VI) BUBBLE DYNAMICS

Results from our earlier work (1) showed that the presence of fines in the slurry had a strong influence on the gas flow behavior in the three-phase system. Increasing the gas velocity normally increases the bed expansion slightly when liquid phase is a pure liquid. However, increasing the gas velocity when fines are present in the liquid (i.e., slurry) can, in fact, result in a slight bed contraction. This matter of bed contraction is discussed further in Section III. A very complex process of bubble coalescence and breakup is occurring in a cold-flow fluidization system. The fluidization occurring in a chemically reacting system is even more complex.

Bubbles are coalescing and growing as they rise through the system. The fines in the slurry are promoting this coalescence, due to their presence on the bubble surface through a complex but poorly understood process. Bubble coalescence is also promoted by the higher viscosity caused by the fines in the liquid. Bubbles break up if they grow through coalescence to a size larger than their maximum stable bubble size. The catalyst particles also promote bubble breakup. It is clear that a better understanding of the bubble dynamics occurring in the H-Coal system is essential.

Because of the complexity of the three-phase system being studied, it was anticipated that development of a quantitative technique and subsequent data collection would be difficult. Therefore, development of three techniques for investigating different aspects of the bubble dynamics was carried out in parallel: 1) A light beam probe was developed which involves passing light from a helium-neon laser through the three-phase system and monitoring the transmitted light intensity; 2) A holographic technique was developed to capture three-dimensional images of bubbles rising through the three-phase system; 3) A resistivity probe was developed to measure bubble size and bubble frequency.

Due to the nature of the liquids chosen in this study on bubble dynamics, very small bubbles were observed within the cold-flow system. These bubbles were smaller than originally anticipated when this project was first proposed. This resulted in the inability to use laser-Doppler anemometry, as was originally planned. Also, the development of the holographic technique required extensive efforts and is only now becoming a useful tool. Nonetheless, significant progress has been made in the development of three quantitative techniques, and important results are presented and discussed.

Experimental System

Description of the Apparatus.--The experimental setup consisted of a three-phase fluidized bed operating in a 6" diameter vertical column. A schematic diagram of the apparatus is shown in Figure 30. The vertical column was constructed using four pieces of glass pipe, each section being 2 ft in length. Carbon-steel spool pieces placed between the glass sections were radially drilled and fitted with threaded pipe connectors, thus providing points of access to the interior of the column. A diagram of the metal spool pieces is given in Figure 31.

The inlet distributor section, shown in Figure 32, was modeled after H-Coal pilot plant distributors and was constructed by Amoco Oil. It consisted of a carbon-steel mixing chamber, into which the gas and liquid flowed; a vertical exit pipe; and a slotted bubble cap. The gas/liquid mixture leaves the mixing chamber through a stainless steel screen spot-welded to the inlet of the exit pipe. From this point, the fluid mixture flows upward to the top of the bubble cap, where it is forced down and out through the slots and into the fluidized bed.

As shown in Figure 30, we see that when the gas phase passes through the inlet distributor, it first comes in contact with the liquid phase approximately 6' before entering the distributor section. This is the method of fluid injection used in the H-Coal process, thus allowing gas/liquid mixing to occur before reaching the reactor. Further bubble breakup occurs as the fluid flows through the screen in our cold-flow system and out the slots of the bubble cap.

The gas flow could be routed either into the liquid inlet pipe leading to the inlet distributor or directly into the column via the bottom-most spool piece and a gas sparger. The sparger was a length of 3/8" diameter copper tubing which spanned the diameter of the column. Five 1/16" diameter holes spaced approximately 1" apart served as a means of injecting relatively large bubbles (a few centimeters or so in diameter) into the bottom of the bed.

At the top of the column, the fluid mixture is separated into its gas and liquid phases using a spiral entrainment separator (see Figure 33). The liquid then flows through an overflow cup, down a return line, and into the holding tank, where it is recycled. The nitrogen gas used is simply vented to the atmosphere. A stainless steel screen is spot-welded to the overflow cup to prevent the entrainment of solids out of the column.

Fluids and Solids.--The initial solids used in the column were pyrex glass cylinders. These were nominally 2 mm in diameter and 5 mm long. The fluid used was a mixture of two organic solvents in a proportion such that the refractive index of the fluid matched that of the glass cylinders. The purpose of this was to make optical measurements possible within the bed. Property values of the constituent fluids and the mixture are given in Table XI.

In addition to the Pyrex glass cylinders, spherical glass balls with diameters of 1, 2, 3, and 5 mm were also used. This was done in order to determine the effect of particle size on bubble sizes. Basic data concerning the particles are provided in Table XII. The settled bed heights of the particles were approximately 70 cm.

Bed Expansion and Pressure-Drop Measurements

Bed Expansion Measurements.--Bed expansion measurements were made for various flow conditions ranging from a fixed bed at low liquid flow rates to a fully fluidized state in which the bed expanded by 120%. It was found that the bed expansion changed by such a small amount while varying the gas flow rate that this effect could be neglected. Therefore, in this section, bed expansion is assumed to be a function of liquid flow rate only. The minimum fluidization velocity was found to be 1.60 cm/sec. This represents the condition of incipient fluidization in which the solid particles are just supported by the fluid.

The average solid volume fraction was calculated using measurements of the average bed height and knowing the total volume of solids within the system. Results are shown in Figure 34. Assuming a homogeneous distribution of solids throughout the bed, it was possible to model the flow according to an empirical drift flux model as given by Wallis (51), based on the work done by Richardson and Zaki (49). The following model relates the average liquid flux, U_l (cm/sec), to the average volume fraction of solids, ϵ_s:

$$U_l = 17.58 \left(1 - \epsilon_s \right)^{2.208} \qquad (12)$$

From this, we see that the terminal velocity of the solid particles is 17.58 cm/sec, which is consistent with theoretical predictions based on balancing the gravitational and drag forces acting on a single particle.

Pressure-Drop Measurements.--Void fractions of the gas phase (gas holdup) above and within the fluidized bed were determined from pressure-drop measurements (48). It was found that for low liquid flow rates, the gas holdup increased with both gas flux and liquid flux. Because some bubbles were generated by passage of both liquid and gas through an inlet distributor, it was concluded that the inlet distributor did not produce bubbles of a constant size. In fact, because of these studies, it is now felt that the design of the inlet distributor may be very important to bubble size distributions found within the fluidized bed. Results indicate that the inlet distributor was able to produce much smaller bubbles than those produced by the breakup of large bubbles when colliding with solid particles inside the fluidized bed.

Generally, the gas holdup within the three-phase region was found to be much higher than that above the bed. This can be attributed to increased accumulation of gas bubbles inside the bed, caused by collisions between the bubbles and solid particles. These collisions impede the flow of bubbles through the bed, thus increasing the gas holdup.

Light Beam Probe and Bubble Breakup Model

Description of the Light Beam Probe.--A new optical technique was developed to measure bubble sizes in the two-phase region above the catalyst bed (47). This new method can be used in flows of spherical bubbles with diameters greater than 120 microns. Implementation involves passing a narrow beam of light through the bubbly flow and monitoring the transmitted light intensity. Light from a helium-neon laser enters and exits the column through thin probes, shown in Figures 35 and 36. (The beam length was typically between 3 and 12 mm.) As a bubble passes between the probe tips, it blocks the light beam, as shown in Figure 37. The basic units of data are the rate at which each bubble blocks off the beam and the duration of the blockage. Figure 38 shows typical signals from the light beam probe. By statistically analyzing a set of such data, bubble size and average velocity distributions were determined. A typical bubble size distribution curve is shown in Figure 39.

