquid-only suspensions. The bubble size increases with an increase in solids holdup.
2. The effect of pressure on the initial bubble size was studied for the constant flow regime and was found to be insignificant for this condition. Standard analysis of bubble formation identifies three different flow regimes: constant flow, intermediate and constant pressure. The effect of pressure could be significantly different in each regime. The constant pressure regime is generally the condition of industrial importance. We plan to measure the effect of pressure in the constant pressure regime in the next quarter.

(The Ohio State University)

Task 6: Data Processing

Prediction of Mixing Times from Tracer Studies

As discussed in previous reports, a fundamental, two-dimensional convection-diffusion model has been developed. This model has been used to interpret the liquid-phase tracer data taken at the LaPorte AFDU during methanol synthesis. The model parameters were obtained, based on the new scaleup methodology (see Task 3). Since CARPT measurements on air-water systems at atmospheric pressure and the estimated radial gas holdup profile at LaPorte were used to estimate parameters from laboratory data, the methodology allows calculation of flow characteristics for industrial situations from laboratory data.

The gas holdup profile at LaPorte was estimated from the Nuclear Gauge Densitometry and the pressure drop measurements taken during the run. The model can provide an estimate of the internal liquid mixing in bubble columns. The values of mixing time for the AFDU were estimated from the model using the new methodology. The mixing times were also inferred from tracer measurements during the trial. The values agree well. Thus, we conclude that this scaleup procedure results in fairly good predictions of the characteristic mixing times within the column, as measured by the radiation detectors at various axial locations. This provides an indirect verification of the model.

(Washington University)

Turbulence Parameters

Analysis of laboratory data for two subjects has been completed. The findings from both these studies will be organized in a report format for the next quarterly report. The two areas are:

1. estimation of the liquid phase turbulent mixing length from CARPT and CT measurements for use in CFD (Computational Fluid Dynamics) simulations of flows in bubble columns. (An estimate of mixing length is needed for the simplest closures for turbulence models used in most CFD solvers, such as the CFDLIB code developed at Los Alamos.)
2. comparison of time-averaged liquid/slurry “turbulent” parameters in gas-liquid and gas-liquid-solid slurry bubble columns.

(Washington University)

The Ohio State University Research

The report from Ohio State University for the period follows:

INTRINSIC FLOW BEHAVIOR IN A SLURRY BUBBLE COLUMN UNDER HIGH PRESSURE AND HIGH TEMPERATURE CONDITIONS

Quarterly Report

(Reporting Period: April 1 to June 30, 1998)

Highlights

- An optical fiber probe was developed to measure the initial bubble size from a single nozzle and bubbling-jetting transition in liquid-solid suspensions. The signals of light intensity from the probe were analyzed to study the bubbling-jetting transition.

- The effects of particle and pressure on bubble formation in liquid-solid suspensions were studied. The presence of particles in liquid had a significant effect on the initial bubble size and motion of bubbles. At identical operating conditions, the bubbles formed in the liquid-solid suspensions were larger than those in the liquid. The bubble size increased with an increase in solids holdup.

- The effect of pressure on the initial bubble size may vary significantly with bubble formation conditions dictated by constant flow condition, intermediate condition and constant pressure condition. In this work, the bubble formation was under the constant flow condition. It was found that the effect of pressure on initial bubble size was insignificant for the constant flow condition.

Work Conducted

Measurement of Initial Bubble Size and Bubbling-Jetting Transition

The optical fiber probe system discussed in the previous monthly report was used for the detection of bubbles or jets in liquid-solid suspensions at high pressures.

To establish objective criteria for the bubbling-jetting transition in liquid-solid suspensions, experiments were first performed in a liquid system over a wide range of orifice Reynolds numbers ($Re_{o,g}=\rho_g d_0 u_0/\mu_g$). Figure 1 shows a series of photographs of the gas flow through the orifice at various $Re_{o,g}$. At $Re_{o,g}$ = 1,075 (case E1), single bubbles are formed from the orifice. With increasing $Re_{o,g}$ to 5,321 (case E2), bubbles being formed at the orifice start to interact with the preceding ones. Bubble coalescence occurs between the two bubbles, sometimes involving more bubbles. In case E3 ($Re_{o,g}$ = 8,809), frequent coalescence of successive bubbles is observed. A jet-like gas plume appears in the photo, which rarely breaks up near the orifice. Case E3 marks the beginning of the bubbling-jetting transition. In cases E4, E5, and E6, it is clear from the photos that the flow is in the jetting regime. Bubbles of various sizes break away from the top of the jets in this regime. It can also be seen that the jet penetration depth increases with an increase in $Re_{o,g}$.

