NOVEL TECHNIQUES FOR SLURRY BUBBLE COLUMN HYDRODYNAMICS

FINAL TECHNICAL REPORT
June 1, 1995 - July 30, 1998
(+ 9 Months No-Cost Extension)

EXECUTIVE SUMMARY

The objective of this cooperative University-Industry research effort between Washington University (WU), Ohio State University (OSU) and Exxon Research and Engineering Company (ER&E) was to improve the basis for scale-up and operation of slurry bubble column reactors for syngas conversion and other coal conversion processes by increased reliance on experimentally verified hydrodynamic models.

The emphasis during this first year of this three year project was placed on developing the experimental techniques that are capable of providing accurate measurements of hydrodynamic quantities such as velocities and holdup distribution at operating conditions of interest. The accomplishments of Year 1 were described in the First Technical Annual Report and can be summarized as follows.

At Washington University (WU) the unique Computer Aided Radioactive Particle Tracking (CARPT) technique was modified and improved. The technique is capable of providing solids (or liquid) velocities at high gas or solids holdup where all other methods fail. Wavelet filtering of particle position versus time data was implemented, and this improved by an order of magnitude the accuracy of the turbulence parameters that the technique can provide. A Monte Carlo based modeling for calculation of detector responses for any position of the tracer was initiated. Data obtained with CARPT for liquid velocities was compared to the velocity estimates generated by heat pulse probe (HP), and it was shown that the later technique (HP) provides reliable local velocity information only in bubbly flows while it underestimates the velocities substantially during turbulent flow, i.e at high gas superficial velocity.

At Ohio State University (OSU) the limitations of the very powerful Particle Image Velocimetry (PIV) technique were explored and it was found that the technique has the potential of providing the full velocity and holdup field in columns of up to 4" in diameter. Experiments were initiated to compare PIV results in such a 4" column to the CARPT data. A new and useful probe for evaluation of local instantaneous heat transfer coefficient was developed at OSU and its capabilities demonstrated at ER&E.

At Exxon Research and Engineering (ER&E) preparations were made for making the high pressure 6" diameter bubble column available for CARPT and other measurements. Necessary modifications to the column were designed. Jointly with WU needed tracer particle strength was determined and a device for calibration was designed. In addition, CFDLIB codes for hydrodynamic modeling have been installed and a preliminary case

study of a two dimensional column was conducted. It was shown that experimental time averaged velocity profiles qualitatively agree with the model predictions.

The emphasis during this second year of the project was: i) on further refinement and development of experimental techniques needed in obtaining reliable flow dynamic parameters in bubble columns, ii) on verification of these techniques, iii) on development of phenomenological models for bubble columns, iv) on testing of fundamental computational fluid dynamics models in predicting liquid flow patterns in bubble columns and v) on preparing a facility for high pressure studies in a six inch diameter column. The accomplishments of Year 2 were described in the Second Annual Technical Report and can be summarized as follows.

The CARPT technique was fully validated by a joint effort between WU and OSU. The comparison of PIV and CARPT data, taken on the same 4" diameter column, revealed that the two techniques generate essentially the same liquid time averaged velocities and the same order of magnitude of turbulence parameters at low superficial gas velocity when gas holdup is low. At such conditions both techniques re reliable. At high gas velocities that lead to churn turbulent flow, the results produced by the two techniques start to diverge somewhat, but the discrepancies can be fully accounted for on physical grounds (e.g. too short an interval of collection of PIV data for proper averaging, errors in PIV due to enhanced gas holdup, inability of CARPT to capture small scales of turbulence). It was concluded that the two techniques complement each other well. The data obtained from CARPT was also compared to Laser Doppler Anemometry (LDA), and Hot Film Anemometry (HFA) data and reasonable agreement was found. This validated the use of CARPT in churn turbulent flows, which are of industrial interest, and in which other techniques cannot provide the desired information.

A light transmittance optical probe was developed at OSU and its usefulness in obtaining information on bubble passing frequencies in opaque flow was successfully demonstrated. An optical reflectance probe was developed by Delft University for the measurement of bubble sizes and was successfully tested at WU in bubbly flows.