Results of the Light Beam Technique.--Measurements made with the light beam probe showed the bubble size distribution to be highly dependent upon the size of the solid particles within the fluidized bed but relatively independent of the gas and liquid flow rates, at least for the low gas velocities employed here. The latter is illustrated in Figure 40, where the gas and liquid fllow rates were varied without significant change in the Sauter mean bubble diameter. Initially, 2 mm x 5 mm cylindrical particles were used, but these were later replaced with glass spheres ranging in size from 1 to 5 mm in diameter. Average results of bubble size measurements are listed in Table XIII.

Equilibrium Bubble Size Model.--Dimensional analysis was applied to the breakup of bubbles passing through the fluidized bed. Based upon the data, a model was constructed of the equilibrium bubble size distribution in a bed of particles of known size. This model relates the dimensionless mean bubble diameter to the Eotvos number as follows:

$$d_m/d_3 = 1.544\ (\sigma/\Delta\rho d_3^2\ g) + 0.143$$

where $\Delta\rho\, d_3^2 g/\sigma$ = Eotvos number.

Gas holdup measurements made both within the three-phase fluidized bed and above the bed in the two-phase gas/liquid region are shown in Figures 34A through 34D. The data show the void fraction within the three-phase region to be higher than that found above the bed in the two-phase region at very low gas rates. In Figure 34A the difference between the two values is approximately equal to the void fraction which accumulates inside the bed. This suggests the bubbles are not allowed to flow freely but are trapped inside the bed. Visual observations of the bubbles tend to confirm this hypothesis. The presence of closely packed solid particles tends to decrease the average bubble velocity due to collisions.

In Figures 34B through 34D we see that increases in both liquid and gas flow rates tend to narrow the difference between the two curves. This suggests the bubbles find less resistance to flow when the bed is expanded, thus behaving more like bubbles flowing through liquid alone. These interesting results complement those presented earlier in Section IV (Table V) where gas

holdup in the dilute phase at higher gas velocities was found to be about 10% higher. A plot of the dimensionless mean bubble diameter versus the inverse of the Eotvos number is shown in Figure 41.

Very little information was found in the literature to test the validity of this model. Lee (46), however, does give a mean bubble diameter of 0.24 cm in a water-fluidized bed of 0.6 cm diameter glass particles. The data point determined from this information is also included in Figure 41. It fits quite well, which is surprising, considering the effects of viscosity have been neglected.

Bubble Breakup and the Critical Particle Size.--The breakup of large bubbles to form an equilibrium bubble size is believed to be dependent upon the solid particle size. Yet, it was observed that the larger particles were far more efficient at breaking up large bubbles than were the small particles. If we assume that when a particle passes through a bubble it strips away a volume of gas approximately equal to its own volume, then to be consistent with our model, there exists a critical particle diameter below which large bubbles will not be broken up efficiently. By equating the particle size with the mean bubble diameter, a critical particle diameter of 1.89 mm is determined. This value is consistent with the data, but it could be argued that reasonably efficient breakup of large bubbles requires a particle size somewhat greater than 2 mm.

A similar analysis can be done for water-fluidized beds by substituting the density and surface tension values for water (listed in Table XIII) into the equilibrium bubble size model. By equating the particle size with the mean bubble diameter, a critical particle diameter of 2.9 mm is determined for water-fluidizing a bed of glass particles. This estimate of a particle size required for efficient breakup of bubbles is in good agreement with literature summarized by Vasalos, et al. (1), from which it appears that the critical particle size in water (assuming a solid density of about 2.5 gm/cm^3) is between 3 and 4 mm.

Holography

Description of Holographic Technique.--Holography was used to capture three-dimensional images of bubbles flowing within the dense-phase region. Pulses of light from an Nd:YAG laser were used to illuminate the test section and form holograms for later analysis. The short duration of the pulses allows an instant of time to be captured on a holographic plate. Figure 42 shows the beam paths and optical arrangement required to form an off-axis hologram. When first entering the test section, the laser light passes through a diffusing plate. This is done to create a more uniform backlighting against which the bubbles are viewed, providing better clarity and definition of the bubbles in the hologram.

When constructing a hologram, the room containing the equipment must be totally dark, so that only the illumination from the laser strikes the holographic plate. After inserting a new plate into the plate holder, the laser is pulsed once, causing the sizes and positions of bubbles within the viewing volume to be permanently preserved. The plate is then developed and allowed to dry before reconstruction and analysis.

Holographic Analysis System.--To view the three-dimensional image contained on the holographic plate, the output from a helium-neon continuous-wave laser is used to retrace the path of the reference beam. As shown in Figure 43, a real image is formed in front of the holographic plate. A video camera is then used to focus on a plane inside the three-dimensional image and display this plane on a video monitor. By attaching the camera to a traversing mechanism, it is possible to change the focal plane remotely and to measure the radial position of the bubbles within the viewing volume.

A digital electronic circuit was designed to superimpose a thin horizontal and a thin vertical line on the video monitor's image from the video camera. The position of these lines on the screen can be changed while being monitored by a computer. By properly calibrating the magnification of the images displayed on the screen, it is possible to use this horizontal/vertical line generator to measure distances across the screen. This aids in the determination of bubble sizes from holograms. Figures 44 and 45 detail the circuitry used in the horizontal/vertical line generator.

Results.--The optical bench and the holographic analysis system took several years to design and build, so that the results obtained to date are limited. However, the technique has proved to be useful in determining bubble sizes and local void fractions. This technique offers an advantage over intrusive probes in that it dispels any doubts about the actual size and shape of bubbles inside the viewing volume.

Results from an analysis of one typical hologram are shown in Figures 46, 47, and 48. Although shown as a histogram plot, the actual bubble sizes were measured individually with better than ± 0.01 mm precision. Such results compare favorably with those found via the earlier mentioned light beam probe.

Resistivity Probe

A resistivity probe with a DC power supply was developed and used to measure bubble sizes within the three-phase fluidized bed (7). Unfortunately, due to the nature of the liquid phase, many of the gas bubbles in the three-phase fluidized bed were too small to be detected by the probe. Quantitative results in the three-phase system thus could not be obtained. However, the distribution of the gas phase was studied qualitatively in the two-phase region above the bed. It was found that the bubbles were distributed more uniformly at higher liquid and gas flow rates.

Although the probe was unable to be properly calibrated so as to provide accurate bubble size measurements, it did adequately measure the bubble frequency, defined as the number of bubbles that contact the probe tip per unit time. Such a probe could be employed in an actual H-Coal reactor to detect flow regime changes, presence of large bubbles, and channeling of the gas phase. For the probe to work properly, the resistivity of the mixture must be below 10^6 ohms/cm^3. This condition is likely to exist at the operating pressures and temperatures of an actual H-Coal reactor.

A schematic diagram of the resistivity probe and associated electronics is given in Figures 49 and 50. Further details of the probe are given by Tsung (7).

Summary

Several new and powerful techniques for measuring bubble sizes and gas holdups were developed and shown to be useful. The narrow light beam probe is a very practical tool for measuring spherical bubbles greater than 120 microns. Holography is capable of determining the exact size, shape, and position of bubbles within some viewing volume. Although it does not provide absolute measurements, the resistivity probe developed during this study can most likely be used in actual H-Coal reactors to determine bubble frequencies and qualitative measurements of the flow regime.

Several interesting results were found during the course of this investigation. First, the design of the inlet distributor was determined to play an important role in the breakup of bubbles initially entering the fluidized bed. It was observed that smaller bubbles were generated by the inlet distributor than could be produced by the breakup of large bubbles by the solid particles within the bed. Second, the volume fraction of solids within the bed was found to be dependent mainly upon the liquid flux and could be predicted by an empirical drift flux model.

