Washington University Research

The following quarterly report from Washington University for the period Jully - September 1995 contains the following brief chapters:

1. Introduction
2. Review of Measurement Methods (Task 1)
3. Tracer Studies (Task 4)
4. CARPT/ CT Modifications (Task 3.1)
5. Development of Phenomenological Models (Task 3.2, Task 6)
6. Computational Fluid Dynamics Simulation (Task 3.2)
7. Research Progress and Timetable (Task 7)
8. References

Slurry Bubble Column Hydrodynamiics

Second Quarterly Report for Contract DOE-FC 22-95 P'C 95051

July - September 1995

1 Introduction

The main goal of the subcontract to the Chemical Reaction Engineering Labboratory (CREL) at Washington University is to study the fluid dynamics of slurry bubble columns and address issues related to scale-up. Experimental investigations to examine the efffect of operating conditions such as superficial gas velocity, solids loading, type of distributor etc. on the fluid dynamics are to be made. The specific objectives that have been set out for the fist year of the project are as follows :

1. Assess the suitability of available experimental techniques for the measurement of global and some local hydrodynamic parameters in an industrial sccale system and make recommendations for the use of these techniques at La Porte.

2. Interpret the existing tracer experiments and make recommendations for future tracer tests on La Porte reactor.

3. Modify the CARPT/CT experimental facility for study of slurry bubble columns.

4. Develop a phenomenological model for the key hydrodynamic features in bubble columns as a basis for an improved reactor model.

5. Introduce appropriate closure schemes and constitutive forms in the hydrodynamic codes to achieve agreements between data and models and test modell reliability.

The activities that have been undertaken in the second quarter (after the initiation of the project in April 1995) towards fulfilling the above mentioned objectives are described in the subsequent sections.

2 Review of Measurement Methods

A survey of all the available methods for the measurement of fluid dynamiic parameters in a high pressure high temperature multiphase chemical reactors has been completed and is

the subject of a separate and complete topical report which will be issued iin the immediate future. The emphasis was placed on techniques that can be utilized under conditions of interest in practice such as high pressure and temperature, large solids' holldup etc. A draft of the topical report resulting from this review has already been submitted! to Air-Products and in it an attempt was made to present a comprehensive review of tthe experimental methods available for multiphase systems. A summary of that report is presented here. The measurement methods for the determination of the following quantities were considered :

- Void fraction and solids' concentration;
- Bubble size distribution and rise velocities;
- Liquid and solid phase velocities.

Gas holdup and solids' concentration measurement methods can be broadly classified into two categories : (1) those providing an overall or global measurement amd (2) those that provide a local or point measurement. The global measuring techniques yield information on the line, area or volume averaged gas or solids' holdup. In general, the measurement of the overall holdup is relatively simple. It provides information regarding what fraction of the system volume is occupied by the phase of interest. The global measuring techniques that were reviewed include the bed expansion method, pressure drop method and quick closing valve method in addition to a few others. In the bed expansion method the holdup is measured based on the initial static height of the bed and the dynamic height of the dispersion. In the pressure drop method the holdup in a section of the flow in between two pressure taps is related to the difference in pressure between the two taps. In the quick closing valves technique the dispersion between two sections of the test loop is isolated and the mass of phases in the isolated section are measured.

Another technique that is often used for the determination of the overall holdup is based on the measurement of impedance of the two phase dispersion. Gas (air) and liquid (water) have significantly different electrical conductivity and permittivity and it is this difference that is exploited by the technique. The variation in the flow structure is accompanied by a variation in the impedance of the two phase mixture which is measured by metallic electrodes introduced suitably in the flow. The void fraction is then estimated by adopting a relative impedance technique.

All the above methods for overall holdup measurement are adequate for use in two phase flow systems. For three phase systems no single method can provide both the solids' and gas holdup, and a combination of the above techniques along with some approximations has to be used for obtaining the relevant information. Upon review of various techniques the recommendation that emerges for estimation of the overall fractional gas holdup is to

measure the pressure profile ahlong the reactor length and provide a rough comparison for it by the bed expansion method.l.

