FOREWORD

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 200-600 ton-per-day H-Coal pilot plant is being constructed next to the Ashland Oil, Incorporated, refinery at Catlettsburg, Kentucky, under ERDA contract to Ashland Synthetic Fuels, Incorporated. The H-Coal ebullated bed reactor contains at least four discrete components: gas, liquid, catalyst, and unconverted coal and ash. Because of the complexity created by these four components, it is desirable to understand the fluid dynamics of the system. The objective of this program was to establish the dependence of the ebullated bed fluid dynamics on process parameters. This will permit improved control of the ebullated bed reactor.

The work performed was divided into three parts: review of prior work, cold flow model construction and operation, and mathematical modeling. The objective of this final report is to outline accomplishments of the project in all three areas.

OBJECTIVE AND SCOPE OF WORK

The overall objective of this work is to improve control of the H-Coal reactor through a better understanding of the hydrodynamics of ebullated beds. The project is divided into three main tasks:

1) Review of prior work in three-phase fluidization.

2) Construction of a cold flow unit and collection of data.

3) Development of a mathematical model to describe the behavior of gas/liquid fluidized beds. The model should be based on information available in the literature and on data generated in the cold flow unit.

Progress made in all three tasks during this project is presented in this report. A list of all publications previously issued under this project is given in Table I.

SUMMARY

This report discusses the results of work aimed at understanding the hydrodynamic behavior of the H-Coal reactor. The scope of the work extended over three areas: review of prior work, data collection, and analysis and model development. Progress in each area is reviewed below.

Review of Prior Work

From the physical description of the H-Coal reactor, it became apparent that the H-Coal reactor can be modeled as a gas/slurry/catalyst system. The review of prior work established that there are several factors which control the bed expansion achieved when gas and/or liquid or slurry flow through a bed of catalyst particles. With liquid flow alone, the bed expansion can be described by several correlations. One of these was selected for correlating the data obtained in this work. This is the Richardson-Zaki correlation describing the volume fraction occupied by the liquid phase as a function of superficial liquid velocity and particle terminal velocity. The properties of the liquid phase and catalyst particle size are implicitly taken into account. When gas is added to the system, it has been generally found that the gas bubble dynamics control the hydrodynamic properties of the three-phase system. With particles greater than 3-4 mm, the gas tends to flow into the form of small bubbles, thus resulting in the so-called ideal bubbly flow behavior. On the other hand, smaller particles tend to promote bubble coalescence, resulting in large bubbles which characterize the churn turbulent flow regime. This flow phenomenon controls not only the bed

expansion for a given flow rate, but also determines the response of the three-phase system to changes in operating variables. For example, for a three-phase system with gas flow in the churn turbulent regime, increasing the gas velocity may lead to bed contraction. Many investigators developed mathematical models to describe their data. Of all these models, two were selected for correlating the data obtained during this work: the Darton-Harrison drift flux and the Bhatia-Epstein generalized wake models. The first model attempts to utilize principles from gas/liquid flow to define whether the system studied operates in the ideal bubbly or churn turbulent regime. For this purpose, the operating conditions--gas and liquid velocity--and the gas and liquid holdups are required. The second model (Bhatia-Epstein) explicitly considers the structure of the bed. The mathematical equations are based on the assumption that the bed consists of three phases: gas bubbles, wakes, and the particulate phase. The particulate phase consists of liquid/solid, and the bubble wakes consist of liquids/solids. The model recognizes that bubble size could vary with the properties of the fluid phases and that the concentration of solids in the wake phase can vary from 0 to that in the particulate phase. The Bhatia-Epstein model was the basis for developing correlations from data obtained under the work discussed in this report.

Data Collection and Analysis

Although the review of prior work established several models which might be applicable for the H-Coal reactor, most of these are based on water systems without addition of fines. It was therefore very important to obtain data in a system which closely simulated the H-Coal reactor. Study of the fluid dynamics in the actual reactor was ruled out by H-Coal participants. The cost of the operation in the process development unit (PDU) operated by Hydrocarbon Research, Incorporated (HRI) is very high for completing a meaningful study. No experimental techniques are reliable which can be used with confidence inside the reactor.

Cold Flow Model.--For the above reason, a cold flow model was constructed which was almost identical in size with the PDU reactor. The reactor ID and length were 6" and 20', respectively. The reactor consisted of four glass sections connected with metal spool pieces. Taps inserted in the spool pieces were used for temperature and pressure measurements. Sample probes could also be inserted through special ports. Since the major goal of this contract was to simulate the PDU reactor, the distributor used in the bottom of the reactor and the cup used for the suction of the recycle pump were identical with those of the PDU reactor. Other design aspects incorporated features previously established by HRI.

The experimental techniques selected for studying the fluid dynamics of the gas/slurry/catalyst system were the gamma-ray technique and differential pressure measurements. In selected cases, direct sampling was used to establish the fines distribution along the reactor. Argon-41 radioactive tracer experiments were also conducted to establish the residence time distribution (RTD) in the reactor.