An equilibrium bubble size model was developed based upon data gathered by the light beam probe. This model relates the Sauter mean bubble diameter to the solid particle diameter, the solid/liquid density difference, and the surface tension of the fluid. From this model, a critical particle size was also determined at which efficient breakup occurs.

Recommendations

After completing this study of a three-phase fluidized bed, it is apparent that much more basic research is needed to better characterize the dynamics of such a system. The equilibrium bubble size model should be tested by varying the surface tension and the solid/liquid density difference to see if the model accurately predicts the Sauter mean bubble diameter. Also, the effect of liquid viscosity should be determined, since the model contains no viscosity term.

Because of the added complexity involved, this study did not look into the effect of using a slurry other than liquid alone to fluidize the solid bed. Such a system will probably have a different bubble size distribution due to the presence of these small particles. The effect of slurry particles should then also be studied in future work.

Since the holographic technique took several years to develop and is only now useful as a measuring tool, it is felt that further tests should be done to make use of this specialized technique. For example, because large bubbles are not generally spherical, as are small bubbles, additional studies regarding the shape and size distribution of large bubbles would be of interest. Ultimately, the holographic technique has the potential to provide insight into the nature of the wakes trailing behind the bubbles.

Last, it is felt that the design of the inlet distributor should be studied to determine how the initial bubble size distribution entering the reactor can be controlled for maximum reaction to take place.

VII) HYDROCARBON RESEARCH, INCORPORATED, PDU EXPERIMENTS

A series of fluid dynamic experiments was performed during PDU Run 10 (2) on HRI's process development unit (PDU). The response of the gas/slurry ebullated bed to changes in operating variables such as gas and liquid velocity or liquid viscosity was measured. The experimental plan included tests during the normal duration of the PDU run (relatively constant flow rates but changing product viscosity) and an additional period where the gas and liquid flow rates were varied over a wider range. The ultimate goal of these experiments was to extend the model development performed under a previous contract (1) to apply to the operating PDU reactor.

Samples and Data Collection

The data required for analysis of the fluid dynamics tests involved obtaining a variety of samples and operating conditions.

PDU Operating Conditions.--The primary variables were the liquid flow rates (both slurry mix tank feed rate and internal recycle rate) and the gas flow rates (both fresh hydrogen and recycled hydrogen-rich gas). These variables were provided by HRI (62) and are summarized in Table XIV.

Liquid Samples.--Sixteen reactor liquid product slurry samples were withdrawn from the bottom of the high-pressure separator (Figure 51) during the test program. The sampling procedure used by HRI has been previously documented (2). Table XV itemizes the sampling dates. Battelle Memorial Institute later measured the viscosity and density of these samples. Four additional samples were also withdrawn for Oak Ridge National Laboratory (ORNL) for viscosity and density determination.

Liquid samples from the clean oil tank and slurry mix tank were also obtained on selected test dates. Slurry mix tank liquid characterizations appear in Table XVI.

PDU Reactor Effluent Composition.--Based on flow rates and inspections of test samples taken at various process locations, HRI routinely back-calculates an estimated reactor slurry liquid composition for material leaving the bottom of the O-1 separator. The data, reported as fractions of unreacted coal, ash, resid, and distillate, appear in Table XVII. This information will be correlated and used later for estimating the density and viscosity for tests where no liquid sample was taken.

Catalyst Samples.--One sample was taken on 8/3/80. Another sample was taken by HRI at the completion of the PDU run on 9/24/80. A linear variation of catalyst density between these two dates was assumed. The density increased

only 10% over the entire duration of the run. A summary of catalyst-soaked particle densities used for data analysis appears in Table XVIII.

Gamma-Ray Scan.--A gamma-ray scintillation detector was mounted on the existing PDU reactor traverse scanner. This system, previously developed by HRI, consisted of a sled mounted on vertical rails which ran parallel to the reactor. By adjusting the elevation of the gamma-ray source and detector and noting changes in the count rate, the catalyst bed height may be determined. A second 1" x 1" detector crystal was positioned opposite the center of the beam from the 500 mc Cs-137 source. This installation was designed to retain the existing HRI bed density monitoring equipment and to provide gamma-ray attenuation data in small, well-defined areas of the reactor.

Side and top views of the assembly appear in Figures 52 and 53, respectively. A schematic description of the electronics train appears in Figure 54. The criteria for setting the gain of the amplifier and discriminator to bracket the Cs-137 photopeak are presented in Figure 55. Because the apparent peak energy shifted with detector temperature (Figure 56), retuning the electronics was necessary before each fluid dynamics test.

The first scan performed was a "zero" scan on an empty reactor. The no-flow gamma-ray absorption data and subsequent data under operating conditions could then be used to calculate the average density at each point. As will be detailed in a later section, knowledge of the catalyst bed expansion and liquid density then allows calculation of all component phase volume fractions.

On 7/11/80, the empty reactor (zero) gamma-ray scan was taken. After tuning the detector electronics, absorption measurements were obtained at 2" axial intervals, with some 1" interval data near the bottom of the column. The density monitoring system was able to scan elevations between approximately 3 and 14 ft above the bottom flange of the PDU reactor.

Figure 57 illustrates the variation in count rate with detector elevation. At each location, the count rate displayed is the average of at least two separate measurements taken for a 20-second interval. Standard deviations were on the order of ± 1%. At elevations of 4 ft and 6 ft, sharp drops in the average zero count rate may be seen. These were caused by external pipes and flanges connected to the reactor. Data taken here will be most sensitive to errors in traverse position during the operating test scans. Therefore, results from these locations may possibly not be suitable for use.

Gamma-ray scans on the operating PDU were taken at 14 test periods during the run. Because the reactor now contained dense, absorbing material, the average gamma-ray counts dropped. The average gamma-ray count rate was taken over 30-second test intervals, again with at least two measurements at each elevation. Three scans were also taken during the shutdown sequence (two on 9/24/80 at 2:12 PM and 7:52 PM, and one on 9/25/80 at 11:08 AM) to measure the absorption of the settled catalyst bed.

In addition, some interference was encountered when the sled was at the top of the column and was exposed to a second gamma-ray source (the level detector of the high-pressure separator) which was being used. This marked increase in count rate with elevation was not observed in any of the PDU tests or shutdown scans. Therefore, a modified zero scan, also shown in Figure 57, was used in the data analysis.

Density and Viscosity of PDU Reactor Liquid Slurry Samples

Sixteen PDU reactor liquid slurry samples were characterized at Battelle Memorial Institute to measure their densities and viscosities at process conditions. Battelle's experimental apparatus appears in Figure 58. Liquid was charged from the sample bomb into the viscometer while maintaining the pressure at 2,000-3,000 psig. A bob (Item 9) was alternately raised and lowered through the liquid-filled chamber. A magnetic coupling (Items 4 and 5) eliminated leaks or mechanical interference from shaft seals. Full details of the testing technique appear in Battelle's final report (Appendix C). During the several months that the experiments were performed, Battelle made some modifications to the viscometer to improve its accuracy and reliability. After these improvements were completed, several samples were rerun. For all these repeated cases, the later data were judged to be more accurate and were therefore used in our analysis.

The slurry samples were generally charged to the viscometer at a low temperature (around 400°K), just sufficient to allow the liquid to flow easily. The viscometer temperature was maintained at a constant value while the bob was moved up and down by the magnetic drive at different speeds. Measurement of the force to move the bob at a given velocity allowed calculation of the shear rate and the shear stress. Because the bob moved vertically, the density of the slurry could be calculated from the buoyancy effects. The temperature was then raised to the next measurement value up to a maximum of 725°K (845°F), slightly above the PDU reactor normal operating temperature.

