

ABSTRACT

The fluid dynamics occurring in HRI's H-coal process development unit (PDU) coal liquefaction reactor during Run PDU-10 were measured and compared with Amoco Oil cold-flow fluidization results. It was found that catalyst bed expansions and gas holdups are higher in the PDU than those observed in the cold-flow tests for slurries having the same nominal viscosity. Comparison of PDU results with cold-flow results shows that the bulk of the operating reactor gas flow lies in the ideal bubbly regime. It also appears that the gas bubbles in these PDU tests are rising quite slowly. Only two of the operating points in our test program on the PDU were found to lie in the churn turbulent regime. Existence of churn turbulent behavior during these two experiments is consistent with trends observed in earlier cold-flow experiments.

Two- and three-phase fluidization experiments were carried out in Amoco's cold-flow fluid dynamics unit. The data base now includes fluidization results for coal char/kerosene slurry concentrations of 4.0, 9.8, and 20.7 vol% in addition to the 15.5 and 17.8 vol% data from our earlier work. Both HDS-2A and Amocat-1A catalysts were used in the tests. Bed expansion is primarily a function of slurry velocity, with gas velocity having only a weak effect. Bed contractions have been observed in some cases at sufficiently high gas velocity. Gas and liquid holdups were found to be uniform across the cross-section of the Amoco cold-flow fluid dynamics pilot plant.

A viscometer was adapted for measurement of the viscosity of coal slurries at high temperature and pressure. Three techniques were developed for the study of the bubble dynamics occurring in three-phase fluidized beds: 1) A laser light beam probe for measuring the behavior of bubbles greater than 120 microns; 2) A laser holographic technique for determining the size, shape, and position of bubbles in the bed; and 3) A resistivity probe for determining bubble frequencies and a qualitative measure of the flow regime. The bubble size studies showed that the design of the inlet distributor played an important role in the breakup of bubbles entering the fluidized bed. It was shown that smaller bubbles can be generated by this particular inlet distributor than could be produced by the breakup of bubbles by solid particles within the bed.

Based on experiments carried out in the Amoco cold-flow unit, a significant degree of backmixing was found to occur in the H-Coal system. Dispersion coefficients and Peclet numbers were found to lie in the ranges 70 to 130 cm²/sec and 3.6 to 9.9, respectively.

I) INTRODUCTION

The H-Coal process, developed by Hydrocarbon Research, Incorporated (HRI), involves the direct catalytic hydroliquefaction of coal to low-sulfur boiler fuel or synthetic crude oil. The process has been demonstrated in a 3 T/D process development unit (PDU) at HRI's Trenton, New Jersey, facilities and in a 600 T/D pilot plant at Catlettsburg, Kentucky.

The HRI H-Coal process involves the use of an ebullated-bed reactor, as shown in Figure 1. The catalyst in the reactor is fluidized by the flow of slurry and hydrogen gas passing up through the reactor. The slurry stream entering the reactor consists of fresh coal slurry feed and a recycle slurry stream. The gas stream consists of recycled hydrogen and fresh make-up hydrogen.

The design, operation, and control of the reactor requires a detailed understanding of the fluid dynamics of this complex reacting system. Factors affecting the performance of the reactor and information needed for design include the following:

- A) Catalyst bed expansion.
- B) Void fraction (phase holdup) of the gas, slurry, and catalyst phases and their variation along the reactor length.
- C) Bubble dynamics, including wake effects.
- D) Mixing parameters: Axial and radial mass dispersion of the gas, slurry, and catalyst phases, and axial and radial dispersion of heat.
- E) Transport parameters: Gas/liquid and liquid/solid mass transfer coefficients and gas/liquid interfacial area.
- F) Thermodynamic parameters: Vapor/liquid thermodynamic data, specific heats, thermal conductivities.
- G) Reaction data: Kinetics, conversion, product yields, and product qualities.

Our previous research on the fluid dynamics of the H-Coal system (1) addressed several of the above items (A, B, and part of C and D). A cold-flow pilot plant was built which is dimensionally identical with the H-Coal PDU reactor shown in Figure 1. Cold-flow fluidization experiments were carried out using kerosene, mineral oil, and several coal char/kerosene slurries. The gases included nitrogen and helium. A standard (1/16" x 3/16") catalyst, Cyanamid HDS-2A, was used in the majority of the experiments. Direct sampling of the slurry at various points throughout the system showed that the concentration of the slurry was uniform throughout the system and thus could be treated as a single phase.

Bed expansion data and phase holdup data were collected for a variety of gas and liquid (slurry) flow rates. Although the majority of the data were collected using pure fluids of the appropriate viscosity, some data were also collected using coal slurries. Analysis determined that the Richardson-Zaki correlation describes the effect of slurry flow rate on catalyst bed expansion. Bed expansion is primarily a function of liquid (slurry) velocity, with gas velocity having a secondary effect. When a lower-viscosity pure fluid such as kerosene is the fluid phase, an increase in gas velocity always results in an increase in bed expansion. However, when a higher-viscosity fluid is used such as mineral oil or a coal char/kerosene slurry, a slight contraction of the fluidized catalyst bed can occur above a sufficiently high gas velocity.

The experimental data were used to develop a modified Bhatia-Epstein model which can predict bed expansion and phase holdups for the various phases as a function of gas and liquid (slurry) superficial velocities. It was shown that the bed could be described by the gas bubbles, wakes following the bubbles, and the slurry/catalyst particulate phase, as shown in Figure 2. Correlations were derived describing bubble rise velocity, wake volume, and solids holdup in the wake as a function of operating conditions.

The nature of the bubble dynamics for the three-phase system was studied using the Darton-Harrison drift flux model. Analysis showed that ideal bubbly flow occurs in the ebullated bed over much of the operating range when pure kerosene is used as the liquid phase. Above a certain velocity, however, the gas flow will change from small bubbles (ideal bubbly) to large bubbles (churn turbulent). The latter type of flow results in an unsteady flow behavior in the bed. When a high-viscosity fluid such as mineral oil or a coal char/kerosene slurry is the liquid phase, the gas flows in the form of large bubbles over much of the gas velocity range. Mass transfer effects and chemical reactions depend strongly on bubble size distribution and residence time; and therefore, knowledge of the type of gas flow occurring in the system is very important.

Radioactive gas tracer studies were also carried out to investigate the nature of the bubble dynamics and determine the extent of gas dispersion in the H-Coal system. It was determined that the flow of gas through the catalyst bed is intermediate between plug flow and backmixed. It was also concluded that at sufficient gas velocity, large bubbles having a higher rise velocity are flowing up the center of the column with a flow of smaller bubbles down along the wall.

Reaction data (Item G), including conversion, product yields, and product qualities, have been obtained by HRI (2) during operation of the 3 T/D PDU under a variety of operating conditions. Micro-autoclave data have also been obtained by HRI as a supplement to the PDU data and as an operational aid.

Although the above results provided a better understanding of the fluid dynamics in the three-phase fluidized H-Coal system, several major and important areas remained unexplored and required investigation. This, then, is the basis for our current work, and the results are discussed in this report.