Techniques based on the uuse of radiation have also been reviewed in the report. This method can also be used for pproviding a chordal average measurement or a point measurement depending on the compplexity of the measurement process. The basis for the measurement is the attenuation oof radiation by matter. The absorption of a narrow beam of radiation of initial intensity I_o l by a homogeneous material with a mass attenuation coefficient μ is expressed as :

$$I = I_0 \, exp(-\rho \, \mu \, l) \tag{1}$$

where I is the intensity of radiaation detected after the beam has traveled a distance l through the absorbing medium. For aa mixture of two substances, say a gas and a liquid with an attenuation coefficient μ_g andd μ_l and densities ρ_g and ρ_l, respectively, the corresponding relation is

$$II = I_o \, exp[-(\rho_g \, \mu_g \, l_g + \rho_l \, \mu_l \, l_l)] \tag{2}$$

where l_g and l_l are the path leengths of the beam in the gas and the liquid, respectively. In terms of the measured intensitties I_{tp}, I_f and I_{mt} corresponding to the test section with the two-phase mixture, full of liqquid and completely empty, respectively, the chordal average void fraction is computed fromm:

$$\epsilon = \frac{ln(I_{tp}/I_f)}{ln(I_{mt}/I_f)} \tag{3}$$

If a few chordal measuremeents are obtained then it is only possible to obtain the chordal average holdup along the beamn paths. This is the commonly used radiation technique and is called densitometry. On the otther hand if several such measurements are obtained spanning the section of interest completely then the distribution of the holdup over an entire cross section can be obtained and tthe method is then referred to as Computed Tomography. In systems as large as the La Ponrte reactors a gamma ray densitometer for chordal averaged void fractions is recommended.l.

It is also possible to measunre the local void fraction by means of probes. The commonly used probes are based on eithoer electrical or optical principles. The electrical impedance probes can be further based onn either conductive or resistive or capacitive effects. A conductivity probe makes use of tthe difference in conductivity of the gas and liquid phase and is quite suitable for aqueous gaas-liquid systems. Resistivity probes sense the variation in resistance between two electrodess with the passage of bubbles through the gap between them.

They are more suitable for measurement of solids' concentration. Similarly, a capacitance probe uses the difference in the dielectric constant associated with each phase for phasic discrimination. They can be used in non-polar mediums and have been used more often for solids' concentration measurements in fluidized beds and three phase systems. Depending on the phase that exists at the tip of the probe the signal in principle is binary in nature and the integration of the periods of time corresponding to a particular phase along with the total length of time for the signal gives a measure of the holdup of that phase. Optical probes exploit the differences in the index of refraction of the two phases and rely on the application of Snell's law at the probe-fluid interface. Depending on which phase exists at the probe's tip the light from the tip is reflected or refracted. Once again the holdup of a given phase is obtained on the basis of the ratio of the sum of the time periods of the signal corresponding to the phase and the total time of the signal.

All the above probes can also be used for the measurement of bubble sizes and their velocity. Two such probes can be integrated together such that their tips are vertically aligned and a small distance apart so that from the measurement of the time of flight of a bubble between the two tips the bubble rise velocity can be estimated. Each of the sensors has a binary output signal depending on which of the phases is in contact with the tip. As a bubble passes over each of the tips there is a mutual time delay t between the signals from the two sensors due to the time needed for the bubble to proceed from one probe to the other. The distance d between the probe tips being known, the component of the bubble velocity along the direction defined by the line joining the probe tips can be estimated as :

$$v_x = d/t \tag{4}$$

This velocity along with the knowledge of the mean residence time of the bubble at one of the probe tips t_m can be used to estimate the pierced chord length of the bubble as :

$$l_g = v_x t_m \tag{5}$$

A rather simple method that has found wide acceptance for measurement of bubble velocities, and in turn their sizes, is the dynamic gas disengagement technique. The method requires an accurate recording of the rate at which the surface of the dispersion drops once the gas flow is interrupted. The measured disengagement profile is used to estimate the holdup structure that existed just before gas shut off. In its simplest form the technique assumes one or two dominant bubble sizes. The initial part of the disengagement profile is considered to be dictated solely by large bubbles. The small bubbles disengage only after all of the large bubbles have left the system. The disengagement profile (the height of the two

phase dispersion as a function of time) has two distinct regions, corresponding to the two bubble sizes, which are fitted with straight lines. The slope and intercepts of the straight lines are related to the holdup and the rise velocities of the corresponding bubble sizes. If some relation (correlation) can be assumed between bubble rise velocities and their sizes then, the latter can also be estimated.

The choice of probe to be used for local holdup as well as bubble size measurement depends to some extent on the physical properties of the liquid phase in the reactor. For liquids such as alcohols the conductivity probe is more suitable since they are polar in nature and therefore the use of capacitive probes is problematic. On the other hamd if the liquid phase consists of paraffins and olefins the conductivity is much lower and also these liquids are not as polar as alcohols. Consequently, a capacitance probe would be a better choice. Also, the liquids should have as low a viscosity as possible so that the dewetting time of the probe is small. For the Fischer-Tropsch wax at 250°C this should not be a cause of problems. If only the local gas holdup is of interest then either the conductivity or the resistivity probe (depending on the liquid properties) is the best choice. If however, the solids' concentration is also desired, then either the multi-sensor resistivity probe or the capacitance probe can be used. However, despite all the claims that have been made about the capabilities of these latter probes one would still need to get a sample probe for testing in simullated conditions to determine their appropriateness for the specific application. In addition, it is highly recommended to perform dynamic disengagement studies on the La Porte reactor for the assesment of bubble classes and sizes.