Analysis of initial data indicated that the rheological behavior of these slurries could be modeled by the Bingham model:

$$\tau = \tau(0) + \dot{\gamma} * \text{Plastic Viscosity} \qquad (14)$$

where τ = Shear stress.
$\tau(0)$ = Yield stress.
$\dot{\gamma}$ = Shear rate.
Plastic Viscosity = Rate at which shear stress increases as shear rate is increased.

For a Newtonian fluid, $\tau(0)$ is zero, and the "plastic" viscosity is the normal, shear-independent viscosity.

Figures 59 through 61 illustrate the variance of apparent plastic viscosity, yield stress, and density versus temperature. Attempts to replicate these data at lower temperatures, however, suggested that irreversible changes were occurring in the samples above 700°K. Figure 62 illustrates this phenomenon for Amoco Sample No. 8. After tests at 725°K, the measured viscosity at 700°K

is markedly lower than that obtained during initial heatup. Battelle performed an experiment to estimate the magnitude of this change over time at high temperature. Amoco Sample No. 15 was charged to the viscometer and rapidly heated to 725°K. During the next two hours, the changes in plastic viscosity, yield stress, and density were measured. Figures 63 through 65 present the results. Included for comparison are the values obtained earlier for the sample when the gradual heatup included several tests at lower temperatures.

We speculate that these changes may result from a reaction, preasphaltenes → asphaltenes, which occurs at elevated temperatures. Figure 66 presents data reported in References 55 and 56 contrasting the viscosity of asphaltenes and preasphaltenes. The dashed portions of the lines are extrapolated. Clearly, small decreases in preasphaltene content can translate into significant viscosity decreases. Although the plotted data were developed for Synthoil process liquids, similar trends for viscosity could be expected for the components of H-Coal slurries as well.

The viscosity and yield stress, strongly influenced by intermolecular attractive forces, will be more sensitive to chemical composition than the density. Because Amoco Sample No. 15 had the highest resid content of all the samples tested (31.3%), it is probably the most reactive. Hence, the changes in viscosity observed for this sample in the Battelle viscometer experiments represent an upper limit.

The Oak Ridge National Laboratory viscometer used a different design, as shown in Figure 67. Slurry liquids were again removed from the sample container at low temperature. The liquid was then passed through the preheater coil so that the material reached the test temperature (719°K = 834°F) just as it left the coil array. The liquid then passed through a capillary tube viscometer, where the pressure drop for flow was measured over a known length of tubing. After passing through the viscometer, the liquid sample was set aside and not reused. A previously calibrated gamma-ray apparatus determined the liquid density at the process conditions. Measurement of pressure drop at several flow rates allowed calculation of parameters for a rheological model. ORNL's analysis indicated that either a power law or Bingham plastic viscosity model could adequately describe the data.

Based on the preheater dimensions and ORNL's reported flow rates, a plot illustrating the maximum time the sample spends at different temperatures may be developed. Such a plot appears in Figure 68. By contrast, a time-temperature history for the Battelle test sequence also appears here. Clearly, the ORNL design, with its shorter residence times, is less likely to degrade samples during measurement. ORNL's final report appears in Appendix D.

These reaction effects dictated a new strategy for determining the physical properties at PDU reactor conditions. Figure 69 illustrates Battelle data obtained at lower temperatures for Samples 3 and 4, when experiments were performed to test the effect of changing the hydrogen partial pressure. Within the ± 10% error limits ascribed to the measurement technique by Battelle, no degradation in viscosity is apparent up to 675°K. Therefore,

the following method was selected to estimate the physical properties at process conditions:

Use only data taken at or below T = 650°K. Extrapolate to 719°K using the following models:

Viscosity: Arrhenius model (activated exponential).
Density and Yield Stress: Linear variation with temperature.

For the tests where a sample either was not taken or was lost in processing, correlation techniques were used to estimate the properties. The correlation parameter chosen was the quantity

100% - % Ash - % Coal - % Resid

which corresponds with the extent of conversion to light, low-viscosity products. These correlations are shown in Figures 70 through 72.

Table XIX presents the final values of density and viscosity assigned to the samples. Table XIX also compares the Battelle and ORNL sample results. With the exception of Amoco Sample No. 11 versus ORNL No. 4 (which were taken at low liquid and high gas flow rate), agreement between the two sets of values is reasonable. Note also that the viscosity of the samples as determined by Battelle ranges from 0.9 to 3.4 cp. By comparison, the viscosity of kerosene at 70°F is 1.4 cp, while the viscosity of kerosene/coal char slurries ranges from 3.8 to 4.9 cp.

Analysis and modeling of the PDU-10 data will require use of some fluid dynamics correlations, which will be discussed in a later section. Appendix E documents the method used to combine the plastic viscosity and yield stress into a pseudo-viscosity. The majority of force suspending a catalyst particle will be viscous shear forces rather than yield stresses. This will simplify later analyses by allowing the slurry to be modeled as a Newtonian fluid. This in turn will allow correlations developed for Newtonian fluids such as water to be directly applied to the PDU experimental data.

Data Transformations

The following methods were used to calculate linear gas and slurry velocities from the metered values.

Gas Velocity.--HRI routinely calculates superficial gas velocities at the PDU reactor inlet and outlet for its daily material balance reports. These calculations assume ideal gas behavior. The average of these two velocities (which typically were within 10% of each other) was used for data analysis.

Slurry Velocity.--The superficial liquid velocity is computed by summing its two components, slurry feed and slurry recycle:

$$U_l = \frac{\dot{V}_{\text{Slurry Feed}} + \dot{V}_{\text{Internal Recycle}}}{A} \qquad (15)$$

where $\dot{V}$ = Volumetric flow rate, $ft^3/sec.$
U_l = Slurry superficial velocity, ft/sec.
A = PDU cross-sectional area, $\pi/4\ (8.5")^2$

At reactor conditions, the fresh slurry feed is assumed to have the same density as the recycle slurry. This assumption was based on comparison of room temperature densities of the slurry mix tank (Table XVII) with ORNL measurements of recycle slurry densities at room temperature (Table 5 of Appendix D). Densities for both materials were comparable.

Then:

$$\dot{V}_{\text{Slurry Feed}} = \frac{\text{Slurry Feed Rate, Lb/Hr}}{\rho \times 3{,}600} \qquad (16)$$

The internal recycle rate was monitored by a Venturi meter, previously calibrated with water and then adjusted to an assumed slurry density of 0.85. After slurry densities became available from Battelle and ORNL, the flow rate was adjusted.

$$\dot{V}_{\text{Internal Recycle}} = (\text{Meter gpm/ft}^2)\sqrt{0.85/\rho_{\text{Actual}}} \cdot A \cdot 2.228 \cdot 10^{-3}\ \frac{Ft^3/Sec}{GPM} \qquad (17)$$

Catalyst Bed Expansions

Catalyst bed expansions were determined by gamma-ray absorption measurements. An increase in the ratio (I/I_o), the number of gamma-ray counts under test conditions divided by the comparable count rate at that elevation for an empty reactor, corresponded to the interface between the dense catalyst bed and the less dense dilute phase. In some cases, this interface was quite sharp (Figure 73); for some other tests, the catalyst density slowly decreased with higher elevation, producing a diffuse dense-/dilute-phase interface, as shown in Figure 74. This phenomenon was also observed during some cold-flow tests using Amocat-1A catalyst, as was discussed in Section III.