The signals of light intensity from the probe corresponding to cases E1, E2, E3, and E6 are shown in Figure 2(a), along with their power spectra [Fig. 2(b)]. The signal for E1 shows sharp and discrete peaks in time domain. The shape of the peaks is regular, and the height and width of the peaks are uniform. Distinct peaks also appear in frequency domain, with the bubbling frequency equal to the dominant frequency. When bubble interactions start (E2), most apparent peaks in the signal include several auxiliary peaks, corresponding to interacting bubbles. Most peaks in this case are regular in shape, height, and width, although some irregular peaks appear, which induce the widening of the frequency spectrum. However, the range of the dominant frequencies is still relatively narrow. The signal for E3 loses almost all the regularities in shape, height, and width of the peaks. The corresponding power spectrum also shows a wide distribution of dominant frequencies. All these characteristics signify the formation of jetting. The characteristics of the signals are similar once the flow is under the jetting condition, i.e., irregular in shape, height, and width of the peaks in the signal and a wide range of dominant frequencies or disappearance of dominant frequencies, as in case E6.

Effect of Particles on Initial Bubble Size in Liquid-Solid Suspensions

For high-pressure systems, some studies in the literature examine the initial bubble size in liquids, but little is known about the initial bubble size in the presence of particles. However, a reasonable estimation of the initial bubble size is important for the design of slurry bubble column reactors in which catalytic particles are present.

Our experiments found that the presence of particles in liquid has a significant effect on the motion of bubbles. Flow visualization of bubble formation in the liquid, with or without continuous liquid flow, reveals that all the bubbles follow the same trajectory. Once the probe is properly aligned with the orifice, the trajectory of the bubbles intercepts the tip of the probe, yielding consecutive steady peaks in the light intensity signals in the bubbling regime. However, the bubbles in liquid-solid suspensions have varied trajectories. The probe can only detect bubbles periodically. Flow visualization also confirms that the bubbles emerge from the bed surface at different locations. The unsteady bubble trajectories in liquid-solid suspensions are due to the heterogeneous nature of the suspension.

Figure 3 shows the effect of particles on the initial bubble size. The bubble size is obtained at the same distance above the orifice in the liquid and the liquid-solid suspension. Various solids holdups in the suspension are obtained by varying the fluidizing liquid velocity. The fluidizing liquid velocity is so small that the superficial liquid flow does not significantly alter the bubble formation behavior. At both ambient and 4.2 MPa pressures, the bubbles formed in the liquid-solid suspension are larger than those formed in the liquid for a given u_0. The bubble size increases with an increase in solids holdup. The experimental data of Massimilla et al. (1961) showed a similar trend. The experimental data shown in the figure clearly indicate that models or correlations obtained in liquid will significantly underestimate the initial bubble size in liquid-solid media.