In all cases, a data acquisition system tied to a ModComp II computer was utilized. The computer operated an elevator where the gamma-ray equipment was mounted. All data inputs were used to prepare a preliminary analysis.

Data Collection.--Meaningful application of this work for the H-Coal process is possible only if liquid, gases, and solids used in the cold flow studies have properties similar to the H-Coal liquids at actual operating conditions. At the beginning of this program, some information was available from HRI which indicated that kerosene/coal char slurries closely resembled the behavior of H-Coal slurries. Since the physical properties of the H-Coal slurries were not known at the start of the program, this work followed HRI's suggestion in selecting coal char/kerosene slurries for cold flow studies. The coal char has a density and particle size distribution which is about the same as the unconverted coal and ash in the H-Coal reactor recycle stream. Kerosene with 1, 5, 10, 15, and 17.8 vol% coal char was studied. Although kerosene was the base liquid, water and mineral oil were also used to scope the effect of liquid properties.

Nitrogen, helium, and Freon-12 were the gases selected. For most of the experimental program, standard H-Coal catalyst was used with dimensions of 1/16" ID, 3/16" particle length.

As part of this program, a sampling system was designed for obtaining samples from the PDU reactor for viscosity measurements. These were carried out by Battelle Labs along with coal char/kerosene slurries with 5.1, 10.5, and 17.8 vol% coal char. An analysis of these results demonstrated that the H-Coal liquids and coal char/kerosene slurries behave as Bingham fluids. The viscosity of the H-Coal liquids at PDU conditions is about the same as the viscosity of the coal char/kerosene slurries. Further work is required in the future to establish the effect of the non-Newtonian behavior on the fluid dynamics of the H-Coal reactor.

Data Analysis.--In analyzing bed expansion with liquid or slurries, it was found that the latter behave in the same way as clear liquids of equal viscosity. Thus, bed expansion with mineral oil (175°F) is the same as with a 17.8 vol% coal char/kerosene slurry of about equal viscosity when operated at the same liquid velocity. Hence, coal char and kerosene can be treated as a single phase. This was supported by direct sampling data which showed that coal char is uniformly distributed throughout the reactor. The Richardson-Zaki correlation was validated with slurry catalyst data. With gas added to a liquid, it was generally found that the viscosity of the fluid medium has a significant effect on gas/liquid/solid holdup. For kerosene, addition of nitrogen results in an increase in bed expansion. Application of the Darton-Harrison model indicated that in this case most of the gas flow is in the ideal bubbly regime. When the viscosity of the kerosene is increased by the addition of coal fines, or when a clear liquid with high viscosity is used, addition of gas may result in bed contraction. The Darton-Harrison drift flux model indicates that in this case most of the gas flows in the form of large bubbles.

Model Development

The experimental data were used to develop parameters for a modified Bhatia-Epstein type model with the capability of predicting not only bed expansion, but also the volume fraction occupied by the various phases. For the successful development of this model, it was necessary to derive correlations which describe a) the bubble terminal velocity as a function of gas and liquid rates; b) the wake volume as a function of liquid velocity; and c) the solids holdup in the wake as a function of bubble terminal velocity. It was generally found that high viscosity due to either clear liquids or addition of fines in kerosene increased the bubble terminal velocity and the size of the wake volume.

Recommendations

Additional measurements of the viscosity of H-Coal liquids from the PDU reactor are recommended to establish whether indeed the H-Coal liquids are Bingham fluids. These measurements should be carried out concurrently with gamma-ray scans of the H-Coal reactor to establish the response of the bed height to changes in liquid and gas flow rates. A comparison of these data with cold flow studies will give information regarding the significance of the results from this work.

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 (1,2). The process has been demonstrated by HRI in a process development unit (PDU) at Trenton, New Jersey. This unit is capable of processing up to three tons of dried coal per day. A schematic of the PDU reactor appears in Figure 1. A 200-600 ton/day pilot plant is under construction at Catlettsburg, Kentucky (2).

In the H-Coal process, coal is dried, pulverized to approximately 100-mesh size, and slurried with coal-derived oil. This slurry is charged with hydrogen to an ebullated bed reactor containing a hydrogenation catalyst (3). The reactor operates between 427-482°C and 136-218 atm.

The unique feature of HRI's H-Coal process is the use of an ebullated bed as the means of achieving good mixing between the catalyst particles, the slurried coal, and the hydrogen gas. The catalyst is in extruded form and is similar to those used in petroleum hydrotreating processes. Typical catalyst dimensions are 1.6 mm diameter and 4.8 mm length.

In the ebullating bed, the upward flow of the slurried coal and gases forces the catalyst bed to expand. The expansion of the bed allows fine particles such as coal and ash to pass through the bed and out with the liquid stream, leaving the catalyst bed within the reactor.