Per cent catalyst bed expansions were calculated using the formula

$$\% \text{ Bed Expansion} = 100 * \frac{\text{(Expanded Bed Height - Initial Bed Height)}}{\text{(Initial Bed Height - 8")}} \qquad (18)$$

Figure 75 details how elevation values were defined for the PDU reactor. Zero was chosen to be the flange line at the bottom of the reactor vessel. The 8" offset accounts for the height taken up by the plenum chamber and distributor plate (2).

On startup, the PDU reactor was charged with presulfided Amocat-1A catalyst to an initial settled bed height of 96". At the end of the standard PDU test period (Period 41-A), approximately 17 lb of slurry-soaked catalyst was withdrawn from the system. This reduction of inventory was selected

to allow safe operation over a wider range of process variables. The settled initial bed height for all the fluid dynamics tests taken during Periods 41-B and later was 78", as determined by the gamma-ray scans performed during the shutdown sequence.

A listing of catalyst bed expansions for the 14 tests is in Table XX. Also included are the liquid and gas flow rates and the estimated viscosity and density of the reactor slurry liquid.

By choosing selected tests, consistency checks on the data are possible. Table XXI compares four tests where the nominal slurry viscosity was roughly constant at 2.6 cp. In going from Test 4 to Test 6 to Test 14 at constant gas velocity, the expected trend of increasing bed expansion with increasing liquid velocity is apparent. Comparing Test 14 with Test 13, where the liquid velocity remained constant and the gas velocity was decreased, the bed expansion decreased from 96% to 73%. Both these responses have been observed in cold-flow studies, as discussed in Section III.

Comparison of the magnitude of PDU catalyst bed expansions with those observed in cold-flow studies, however, reveals that the expansions in the PDU are significantly higher than other pilot plant tests with fluids of the same nominal viscosity. Run 224 was performed in the Amoco fluid dynamics cold-flow pilot plant to study the fluid dynamics of the system using Amocat-1A catalyst and a 25 wt% slurry of coal char fines in kerosene. The estimated slurry viscosity is 4.5 cp. Table XXII compares the observed bed expansions during PDU-10 with those for cold-flow Run 224. In all cases, the PDU bed expansions are equal to or (usually) are greater than the cold-flow test bed expansions, even though the maximum measured PDU slurry viscosity was roughly 3.5 cp. Discussions of reasons for this discrepancy will be presented shortly.

Phase Volume Fractions

A primary goal of the experimental program was to measure the volume fractions (holdups) occupied by the gas, liquid slurry, and catalyst phases for model development. The gamma-ray elevator described earlier was the chief instrument to obtain these data.

When a collimated beam of gamma-rays passes through matter, it is attenuated at an exponential rate (64):

$$I/I_o = e^{-\rho \mu L} \qquad (19)$$

where I = Attenuated intensity.
I_o = Original intensity.
ρ = Material density.
μ = Material mass absorption coefficient.
L = Material thickness.

For gamma radiation from a Cs-137 source, the mass absorption coefficients for most elements are constant. However, hydrogen, because of its higher number of electrons per unit mass, has a mass absorption coefficient roughly twice as large as other elements (64).

Mass Absorption Coefficients for the PDU Fluid Dynamics Tests.--Mass absorption coefficients for the PDU catalyst and liquids were determined by combining the empty reactor zero gamma-ray scan data with the data taken during the shutdown (9/24/80 at 7:52 PM). Because temperatures were close to ambient, the following formulas could be used:

In the Catalyst Bed:

$$\frac{-\ln(I/I_o)}{L} = \rho_L \varepsilon_L \mu_L + \rho_c \varepsilon_c \mu_c \qquad (20)$$

Above the Catalyst Bed:

$$\frac{-\ln I/I_o}{L} = \rho_L \mu_L \qquad (21)$$

HRI provided the temperature history for this shutdown period and the density versus temperature relation for the shutdown oil. Figure 76 illustrates this relationship, with annotation of the three shutdown test scan times. Table XXIII details the I/I_o versus elevation profile of the 9/24 PM test. Tables XXIV and XXV present the calculation of the liquid and catalyst mass absorption coefficients. Because ORNL studies on various pure liquids and coal-derived slurries found no significant variations of mass absorption coefficient with composition (65), these coefficients were judged applicable to all the PDU tests.

Volume Fractions.--With the mass absorption coefficients determined in the previous section and densities available (from Tables XVIII and XIX), the following relationships were used to compute the volume fractions occupied by gas, catalyst, and slurry liquid:

$$\ln (I_t/I_o) = -L(\rho_{sl}\mu_{sl}\varepsilon_{sl} + \rho_g \mu_g \varepsilon_g) - \rho_{st}\mu_{st}L_{st}\left[\frac{1}{(1+\alpha\Delta T)^2} - 1\right] \qquad (22)$$

The second term on the right-hand side of the equation (very small compared with the first) arises from the fact that the zero scan is performed at ambient temperature, while the operating scans were performed at elevated temperatures.

The gas density was calculated from the ideal gas law to be 0.42 lb/ft^3. The gas mass absorption coefficient was taken to be twice the liquid value.

The catalyst volume fraction ε_c is determined from the catalyst bed expansion:

$$\varepsilon_c = \varepsilon_{co}\left(\frac{100}{100 + \%\ \text{Bed Expansion}}\right) \qquad (23)$$

where ε_{CO} (the volume fraction at settled conditions) is determined by the ratio of dry settled density to dry particle density:

$$\varepsilon_{CO} = \rho_B/\rho_P \tag{24}$$

The gas holdup is computed by difference:

$$\varepsilon_g = 1 - \varepsilon_C - \varepsilon_{sl} \tag{25}$$

Simultaneous solution of Equations 22, 23, and 24 provides values for ε_C, ε_{sl}, and ε_g.

Assorted experimental values used in calculating these holdups appear in Table XXVI. Calculated holdups for the dense phase (gas/slurry/catalyst) region of the reactor are tabulated in Table XXVII. Also included is the Darton-Harrison gas drift flux, defined by

$$V_{CD} = \varepsilon_g\ (1 - \varepsilon_g)(U_g/\varepsilon_g - U_s/\varepsilon_{sl}) \tag{26}$$

Corresponding gas/slurry dilute-phase holdups appear in Table XXVIII.

Discussion

Examination of the holdups in Table XXVII indicates several unexpected results. For Tests 1 and 2, the gamma-ray attenuation was such that the projected volume fraction of the liquid was excessively high. This caused the calculated holdup of the gas to be less than zero, clearly not a physically realistic situation.

The drift flux, V_{CD}, is a measure of how rapidly the gas bubbles are rising relative to the average liquid flow rate. A negative drift flux suggests that the bubbles are moving down rather than up. The apparent negative value for the drift flux in Test 5 is as physically unrealistic as the sub-zero gas volume fractions in Tests 1 and 2.

These errors could have been caused by

A) Incorrect assignment of the liquid density: If the liquid were more dense, less volume would be required to provide the proper gamma-ray attenuation. For Test 1, however, two duplicate samples were taken from the separator bottoms. Battelle's independent density measurements agreed within 1% of each other.

B) Incorrect catalyst density: At the beginning of the run, the catalyst was presulfided but was assumed to be free of contaminant ash deposits.

C) Incorrect liquid mass absorption coefficient: For startup, atmospheric still bottoms were used as slurry liquid until sufficient recycle solvent

had been generated in the run. These atmospheric still bottoms had a high hydrogen content and a higher hydrogen donor ability than later solvent (Tables 8 and 29, Reference 2). However, this higher hydrogen content should not have caused a significant (greater than 10%) increase in the mass absorption coefficient.