Effect of Pressure on Initial Bubble Size in Liquid-Solid Suspensions

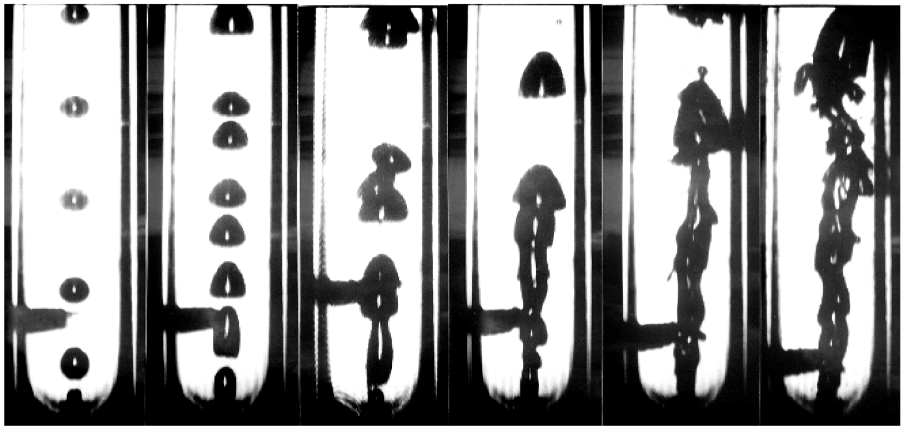
Numerous experimental and modeling studies have been conducted over the past decades on bubble formation from a single orifice or nozzle submerged in liquids, mostly under ambient conditions. Only a few studies were conducted at elevated pressures. The high-pressure studies indicate that an increase in gas density reduces the size of bubbles formed from single orifices. However, these results were limited to water systems only. The effect of pressure on the initial bubble size in hydrocarbon liquids systems is not understood. Furthermore, it is known that the volume of the gas chamber connected to the nozzle is an important factor in determining the initial bubble size. Clearly, the effect of pressure on initial bubble size may vary significantly with the bubble formation conditions dictated by three conditions, i.e., constant flow, intermediate and constant pressure conditions. In this work, the volume of the gas chamber is zero and the bubble formation can be considered as under constant flow conditions.

Our work shows that the effect of pressure on initial bubble size is insignificant in the liquid-solid suspension, as well as in the liquid (as shown in Fig. 4), under constant flow conditions. Increasing pressure does not significantly change the bubble sizes for a given solids holdup, orifice gas velocity, and temperature.

It is well known that the initial bubble size is determined based on the balance among various forces acting on the bubble formed at the nozzle. In a liquid, the upward forces include buoyancy and gas momentum forces; the downward forces include liquid drag, surface tension, bubble inertial force, and Basset forces. The presence of particles induces two additional downward forces on the bubble: particle-bubble collision force and liquid-solid suspension inertial force. The increase in the particle-bubble collision force and liquid-solid suspension inertial force with increasing solids holdup leads to an increased initial bubble size. Considering all the forces, a mathematical model has been developed to quantify the initial bubble size in liquid-solids suspensions. The model will be described in future monthly reports. This model reveals that for the current experimental system, the effect of pressure on the overall upward forces and overall downward forces is comparable, leading to an insignificant net effect of pressure on the initial bubble size.

Reference

Massimilla, L., A. Solimando, and E. Squillace, *British Chemical Engineering*, April, 233 (1961).



(a) E1 (b) E2 (c) E3 (d) E4 (e) E5 (f) E6

Figure 1. A series of photographs showing the bubbling-jetting transition (P = 4.24 MPa, T = 28ºC). (a) E1: u_0 = 0.27 m/s, $Re_{o,g}$ = 1075; (b) E2: u_0 = 1.35 m/s, $Re_{o,g}$ = 5321; (c) E3: u_0 = 2.23 m/s, $Re_{o,g}$ = 8809; (d) E4: u_0 = 2.60 m/s, $Re_{o,g}$ = 10243; (e) E5: u_0 = 3.99 m/s, $Re_{o,g}$ = 15759; (f) E6: u_0 = 6.42 m/s, $Re_{o,g}$ = 25355.