II) OBJECTIVES AND SCOPE OF PRESENT WORK

Task 1: Model Validation

A major question in fluidization research is how well cold-flow experiments conducted at ambient temperature and pressure simulate the fluid dynamics occurring in a reacting system under reaction conditions. With regard to our studies, a corollary question involves how well the modified Bhatia-Epstein model based on cold-flow data predicts bed expansions and phase holdups for the H-Coal system. An experimental program was conducted in this current study on the HRI PDU during Test PDU-10 to obtain the necessary fluidization data. The objective was to compare the fluid dynamics measured in the actual H-Coal reactor with those measured in the cold-flow pilot plant experiments. The analysis included a critical evaluation of our modified Bhatia-Epstein model which had been developed based on cold-flow data.

Density, viscosity, and surface tension of the liquid phase play important roles in three-phase fluidization. Samples of the slurry were withdrawn from the PDU reactor by HRI during the PDU-10 experiments for analysis. As far as we know, there is no readily available technique for measurement of the surface tension of coal slurries at high temperature and pressure. Measurement of the density is relatively straightforward. The viscosities of the slurry samples were determined by Battelle Research Institute and Oak Ridge National Laboratory in their viscometers at reaction temperature and pressure. Battelle had successfully used the same technique previously for viscosity measurement of Synthoil products. An objective of our current work was the adaption by Battelle of their viscometer to the measurement of H-Coal reactor samples at reaction temperature and pressure.

Although not funded by this contract, four H-Coal reactor slurry samples were sent to Oak Ridge for viscosity measurement in their pipeline viscometer. The purpose was to make a comparison of the viscosity values from two independent techniques.

An additional objective of Task 1 was to obtain additional bed expansion and phase holdup data for coal char/kerosene slurries having several concentrations. Although the earlier slurry data were valuable, they were limited in scope. This additional fluidization information will enlarge the slurry data base and provide additional confidence to our modeling efforts.

Task 2: Bubble Dynamics Studies

It was clear from our earlier work as well as results in the literature for other experimental systems that a basic understanding of the bubble dynamics in the system is essential. The objective of this task was to determine gas holdup, bubble size distribution, and respective bubble velocities for a given set of operating conditions. A major part of this task to be carried out by Northwestern University researchers was to develop techniques suitable for studying such phenomena in this complex three-phase fluidized bed system. The technical plan included development of a resistivity probe, a narrow-beam laser probe, a laser holographic technique,

and a laser-Doppler anemometric technique. (It turned out that only the laser-Doppler anemometry could not be developed, because the bubbles were smaller than anticipated due to the nature of the fluids chosen in this part of the cold-flow study. This precluded study of the liquid flow patterns using this technique.)

Task 3: Gas/Liquid Mixing

Gas Mixing.--A common assumption in many modeling studies, including our modified Bhatia-Epstein model, involves radial uniformity in the system. Results from the radioactive tracer studies in our earlier work raise some question whether such an assumption is valid in the H-Coal reactor. It was found earlier that for at least certain operating conditions there are larger bubbles having large rise velocities traveling up the center of the column, with downflow of smaller bubbles along the wall. Therefore, an objective of this study was to determine the gas holdup as a function of radial position in the cold-flow column. Because bubble coalescence--i.e., churn turbulent behavior--explains the gas tracer results, coal slurries were chosen as the fluid of interest in this task.

Liquid Mixing.--The liquid residence time distribution is also required to understand the mixing/dispersion phenomena occurring in this three-phase system. Because our earlier results showed that the concentration of coal char fines in the slurry was uniform throughout the system, the slurry can be treated as a pseudo-homogeneous phase. The objective of this task was to determine the amount of slurry mixing/dispersion occurring in the H-Coal system. Our cold-flow pilot plant was used in this phase of the study.

Catalyst Mixing.--As part of a separate DOE contract, Sandia National Laboratories charged Co-60 tagged catalyst to the HRI PDU during PDU-10 to determine the extent of catalyst mixing in the H-Coal reactor. Catalyst withdrawals were made daily, and the level of radioactivity was determined. Sandia found that the reactor is acting approximately as a backmixed system. A maximum in radioactivity level of the withdrawn catalyst was found to occur three days after addition.

Task 4: Model Implementation

The results from the current studies were incorporated into our modified Bhatia-Epstein model. The model is available upon request to users in easy-to-understand format in the form of computer cards or tape.

III) AMOCO COLD-FLOW EXPERIMENTS: BED EXPANSION AND PHASE HOLDUPS

Cold-flow experiments were performed to measure the fluid dynamics properties of the three-phase ebullated bed (catalyst bed expansion and component phase holdups). The goal of these experiments was to study the response of these variables to changes in operating conditions and to provide data to develop parameters for the Bhatia-Epstein model. In order to validate and improve on this model, which was developed under the previous contract (1), these

cold-flow fluidization experiments were carried out to provide a wider range of fluid dynamic data. Kerosene and slurries of 4.0, 9.8, and 20.7 vol% coal char fines in kerosene as the continuous fluid phase were studied with the H-Coal (Cyanamid HDS-2A, 1/16" ID x 3/16" long) catalyst. A 15.4 vol% slurry was also used with the newly developed Amocat-1A catalyst particles. In both cases, nitrogen was used as the gas phase.

Equipment and Procedures

A schematic diagram of the H-Coal cold-flow fluid dynamics pilot plant is shown in Figure 3. The process flow of this pilot plant simulates that of the process development unit (PDU) built by HRI. Detailed descriptions of the equipment have been presented in a previous report (1). The reactor column, gamma-ray equipment, and some changes of piping are briefly described here.

The reactor is constructed from four 6" ID x 5' long glass sections, which are connected via flanges to five metal spool pieces. These spool pieces have connections for sample taps, pressure taps, and thermowells to monitor the system. Samples of the slurry are withdrawn through the spool pieces to establish the concentration of coal char fines along the reactor column.

A computer-controlled sled carries a Cs-137 gamma-ray source and a detector vertically along the reactor. The sled was modified to allow parallel chords at any given elevation to be penetrated by the Cs-137 gamma rays. Gamma-ray scan data obtained during the experiments are used to calculate bed expansions and phase holdups.

Minor piping modifications (shown by dashed-line connection in Figure 3) were made to improve the pilot plant's operability. During Run 221, it was necessary to keep liquid flowing through the recycle cup downcomer line (see Figure 1) to the slurry recycle pump to prevent blockage by the coal fines. Mixing of this stream and a parallel stream from Separator D-3 through the slurry feed pump into the unit was hampered by interaction between the two pumps. The recycle pump, a positive-displacement pump, was overcoming the head of the centrifugal slurry feed pump. Some experiments were performed by draining the separator and recycling all the slurry feed through the recycle cup. Gas entrainment in this recycle line, however, hampered data collection at higher gas velocities. Subsequent runs were conducted using the modified piping system, shown by the dashed line, in which both recycled slurry streams are now drawn from Feed Tank D-1.

For those tests that started with a new batch of catalyst, the catalyst particles were allowed to soak in the kerosene for a period of two days. The coal char fines concentration in the kerosene was then adjusted to the desired value. The gamma-ray scanning technique was employed to find the bed height and volume fractions occupied by the various phases for each run.