D) Reactor liquid homogeneity: If the PDU liquid flow regime were not a perfectly well-mixed CSTR, gradients in coal conversion and physical properties could exist. In this case, the sample from the separator may not be representative of the entire system. Although these property gradients may have occurred in all the tests, the runs with the most reactive conditions (freshest catalyst plus highest solvent hydrogen donor activity) would be expected to exhibit the largest inhomogeneities.

Comparison with Cold-Flow Pilot Plant Tests

Table XXIX compares the physical properties for the PDU-10 tests with corresponding values for four cold-flow pilot plant tests using coal char/kerosene slurries. Amoco Run 224 was performed using Amocat-1A catalyst to measure its fluid dynamic performance with a well-defined slurry having approximately the same fines content as the PDU slurries. Although the HRI slurries are slightly denser, the viscosity (which has a much stronger influence on such variables as catalyst bed expansion) is lower.

Table XXX contrasts the observed gas holdups in the PDU-10 tests with those at corresponding superficial velocities in the Amoco cold-flow tests. Most of the gas holdups in the PDU are significantly higher than corresponding cold-flow values.

Analysis of earlier H-Coal cold-flow pilot plant data developed under a prior contract (1) indicated two effects of liquid slurry viscosity:

A) As slurry viscosity was increased, catalyst bed expansions increased.

B) As slurry viscosity was increased, gas holdups decreased. This was attributed to the effect of increased viscosity in accelerating the coalescence of gas into large, rapidly rising bubbles.

The difference in viscosity of preasphaltene and asphaltene species (Figure 66) may be influencing the PDU fluid dynamic behavior. The HRI PDU slurry recycle system is designed to exclude gas and return only slurry from the reactor top. This material's composition is nominally identical with the high-pressure separator bottoms stream, which was sampled and later characterized. The fresh feed from the slurry mix tank accounted for 10-25% of the total reactor liquid flow. The viscosity of the fresh feed leaving the unit preheater, containing either suspended or partially dissolved coal, may be significantly higher than the recycle stream, which has undergone chemical reaction. If the PDU behaved as a perfectly back-mixed reactor, the separator bottoms would be representative of the material in the reactor. However, if any deviations from CSTR performance exist, then the introduction of the high-viscosity preheater outlet liquid could cause a significant gradient of viscosity and composition across the reactor.

The high incoming viscosity would cause a high bed expansion. A continuous generation of hydrogen and other light gases would ensure a large holdup of small bubbles. If this did occur, then both high bed expansions and high gas holdups could occur simultaneously. Examination of the I/I_o ratio for several tests in Appendix F (3,6,7,8, for example) shows a far wider variation of this ratio with elevation than that observed for the shutdown tests (15 and 16) or the zero scan.

Figure 77 is a conventional plot of the drift flux versus gas holdup in the dense bed (gas/slurry/catalyst) portion of the reactor. The three tests (1, 2, and 5) which exhibited apparently negative gas holdups or drift fluxes are not included on the graph. Also shown for comparison is the line defining ideal bubbly operation developed in the pilot plant cold-flow tests with kerosene (Run 218).

Comparison of the PDU data with the composite cold-flow results illustrates that the bulk of the operating reactor gas flow is in the ideal bubbly regime. Because the slope of the V_{CD} versus ε_g line is related to the bubble rise velocity, the location of most PDU points below the composite line suggests that the gas bubbles in these tests are rising quite slowly. Tests 8 and 10 clearly lie in a churn turbulent area of operation. These two tests were performed at high gas velocity (0.09 ft/sec) and low or medium liquid velocities (0.050-0.075 ft/sec). The transition from ideal bubbly to churn turbulent flow had previously been observed in cold-flow tests under conditions of high gas velocity and low liquid flow rate. Test 12, performed at both high gas and high liquid superficial velocities, lies close to the ideal bubbly line, although it is clearly separated from the other tests. The effect of increasing liquid velocity in delaying the change from ideal bubbly to churn turbulent operation was noted previously (1).

The liquid slurry in Tests 8 and 12 also exhibited the highest viscosities (3.80 and 3.56 cp, respectively, as measured by Battelle). Previous data (1) established the effect of increased slurry viscosity in accelerating coalescence of gas into large bubbles with rapid rise velocities; the corresponding gas holdup decreased. Under these conditions, the calculated drift flux assumes a large value, placing operation in the churn-turbulent regime. However, Test 10, with a (Battelle-measured) viscosity of 2.11 cp, lay further in the churn turbulent region than either Test 8 or 12. It is probable that the high gas velocity influenced the fluid dynamics more than did the slurry viscosity.

VIII) MATHEMATICAL MODEL

A variety of models have been proposed in the literature to describe the fluid dynamics of a three-phase fluidized bed. A review of these models was presented in Reference 1.

No completely theoretical models were found during the literature search. Model approaches developed by other authors have tended to be in one of two categories:

A) Empirical correlation: Phase holdups are correlated against operating variables.

B) Semi-empirical models: Phase holdups are calculated from the results of model calculations. Specific assumptions are postulated about the behavior of bubbles, wakes, and solid phases. However, each model "law" incorporates a parameter which may be described by a correlation approach rather than an a priori calculation.

Review of the Bhatia-Epstein Model

The Bhatia-Epstein model was selected during the initial contract (1) for several perceived advantages over other models:

A) It is able to predict both catalyst bed expansions and bed contractions when gas is introduced into a liquid-fluidized bed.

B) It is able to describe operation both in the ideal bubbly and churn turbulent regimes.

C) Its parameters (bubble rise velocity, wake structure, and wake composition) are amenable to a physical interpretation and may be checked by such experimental approaches as radiotracer techniques.

Figure 78 illustrates the basic features of the Bhatia-Epstein model. The system is divided into three component phases: gas bubbles, wakes behind the bubbles, and particulate (catalyst plus liquid slurry) phases. The equations of the model are presented in Table XXXI. Equation 1a is not strictly an independent equation but may be derived from the other nine. A short discussion after each equation indicates which are definitions and which incorporate specific models or assumptions.

A key independent variable of the model is the bubble rise velocity, U_{tb}. This variable, relating how rapidly bubbles rise relative to the liquid, will control the gas holdup. Additionally, because the bubbles are assumed to drag their wakes behind them at the same velocity, a mechanism is incorporated which allows liquid to be transported through the system at a higher velocity.

A second model parameter is related to the wake structure. This is the wake volume ratio, K_o, the ratio of the wake volume to the gas bubble volume for a single gas bubble at an infinitesimal gas volume fraction. Experimental studies of the relationship between wake and bubble volumes have been performed in liquids. Henriksen and Ostergaard (66) noted that the wake volume behind a single bubble was that which completed the sphere whose upper surface was defined by the bubble top. In turn, the bubble shape was strongly influenced by the viscosity of the liquid. These observations are incorporated in a semi-empirical fashion into the model by defining upper and lower limits to the parameter.

The wake behind the bubbles is assumed to entrain both slurry and catalyst. The amount of catalyst carried by the wake is described by the parameter

X_k. If X_k is 0, the wake is assumed to be composed solely of liquid. For X_k = 1, the ratio of solid content to liquid content in the wake is the same as that existing in the particulate phase, where slurry fluidization is expanding the catalyst bed.

The parameter X_k was discussed by El-Temtamy and Epstein (67) in a paper which appeared after the basic model was published. We find from our cold-flow studies that the Bhatia-Epstein model can describe the experimental results by using the published correlation for X_k. In the PDU-10 modeling, X_k was taken to be an independent variable along with U_{tB} and K_o.