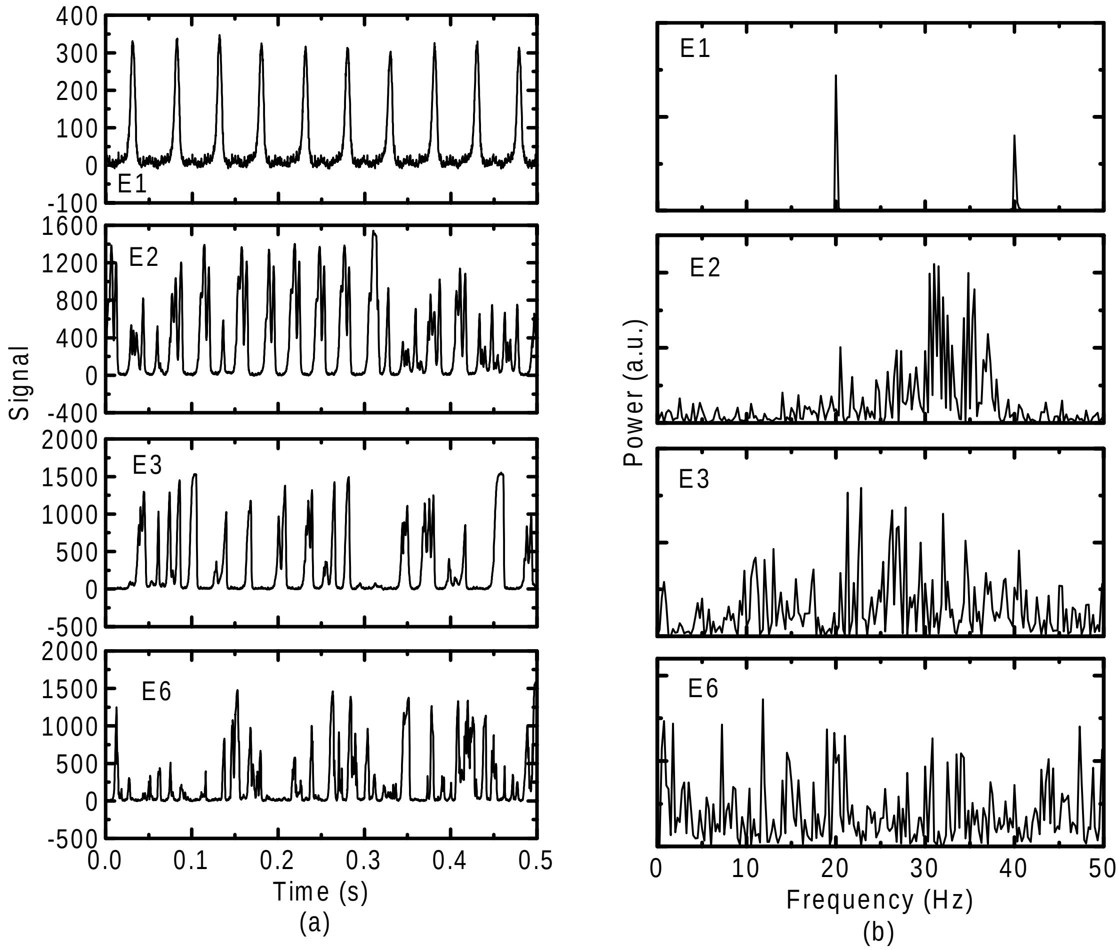


Figure 2. (a) Typical signals and (b) corresponding power spectra.