Techniques that have been commonly used for the measurement of liquid velocities in multiphase systems are essentially the ones used in single phase flow with some modifications in the interpretation of the measured data. These commonly used methods include the simple pitot tube, devices based on the turbine flowmeter, hot wire or film anemometry, Laser Doppler Velocimetry (LDV), Particle Image Velocimetry (PIV), particle tracking and tracer techniques.

The principle of the pitot tube is very well known and is based on measuring the differences in the pressure at the point of interest and the static pressure at the wall. The tube is inserted close to the point of interest in the flow such that its opening faces the flow. The velocity at the point is calculated based on the dynamic pressure measurement. The relationship is

$$\Delta P = \frac{1}{2}\rho v^2 \tag{6}$$

and is applicable for single phase flow. For two phase flow situations the above relation needs to be modified to account for the holdup of the second phase and some additional assumptions need to be made.

In hot wire anemometry a small electrical resistance wire or film (supported on some base) is heated and exposed to the flow stream. Due to the removal of heat by the flowing fluid the resistance changes, and this change is a function of the flow velocity and the physical properties of the fluid. Thus, in single phase flow the heat flux is directly related to the velocity. For two phase flow systems complexities arise due to the presence of a second phase and calls for some rather careful signal processing.

In analogy to tracer techniques used for measurement of the residence time distribution of a phase in a reactor, Lubbert and Larson (1990) have developed a tracer technique for measurement of the local liquid phase velocity. The method relies on using heat instead of electrolytes or dyes as the tracer. Fluid elements are tagged by direct local ohmic heating using a high frequency alternating current between two small electrodes introduced inside the reactor. The dispersion of heat is measured at a small distance away from the source of heat using a hot-film anemometer switched as a temperature detector. The information concerning the time of flow distribution is obtained from the cross-correlation between the input and output signals. A probability density function (p.d.f.) is assumed for the number of tracer particles at a given distance from the source, at a given instant of time after injection. This distribution is assumed to be normal. A nonlinear fit of the measured time of flow data to the assumed p.d.f. provides the mean time of flow as well as certain other parameters related to the local dispersion. From the mean time of flow and the distance between the sensors the local liquid phase velocity can be estimated.

Laser Doppler Velocimetry is considered to be an accurate and reliable method of measuring flow velocities in single phase flow. In a dual beam system two laser beams of equal intensity are focused to cross at a point of interest in the flow field. The measurement volume is a small ellipsoidal region at the intersection of the beams. The fluid is seeded with minute tracer particles which follow the motion of the fluid. When one such particle passes through the control volume, light from each of the beams gets scattered and interferes in space. This is seen as a varying intensity fringe pattern by a detector and is referred to as a Doppler burst. The particle velocity U is then inferred from the Doppler shift frequency.

Particle Image Velocimetry (PIV) (Adrian, 1991) in its simplest form uses a sheet of laser light to illuminate a section of the flow and images of small scattering (tracer) particles are photographed at right angles to the sheet. The concentration of the particles used corresponds to volume fraction of the order of 10^{-8} to 10^{-5} and consequently does not affect the fluid rheology. The velocity field in the plane of the imaged sheet is measured by recording a series of exposures and extracting the mean displacement of the particle image between successive exposures. Similar to other optical techniques PIV is restricted to relatively transparent media. Thus the concentration of suspended solids (if one of the

phases is a solid) has to be low. Even if one resorts to refractive index matching of the solid and the liquid phase, high concentrations of the solids would mean a reduction in the transmission of the scattered light. The use of PIV techniques to bubble columns and gas-liquid-solid fluidized beds has been advocated by L. S. Fan and his group at the Ohio State University (Tzeng et. al. 1993).

Some of the techniques mentioned above for the measurement of liquid phase velocities can also be adopted for the measurement of solid particle velocities. The laser velocimetry and particle image velocimetry methods are applicable for solids' velocity measurements in system with relatively small solids loading, generally about 15 to 20 %. With higher solids' concentration the attenuation and scattering of the light beam or sheet leads to problems in the interpretation of the signal. The radioactive particle tracking technique in our laboratory is ideally suited for the measurement of solids' velocities.