Based on the developed information on gas holdup profiles by Computed Tomography (CT) and based on liquid velocity patterns observed by CARPT, an engineering type model was developed at WU to represent flow, and mixing of each phase (liquid and gas) and transport between phases in bubble columns. The features and governing equations of the model were reported as well as the relationship of model parameters to the existing knowledge base.

The Computational Fluid Dynamics (CFD) work at WU and ER&E resulted in successful simulation of the most often quoted and reported experiments in two-dimensional bubble columns which reveal a meandering structure and two streaks of downward moving vortices on each side of the meandering high holdup structure. It was also shown (WU and ER&E) that the meandering plume observed experimentally in a partially aerated column can be captured by CFD while using relatively simple closure forms.

The planned high pressure experiments in large diameter bubble columns (6") were rescheduled to be conducted at Washington University. This change was necessitated by the fact that it proved impossible for WU to conduct the originally planned radioactive particle experiments at Florham Park due to safety and licensing issues that were not foreseen. To remedy for this, ER&E provided the additional funds needed to design and operate a high pressure facility at WU. The design of this facility was completed during the second year of the project. At that point it also became clear that a nine month no cost extension was needed to install and test the new facility and conduct the planned work on it.

During the final year 3 of the contract, and during the 9 month no-cost extension period, the necessity of which was explained above, the focus of the research was to implement the high pressure facility, collect and interpret data at high pressure and perform a quantitative comparison between CFD model predictions and data. The accomplishments achieved during this final period of the contract are described in this report and are summarized below.

A high pressure 6" diameter column was installed and operated at Washington University. Computer Tomography (CT) measurements revealed that the effect of pressure on gas holdup profiles is negligible in bubbly flows at low superficial gas velocities. At high superficial gas velocities, at which the flow at atmospheric pressure is churn turbulent, increased pressure causes a flattening in gas holdup profiles and a rise in overall holdup. This is apparently caused by the ability to maintain bubbly flow at higher superficial gas velocities at elevated pressures and delay transition to churn turbulent flow. Available correlations that can predict the observed mean holdup at atmospheric and elevated pressures were identified and reported. The details of the holdup measurements are described in Chapter 3 of this report.

The Computer Automated Radioactive Particle Tracking (CARPT) runs at WU in a 6" diameter column at elevated pressure reveal the same single cell liquid recirculation pattern in the time averaged sense, with the liquid rising in the middle of the column and falling down by the walls. The distributor zone where entry effects are notable is, however, much expanded at higher pressure. The magnitude of liquid recirculation velocities in the fully flow developed region is considerably reduced at elevated pressure compared to the results obtained at the same gas superficial velocity at atmospheric pressure. Details are provided in Chapter 4 of this report.

The sophisticated Monte Carlo procedure for calibration of the detectors for the CARPT experiments and for identification of the particle position during the CARPT experiments has been completed at WU and successfully tested against experimental data. Details are provided in Chapter 5 of this report. The availability of the Monte Carlo technique assures more extensive use of CARPT in the future.

Fluid dynamic behavior of small diameter (up to 4") high pressure columns was studied extensively by PIV and other techniques at OSU. Valuable correlations were developed for the bubble rise velocity. The data base for bubble size distributions was augmented and it clearly establishes the narrowing of the bubble size distribution and the shift to smaller bubble sizes at elevated pressure. Sophisticated modeling, based on first principles, of the shape and rise velocity of a bubble in a solid-liquid suspension was successfully completed and good comparison obtained between experimental results and model predictions. A comprehensive description is provided in Chapter 6 of this report.

Our combined effort (WU and ER&E) to describe the flow field in the whole bubble column by computational fluid dynamics (CFD) focused on the use of Euler-Euler k-fluid model in the framework of the CFDLIB code of Los Alamos. Only the Euler-Euler based code was deemed practical for description of the flow field in the whole column. In this project we wanted to demonstrate that Euler-Euler approach is capable of predicting experimental observations even at low gas holdup in bubbly flow, which is the most severe test of the model since by definition the Euler-Euler approach is ill-suited for predicting the flow field caused by a single bubble or at vanishingly small gas holdup. At such low holdup conditions the predictions of CFDLIB (WU and ER&E) were compared to the best PIV data available (OSU) in two dimensional beds. Very good comparison was obtained between predicted and measured time averaged liquid velocities. All the observed dynamic features of all the large structures of the flow were predicted well. The computed turbulence parameters were of the same order of magnitude and exhibited the same trends as data and the existing discrepancies can be fully rationalized based on physical arguments. This successful verification of CFD Euler-Euler approach for bubbly flow provides an increased motivation for its implementation for 3D simulations and for churn-turbulent flows. The detailed comparison between CFD predictions and PIV data in 2D columns is presented in Chapter 7 of this report.