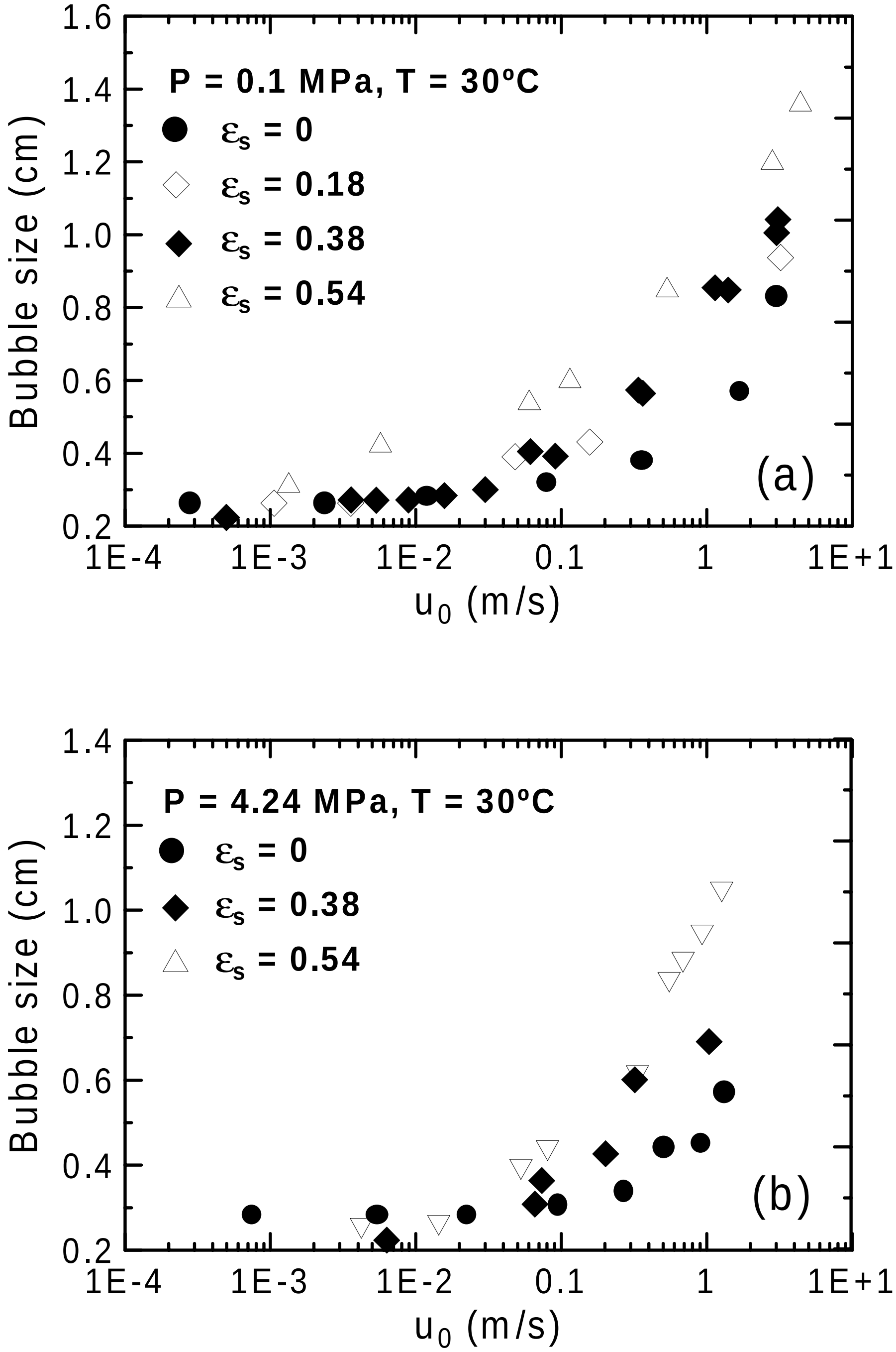


Figure 3. Effect of particles on the initial bubble size at various orifice gas velocities: (a) P = 0.1 MPa; (b) P = 4.24 MPa.

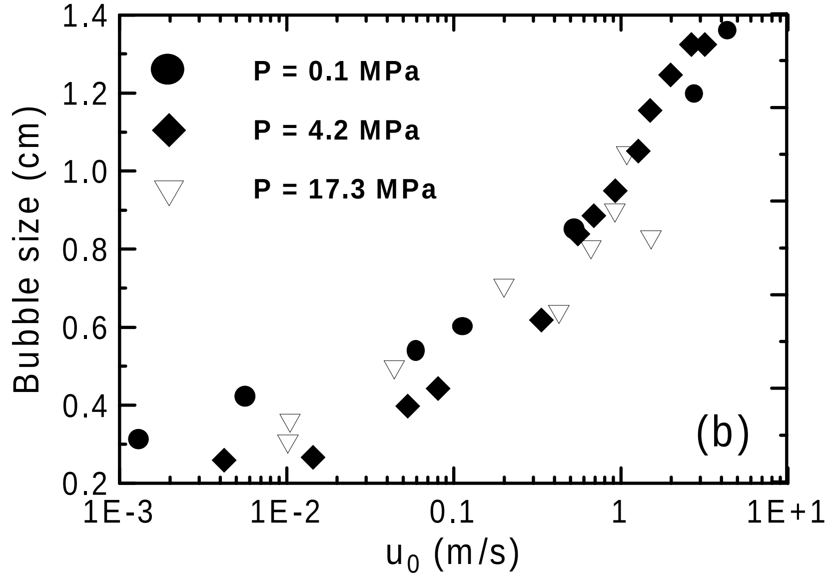


Figure 4. Effect of pressure on the initial bubble size in liquid and liquid-solid suspensions under constant flow conditions: (a) solids holdup = 0; (b) solids holdup = 0.54.

Washington University in St. Louis

The report for Washington University for the period follows.