The conclusion of the review is that the presently available instrumentation for measurement of the hydrodynamic parameters are by and large cumbersome to be used in a slurry bubble column on the scale of a pilot plant. However, some gross features of the flow in such a system are still measurable. The measurement of the overall gas holdup can be achieved by means of the bed expansion method and/or by the pressure drop measurement. The bed expansion can be conveniently measured by using the gamma densitometer already in use at La Porte. It is also recommended to install a series of pressure taps allong the column height which would enable the measurement of the sectional holdup in the system. They can also be used in the estimation of bubble sizes by means of the dynamic gas disengagement technique. Installation of an Americium - 241 source in addition to the Cesium - 137 source is also recommended to provide some chordal average measurements of the solids holdup by means of dual energy densitometry principles (Bukur et. al., 1996). Measurement of center-line phase velocity can also be accomplished by means of a suitably calibrated pitot tube. Tests in using the heat pulse probe of Lubbert to provide some measure of the dispersion coefficient and in turn the velocity of the phases is also recommended.

3 Tracer Studies

During this quarter all the raw data collected by Tracerco for Air Productts was assembled and analyzed for consistency. Correct operating conditions for all the experiments as well as the exact positions of all detectors were recorded. The mode of tracer injection was analyzed. Preparations were made to fit the data with the existing axial dispersion based models via a parameters estimation program developed at our Chemical Reaction Engineering Laboratory (CREL). Model development was also initiated for phenomenological interpretation of the

fluid dynamics that does not rely on axial dispersion concepts.

Since it is anticipated that all the tracer studies will be completed duriing the first year of the contract as planned, a detailed technical discussion of the approach used and results obtained will be presented in a subsequent quarterly report in full followed by a topical report.

4 CARPT / CT Modifications

Work was initiated in this area for improving the accuracy of the CARPT technique via wavelet filtering and in extending the auxiliary instrumentation for our sllurry bubble column facility to include sectional differential pressure measurements and viideo camera flow visualization.

The above work should be completed during the subsequent quarter and all technical details will be presented then.

5 Development of Phenomenological Models

Work is in progress on using the CARPT/CT findings in developing a liquid recirculation model and gas backmixing model. Technical details will be shown in the next quarterly report.

6 Computational Fluid Dynamics Simulation

Work is in progress on the simulation of a two dimensional bubble column utilizing the CFDLIB codes of Los Alamos. Results and comparison to experiments willl be presented in the next quarterly report.

7 Research Progress and Time Table

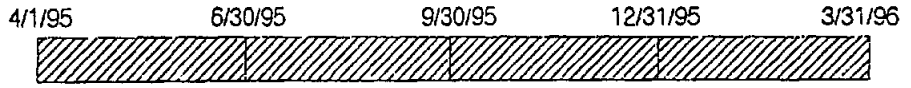
The time table for the work proposed for the first year of the contract is shown below together with the portions of the work actually accomplished. Clearly the work is progressing well on schedule.

DOE-AIR PRODUCTS SUBCONTRACT TO WASHINGTON UNIVERSITY

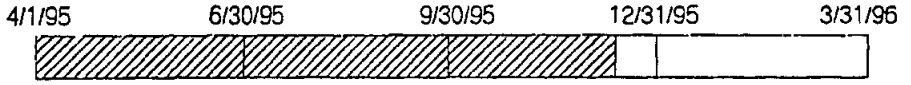
SBCR HYDRODYNAMICS

Objectives set for the first year (4/1/95 - 3/31/96) and accomplishments to date (9/30/95)

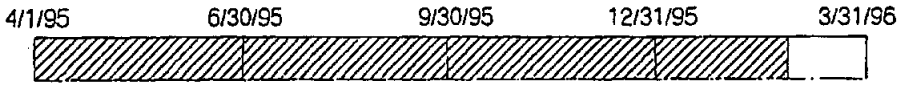
1. State of the Art Review of Measurement Methods



2. Interpretation of Tracer Runs at La Porte

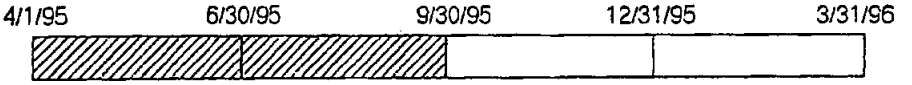


3. Modification of CARPT/CT for Slurry Systems



4. Phenomenological Model for Liquid Recirculation

5. CFDLIB Codes Testing and Simulation



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Tzeng, J. W., Chen, R. C. and Fan, L. S., 1993, Visualization of Flow Characteristics in a 2-D Bubble Column and 3 Phase Fluidized Bed, AIChE, J., 39, 733-744.