1. INTRODUCTION AND MOTIVATION

Synthesis gas from coal is one of the most abundant and reliable sources of energy and chemicals. From the heat transfer and high volumetric productivity viewpoints, bubble columns (BC) and slurry bubble columns (SBC) operated at high gas velocities in churn turbulent flow regime are the reactors of choice for a variety of synthesis gas conversion processes.

There is hardly any data in the flow regime, neither are there any fundamentally based design and scale-up procedures. In response to this need the Department of Energy (DOE) initiated the Fluid Dynamics Initiative which focuses on advancing the state of the art in understanding the fluid dynamics of bubble columns/slurry bubble columns and replacing empirical design methods with a more rational approach.

As part of this initiative DOE awarded the DE-FG22-95 PC 95212 grant for a collaborative effort between the Chemical Reaction Engineering Laboratory (CREL - Washington University (WU), Ohio State University (OSU) and Exxon Research and Engineering (ER&E) to advance the understanding of the hydrodynamics of BC/SBC with the overall objectives to:

i) develop and verify accurate experimental techniques for obtaining important fluid dynamics parameters,
ii) generate database and expand it to high pressure systems,
iii) develop a set of phenomenological models and correlations to utilize this database,
iv) utilize the data to verify the fundamental computational fluid dynamics (CFD) models by testing different closure forms.

This grant enabled a unique integration of the expertise of the two universities (WU and OSU) and industry (ER&E) toward achieving the goals set by the DOE Hydrodynamics Initiative. This study complemented well the work in progress at CREL-WU and OSU, Conract NO. DE-FC22-95 PC 95051, related to the LaPorte Advanced Fuels Demonstration Unit (AFDU) operated by Air Products with Department of Energy funding.

1.1. Bubble Columns (BC/Slurry Bubble Columns (SBC)

A bubble column (BC), shown in Figure 1.1, is a cylindrical vessel in which gas is sparged through a batch liquid or a cocurrently or countercurrently flowing liquid. The gas superficial velocities are order of magnitude larger than those of the liquid so that it is the gas that governs the fluid dynamics of the system. Small slurry particles in case of slurry bubble column (SBC), follow the liquid. It is well known that fluid dynamics (phase velocities and holdup distribution) affect the phase mixing and transport between the phases (gas-liquid interfacial area, transport coefficients) and, hence, to a large extent affect the 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 as illustrated in the flow chart shown in Figure 1.1. Thus, quantitative knowledge of fluid dynamics is essential for rational and predictive reactor design and scale-up.