ENGINEERING DEVELOPMENT OF SLURRY BUBBLE COLUMN REACTOR (SBCR) TECHNOLOGY

Thirteenth Quarterly Report
for
April 1 - June 30, 1998

(Budget Year 3: October 1, 1997 – September 30, 1998)

Submitted to

Air Products and Chemicals

Contract No.: DE-FC 22 95 PC 95051

Chemical Reaction Engineering Laboratory
Chemical Engineering Department
Washington University

Objectives for the Third Budget Year

The main goal of this subcontract from the Department of Energy via Air Products to the Chemical Reaction Engineering Laboratory (CREL) at Washington University is to study the fluid dynamics of slurry bubble columns and address issues related to scaleup and design. The objectives for the third budget year (October 1, 1997 – September 30, 1998) were set as follows:

- Further development of phenomenological models for liquid and gas flow.
- Testing of the models against available data from the LaPorte AFDU.
- Evaluation of turbulent parameters in 18-inch-diameter columns, with and without internals, using collected CARPT data in these columns.
- Development of relationships between fundamental and simpler practical models for industrial use.
- Further improvement in fundamental computational fluid dynamics models and testing of the models against the CARPT/CT data.
- Preliminary assessment of differences in gas-liquid and gas-liquid-solid systems.
- Testing the effect of the gas distributor on flow patterns.

In this report, the research progress and achievements accomplished in the thirteenth quarter (April 1 - June 30, 1998) are discussed.

Outline of Accomplishments

- **Scaleup Procedure for Turbulent Eddy Diffusivity**

  Correlations for the estimation of turbulent eddy diffusivities are developed based on the existing limited database for scaleup and design of bubble columns. The cross-sectional averaged axial and radial turbulent eddy diffusivities obtained by the Computer Automated Radioactive Particle Tracking (CARPT) in air-water in three column sizes (14, 19 and 44 cm diameters) are used to develop these correlations. Because of the limited number of data points available, the developed correlations represent a preliminary attempt to describe the effects of scale and superficial gas velocities on the turbulent diffusivities. The developed scaleup procedure for the gas holdup, liquid recirculating velocity (reported in the 12th quarterly report) and eddy diffusivities enables the estimation of these parameters in systems of industrial interest. Accordingly, such a procedure allows us to utilize the developed fundamental two-dimensional convection-diffusion model to interpret the liquid tracer data obtained in the AFDU at LaPorte.

- **Interpretation of the Liquid Phase Tracer Data During Methanol Synthesis at the LaPorte AFDU using the Fundamental Two-Dimensional Convection-Diffusion Model**

  The fundamental two-dimensional convection-diffusion model has been developed to interpret the liquid phase tracer data taken at the LaPorte AFDU during methanol synthesis. Based on the developed scaleup methodology, model parameters were obtained from CARPT measurements using as input the estimated radial gas holdup profile at LaPorte. The gas holdup profile at LaPorte was estimated from the Nuclear Gauge Densitometry and pressure drop measurements. The results show that the model provides a good representation of the internal liquid mixing in bubble columns. The developed scaleup procedure for evaluating the model parameters in the AFDU slurry bubble column reactor during methanol synthesis results in fairly good predictions of the characteristic mixing times within the column, as measured by the radiation detectors at various axial locations.

  During this quarter, the work has been completed on i) estimation of the liquid phase turbulent mixing length from CARPT and CT measurements for use in simulation with the CFDLIB code and ii) comparison of time-averaged liquid/slurry “turbulent” parameters in gas-liquid (G-L) and gas-liquid-solid (G-L-S) slurry bubble columns. However, the findings have not yet been organized in a report format. This will be accomplished during the next quarterly report.

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1. Scaleup Procedure for the Turbulent Eddy Diffusivity

Studies of the effects of equipment scale and operating conditions on fluid dynamic parameters using experimental data obtained by Computer Automated Radioactive Particle Tracking (CARPT) and Computed Tomography (CT) and from the literature will aid in the design and scaleup of bubble column reactors. Our specific focus is on utilizing the available hydrodynamic information to model liquid mixing in bubble columns in the churn-turbulent flow regime. In this regard, the fluid dynamic parameters of interest are the gas holdup and radial holdup profile, the liquid recirculation velocity and liquid turbulence, which can be quantified by the turbulent eddy diffusivities.

Therefore, a scaleup methodology for gas holdup, liquid recirculating velocity and turbulent eddy diffusivity has been developed. The aim is to develop a basis for the approximate characterization of churn-turbulent bubble columns that enables the estimation of these fluid dynamic parameters in industrial-scale units based on measurements of these parameters in air-water atmospheric systems. This should enable the use of the developed two-dimensional convection-diffusion model to interpret liquid phase tracer data taken at the Alternative Fuels Development Unit (AFDU) at LaPorte, Texas during methanol synthesis.