The variation in feed composition and slurry concentration causes significant variation in the feed viscosity. These viscosity changes affect bed expansion and lead to a range of operating conditions, as shown below (4):

% Expansion of Catalyst Bed	63-80
Reactor Slurry Composition:	
% Resid	13-18
% Solids (Coal and Ash)	9-22
Slurry Properties (at Reactor Conditions):	
Viscosity, cp	0.2-0.5
Specific Gravity, g/cc	0.6-0.7
Gas Velocity, cm/sec	1.2-4.9
Slurry Velocity, cm/sec	2.1-5.5

From the description of the H-Coal reactor, it becomes evident that bed behavior is a complex function of process variables and gas, liquid, and solid properties. Several models have been proposed in the literature for describing the behavior of three-phase fluidization as a function of these variables. However, these models have been tested only for air/water fluidized beds. Only limited data exist for liquids other than water, and no data exist for slurries which are of primary importance to the H-Coal process. In order to fill these voids, Amoco Oil has contracted with DOE to study ebullating beds. The ultimate objective is the development of an improved mathematical model which will accurately describe the behavior of the ebullating beds in the H-Coal reactor.

The work was divided into three main tasks: 1) review of literature; 2) construction of a cold flow model to simulate the PDU reactor; and 3) data collection and analysis. This report will present results from all areas. It will be shown that based on experiments conducted with slurries of coal char in kerosene, a model has been developed which describes the holdups of the various phases when the operating conditions and the physical properties of the fluids are known.

Reviewing available data, correlations, and models of three-phase systems is the first important task of the contract. For convenience and reasons which will be understood later, this task has been subdivided into the following areas:

1) Liquid/solid fluidization.

2) Vertical gas/liquid flow.

3) Gas/liquid/solid fluidization.

The review of these areas served as a basis for the development of a mathematical model to correlate three-phase fluidization data.

According to Darton and Harrison (5), a three-phase fluidization model should fulfill the following criteria:

1) As the volume fraction of gas approaches zero, the model should describe liquid/solid fluidization behavior.

2) As the volume fraction of solids approaches zero, the model should reduce to a suitable gas/liquid model.

3) The sum of the volume fraction of the various phases should add up to one.

4) The model should predict whether the bed will expand or contract upon the addition of gas.

In addition to the first criteria, understanding the liquid/solid fluidization is also important in predicting the H-Coal reactor bed height when there is an unexpected interruption in the hydrogen flow. Numerous correlations for predicting the bed height in liquid fluidized systems have been reported. These are summarized in Table II. The most widely used method of correlating liquid/solid fluidization data is that of Richardson and Zaki (6). Their analysis recognizes the similarities between sedimentation and fluidization. They observed that the settling velocity of a suspension relative to a fixed horizontal plane was equal to the upward liquid superficial velocity needed to maintain the suspension at the same concentration.

As shown in Table II, the correlation relates the liquid volume fraction (ϵ_l) due to bed expansion to the ratio of the superficial liquid velocity (ϵ_l) to the terminal velocity of a single particle (U_t):

$$\epsilon_l^n = U_l/U_t \qquad (1)$$

For spherical particles:

$$n = f(Re_t, d/D) \qquad (2)$$

where: Re_t = $d_p\ \rho_l U_t/\ \mu_l$, Reynolds number
d = particle diameter
ρ_l = liquid density
μ_l = liquid viscosity
D = bed diameter

The functional form of Equation 2 for different ranges of Reynolds number is given in Table II. It should be noted that for values of Re_t less than 0.2 or greater than 500, the exponent n in Equation 1 is independent of liquid viscosity.

For non-spherical particles, the exponent n in Equation 2 also becomes a function of particle shape. For turbulent Reynolds numbers (greater than 500), Richardson and Zaki found:

$$n = 2.7\ K^{0.18} \quad (3)$$

where: $$K = (\pi/6)d_s^3 d_p^3 \quad (4)$$

The constant K is the particle shape factor proposed by Heywood (7). In Equation 4, d_s is the diameter of a sphere with the same volume as the particle, and d_p is the diameter of a circle of the same area as the projected particle when lying in its most stable position.

The addition of gas to a liquid fluidized bed increases the complexity of the system considerably. Kim, Baker, and Bergougnou (11) and Darton and Harrison (5) reported that for the range of superficial gas and liquid flow rates of interest to the H-Coal process, two gas/liquid flow regimes exist in the catalyst bed: 1) the ideal bubbly or bubble disintegration regime, in which the bubbles rise as a uniform, steady cloud with little interaction; and 2) the churn turbulent or bubble coalescing regime, which is a transition between ideal bubbly flow and fully developed slug flow. The churn turbulent regime is dominated by bubble coalescence. Hence, the bubble size is larger than in the ideal bubbly regime, bubble wake effects become important, and the flow is unsteady.