A zero gamma-ray scan was then carried out to find the bed height (H_0) at zero gas and liquid flow. For each subsequent set of operating conditions,

the gamma-ray elevator was used to scan the reactor length. These data, obtained by the Modcomp II computer, were stored for subsequent analysis and plotting. An example of gamma-ray scans of various gas velocities is shown in Figure 4. From plots of this type, a bed height was established for each test. Based on this bed height, the initial catalyst charge, and the gamma-ray measurements along the reactor, the holdup of each individual phase was determined. When coal char fines are present, additional information is needed. This was established by measuring the fines distribution along the reactor by direct sampling through the spool pieces. Details of the calculation procedures to establish the holdup of each phase can be found in an earlier report (1).

Experimental Conditions and Data Collection

Table I summarizes the operating conditions and other information for all experimental cold-flow runs conducted during this program. All of these tests were performed at ambient temperature and pressure. Gas, catalyst, and slurry volume fractions were determined at various gas and slurry velocities within the respective gas and slurry flow rate ranges listed in Table I.

Run 218, which involved kerosene, was performed to measure unit performance and provide a base line for the subsequent runs. After the cold-flow pilot plant was configured as shown in Figure 3, fluid dynamics tests were conducted with various coal char/kerosene slurries. Slurry samples were taken at each spool piece during Runs 221-224 under flow conditions to check the coal char concentration in the kerosene.

Bench-scale experiments were also performed to measure the apparent density of the coal char fines, the soaked particle density of the catalysts, and the average dimensions of the catalyst particles. These results are summarized in Table II. Runs 219 and 220 were exploratory runs and are not presented in Tables I and II.

Data Analysis and Results

Appendix A contains experimentally determined catalyst bed expansions and phase holdup results for both the catalyst bed itself (dense phase) and the region above the bed (dilute phase).

Phase Holdup and Other Hydrodynamic Parameters.--The coal char/kerosene slurry is considered here as a pseudo-homogeneous liquid. This is supported by the uniformity of the coal char fines concentration as measured throughout the pilot plant system. For a two-phase liquid/solid system, the liquid (or slurry) holdup can be correlated with operating conditions and the physical properties of the system by the application of the Richardson-Zaki correlation:

$$\epsilon_{sl}^n = U_1/U_t$$

where $\epsilon_{sl} = \epsilon_l + \epsilon_f$.

Slurry holdup, ϵ_{s1} , can be calculated from the gamma-ray data or from catalyst bed expansion ($1 - \epsilon_c$). When plotting ϵ_{s1} versus U_1 on log-log paper (see Figure 5), the Richardson-Zaki parameters n and U_t can be determined from the slope and the intercept of the straight line that best fits the data. Table II summarizes the Richardson-Zaki parameters determined for each of the experiments. For purposes of comparison, the fines concentrations and corresponding viscosities are also presented in Table II.

Three-phase gas/liquid/solid fluidization data may be correlated using the drift flux approach of Darton and Harrison (3), where the drift flux, V_{CD} , is defined as

$$V_{CD} = U_g (1 - \epsilon_g) - U_l \epsilon_g (1 - \epsilon_g) / (\epsilon_l + \epsilon_f) \quad (1)$$

The drift flux physically represents the volumetric flux of gas relative to a surface moving at the average gas plus liquid velocity. A schematic diagram of V_{CD} versus ϵ_g is shown in Figure 6 to illustrate characteristics of different flow regimes defined by the V_{CD} and ϵ_g relationships. Two distinct flow regimes are identified and described by Darton and Harrison (3): 1) The ideal bubbly regime, in which the bubbles are relatively small and rise uniformly with little interaction; and 2) The churn turbulent regime, in which bubble coalescence is dominant; the bubble size is larger, and the flow is unsteady. There exists also a transition region between the ideal bubbly and fully developed churn turbulent flow regimes.

Data from Run 218 were analyzed according to Equation 1, resulting in a linear relationship between V_{CD} and ϵ_g , as seen in Figure 7. Because Run 218 used straight kerosene, the bubble size and the bed behavior could be observed visually at the glass column wall. Within the range of superficial gas and liquid velocities investigated in Run 218, ideal bubbly behavior was observed at the wall. The plot in Figure 7 substantiates the fact that the flow is ideal bubbly, as defined by the Darton-Harrison drift flux analysis. These results for Run 218 will define the ideal bubbly operating line for all subsequent cold-flow results involving slurries.

Drift flux plots for Runs 221 through 224 are presented in Figures 8-11. The solid line represents the ideal bubbly flow regime as defined by Run 218 experimental data. The dashed lines represent bed behavior in a transition regime between the ideal bubbly and fully developed churn turbulent states. A comparison of the drift flux results for Runs 221, 222, and 223 indicates that increasing the viscosity of the slurry phase tends to enhance the churn turbulent behavior of the bed at comparable operating conditions. This is consistent with the theory that a higher liquid viscosity promotes bubble coalescence, resulting in larger bubbles, which are characteristic of the churn turbulent flow regime. Higher slurry velocity has a common effect of reducing the magnitude of V_{CD} . Therefore, increasing slurry velocity will stabilize the bed behavior. A comparison of drift flux results for Runs 222 and 224 (Figure 9 versus Figure 11) indicates that drift flux correlations for the Amocat-1A (Run 224) catalyst are comparable to those for HDS-2A (Run 222).

Gas holdup is one of the most important variables characterizing the fluid dynamics of three-phase fluidization and is sensitive to the properties of the liquid phase, particularly the viscosity. The gas holdup in the catalyst bed, plotted versus the superficial gas velocity in Figure 12, increases with increasing velocity in all cases at low velocities. There is a velocity in each case, however, above which the gas holdup becomes constant. Alternatively stated, the average bubble rise velocity is increasing at the same rate that the gas superficial velocity is increased. Note in this latter region that the gas holdup decreases as the concentration of fines in the slurry is increased. This indicates that the higher concentration of fines, and thus higher slurry viscosity, promotes the coalescence of gas bubbles into larger bubbles having larger rise velocities. The end result is a decrease in the gas holdup.

Catalyst Bed Expansion.--Knowledge of catalyst bed expansion effects as a function of operating conditions is required for proper design and operation of the reactor. As the superficial slurry velocity is increased, the bed expansion increases, as shown in Figures 13 and 14 for 0.0 and 0.10 ft/sec superficial gas velocity, respectively. Addition of a small amount of fines does not affect the bed expansions to any great extent. Addition of larger amounts of fines does affect the bed expansions significantly due to the increased viscosity of the resulting slurries.

Bed expansions as a function of superficial gas velocity are presented in Figures 15 and 16. Note that the bed expansion is a much weaker function of gas velocity (Figures 15 and 16) than of slurry velocity (Figures 13 and 14). The results also show that there is apparently a slight bed contraction in some cases at sufficiently high gas velocity. Bed expansions of about 60% are observed in Figure 15 for a nominal H-Coal reactor slurry velocity of 0.10 ft/sec. This will be compared later with bed expansions measured on the actual H-Coal reactor in Section VII.