Model Parameters

Because it is assumed in the Bhatia-Epstein model that the slurry-fluidized portion of the bed may be described by the Richardson-Zaki correlation, values of the catalyst terminal velocity (U_t) and the index (n) must be available. In the cold-flow pilot plant, these two parameters are obtained by performing a series of two-phase slurry fluidization experiments, where changes in the bed expansion are measured as a function of superficial slurry velocity. The slurry composition remains constant for the tests, and there is no gas flow.

During the PDU experimental program, however, it was not possible to determine these parameters. The density and viscosity of the liquid slurry changed as reactor operating conditions were altered during the test period. Additionally, failure to maintain continuous gas flow through the inlet lines would have resulted in coking and blockage.

A parallel analysis of liquid/solid fluidization data in the literature was performed under this contract. A major conclusion of the study (full report attached in Appendix G) is that the relation between the drag coefficient and Reynolds number for an infinitely long cylinder can be applied to the finite catalyst extrudates used in the PDU reactor by proper choice of the diameter, d. Use of these correlations then allows the two Richardson-Zaki parameters (U_t and n) to be estimated solely from physical properties.

The slurry density (ρ_l) and viscosity (μ), along with the catalyst density (ρ_s) and diameter (d) are used to calculate the Galileo number Ga:

$$Ga = \frac{d^3 \cdot \rho_l \cdot g \cdot (\rho_s - \rho_l)}{\mu^2} \qquad (27)$$

Although the slurry is a Bingham plastic (58), the analysis in Appendix E indicated that the bulk of the forces are related to the fluid's plastic viscosity. Hence, this treatment (which is strictly proper only for a Newtonian fluid) is valid here. With the Galileo number established, the terminal Reynolds number for a single particle is calculated. The literature correlation for this is shown in Figures 2 and 8 of Appendix G.

The Reynolds number, Re, is defined by

$$Re = \frac{d * u_t * \rho_l}{\mu} \qquad (28)$$

The particle terminal velocity, u_t, is the only unknown.

Richardson and Zaki presented a correlation for the index n of spheres in their original paper. Figures 9 and 10 of Appendix G show that this correlation is applicable to cylinders as well.

These calculations were performed for all 14 PDU-10 tests. Table XXXII tabulates the individual fluid properties (density and viscosity) and the predicted terminal Reynolds numbers, terminal velocities (U_t), and Richardson-Zaki index (n) for each test in the PDU experimental program.

Figure 79 illustrates the range of Galileo numbers over which the Reynolds number was predicted using the literature correlation. Some experimentally measured cold-flow pilot plant data points are included for comparison. Since both the Galileo number and the terminal Reynolds number can be measured for these cold-flow tests, the individual data points may not fall precisely on the line summarizing prior literature data. Figure 80 is the corresponding prediction for the index n in the PDU-10 tests. Again, experimentally determined values of n for four cold-flow slurry fluidization tests are also presented. With the exception of Test 221, the agreement between the slurry data points and the Richardson-Zaki correlation for n is quite good.

Because the Bhatia-Epstein model incorporates the predicted parameters U_t and n in Equation 4 of Table XXXI, the sensitivity of the model predictions to changes in the fluid viscosity was also explored. As was speculated in Section VII, the average viscosity in the catalyst bed could have been higher than that assigned to the separator bottoms liquid. Therefore, each viscosity was multiplied by 4. This magnitude of increase was selected to raise the PDU liquid viscosities to the maximum level tested during the prior contract (1). Test Series 420 and 427, documented in Reference 1, employed mineral oil at a temperature of 100°F to fluidize a catalyst bed. The liquid viscosity was 14.6 cp. Comparison of these bed expansions with those for a 4 cp fluid indicated that the viscosity increased bed expansion by a factor of about 2.5. This is roughly the maximum discrepancy between cold-flow and PDU tests noted in Table XXII.

Increasing the viscosity affects the Galileo number (Equation 27). Hence, the terminal Reynolds number and the Richardson-Zaki index n will need to be recalculated for the high-viscosity case. Table XXXIII summarizes the new calculations, which assume each test's viscosity to be the Battelle value multiplied by 4. Figures 81 and 82 compare the newly calculated Galileo numbers for the PDU tests and include the cold-flow tests as a point of reference. As expected, increasing the viscosity does decrease the terminal velocity.

Cold-Flow Tests

The Bhatia-Epstein model was applied to analysis of the data from both the cold-flow (kerosene/coal fines slurry) tests and the HRI studies. Tables XXXV

to XXXVIII contain the observed volume fractions and X_k wake concentration ratio (predicted by correlation in Reference 67) and list the two parameters U_{tb} (bubble terminal velocity) and K_o (the single bubble wake volume ratio) for the cold-flow, three-phase fluidization tests.

The quantity Δ is a measure of how well the model is able to fit the data and is defined by the relation

$$\Delta = \text{Sum}[\varepsilon(\text{Predicted}) - \varepsilon(\text{Observed})] \qquad (29)$$

A zero value of Δ indicates that when given the listed parameters, the model was able to predict the observed volume fractions with no error.

Bubble diameters were calculated from the bubble rise velocities using the Peebles and Garber correlation (68). Table XXXIV presents the four equations for the effective radius r(e) which apply for different Reynolds number ranges. The data were developed for single isolated bubbles in a liquid medium. The correlation predicts a maximum bubble velocity at the boundary between Regions 2 and 3 (where further increases in the bubble diameter cause the bubble to rise more slowly).

The rise velocity of single bubbles is controlled by a balance of the viscous drag or surface tension force and buoyancy effects. Application to the three fluidized-bed experiments introduces two additional unknowns:

A) The Peebles and Garber experiments did not address the effect of a finite gas holdup upon the average bubble rise velocity. Bhatia (69) correlated a variety of experimental work and suggested that swarms of bubbles rose less rapidly than single bubbles. Other authors (50) have postulated the opposite effect, with non-zero gas holdups increasing the gas/liquid relative velocity.

B) In their correlations, Peebles and Garber use fluid properties such as the density, viscosity, and surface tension. In the three-phase system, choice of values to assign these variables is somewhat arbitrary. A macroscopic viscosity may be assigned to the fluidized catalyst bed based on its resistance to stirring. However, the drag that individual particles exert on a gas bubble may be distinctly different than that of a liquid which is homogeneous on a molecular scale.

These effects are visible upon examination of the predicted bubble diameters in Tables XXXV to XXXVIII. For cases where bubble rise velocities above 0.8 ft/sec were predicted, no corresponding diameter could be calculated by the Peebles and Garber correlation. Previous radiotracer experiments with kerosene/coal char fines (1), however, indicated that some gas bubbles were rising through the bed with velocities of approximately 1 ft/sec.

Figures 83 through 86 correlate the terminal velocity versus $(U_g - U_l)$ for the four cold-flow test series. This choice of the form for the independent correlating variable was discussed in Reference 1. Briefly, previous experiments had noted that as gas velocity was increased in catalyst beds fluidized

either by kerosene/coal char slurries or by high-viscosity fluids such as mineral oil, the nature of the gas flow changed from ideal bubbly to churn turbulent. Increasing the liquid velocity tended to stabilize the flow against this transition. Because churn turbulent operation is characterized by rapid bubble coalescence and high bubble rise velocities, the Bhatia-Epstein model bubble rise velocity U_{tb} could be expected to increase in this region. Prior data (1), as well as the currently developed data shown here, support this view.