The effect of catalyst particle size on the rate of bubble coalescence has been pointed out by Kim, et al. (11) and by Ostergaard (12,13). Small particles promote the churn turbulent (bubble coalescence) flow regime and reduce gas holdup. Large particles, on the other hand, tend to favor the bubble disintegrating (ideal bubbly) flow regime and increase gas holdup.

Numerous investigators (17,18,19,20,21) have found evidence to support this effect of catalyst particle size. The experimental conditions used by these investigators are listed in Table III. From these studies it follows that the critical particle size between coalescing and non-coalescing beds is between 3 and 4 mm for the systems studied.

In a related phenomenon, Turner (14) was the first to report the contraction of some liquid fluidized beds upon the addition of gas. Kim, et al. (11), proposed that this contraction will occur only in beds in the bubble coalescing flow regime. According to Stewart and Davidson (15), contraction occurs because of the formation of liquid wakes behind the bubbles which move through the bed at the bubble velocity. This action reduces the interstitial liquid velocity in the rest of the bed, thus causing its contraction. Ostergaard has also offered a similar explanation.

A summary of additional experimental studies on three-phase fluidized beds is shown in Table IV. Many of these investigators have correlated their data in various forms, as shown in Table V. It should be emphasized that these correlations should be used only for the specific system and range of variables from which they are developed.

where U_s is the gas liquid slip velocity defined below:

$$U_s = \frac{U_g}{\epsilon_g} - \frac{U_l}{\epsilon_l} \tag{8}$$

Combining Equations 7 and 8, the following relationship is obtained:

$$V_{CD} = U_g(1 - \epsilon_g) - \frac{U_l \epsilon_g}{\epsilon_l}(1 - \epsilon_g) \tag{9}$$

Darton and Harrison analyzed the fluidization data of Michelsen and Ostergaard (23) in terms of Equation 9. A plot of V_{CD} vs. ϵ_g revealed two flow regimes: the ideal bubbly and the churn turbulent. This plot is reproduced in Figure 2. As can be seen from the figure, the data from the two flow regimes fall along different lines. Typical transition data are indicated by the dashed lines between the two regimes. The data for the ideal bubbly regime fall on the line given by:

$$V_{CD} = 180\ \epsilon_g (\text{mm/sec}) \tag{10}$$

The condition for bed contraction can be found by adding ϵ_g to both sides of Equation 6 and then taking the derivative of $(\epsilon_l + \epsilon_g)$ with respect to U_g as $\epsilon_g \rightarrow 0$. The condition for contraction is:

$$(1 + \bar{k})(1 - \epsilon_l + \frac{\epsilon_l}{n}) < \epsilon_l \frac{\bar{k}}{n} \frac{U_b}{U_l} \tag{11}$$

Epstein (32) has proposed a similar criterion for bed contraction.

Bhatia and Epstein (33) have also proposed a generalized wake model. Following Ostergaard (16), they also propose three phases to the bed: gas, wake, and the liquid particulate phase. The solids concentration in the wake phase can be varied between zero and that in the particulate liquid/solid fluidized phase by means of an adjustable parameter, X_k. The value of X_k ranges between 0 and 1. The wake volume is related to that of the gas by an empirical function. The particulate liquid/solid phase is described by a Richardson-Zaki type model. The model also takes into consideration the possible existence of two gas/liquid flow regimes in the bed. Bhatia and Epstein develop eight equations for the eight unknowns in the system. These equations are listed in Table VI. The equations must be solved by an iterative procedure.

The authors test this model vs. the data of Michelsen and Ostergaard (23) and data of their own. Their data are for the fluidization of spheres ranging in size from 0.25 to 3.0 mm. The solid density ranged from 2.5 to 11.1 mg/m^3. The gas used was air, and the liquids were water (1 cp), aqueous glycerol (2.1 cp), or aqueous polyethylene glycol (63 cp). The experiments were carried out in 20 and 50 mm columns. Best agreement between the model and data was obtained when $X_k = 0$ (solids-free wake).

This model, along with the contraction criteria proposed later by Epstein (32), also meets the four criteria suggested by Darton and Harrison. In many respects Bhatia and Epstein's wake model is very similar to the model proposed by Darton and Harrison.

The literature search has indicated that the gas flow behavior is very critical in determining not only bed expansion, but also conditions under which bed contraction occurs. It therefore follows that a successful model should take into account the bubble size and wake volume following the bubbles. Only semi-theoretical models such as Ostergaard's (16), Darton and Harrison's (5), and Bhatia and Epstein's (33) are considered promising in correlating experimental data.