Bed contractions at higher gas velocities have been observed before (1), and a theory has been advanced (4,5) which has phenomenological appeal. The wake trailing a rising bubble contains pure liquid or slurry, which is traveling at the velocity of the rising bubble. This slurry in the wake is not available in the slurry proper for fluidization of the catalyst particles. Thus, the effective velocity of the slurry regarding fluidization of the catalyst is less, and thus the bed contracts. The bed contractions observed here are slight but real. Although bed contraction effects will not influence reactor design to any great extent, these effects are of considerable importance in the fundamental understanding of three-phase fluidization.

A qualitative difference between HDS-2A and Amocat-1A was observed in the nature of the catalyst bed/dilute phase interface. At certain combinations of liquid and gas flow rates, the Amocat-1A catalyst formed a very diffuse interface. By contrast, the interfaces measured with HDS-2A catalyst were distinct. Examples of sharp and diffuse interfaces are illustrated in Figures 73 and 74, respectively. Catalyst bed expansions for Run 224 are shown in Figure 16, along with the line delineating sharp and diffuse catalyst bed interfaces.

IV) AMOCO COLD-FLOW EXPERIMENTS: RADIAL UNIFORMITY OF PHASE HOLDUPS

It has been assumed in many studies that gas holdup is uniform across the reactor diameter. It is reasonable to make this assumption when the flow regime can be described as bubbly. In the churn turbulent flow regime, however, gas holdup becomes non-uniform, due either to large eddies or bubble coalescence or both. In the transition zone between the bubbly and the churn turbulent flow regimes, the degree of non-uniformity of gas holdups varies, depending on the physical properties of the continuous phase and on the velocities of both phases.

It was assumed in the previous contract work (1) that the phase holdups were constant across the diameter of the reactor. Some very limited experimental data indicated that this was a reasonable assumption. In order to verify this assumption, gamma-ray scanning experiments were carried out in this current contract at various parallel chords across the column.

Review of Prior Work

In spite of the wide variety and the vast amount of literature in the field of gas/liquid and gas/liquid/solid fluidization, few publications deal with the matter of whether the phase holdups vary in the radial direction. Only a few papers have dealt specifically with this question for two- and three-phase systems.

Hills (6) studied radial non-uniformity of gas holdup and liquid velocities in a 13.8 cm diameter column, two-phase, air-water system operated in a batch fluidization mode. Three different gas distributors were tested in order to evaluate their effects on radial gas holdup distribution. The results showed that for gas distributors having a large number of small holes (0.4 mm) evenly distributed on the plate, the gas holdups are radially uniform at gas velocities below 3.0 cm/sec. At higher gas velocities, there is an increasingly sharp central maximum in gas holdups. Results obtained with a seven-large-hole (1.6 mm) gas distributor showed the central maximum in the gas holdup at all gas velocities.

Recently, Linnweber and Blass (8) extended Hill's work to three-phase fluidization and measured both the gas and solid holdups as a function of radial position. Gas holdup profiles were measured for the air-water-plastic slurry and the air-water-glass slurry systems. The gas holdup profiles were essentially the same as those measured in the two-phase air-water system. The absolute values of gas holdup, however, were slightly less than those measured in the air-water system, due mainly to the addition of the solid phase. These authors also showed that at higher gas rates ($U_g \geq 3$ cm/sec), gas holdups increase toward the center of the column. The solid holdup profiles were relatively more flat in comparison with the gas holdup profiles, and they did not show obvious radial non-uniformity up to the gas velocity of 3.0 cm/sec. The results also showed that the gas and solid holdups are compensating each other--i.e., while gas holdups tend to increase toward the center of the column, the solid holdups decrease correspondingly toward the center of the column. As a result of this solid-gas compensating effect, liquid holdups are expected to be radially uniform at velocities less than 3.0 cm/sec.

Tsung (7) studied the radial gas holdup distribution in a gas-liquid bubble column while the system was operating in a steady-state flow condition. Nitrogen was used as the gas phase, and the liquid phase was a mixture of 66% dipropylene glycol monomethyl ether and 34% diphenyl ether. A single bubble cap distributor was used to disperse both the gas and liquid flows. The qualitative results obtained with a resistivity probe showed that the gas holdups vary with radial position, gas velocity, and liquid velocity. Gas holdups obtained from experiments conducted at low gas and liquid velocities ($U_g \leq 0.44$, $U_l \leq 1.34$ cm/sec) showed parabolic distributions with the maximum values at or near the center of the column. It was noted that the gas holdup distribution curve becomes flatter at a higher liquid flow rate. Nevertheless, the results are of limited value to our study because of the low gas and liquid velocities used in his study, and also because the probe could not be calibrated.

The limited data in our earlier contract work (1) are the only prior data available of direct relevance to the three-phase H-Coal system. Gamma-ray scans through parallel chords of the cross-section of the column indicated that the gas-phase holdups were essentially constant.

Equipment and Procedure

The Amoco H-Coal cold-flow pilot plant was modified so that gamma-ray scanning could be performed at various radial positions. This involved remounting the gamma-ray source and the detector on a traversing support structure and realigning the gamma-ray instrument. A detailed description of the pilot plant, the gamma-ray equipment, and the operating procedure was presented in a previous report (1).

Experimental Conditions and Data Collection

The gamma-ray scan experiments were carried out on the Amoco cold-flow pilot plant using a 25 wt% coal char fines in kerosene slurry system. The physical properties of the system are listed in Table II. The nitrogen gas flow rates were 0.08 and 0.10 ft/sec, while 0.10 and 0.125 ft/sec were selected for the slurry flow rates. Catalyst bed height data were taken during each test.

Gamma-ray scans were conducted at 30, 50, 70, 90, and 110" above the bubble cup distributor. At each elevation, gamma-ray scanning data were taken manually at seven chord positions across the reactor column. As shown in Tables IV and V, the chord position is indexed by R_i/R_0 , where R_i is the corresponding radius of gamma-scan position with respect to the column center and R_0 is the radius of the column. Negative R_i/R_0 values designate chord positions to the left of the column center, and a zero index value designates the column center position.

A zero gamma-ray scan of the reactor filled with kerosene only was also carried out at zero gas and liquid flow. The data acquisition procedure for the zero gamma-ray scan was the same as that for the 25 wt% coal char in kerosene system. These data are necessary in order to calculate the various phase holdups.

Data Analysis

The average of five gamma-ray scans is used in the phase holdup calculations.

The volume fraction of the catalyst is calculated from measurement of the bed height and from the mass of catalyst in the reactor:

$$\epsilon_c = \frac{M}{\rho_{CAH}} \quad (2)$$

The volume fraction of liquid can then be calculated using gamma-ray scan data:

$$\epsilon_l = \frac{(\ln \frac{I_l}{I_m})/d - \mu_c \rho_c \epsilon_c + \mu_l \rho_l}{\mu_l \rho_l + \frac{\mu_f \omega_f \rho_l}{(100 - \omega_f)}} \quad (3)$$

The volume fraction of coal char fines is calculated from the known concentration of fines:

$$\epsilon_f = \frac{\omega_f \rho_l \epsilon_l}{(100 - \omega_f) \rho_f} \quad (4)$$

The volume fraction of gas is then calculated from the difference:

$$\epsilon_g = 1 - \epsilon_c - \epsilon_l - \epsilon_f \quad (5)$$

In the dilute phase above the catalyst bed, ϵ_c is set to 0.0, and the same equations are solved.