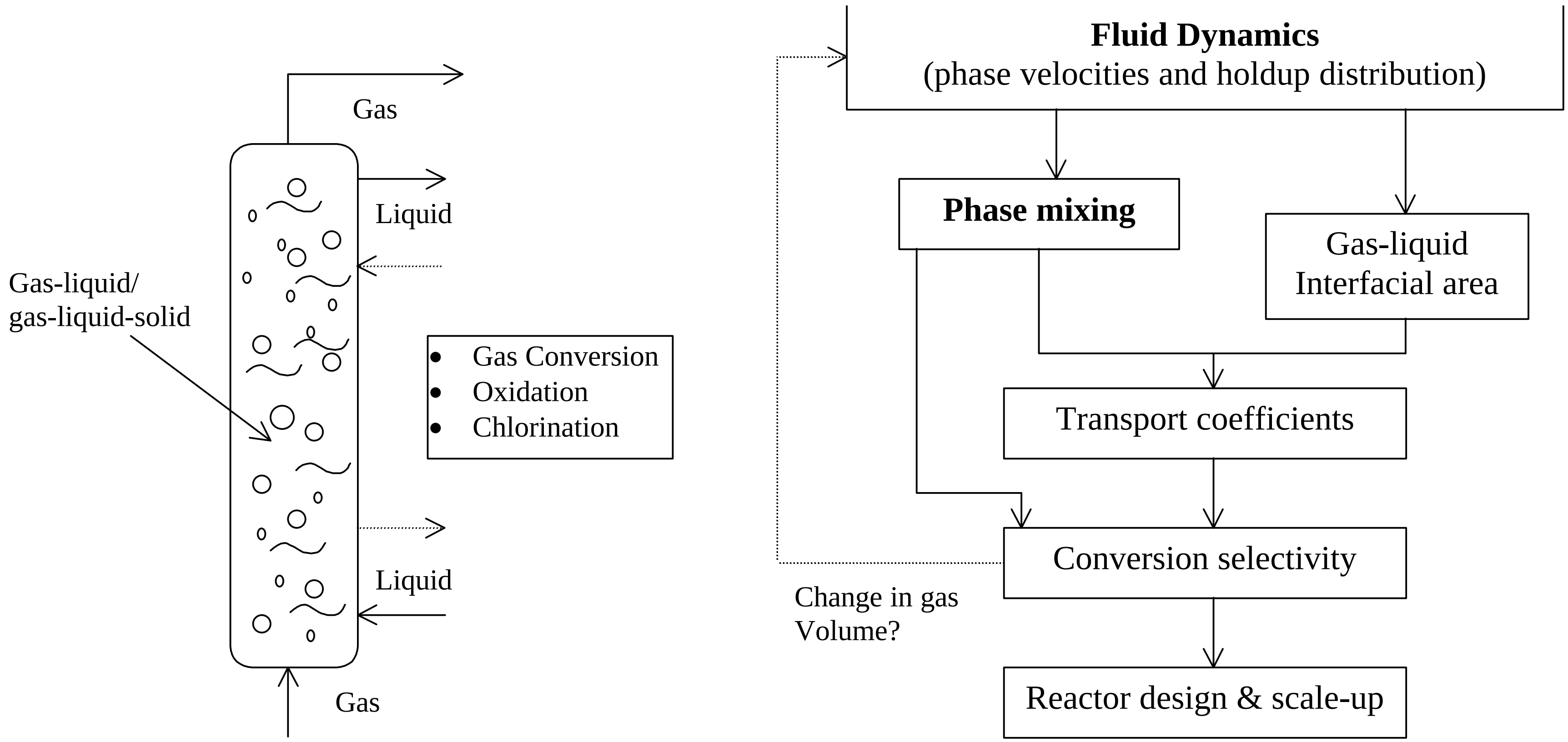


Figure 1.1: Bubble Columns and Slurry Bubble Columns

1.2. Focus of the Investigation

The many phenomena occurring in BC/SBC, as illustrated in Figure 1.2, are very complex due to the intrinsic dynamic behavior of the dispersed phase motion and associated interactions. Physical and thermodynamic properties, operating variables and design parameters affect the fluid dynamics and transport phenomena in the column which together with kinetics determine the reactor performance (conversion and selectivity).

Our goal is to provide the macro scale description of the fluid dynamics phenomena in BC/SBC and to attempt to relate the micro scale and the effects of various operating and design variables on the macro scale circulation, mixing and transport.