CONSTRUCTION OF COLD FLOW UNIT AND DATA COLLECTION

From the literature search it becomes evident that although significant contributions have been made in the field of gas/liquid/solid systems, no data exist for slurry systems directly applicable to the H-Coal process. For this reason, equipment was constructed in Amoco Oil which is almost on scale with the 3 T/D coal liquefaction process development unit (PDU) operated by HRI for several years. Data were obtained at ambient conditions with gas/slurry/catalyst systems having similar physical properties to the H-Coal liquids at reactor conditions. Equipment details and properties of the liquid/slurry used will be described in this section.

Equipment

A schematic diagram of the H-Coal fluid dynamics unit is shown in Figure 3. Design and construction of this unit were coordinated by the systems design group of Amoco Oil. Systems design incorporates elements of process design, mechanical design, instrumentation, automation, and computerization. Details on the work done in each area are given in Reference 34.

The process flow of the H-Coal fluid dynamics unit resembles that of the process development unit (PDU) built by HRI. The reactor consists of a 6" ID vessel 20' in length. The slurry is prepared in a 60-gallon tank. Other major vessels include one 100-gallon feed tank, and a 60-gallon gas/liquid separator with a mist eliminator.

Three pumps are needed for the operation of the unit: 1) a transfer mixing pump for facilitating the slurry preparation and the transfer of the slurry to the feed tank; 2) a slurry feed pump for supplying the slurry to the reactor; and 3) a slurry recycle pump for internal slurry circulation in the reactor vessel. Under typical test operating conditions, the slurry feed rate amounts to 3 gallons per minute, while the slurry recycle pump supplies 15 gallons per minute. Gas and liquid overflow from a 1" ID pipe located at the same level as the top of the rcycle cup. The gas is separated from the liquid in the separator, D-3. Entrained liquid droplets accumulate in a demister located on top of the separator. The gas passes through Cooler E-2, and it can either be vented to the atmosphere through Valve PV-1 or can be directed to a gas recycle compressor. This flexibility is necessary to use gases such as Freon-12 and helium. The gas combines with the total liquid stream before it returns to the reactor. Some details on selected equipment items are given in Appendix A.

The reactor is constructed from four glass sections 6" in ID and 5' in length. The glass sections are connected through flanges to five metal spool pieces. The spool pieces have entries for sample taps, pressure taps, and thermowells to monitor the system. Samples of the slurry withdrawn here are used to establish the coal fines concentration along the reactor.

An elevator is designed so that a ten-millicurie Cs-137 gamma-ray source and detector can travel vertically along the reactor.

Physical Properties of Liquids and Solids

The objective of the cold flow modeling studies was to establish the hydrodynamic properties of the H-Coal reactor. The usefulness of these studies will greatly depend on the selection of the fluids which at ambient temperatures have physical properties similar to those of the H-Coal liquids at actual operating conditions. For this reason, as part of this contract samples of H-Coal liquids from the HRI PDU were sent to Battelle for viscosity measurements. As will be discussed later, it was found that the viscosity of H-Coal fluids is in the range of 1 to 2 cp at actual operating conditions.

In this work, fluids and slurries were used which at about room temperature have properties similar to those of H-Coal slurries in the H-Coal reactor. Water, kerosene, and mineral oil were selected with properties shown in Table VII. The properties were measured as a function of temperature. A Fischer Scientific surface tensiometer (Model 20) was used. The results indicate that surface tension varies linearly with temperature over the range studied. Since during this study He, N_2, and Freon-12 gases were used, it was essential to establish the effect of these gases on interfacial tension. Results from these measurements are reported in Table VIII. It can be seen that there is a small variation of surface tension with gas type, most likely due to gas dissolving in the liquid phase.

In addition to the viscosities of the pure liquids reported in Table VII, the viscosities of kerosene/coal char slurries were measured. Initially, a capillary tube viscometer having an ID of 1.6 mm and a length of 100 cm was constructed and connected to the unit as shown in Figure 4. The viscosities of 11.9 and 15.5 vol% coal char/kerosene slurries were found to be 3.5 cp at 72°F and 3.7 cp at 77°F, respectively. Operation of this viscometer was hampered by frequent settling of the fines and subsequent plugging. Samples of kerosene slurried with 5.1, 10.4, and 17.8 vol% coal char were also sent to outside laboratories for viscosity measurements. These measurements are reported in Table IX. From the comparison of these data with the viscosity of H-Coal liquids at actual operating conditions (see Appendix D), it is concluded that the viscosity of kerosene slurries with 10 to 15 vol% coal char have about the same viscosity as the H-Coal reactor liquids at actual operating conditions.

Another important factor for the successful completion of the cold flow modeling studies is the selection of the fines used for the preparation of the slurries. Previous studies by HRI (35) have established a material (coal char) which has similar properties to those found in the recycle stream of the H-Coal reactor. Results from these measurements are reported in Table X. For a more detailed particle size analysis of the coal char, measurements were conducted by the Illinois Institute of Technology using an optical microscope interfaced with a Quantimet 720 computerized image analyzer. The distribution is reported in Table XI. The geometric mean particle diameter is 3.4 microns.