Results and Discussion

The average gamma-ray scan data presented in Table III and Table IV are average values of five scans taken at each radial position. Phase holdups calculated from these scan data are presented in Table V. The averaged gamma-ray scan data were found to have less than 4% standard deviation, which translates to a maximum of about 5% standard deviation in the calculated liquid holdups. The gas holdups, calculated from the other phase holdups, do not vary significantly across the reactor diameter except for those data at $R_i/R_o = 0.75$. The phase holdups at this radial position are

affected by the presence of a 1" diameter slurry recycle line. The phase holdup data at this radial position are not used in this evaluation. Therefore, the gas and liquid holdups shown in Table V are essentially constant across the reactor diameter within experimental error. The results show that the gas and liquid holdups in both the dense and dilute phases do not vary significantly across the reactor diameter at the gas and slurry velocity ranges tested.

The radial uniformity of gas and liquid holdups does not necessarily imply that the bubble sizes across the reactor are constant. Based on gas tracer tests carried out in the earlier contract (1), it was concluded that large bubbles having a high rise velocity were traveling up the center of the column while smaller bubbles were traveling down at the walls. Although these earlier gas tracer results and the current radial scanning results appear to contradict each other, some interesting conclusions can be drawn from the results.

The gamma-ray scans, which cut horizontally across parallel chords of the column at a given elevation, cannot discriminate between upflowing and downflowing bubbles. The gamma-ray scan sees an average density. Even though the bubble size distribution varies across parallel chords of the column, the average gas and slurry volume fractions are essentially constant.

Table V also shows that the gas and slurry holdups do not vary axially within the bed or dilute phase. The gas holdup is about 10% higher in the dilute phase than in the bed. These results have large implications for our understanding of the heat and mass transfer processes as well as chemical reactions taking place in the reactor, all of which depend strongly on the bubble size distribution.

V) AMOCO COLD-FLOW EXPERIMENTS: AXIAL SLURRY DISPERSION

Background

Gas- and liquid-phase mixing have been studied extensively in gas/liquid fluidized systems but to a much lesser extent in gas/liquid/solid systems. Several review articles [Wen and Fan (9); Shah, et al. (10); Ostergaard (11); and Bischoff (12)] have summarized the mixing characteristics of various types of multi-phase reactors. It is clear from these literature surveys that axial mixing within the different phases has not been adequately studied for the gas/liquid/solid fluidized system. No published data exist for liquid-phase and gas-phase mixing in a three-phase fluidized system where the liquid phase contains suspended fine particles such as the unreacted coal and mineral particles in the H-Coal ebullated bed reactor. Backmixing, recirculation, and other transport mechanisms such as bubble coalescence complicate the nature of gas-, liquid-, and solid-phase motion.

Recently, Bickel and Thomas have studied solid-phase (catalyst) mixing in the H-Coal PDU reactor during a 28-day steady-state operation using Kentucky high-sulfur coal (13). The catalyst residence time distribution was determined

by tracking the radioactivity of daily withdrawn ^{60}Co -tagged American Cyanamid HDS-1442A CoMo catalyst samples from the PDU reactor. The Peclet number was calculated to be 0.45 based on an axial flow dispersion model. This indicates that catalyst mixing in the H-Coal PDU is expected to be well mixed after an initial period.

Amoco's contract tasks include characterizing the gas- and liquid-phase dispersion using the cold-flow unit. Gas-phase mixing experiments were completed in 1980 under the first H-Coal contract (1) using argon-41 as a radioactive tracer. A circulation model consisting of completely mixed tanks in series with a recycle flow was proposed (1). This model is in agreement with visual observation of the cold-flow unit, where small gas bubbles were seen flowing down along the outer walls. Liquid-phase mixing experiments were carried out under the current contract. Experimental results and a one-parameter dispersion model are presented here.

Review of Liquid Dispersion in Multi-Phase Fluidization

Axial liquid dispersion data in the literature for gas/liquid, gas/slurry, and gas/slurry/solid fluidization are summarized and compared. The effects of various independent system variables (gas velocity, liquid velocity, particle size, column diameter, and fluid properties) on the extent of liquid dispersion are discussed in this section.

Gas/Liquid Systems.--The subject of axial mixing characteristics in a vertical bubble fluidization column has been extensively studied and was recently reviewed by Joshi (14), Bischoff (12), and Shah, et al. (10). In general, the liquid dispersion coefficient (D_1) can be correlated with a characteristic velocity and the column diameter (d_t), assuming the column has a large length-to-diameter ratio. A majority of the investigators agree that whereas the dispersion coefficient is a strong function of the gas velocity, it is essentially independent of the liquid flow rate. Therefore, the following model is often used to correlate results:

$$D_1 = a u_g^b d_t^c$$

Table VI summarizes the liquid dispersion coefficient correlations derived by various investigators.

In an attempt to develop a unified model for axial mixing in a multi-phase fluidized reactor, Joshi (14) concluded that the liquid circulation velocity could be used to correlate all experimental dispersion data. The liquid circulation velocity is based on the multiple circulation cells theory in a bubble column and can be calculated from the energy balance. Although it uses liquid circulation velocity, Joshi's unified model (see Table VI) is inherently related to the gas bubble rise velocity. For all practical purposes, gas velocity is by far the most popular characteristic velocity used in liquid dispersion correlations.

Effect of Liquid Properties: The effect of liquid viscosity on the axial dispersion coefficient is not completely resolved in the literature. Cova

(23) and Alexander and Shah (24) reported that the axial dispersion coefficient is independent of the viscosity and surface tension of the liquid. Pilhofer, et al. (26) and Hikita and Kikukawa (19), on the other hand, found that the axial dispersion coefficient is dependent upon liquid viscosity (see Table VI). Their conclusion was derived, however, from a much larger liquid viscosity range (20-200 cp). Cova and Alexander's conclusion was derived from experimental results obtained in a smaller liquid viscosity range (1-15 cp).

The effect of liquid density on the axial dispersion coefficient was studied by Cova (23) using small-diameter columns (less than 1.8" ID). The results indicate that an increase in liquid density increased the dispersion coefficient.

Gas/Slurry Systems.--Liquid-phase dispersion in the gas/slurry system has been studied to a lesser extent, and consequently, fewer liquid-phase dispersion coefficient correlations have been derived. Published liquid dispersion models are summarized in Table VI.

An interesting and important effect of introducing fine suspended solid particles to the gas/liquid system is the enhancement of bubble coalescence which would be expected to enhance the axial liquid dispersion. The fine solid particles suspended in the liquid constitute a pseudo-homogeneous fluid of higher density and viscosity than the pure liquid. The increased viscosity is known to promote bubble coalescence (1).