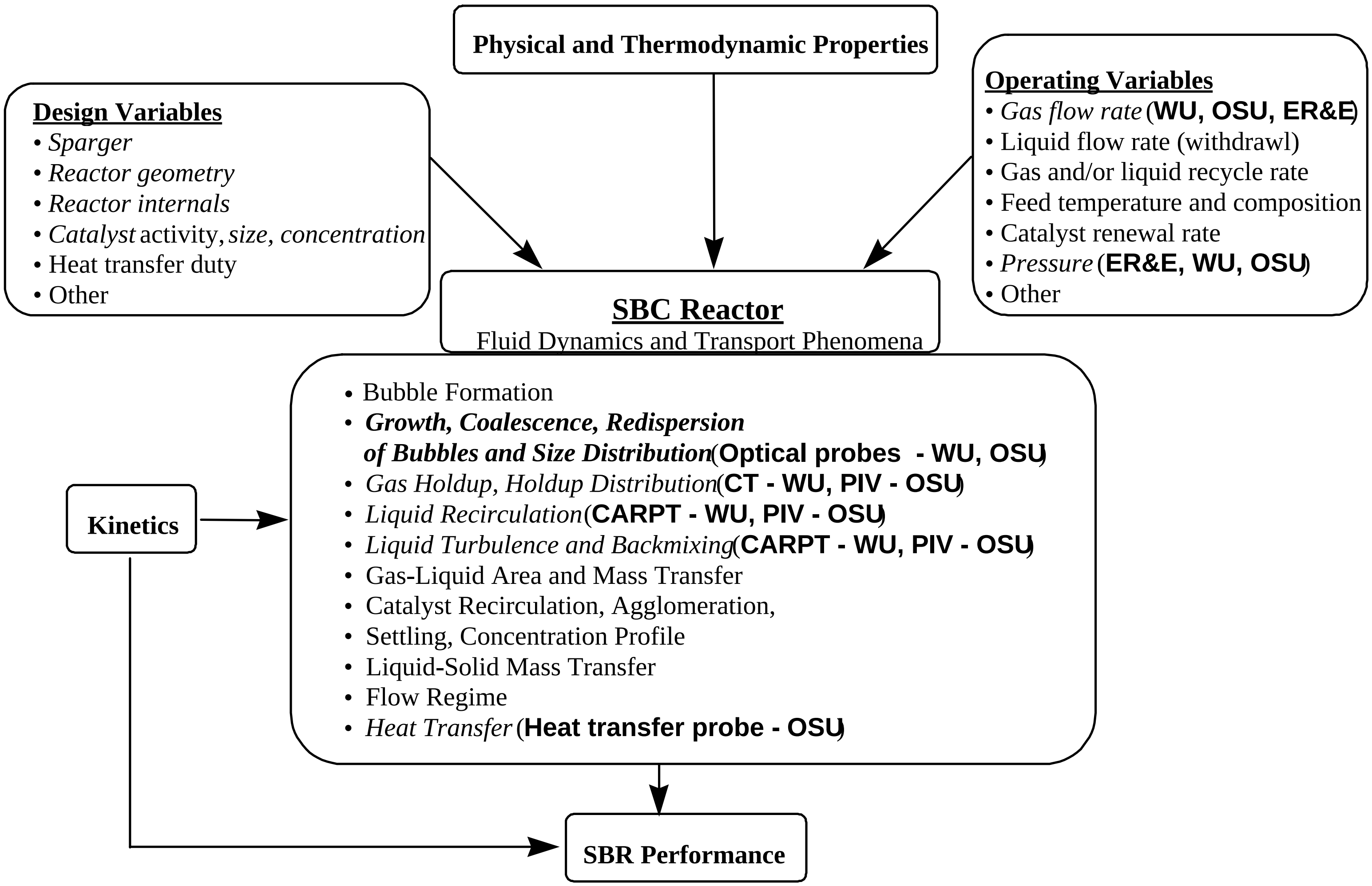
The investigated parameters and the techniques used to study the macro scale hydrodynamics under this grant are illustrated in Figure 1.2. This figure demonstrates also the integration of the expertise of Washington University, Ohio State University and Exxon Research & Engineering in studying the fluid dynamics of bubble columns.

According to the main objectives of this grant, the following phenomena and variables need to be investigated (Figure 1.2) and quantified:

- Holdup distribution via Computer Tomography, CT - (WU) and Particle Image Velocimetry (PIV) - (OSU).
- Liquid circulation via Computer Automated Radioactive Active Particle Tracking, CARPT - (WU) and PIV - (OSU).
- Liquid turbulence and backmixing via CARPT - (WU) and PIV - (OSU).
- Bubble size distribution and bubble rise velocity via optical probes - both reflectance (WU) and transmittance (OSU) probes.
- Effect of gas flow rate and reactor pressure on the fluid dynamics (WU, ER&E, OSU).

Along the process of developing and verifying the advanced experimental techniques used in this research, the database for the fluid dynamic parameters is expanded and utilized to build phenomenological models, correlations and also test fundamental fluid dynamics models. Along these lines we have focused on the following:

- Development of a phenomenological model for gas phase mixing (WU).
- Testing different closure forms by comparing the predictions of computational fluid dynamics models (CFD) with the data obtained by the above techniques (WU, ER&E).