The catalyst particle size distribution has also been measured. The nominal length of catalyst chosen for this study is 4.8 mm; the diameter of the catalyst is 1.6 mm. This catalyst is similar to the one previously used by HRI in PDU studies. The catalyst has been supplied by American Cyanamid. Cyanamid has guaranteed that the variation of length does not exceed ± 20% for each of the above lengths. The length distribution and other catalyst properties are listed in Table XII.

Unit Data

A summary of all experimental runs conducted during this program is reported in Table XIII. For each run, a given amount of catalyst was loaded to the reactor filled with liquid. The catalyst particles were allowed to soak in the test fluid for a period of over 24 hours. The experimental techniques then used to find the volume fraction occupied by the various phases for each run were pressure drop measurements, gamma-ray scans, and radioactive gas tracers.

Pressure Drop Data.--These can be used to supplement the other techniques for calculating the volume holdup of the various phases. The method of calculation is outlined in Appendix B. Because pressure taps through the spool pieces were used, only average holdups through the bed can be calculated.

Gamma-Ray Scan Data.--A zero gamma-ray scan was then carried out to find the zero bed height (H_o) 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 5. From plots of this type, a bed height was established for each test. Based on this bed height, the initial catalyst charge, the pressure drop, and the gamma-ray measurements along the reactor, the holdup of each individual phase was determined. When 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 are reported in Appendix B.

Tracer Data.--The use of a radioactive gas tracer (argon-41) has previously been considered by Michelsen (23). The gas tracer used for this work was also argon-41 (A^{41}), a gamma-ray emitter which is prepared by neutron bombardment of highly purified argon-40. Enough radioactive A^{41} was prepared so that the activity was at least 150 mc when injections into the trcer were started.

The tracer was injected through a tube inserted into the reactor above the bubble cap distributor. A previous study (23) using A^{41} as the tracer noted difficulties in analyzing tracer data due to the interphase transfer of argon. Therefore, non-radioactive A^{40} was used to presaturate the system before injection of the tracer. Experiments also showed that absorption of argon by coal fines is negligible.

Six scintillation detectors were mounted on the system as shown in Figure 6. The spacing between detectors is shown in the same figure. Detectors 1 and 2 always viewed the catalyst bed. Detectors 3 and 4 always viewed the liquid or slurry phase above the bed. Detectors 5 and 6 viewed gas lines only and were used primarily to estimate the overall gas holdup in the system.

All the detectors were highly collimated with lead shielding to view only a thin vertical segment, but to cover the entire reactor cross-section. The detectors were connected to an electronics train which included high-speed data recorders and a data acquisition system linked to a ModComp II mini computer. The data were collected at two rates--either 5 or 10 measurements per second

Tracer tests and gamma-ray scans of the reactor were obtained at the same system conditions. Reactor holdups determined by the scans were compared with tracer results. The tracer data analysis procedure is reported in Appendix C.

Catalyst and Coal Fines Settling Data.--These were determined by shutting off all gas and liquid flows and using the gamma-ray equipment to determine gamma-ray attenuation at a specific location of the reactor as a function of time. Successive runs of this type were used to measure the settling rate of the fines and the catalyst particles from the slope of the graph of interface height vs. time.

A summary of the settling data for HDS-2A catalyst (1/d = 3) fluidized by nitrogen appears in Table XXIV. A plot showing the settling of the fines appears in Figure 7.

Physical Properties of H-Coal Liquids

Measurement of the viscosity of H-Coal liquids is very important to determine if the viscosity of H-Coal liquids at reactor conditions is in the same range as the viscosity of the liquids and slurries studied in the cold flow unit. As a part of the contract, this comparison was made. With the assistance of HRI, H-Coal liquid samples were obtained from the PDU at HRI. The viscosities of these samples, along with three char/kerosene samples, were determined by Battelle Labs (Columbus, Ohio) under a subcontract. Battelle was selected to make these measurements because they had previous experience in measuring the viscosity of Synthoil liquids.

PDU Sampling Technique.--The H-Coal samples were taken during PDU Run 8 in July, 1979. The sampling point on the PDU was located between the external separator and the two-stage pressure letdown valves. The sampling technique is described in Appendix D.

Four H-Coal samples were obtained. The reactor temperature and pressure during the sampling were approximately 850°F and 2600 psig, respectively. Additional data for the samples are contained in Table XIV.