The nature of the pseudo-homogeneous fluid was illustrated by Kato, et al. (34) in evaluating the magnitudes of liquid- and solid-phase dispersion in a three-phase fluidized column. Kato, et al., studied liquid and solid dispersion using various types of fines ranging from 53 to 250 microns and with particle densities ranging from 1.0 to 9.0 g/cm³. Experiments were carried out with different diameter columns and with fines concentrations up to 20%. The significant conclusions of these studies was that the longitudinal dispersion coefficient for the fines in the gas/slurry fluidized bed is the same as that for the gas/liquid. These results imply that the fines in the liquid phase are homogeneously mixed. Vasalos, et al., (1) also found that the concentration of their coal slurry was uniform throughout their three-phase system.

Axial mixing in the bulk of the bed is contributed mainly by wake formation and shedding. These wakes, carrying appreciable solids, contribute to the solid mixing. Therefore, for fine solids ($d_p < 0.1$ mm), there is little difference between axial dispersion coefficient for the solid and the liquid phases.

Effect of Liquid Velocity: Whereas in the gas/liquid system the liquid dispersion coefficient is dependent only upon the gas velocity, the axial liquid dispersion coefficient in gas/slurry fluidization is also dependent upon the liquid (or slurry) velocity. Experimental data in the literature (14,34,35,37) indicate that the liquid dispersion coefficient increases with increasing gas velocity but decreases with increasing liquid (or slurry) velocity. The magnitude of the effect of liquid (or slurry) velocity on

axial liquid dispersion depends on the gas velocity and the solid particle size. A valid approach for evaluating the liquid velocity effect is to compare the Darton and Harrison (3) drift flux values for determining bed behavior (bubbly or turbulent) as a function of liquid (or slurry) velocity. Data obtained from the present work show that a higher liquid velocity suppresses the churn turbulent behavior of the system, resulting in the reduction of liquid mixing. Liquid dispersion coefficient models proposed by other investigators (see Table VI) further quantify the magnitude of the liquid velocity effect.

Gas/Slurry/Solid System.--The effect of adding fluidized coarse, solid particles (such as catalysts) to a gas/liquid or gas/slurry system is quite different from that of adding fine, solid particles to a gas/liquid system. In the gas/slurry/solid case, the liquid dispersion coefficient decreases relative to a gas/liquid system under the same flow conditions, due to bubble disintegration by the large solid particles. The disintegration causes a reduction in bubble size and rise velocity, resulting in less axial dispersion.

Effects of independent variables (gas and slurry velocities and the diameter of the column) upon liquid dispersion are similar to those discussed in the gas/liquid and gas/slurry systems.

Effect of Solid Particle Size: Analysis of liquid dispersion coefficients reported by various investigators (30,31,32,39), including data obtained from this work, indicates that liquid dispersion coefficients decrease as the particle diameter is increased. Ostergaard (31) measured bubble sizes in a three-phase system and reported that bubble sizes in beds composed of 1 mm particle size were one order of magnitude larger than bubble sizes in beds composed of 6 mm particles. Ostergaard's dispersion coefficient data appear much higher than those reported by others (see Figure 17). The use of the larger diameter column may have contributed to this high result. Discrepancies appearing at the low gas velocities may be due to the differences in distributors used in these studies.

Reacting Systems.--Numerous data exist in the literature, and many models are available for predicting the dispersion coefficient for non-reacting two-phase and three-phase systems. However, application of these data and models to predict real system performance is still inadequate. For instance, Panvelker, et al., (38) reported that the liquid dispersion coefficient predicted by the application of the Deckwer, et al., (16) correlation was more than ten times higher than that which was actually measured from a solvent refined coal (SRC) dissolver. It is clear that further study of liquid (slurry) phase mixing is needed with systems more closely simulating both the physical properties and operating conditions of industrial systems.

A recent study of the effect of vertical alignment of the column on liquid axial dispersion indicated that axial mixing can be reduced significantly if the column is well aligned (40). Although the study was conducted in a liquid/liquid spray column, it may have significance for the H-Coal system

and other three-phase systems. The effect of reactor internals also needs further investigation.

Tracer Selection

A suitable liquid tracer is necessary to measure the extent of dispersion in the liquid phase. A number of tracers and techniques have been used to obtain residence time distribution (RTD) functions for chemical reactors. In this particular case, the purpose is to obtain an RTD for the slurry phase of the gas/slurry/solid system under non-reacting conditions.

The criteria for a suitable tracer include 1) little or no absorption by the coal char fines and the catalyst for a short period of time (30-60 minutes); 2) good miscibility in kerosene; 3) accurate determination of the tracer concentration in the liquid; and 4) good chemical and physical stability. These criteria were used to determine the choice of a tracer for our liquid dispersion experiments.

Examples of experimental techniques prior to 1980 are reviewed in several books and review articles. Three-phase experiments are discussed in Shah's text (45). A table of commonly used tracers is included.

Wen and Fan (9) divided liquid-phase experiments into categories by tracer method. Four classes were indexed: conductivity salts, color and UV dyes, radioactive tracers, and titrations. Forty-five papers are included, eight of them dealing with fluidized beds.

Shah, Stiegel, and Sharma (10) give a more extensive review of experimental work, indexed by reaction system. Twenty-eight examples of liquid-phase tracers from the literature are given, with three citations for work in three-phase fluidized beds, but no additional tracer techniques are presented. Other review articles covering the earlier development of tracer methods and mixing models include Ostergaard (5) and Bischoff (12).

Most of the liquid dispersion studies reported in the literature have used clear liquids and non-porous solid particles. Air, water, and glass balls are the most popular components used by the investigators. For these systems, conductivity salts, UV and visible dyes, and radioactive chemicals are equally effective as tracers. These tracers and techniques, however, are generally not suitable for our three-phase system involving a coal char fines/kerosene slurry phase. This is primarily due to the physical properties of the slurry. The coal char/kerosene slurry is essentially non-conductive and opaque. Furthermore, the coal char fines in the slurry are porous particles and strongly absorb certain chemicals. Potential liquid tracers in each category were evaluated in order to select the best qualified liquid tracer for this study.

While conductivity salts are normally used in the air/water systems, additives are available that can increase the conductivity of normally non-conductive organic liquids. These additives are miscible organic salts and are detectable

in the ppm range by using a conductivity probe. The disadvantage of the conductive salt is that an invasive conductivity probe in the fluid may cause flow disruptions. A possible complication of conductivity measurement also exists due to gas bubbles adhering to the probe.

Organic soluble dyes are also available from a number of sources and have been used in studying mixing of clear liquids. UV and visible dyes offer the advantage that the detector is located outside the reactor and will not disturb the flow. The disadvantage is that the dye method requires the use of a transparent system. This is incompatible with coal char fines used in the H-Coal fluid system.

Use of radioactive tracers is an attractive experimental technique, because the tracer concentration can be measured non-invasively. Unfortunately, use of radioactive tracers generates large amounts of low-level radioactive waste for disposal. Therefore, radioactive tracers were excluded from consideration for this task.