In these four figures, the correlation line developed for cold-flow tests with a 17.8% coal char fines/kerosene slurry from Reference 1 are presented for comparison. Bubble rise velocities from kerosene-only fluidization tests are also shown. For all four tests (221-224), the behavior of the bubble rise velocity versus $(U_g - U_l)$ appears to track that previously determined for the 17.8% coal char slurry. As slurry content increases, average bubble rise velocities also increase. This is consistent with the effect of the increased viscosity in promoting coalescence of gas bubbles. The magnitude of the scatter in the model fit becomes evident with the larger number of tests to which the model was applied.

The wake structure effects are described by the wake volume ratio, K_o. If the wake volume is large and the bubbles are rapidly rising, the wake may entrain significant amounts of catalyst and liquid. This in turn may cause the catalyst bed to contract with further increases in gas velocity (4). Alternatively, when the wake volumes are small and the bubble rise velocities are low, wake effects will not be as important in determining the component holdups.

Figures 87 to 90 present the model's single bubble wake volume ratio data plotted against liquid superficial velocity for the four cold-flow tests. The choice of liquid velocity for this relationship was developed in Reference 1. Following the approach of Henriksen and Ostergaard (66) and El-Temtamy and Epstein (67), the wake volume ratio behind the gas bubbles is expected to be controlled by the "viscosity" of the fluidized bed. As the catalyst bed expands, its viscosity decreases (66). The primary variable controlling bed expansion is the liquid velocity (U_l), with gas velocity playing a secondary role. For this reason, the wake volume ratio is correlated against liquid velocity.

The wake volume ratio correlation line developed for cold-flow tests with a 17.8% coal char fines/kerosene (1) slurry are presented in Figures 87 through 90 for comparison. Some wake volume ratios from the four cold-flow tests are lower than those developed in Reference 1. However, the trend of increasing wake volume with increasing liquid velocity is observed.

Application to HRI PDU-10 Tests

This model was then applied to the PDU tests. Of the 14 three-phase tests performed, three were not suitable for analysis by the Bhatia-Epstein model:

Tests 1 and 2, where the gas volume fractions were less than zero.

Test 5, where the drift flux was less than zero. The structure of the model requires bubbles to rise faster than the surrounding liquid, and a negative drift flux corresponds to the case of the bubbles rising more slowly.

The Richardson-Zaki parameters U_t and n are used in the Bhatia-Epstein model to describe the effect of liquid velocity on catalyst bed expansion. These therefore will have a major effect on predictions of total catalyst and liquid volume fraction, while the bubble rise velocity will primarily control the gas-phase holdup. Hence, if the model is able to predict gas volume fractions properly but cannot match the observed catalyst and liquid fractions, one solution may lie in modifying U_t and n.

The model was initially tested using the first set of Richardson-Zaki parameters in Table XXXII. These values were derived by assuming that the characterizations of separator liquid samples developed by Battelle could model the liquid present in the reactor's catalyst bed. Table XXXIX (with significant non-zero values for Δ in Tests 9, 10, and 11) illustrates that the model was unable to match many of the observed catalyst and liquid volume fractions. These failures, coupled with the catalyst bed expansions which were higher than expected, led us to examine the effect of increasing the viscosity in the reactor.

To simulate the added viscosity of the incoming slurry from the preheater, the product viscosities were multiplied by 4. New values of the Richardson-Zaki parameters corresponding to the higher viscosity are presented in Table XXXIII. Incorporating this assumption proved much more successful in matching the bulk of the observed data. Table XXXX compares the model predictions with observed volume fractions. The model is now able to match successfully all the experimental points.

As was consistent with the drift flux plot (Figure 77), most of the PDU runs may be characterized by a low bubble rise velocity, low wake effects, and a high gas holdup. Only two tests (Tests 8 and 10) exhibit high bubble rise velocities and large values of $X_{\dot{K}}$, indicating significant wake effects. A direct comparison of the single bubble wake volume ratios, K_o, does not show significant differences; however, the exponential weighting with gas holdup (Equation 8 of Table XXXI) occurs over a much wider range than was previously noted in cold-flow tests. Table XXXXI illustrates the effect. Tests 8 and 10 have a significant ratio of wake volume to bubble volume. These two tests were previously identified as being in the churn turbulent regime by the drift flux analysis. Test 13 has a high ratio of wake volume to bubble volume; however, because the bubble rise velocity for this test is low, wake effects will not be significant.

For tests such as Tests 9, 11, and 13, the predicted bubble rise velocities are of the same order of magnitude as $2U_g$. At these low bubble velocities, the applicability of Equation 9 of Table XXXI may be questionable. Other variants of model for relative gas/liquid velocity were briefly tested; however, they all predicted relatively low bubble rise velocities for these high-gas-holdup tests.

Comparison of PDU and Cold-Flow Pilot Plant Model Results

A primary aim of this investigation was to assess the suitability of the Bhatia-Epstein model for description of the fluid dynamics in the PDU reactor and to relate the model parameters for the operating reactor to those developed for the cold-flow pilot plant. The bubble rise velocities calculated using the high (4x) viscosity Richardson-Zaki numbers from the PDU tests are contrasted in Figure 91 with those developed in the cold-flow unit Run 224. Figure 92 enlarges the region where the bulk of the PDU and pilot plant data lie.

Cold-flow Run 224 was selected to be most representative of the PDU-10 test, since it used Amocat-1A catalyst and had 25 wt% fines suspended in the liquid, a concentration comparable to the PDU reactor slurries. Two data points from the PDU run (Test 8, in the churn turbulent regime, and Test 13, where the gas flow was judged to be ideal bubbly) do lie within the data range for the cold flow results. Test 10 lies far above the cold-flow data range. The slurry from Test 8 did have the highest viscosity observed (3.8 cp from Battelle's measurements), but several other tests had viscosities higher than the 2.74 cp of Test 13. Test 10, with the highest bubble rise velocity, had an intermediate viscosity value of 2.11 cp. The bulk of the PDU tests exhibit lower bubble rise velocities. This is a direct consequence of the higher gas holdups and lower drift fluxes of the PDU tests as compared with the cold-flow (Run 224) tests.

The failure of the bubble rise velocities to correlate with changes in viscosity again suggests that chemical reaction or other factors unique to the PDU reactor are exerting a strong influence on the gas bubble behavior. For example, reactor pressure alone can affect bubble coalescence phenomena and therefore bubble size and gas holdup. Recent data obtained for the EDS coal liquefaction process (70) showed that gas holdup in the actual reactor operated at higher pressure is significantly higher than the gas holdup in the lower-pressure cold-flow unit. The gas holdup observed in their cold-flow pilot plant was approximately 60% higher at 75 psig versus 20 psig. Although these results (70) were obtained for a gas-slurry reactor with no catalyst present, the trends observed are in agreement with our findings of higher gas holdup in the PDU (high pressure) reactor as compared with the atmospheric pressure cold-flow unit.

Values of K_o (the single bubble wake volume ratio) from the PDU tests are contrasted with cold-flow Run 224 tests in Figure 93. A direct comparison of these two items, however, must be tempered by including the effect of the gas holdup. The ratio of wake volume to bubble volume will be decreased by the exponential weighting in Equation 8 of Table XXXI. Because the PDU runs generally exhibited higher gas volume fractions than those of the cold- flow tests, the wake volumes will be decreased more.

Wake volume ratios for Run 224 calculated by Equation 8 of Table XXXI are listed in Table XXXXII and may be compared with the PDU data in Table XXXXI. With the exception of the three PDU-10 experiments (8, 10, and 13), the wake volume to bubble volume ratios for Run 224 appear comparable to those for the PDU tests.