Viscosity Measurements.--Battelle measured the viscosity of the H-Coal samples and the char/kerosene samples (5.1, 10.4, and 17.8 vol%) in a viscometer described in Appendix D. It was found that both the H-Coal samples and the char/kerosene samples could be characterized as Bingham fluids. The relationship between the shear stress and the shear rate for a Bingham fluid is illustrated in Figure 8. As can be seen from the figure, the relationship is linear, with slope of the line being equal to the viscosity. However, unlike a Newtonian fluid, a certain stress must be overcome before deformation of the fluid occurs. This stress is known as the yield stress and is illustrated by O in Figure 8.
Thus, the relationship between shear stress and shear rate for a Bingham fluid can be summarized as:

$$\dot{\gamma} = 0 \qquad \sigma < \theta \tag{12}$$

$$\sigma = \theta + \eta' \dot{\gamma} \quad \sigma > \theta \tag{13}$$

where: η' = viscosity
$\dot{\gamma}$ = shear rate

The measured values of η' and θ for the H-Coal samples and the char/kerosene samples are listed in Appendix D. The viscosity of the H-Coal samples is plotted as a function of temperature in Figure 9. It is shown that the viscosity is a strong function of temperature. It was found that the pressure had very little effect on viscosity. A comparison of the viscosities of H-Coal liquids with the viscosity and yield stress of coal char kerosene samples at ambient conditions shows that both are in the same range. The viscosity of the H-Coal samples decreased from 48 to 1.2 cp as the temperature increased from 340 to 831°F. The viscosity of the char/kerosene samples displayed the same behavior, decreasing by a factor of approximately 2 as the temperature increased from 70 to 140°F.

The overall effect of the yield stress on the fluid dynamics of the ebullated bed would be to reduce mixing in the bed. This is because before any relative motion could occur between the liquid and gas or solid phases, the yield stress would have to be overcome. For instance, for a bubble rising in the bed, the velocity would be slower because the buoyancy forces would have to overcome the yield force in addition to drag forces. The magnitude of the effect would of course depend on the relative magnitude of the buoyancy and yield forces. For instance, for a yield stress of 10 dynes/cm^2 (1000 milli N/m^2), typical value found by Battelle, the yield force on a bubble 1 cm in diameter would be 7.85 dynes. If the bubble were rising in a fluid of density 1 gm/cc (neglecting gas density), the buoyancy force would be 512 dynes. Thus, the effect of the yield stress would be expected to be small. However, as the bubble size decreases, the effect will become more important.

The effect of non-Newtonian liquids on the behavior of bubbles and solids in them is a subject which has received relatively little attention in the literature. However, it is a subject which could be of great potential interest in further understanding the hydrodynamics of ebullating beds, and it should be studied further.

The principal conclusions which can be drawn from the viscosity measurements are:

1) H-Coal liquids are characterized as Bingham fluids.

2) The viscosity of H-Coal liquids is a strong function of temperature.

3) The viscosity of H-Coal liquids is a weak function of pressure.

4) Char/kerosene slurries used in the cold flow unit are also characterized as Bingham fluids.

5) The viscosity of char/kerosene liquids used in the cold flow unit is in the same range as the viscosity of H-Coal liquids at reactor conditions.

Data Analysis

In this section, cold flow model data are presented which establish the effect of coal char concentration, temperature, gas and slurry velocity, viscosity, and gas type on bed behavior. The effect of the various parameters on bed expansion, catalyst holdup, and gas holdup will be presented. Following these data, correlations will be considered developed on the basis of existing models described in the literature section. However, it is important first to establish whether or not the coal char fines are uniformly distributed throughout the reactor.

Fines Distribution.--The distribution of coal char in the fluid dynamics reactor has been measured by taking samples from the unit. The weight per cent of coal char in the slurry sample is found by millipore filtration. The results of this analysis for 5.1, 10.4, 11.9, 15.5, and 17.8 vol% coal char/kerosene slurries are given in Table XV. In general, little variation in char concentration with position in the reactor is observed. Particle size analysis was also done on reactor bottom and 451-cm-level samples from Test 211-07. These results are given in Table XVI. This analysis was performed to determine if there were particle segregation in the reactor due to flotation effects. The results for the samples taken at the top and bottom of the reactor are similar; however, the sample from the top of the reactor has a smaller average particle size, 2.8μ vs. 3.5μ.

Comparison of the coal char concentration at the reactor bottom and the 457 cm level for Tests 206-5 and 211-11 in Table XV also is a check of coal fine segregation due to flotation. If small fines are preferentially rising to the reactor top with gas bubbles, the difference between concentrations should be greatest for the high gas flow rate case. However, Table XV indicates no effect of gas flow rate. It therefore follows that assuming that the fines are well mixed throughout the reactor is a good approximation. Other investigators have established the validity of the same assumption (37).

Liquid or Slurry Fluidization.--The effect of coal fines concentration on bed expansion at zero gas flow is shown in Figure 10. Increasing the coal char concentration in kerosene from 0 to 17.8 vol% (viscosity increase

from 1.4 cp to 8 cp at 75°F) increased bed expansion by a factor of 2 in most cases over the range of slurry velocities studied. The effect of slurried temperature on bed expansion is also presented in Figure 10. Changing the temperature from 148°F to 85°F (viscosity change from 6.8 to 8.0 cp) also increases the bed expansion. However, the increase in bed expansion is lower because there is a smaller viscosity change.