Three potential tracers and their detection methods were selected for further experimental evaluation:

<u>No.</u>	<u>Tracer for Liquid Mixing</u>	<u>Tracer Detector or Analytical Method</u>
1	Du Pont Stadis-450 (An Oil-Base Conductivity Improver)	Conductivity Cell
2	Mobay Chemical Blue ZV (An Oil-Base Blue Dye)	Visible Light Absorbance
3	Thiophene	Sulfur Analysis by X-ray Fluorescence

Bench-Scale Experiments and Results

Du Pont Stadis-450.--Stadis-450 is a jet fuel additive currently marketed to reduce static hazards by making the hydrocarbon fluid electrically conductive. According to the manufacturer, the resulting conductivity of the mixed liquid is a function of the additive concentration. Therefore, in each of the tests, a known amount of the additive was introduced into a mechanically stirred vessel which contained kerosene or a kerosene/coal char slurry. A meter measured the resulting electrical conductivity, which was recorded as a function of mixing time. Du Pont Stadis-450 absorption by HDS-2A catalyst was also tested separately. In these experiments, catalyst particles were first soaked in kerosene before the absorption rate tests were carried out.

Results from these tests showed that over a limited range, Stadis-450 concentration and electrical conductivity were related. However, when kerosene slurried with fully soaked coal char was tested, the Stadis-450 was strongly absorbed by the char. A rapid decrease in conductivity was observed after the additive was injected into the solution. The rate of absorption of the

additive seems to be affected by its concentration in the slurry. Below 1,500 ppm, Stadis-450 is consistently absorbed at a high rate. Above 1,500 ppm, it is still being absorbed at a high rate for the first five minutes. This is then followed by a slower absorption.

The phenomenon of absorption by HDS-2A catalyst is quite different. The conductivity increased initially for almost 20 minutes, indicating little or no absorption by catalyst particles in this period. The catalyst particles absorb Du Pont Stadis-450 additive at an insignificant rate compared with that of the coal char fines.

Due to the consistently fast absorption of the additive by the coal char fines, Du Pont Stadis-450 and the conductivity detection method were judged to be not suitable for the liquid mixing experiments.

Mobay Blue ZV Dye.--A saturated solution of this dye was used as a tracer in visible light absorptivity tests. The experimental procedure for these tests was similar to that used for the conductivity experiments. Mobay Chemical Blue ZV dye was added to kerosene or kerosene/coal char slurries to produce a known concentration. Spectro-photometric absorption was then used to correlate signal attenuation with concentration. A linear correlation between dye concentration and its light absorption was established satisfactorily with pure kerosene. This method failed, however, to give a definite correlation between dye concentration and absorptivity with the slurry system. Since the inclusion of the coal char fines is necessary to model the H-Coal reactor, this method was also discarded.

Thiophene.--Thiophene was investigated as a candidate liquid tracer, because it is miscible with kerosene, and because it can be accurately detected by gas chromatography or by x-ray fluorescence. Thiophene tests were also run by mixing a known amount of thiophene with well-stirred kerosene/coal char and kerosene/coal char/catalyst slurries. The mixtures were separated by filtration, and the liquid was analyzed for sulfur content by x-ray fluorescence. Thiophene was absorbed at a very slow rate by coal char fines. As shown in Figure 18, there was practically no absorption in the first 90 minutes. Additional absorption tests were carried out up to 180 minutes' mixing time, and results indicated insignificant absorption.

Thiophene absorption by the HDS-2A catalyst particles was also investigated. No appreciable absorption was evident up to 180 minutes' mixing time. Hence, thiophene meets the most important criteria for a liquid tracer--i.e., little or no absorption by coal char fines and catalyst. In addition, thiophene is readily miscible with kerosene and is chemically stable in the solution. Therefore, thiophene has successfully met all four criteria required for a good liquid tracer.

Pilot Plant Testing of Thiophene as a Tracer

Based on the encouraging bench-scale results from using thiophene as a tracer material, scoping experiments were carried out using the H-Coal cold-flow

pilot plant. The objectives of these tests were to gain experience in obtaining the tracer C-curve (concentration versus time curve) at different positions in the fluidized bed and to gain some insight into how best to design the equipment for the tracer injector and the continuous-flow sampling device.

Results of pilot plant liquid mixing tests are presented in Figure 19. The input pulse curve (lower curve) and the resulting downstream C-curve (upper curve) were successfully obtained when 2 cc of the thiophene was injected into the reactor at the slurry inlet port. The extent of liquid dispersion can be estimated based on the data derived from these C-curves. Based on these pilot plant test results, it was concluded that thiophene is a suitable liquid tracer and was therefore selected for the liquid mixing experiments.

Equipment

Liquid-phase dispersion experiments were carried out in the Amoco H-Coal cold-flow pilot plant by injecting a pulse of thiophene at the slurry inlet port and measuring thiophene concentration as a function of time at two locations in the dense-phase region and at two locations in the dilute-phase region. The pilot plant has been modified to include the additional equipment and instrumentation necessary for liquid-phase dispersion tests. The following sections give a description of equipment and experimental procedure used to obtain liquid dispersion test data.

A detailed description of the Amoco H-Coal cold-flow pilot plant and its operation was presented in a previous report (1). A schematic diagram of the pilot plant is shown in Figure 20. A description follows for the equipment added to the pilot plant in order to perform liquid-phase mixing experiments.

- 1) Continuous-flow slurry sampling device: Located just below the slurry sampling probe outlet, this automatic sampling device holds twenty-five 20-cc glass vials evenly distributed around the circumference of a vial tray. The tray rotates at 0.5 RPM, giving a 4.8-second sampling time per vial. Figures 21 and 22 show the detailed mechanical design of the continuous-flow slurry sampling system.

Four identical sets of the continuous-flow sampling device were built. They were installed at the four spool pieces on the fluidization column. The lower two sets receive samples at elevations from within the fluidized bed (dense-phase region), whereas the upper two sets receive samples at elevations from within the dilute phase.

- 2) Constant-volume tracer injector: Constructed of stainless steel material, the injector is a 2-cc, precision-bore barrel with an O-ring sealed plunger. A 200-cc liquid tracer reservoir is connected to the injector's inlet port to provide an adequate supply of liquid tracer. The injector is installed at the slurry inlet line leading to the calming section of the fluidized column. A drawing of the tracer injector assembly is shown in Figure 23.

- 3) Reactor slurry sampling probe: Inserted into the reactor through each spool piece, the sampling probe is equipped with an adjustable tube which can be positioned at any radial position. A drawing of the sample probe assembly is shown in Figure 24.
- 4) Automatic continuous-flow sampling system: The system consists of a programmable controller which controls the rotational movements of the four continuous-flow slurry sampling devices. Each device consists of a synchronous motor and a limit switch. The switch serves a dual function in that it provides a starting position for the vial tray and also stops the vial tray movement after one complete revolution. Each device is provided with a start pushbutton and a starting position indicator on the instrumentation panel board located in the control room. There is also an overall start button which is mounted on the pilot plant structure which controls the initiation of automatic sampling sequence.

The controller logic is such that a sampling sequence cannot be started unless all four vial trays are at the starting position. Once the sampling sequence has been started, the automation controls the time delays between the start and stop of all vial trays. Based on the planned experimental slurry feed rate range (1-5 cm/sec) and on the assumption that the average linear slurry velocity will not exceed 10 cm/sec, the proper time delays for Tray Nos. 2, 3, and 4 can be calculated by dividing the distance between two sampling ports by the maximum linear slurry velocity. It was determined that a 15-second delay for the second tray and subsequent 10-second delays for the third and fourth trays are appropriate.