*** Closure schemes for CFD simulations validated against experimental data from above techniques.**

Figure 1.2: The Phenomena Occurring in Bubble and Slurry Bubble Columns

2. OBJECTIVES

2.1. Review of the Overall Objectives and Accomplishments

The overall objectives of this cooperative University (Washington University and Ohio State University) - Industry (Exxon Research and Engineering) research was to improve the basis for scale-up and operation of bubble column/slurry bubble column reactors (BC/SBC) by an increased reliance on phenomenologically or fundamentally based hydrodynamic models which are experimentally verified. The research under this grant focused at providing experimental tools for measurement of important fluid dynamic quantities.

Specifically the following were set:

Task 1. Develop computerized mathematical procedure (based on physics of radiation and Monte Carlo calculations) for calibration of the Computer Automated Radioactive Particle Tracking Technique (CARPT) that will eliminate the currently lengthy in-situ and allow the technique to be used in the field on high pressure units. Specifically, plan the use of CARPT on a high pressure bubble column.

Task 2. Improve the accuracy of the CARPT technique and compare the velocities obtained by CARPT in certain regions of the column to those determined by the Heat Transfer Probe and Particle Image Velocimetry (PIV).

Task 3. Develop local probes for accessing velocities, holdups and heat coefficients in bubble column reactors.

Task 4. Collect velocity and voidage profile data in gas-liquid and gas-liquid-solid systems at different solids loadings and at close to atmospheric pressure.

Task 5. Use state-of-the-art hydrodynamic models and codes to predict velocity and holdup fields under conditions studied experimentally. Search for more suitable constitutive forms (e.g., lift, drag, turbulence closure models, etc.) to reach agreement between calculated and experimentally observed values. Try to assess the effect of elevated operating pressure on various constitutive forms.

Task 6. Collect the data in a high pressure 6" diameter bubble column and compare to model predictions. Also collect high pressure PIV data in slurry systems at Ohio State University. Refine models if needed.

All of the above tasks have been successfully completed as documented in our technical reports. The development of the Monte Carlo scheme and algorithm (Task 1) for calibration of the CARPT technique proved to be a rather involved and sophisticated endeavor. The progress made at Washington University (WU) in developing this powerful approach was summarized in the Second Annual Technical Report and the final implementation is summarized in this report. CARPT has also been employed successfully in a 6” high pressure bubble column as planned. The interactions with Exxon Research and Engineering (ERE) were invaluable in developing the high pressure system.

The accuracy of the CARPT technique was improved early on by implementing a wavelet based filter for minimization of the spurious velocities (First Annual Technical Report) and an additional gain in accuracy is achieved now with the final implementation of the Monte Carlo calibration procedure (this report). This work was completed at Washington University (WU). The liquid velocity data obtained by CARPT at WU at a specified set of operating conditions in bubbles columns of different diameter were compared with the data collected by PIV on the same columns at identical conditions at Ohio State University (OSU). Very good agreement between ensemble (time) averaged velocities obtained by the two techniques was found and reported (Second Annual Technical Report). It was also shown that the two techniques well complement each other with PIV providing more detailed information about the microstructure of the flow and CARPT providing information on the large scale flow pattern and large scale turbulence. The limited range over which the heat transfer probe can be used to locally determine the liquid velocity was also reported. This work at WU and OSU completed the objectives set by Task 2.

The development and testing of local probes for assessment of velocities, holdups and heat transfer coefficients was pursued through out the duration of this grant at OSU, WU and ER&E. The results obtained with the heat transfer probe and various optical probes were reported in the previous two annual technical reports fulfilling Task 3.

A database for liquid velocity profiles and voidage profiles was expanded (Task 4) both at OSU at column diameters lass than 4” and at WU in columns between 4” and 18” in diameter.