In the last quarterly report (no. 12) the scaleup procedures developed for the gas holdup and liquid recirculating velocity were reported. Here, a scaleup procedure for the turbulent eddy diffusivities is outlined.

1.1 Turbulent Eddy Diffusivities

The cross sectionally averaged axial and radial turbulent eddy diffusivities are defined, respectively, as

$$\overline{D}_{zz} = 2\int_0^1 D_{zz}(\xi)\xi d\xi \tag{1.1}$$

$$\overline{D}_{rr} = 2\int_0^1 D_{rr}(\xi)\xi d\xi \tag{1.2}$$

In this case, the current results from CARPT data in an air-water system and three column sizes, 14, 19, and 44 cm, are considered. The data points shown in Figures 1.1 and 1.2 for D_{zz} and D_{rr}, respectively, are limited in their range of superficial gas velocity, U_g. Therefore, this represents only a preliminary attempt at scaling (extrapolating) D_{rr} and D_{zz}, and needs to be substantiated with further experimental data at higher gas velocities, especially in the largest diameter (44 cm) column.

The following dependencies have been observed for $\overline{D}_{zz}$ and $\overline{D}_{rr}$ (based on CARPT data), and apply to large-diameter columns (>10 cm) in the churn-turbulent flow regime (U_g >5).

$$\overline{D}_{zz}(cm^2/s) = -\frac{2325}{D_c^{0.8}} + 106.6 D_c^{0.3} U_g^{0.3} \quad (1.3)$$

$$\overline{D}_{rr}(cm^2/s) = -\frac{350}{D_c^{0.8}} + 13.0 D_c^{0.3} U_g^{0.3} \quad (1.4)$$

It is emphasized again that, due to the limited number of data points available, these equations represent only a preliminary assessment of the effects of scale and superficial gas velocity on turbulent diffusivities.

CARPT results for the average radial and axial eddy diffusivities in the churn-turbulent flow regime indicate that the radial profiles of the turbulent diffusivities can be approximately expressed as follows:

$$D_{zz}(\xi) = \overline{D}_{zz} P_4$$

where $P_4 = -3.4979\xi^4 + 3.2704\xi^3 + 0.4693\xi^2 + 0.005035\xi + 0.5847$ (1.5)

$$D_{rr}(\xi) = \overline{D}_{rr} P_2$$

where $P_2 = -5.0929\xi^2 + 5.0717\xi + 0.1653$ (1.6)

P_4 and P_2 are fourth-order and second-order polynomials that are independent of gas velocity and column diameter. This is illustrated in Figures 1.3 and 1.4, which show the profiles evaluated using Equations 1.5 and 1.6. The reasonably good comparisons suggest that Equations 1.5 and 1.6 in combination with Equations 1.3 and 1.4 can be used to estimate the profiles for the axial and radial eddy diffusivities as a function of column diameter, D_c, and superficial gas velocity, U_g, in air-water bubble columns operating in the churn-turbulent flow regime.

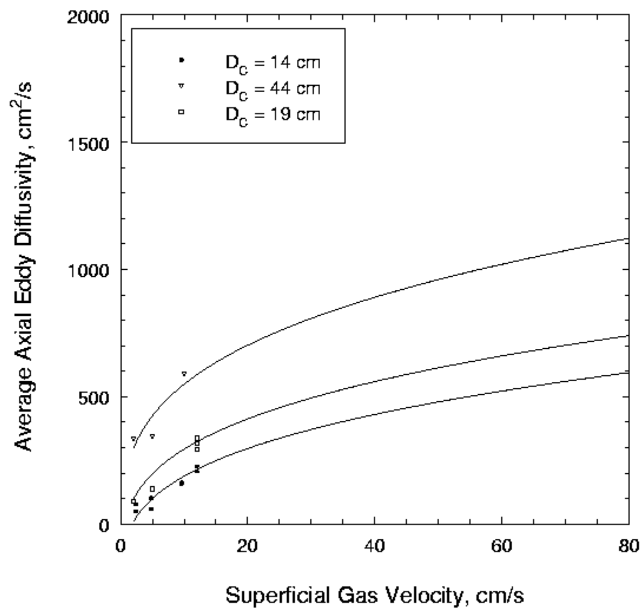

Figure 1.1 Effect of Superficial Gas Velocity and Column Diameter on the Average Axial Eddy Diffusivity