Pilot Plant Results and Discussion

The experimental plan consisted of four series of tests. Data collected from these tests have been processed according to the liquid mixing data analysis program (Appendix B). Liquid mixing parameters--Peclet number, and the dispersion coefficient--are presented in this section. Correlation of the Peclet number as a function of process variables was developed.

Data Collection.--The first series of tests, consisting of Runs 490-6 through -8 and Run 490-21, was designed to investigate the effect of gas flow rate on the liquid dispersion coefficient (or Peclet number). The nitrogen flow was varied from 0 to 4.57 cm/sec (0-0.15 ft/sec), while the slurry flow rate was controlled at 3.05 cm/sec (0.10 ft/sec). Slurry samples were continuously drawn from the reactor at the reactor wall for 4.8 sec/sample. In most cases 25 samples were taken per tray.

The second series of tests, consisting of Runs 490-10 through -16, was designed to investigate the variation of liquid dispersion across the reactor diameter. These tests were carried out under both the nominal H-Coal gas and liquid flow conditions and under zero gas flow conditions.

The third series of tests, consisting of Runs 490-17 through -20 and Run 490-23, was carried out to investigate the effect of gas flow rate on liquid dispersion at the center of the reactor.

The last series, consisting of Run 490-9 and Run 490-22, was intended to evaluate the effect of increasing slurry velocity on liquid dispersion.

Samples from Trays III and IV, located above the fluidized bed in the dilute phase, were also collected and analyzed in order to determine the dilute-phase liquid dispersion coefficient. Unfortunately, good-quality RTD curves could not be obtained, due to the large amount of liquid mixing occurring in this phase. The experimental concentration/time distribution data (C-curves) have been presented in quarterly progress reports (43,44). Figures 25 and 26 contain an example of a C-curve plot and a logarithmic probability plot of cumulative area under the C-curve, respectively. The mean (first moment) and the standard deviation (second moment) of a normal distribution curve which plots as a straight line on probability paper are derived from the fiftieth and eighty-fourth percentile points.

Data Analysis.--Various correlations were tested involving gas and slurry velocities and radial position. In general,

$$Pe = A + [B + D (U_g - 0.1) + E (R)] \exp (-c U_g) \quad (5)$$

1) Liquid Dispersion Coefficient: Experimental data were processed according to the liquid dispersion data analysis program (Appendix B). Liquid mixing parameters--Peclet number and dispersion coefficient--are presented in Table VII.

2) Liquid Dispersion Correlations: The liquid dispersion results were used to develop a correlation between the liquid dispersion coefficient (or the Peclet number) and the process variables such as superficial gas and slurry velocities. Least squares curve-fitting methods were used to evaluate a variety of correlation formulas. Radial position dependency was also assessed.

As seen in Table VIII, the least squares analysis results for Correlation 1 indicates that there is no significant effect of radial position on the Peclet number. Consequently, the term representing the radial position (R) was excluded from other correlations. Statistical results for Correlation 1 and 2 show that the slurry velocity effect is insignificant within the ranges of gas and slurry velocities employed for these liquid dispersion experiments. The conclusion is that the Peclet number is not a function of the radial sampling position but is a strong function of superficial gas velocity, U_g .

Correlations 3 and 4, which both adequately fit the data for the nominal H-Coal slurry velocity (approximately 0.1 ft/sec) in the superficial gas velocity range from 0 to 5 cm/sec (0-0.164 ft/sec), are

$$\text{Correlation 3: } Pe = 5.05 + 42.91 \exp (-35.48 U_g) \quad (6)$$

and

$$\text{Correlation 4: } Pe = 40.26 \exp (-17.83 U_g), \quad U_g \text{ in ft/sec} \quad (7)$$

Correlation 4 implies that $Pe \rightarrow 0$ when $U_g \rightarrow \infty$, while Correlation 3 implies that $Pe \rightarrow 5.05$ when $U_g \rightarrow \infty$. Correlation 3 fits the data better at $U_g = 0$ (see Figure 27) and gives an overall better fit to the experimental data (see Tables IX and X). Figure 27 compares the experimental results with both correlations.

3) Application of the Dispersion Model: Using Correlation 3, the C-curve for the second sampling tray can be reconstructed based on the C-curve data (from Tray I) and the process conditions (U_g and U_1) for that run. The following procedure is employed:

A) Estimate Pe for a given U_g :

$$Pe = 5.05 + 42.91 \exp(-35.48 U_g), \quad U_g \text{ in ft/sec} \quad (8)$$

B) Estimate σ according to the dispersion model:

$$\frac{\Delta\sigma^2}{T_m^2} = \frac{2}{Pe}, \quad \text{where } T_m = \frac{L}{U_1} \quad (9)$$

C) Calculate the log normal variance s using the conversion formula:

$$s_i = \sqrt{\frac{\sigma_i^2}{\ln^2 + 1}}, \quad \text{where } i = 1 \text{ and } 2 \quad (10)$$

4) Calculate tracer concentration versus time profile using the following equation:

$$C = C_0 + \frac{Q}{T \cdot s \cdot \sqrt{2\pi}} \exp\left[-\frac{(\ln T - \ln T_m)^2}{2s}\right] \quad (11)$$

where Q = Amount of tracer injected into the reactor.

C_0 = Initial tracer concentration (i.e., Tray I curve).

Run 490-17 data were used as an example to demonstrate the validity of the proposed Correlation 3 and the calculation procedure. The predicted C-curve compares reasonably well with the experimental data, as can be seen in Figure 29.

4) Tracer Material Balance: The amount of thiophene calculated from the tracer C-curve and the amount of thiophene actually injected into the system are compared and shown in Figure 28. While reasonable thiophene material balances were obtained for most of the runs (within $\pm 10\%$ of actual injection amount), several runs showed that the calculated amount of thiophene was lower than that actually injected.

The x-ray fluorescence analytical precision (± 15 wppm) accounts for a part of the thiophene imbalance. The fluctuant tail of the C-curve contributes the other part of the error.

The effect of thiophene imbalance upon the liquid dispersion results is not significant, because the amount of axial mixing is estimated from the relative change of variance of the two RTD curves. Application of the method of moments to estimate the dispersion coefficient and its sensitivity upon parameters such as the mean and variance of the tracer C-curve is discussed in Appendix B.

Conclusions

The amount of slurry-phase axial dispersion occurring in a cold-flow system simulating the HRI PDU reactor was studied. Slurry-phase axial dispersion coefficients were obtained for the fluidized-bed phase of the ebullated bed reactor. Typical values of the slurry dispersion coefficient at nominal H-Coal operating conditions are in the range of 70-130 cm²/sec. Peclet numbers range from 3.6 to 9.9. This represents a significant degree of backmixing in the reactor. A correlation was developed which relates the axial Peclet number to the superficial gas velocity. Application of this correlation in conjunction with the axial dispersion model to predict residence time distribution information is demonstrated.