Our ultimate goal (Task 5) was to test the available models for computational fluid dynamics based on the ensemble averaged Eulerian k-fluid model to predict the experimental data for liquid velocities and gas holdup profiles. This task has been arduous as it requires considerable computational time and testing of various closure forms for interphase interaction terms. While pursuing this work we have also outlined as an interim solution a phenomenological model for assessing gas and liquid phase mixing (Second Annual Report). This model relies on the collected liquid velocity – gas holdup profiles data base and uses extrapolations to operating conditions. We at WU have also established a successful comparison of the predictions of the Euler k-fluid model, run in the CFDLIB framework of Los Alamos, and PIV data collected at OSU. This comparison is summarized in this report. The validity of CFD models for churn-turbulent and gas-liquid-solid flows remains to be established.

High pressure data have been collected at OSU in a slurry system and the findings are presented in this report. At WU the high pressure facility for a 6” diameter column has been completed and holdup and CARPT data at elevated pressure was obtained and is presented in this report. This accomplishes Task 6.

2.2. Accomplishments During Year 1

The first year was dedicated to the development and improvement of experimental techniques. Major accomplishments were as follows:

- The modification of Computer Automated Radioactive Particle Tracking (CARPT) and Computed Tomography (CT) facilities for improved accuracy (**WU**). This included the implementation of wavelet based filtering and energy thresholding for significant reduction in spurious velocities.

- The work on Monte Carlo based simulation of CARPT calibration and the mathematical approach worked out (**WU**).

- The development of a special heat transfer probe to measure instantaneous local heat transfer coefficients in the vicinity of a single large passing bubble (**OSU**). This included:

  - Validation of the reliability and accuracy.
  - Evaluation of probe's capabilities for use in high pressure systems.

- The Computational Fluid Dynamic (CFD) simulation of a 2D bubble column was performed based on the CFDLIB algorithm of Los Alamos and utilizing the ensemble averaged Eulieran k-fluid model. (**WU and ERE**). This included:

  - The evaluation of various closure schemes in CFD modeling such as mixing length and kinematic turbulent viscosity.

- The preparation for experiments in a high pressure 6" slurry bubble column. (**ER&E and WU**) was initiated. This included:

  - The computation of particle source strength for CARPT experiments.
  - The design of a calibration device for the high pressure 6" slurry bubble column.

2.3. Accomplishments During Year 2

During Year 2, the focus was on further improvement of experimental techniques and their testing; comparison of velocity, holdups and other fluid dynamic quantities obtained by different techniques; further model development and CFD; and preparations for high pressure studies. The accomplishments were as follows:

- The development of the extensive programs required for Monte Carlo calibration of CARPT was completed (**WU**).

- The comparative hydrodynamic study of a 4" bubble column using CARPT and Particle Image Velocimetry (PIV) was completed (**WU, OSU**).

- CARPT measurements were compared with other techniques such as Heat Pulse Anemometry (HPA) , Laser Doppler Anemometry (LDA), Hot Film Anemometry (HFA) **(WU)**.

- Progress was made in the development of transmittance and reflectance optical probes for gas holdup and bubble size measurements in churn turbulent flows (**OSU, WU, ER&E**).

- Various closure schemes in the CFD simulations of bubble column fluid dynamics were tested **(WU, ER&E)**.

- The design of a high pressure 6” slurry bubble column at Washington University was completed **(WU, ER&E)**.

2.4. Objectives and Accomplishments for Year 3

The objectives set for Year 3 were as follows:

- Develop high pressure/high capacity 6" slurry bubble column facility at Washington University **(WU, ER&E)**.

- Monte Carlo simulations for CARPT calibrations and experimental verification **(WU, ER&E)**.

- Expand the experimental database to high pressure systems and gas-liquid-solids systems using PIV **(OSU)** and CARPT/CT **(WU)**.

- Test the predictions from CFD models against CARPT, CT and PIV data using various closure schemes **(WU, ER&E)**.

To facilitate the review of the accomplishments achieved during year 3 (and during the follow-up no-cost extension period) of the grant the rest of this report is organized in self contained chapters.

Chapters 3 and 4 describe the work done to extend the CT/CARPT techniques to high pressure 6” diameter bubble columns **(WU, ER&E)**, respectively.

Chapter 5 outlines in detail the implementation of the Monte Carlo procedure for CARPT **(WU)**.

Chapter 6 presents the slurry data at high pressure in smaller diameter columns **(OSU, ER&E)**.

Chapter 7 outlines the comparison of data and CFD predictions for bubbly flows **(WU, ER&E)**.