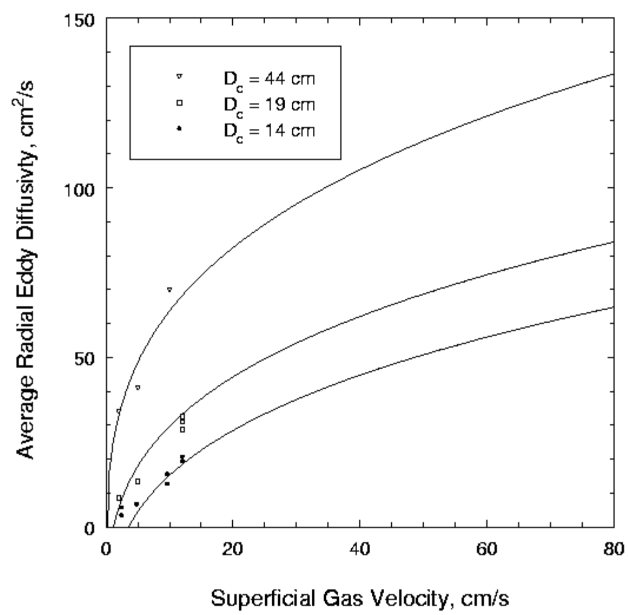

Figure 1.2 Effect of Superficial Gas Velocity and Column Diameter on Average Radial Eddy Diffusivity

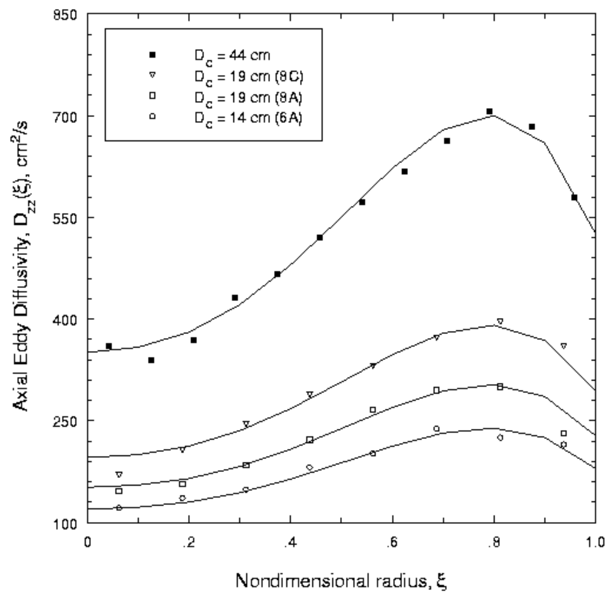

Figure 1.3 Radial Profile (P_4) of the Axial Eddy Diffusivity

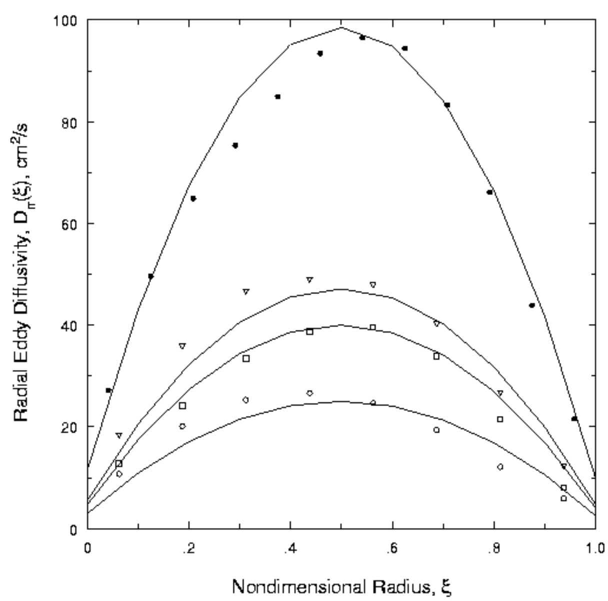

Figure 1.4 Radial Profile (P_2) of the Radial Eddy Diffusivity