The need to establish whether the increased bed expansion from coal char/kerosene slurries is due to their high viscosity necessitated experiments with mineral oil. The viscosity changed with temperature from 4.2 cp at 175°F to 22.4 cp at 70°F. Bed expansions at various liquid velocities with the temperature as a parameter are plotted in Figure 11. It is again observed that increasing the viscosity has a large effect on bed expansion. In fact, Figure 12 indicates that coal fines/kerosene slurries and mineral oil result in the same bed expansion.

The effects of the catalyst properties on bed expansion are shown in Figure 13. For 1/16" diameter catalyst particles, decreasing the length from 3/16" to 2/16" will increase the bed expansion by about 20%. No studies with fines were performed using the 1/d = 2 catalyst; however, the conclusions drawn from Figure 13 are expected to be applied in these cases. A much smaller increase in bed expansion was obtained when for particles with 1/d = 3, the bulk density increased from 0.7 to 0.9. The high-density catalyst was used in a PDU run, and as a result it had a low pore volume ($\sim$0.25 cc/gm) compared with 0.66 of the catalyst used for most of the tests in this study. Since the bed expansion depends on the density of the catalyst particles soaked with liquid, the small bed increase is due to the fact that both catalysts have about the same soaked catalyst density.

For a liquid/solid system, the liquid holdup can be calculated from the bed expansion with the procedures described in Appendix B. Correlation of the liquid holdup with operating conditions and the physical properties of the system can be done by the application of one of the several models listed in Table II. The Richardson-Zaki (6) model was used during this work because of its successful application in previous studies with systems similar to those considered in this contract (45). Following Equation 1, plotting $_l$ vs. U_l in log-log paper or a plot of ln $_l$ vs. lnU_l in linear paper should result in a straight line with slope equal to the reciprocal of the Richardson-Zaki index n. This is illustrated in Figure 14, where the liquid volume fraction is plotted as a function of superficial slurry velocity. Utilizing standard linear regression techniques, the experimental Richardson-Zaki index n was determined for each run. These are reported in Table XVII. The results indicate that the Richardson-Zaki index changed very little with either coal char fines concentration or viscosity. This is expected because the index n is insensitive to changes in this Reynolds number range. For example, according to the correlation for n given in Table II ($1 \leq Re \leq 200$), to obtain a 10% change in n, the viscosity must change by a factor of 3.

Three-Phase Fluidization--Gamma-Ray.--Bed expansion for both kerosene and kerosene slurries is presented in Figure 15. It has been previously discussed that the addition of gas in a liquid fluidized bed will generally result in an increased bed expansion. Figure 17 indicates that although this is generally true for the nitrogen/kerosene/catalyst system with zero fines, bed expansion is in fact reduced in some cases with a 17.8 vol% coal fines/kerosene slurry. It is believed that the fines uniformly distributed throughout the reactor increase the viscosity of the kerosene, promoting the formation of large bubbles due to coalescence. Each large bubble carries with it a substantial wake of liquid/catalyst. As a result, the liquid in the wake goes through the bed much faster than if it flowed through the interstices between the particles when the gas was absent. Thus, the wake liquid may be regarded as not flowing through the bed at all and, as far as the particles are concerned, the liquid flow is reduced and therefore the bed contracts. The effect of temperature on bed expansion with a 15.5 vol% coal char slurry system is illustrated in Figure 16. It is shown that as the temperature is reduced, the bed expansion increases due to the viscosity increase of the coal char/kerosene slurry. A comparison of Figures 15 and 16 establishes again that the effect of coal fines is mainly due to the increased viscosity of the liquid kerosene. Figures 15 and 16 illustrate the conditions under which bed contraction occurs. As Ostergaard (30) has reported previously, the bed contraction is a function of the relative liquid/gas flows and the viscosity of the liquid phase.

In an effort to compare a coal char/kerosene slurry with mineral oil of about the same viscosity, bed expansions are plotted in Figure 17 for both systems at different operating conditions. Kerosene liquid data are also presented for comparison. The comparisons shown in Figure 17 indicate that coal char/kerosene slurries behave in a manner similar to a pure liquid of the same viscosity. A similar conclusion was reached in an earlier section with a two-phase slurry/catalyst system.

Although the viscosity has great influence on gas flow behavior, it has also been reported previously (20,21,18,19) that small particles promote bubble coalescence and hence favor conditions under which bed contraction takes place. Large particles, on the other hand, tend to reduce bubble coalescence. It has been found that the critical particle size is between 3 and 4 mm for conditions reported in the literature. In an effort to establish the effect of particle size on gas flow behavior, the fluidization of catalyst particles with nitrogen and kerosene was studied when the l/d ratio of the particles changed from 3 to 2. Figure 18 indicates that over the range of particle dimensions studied, the gas flow behavior remains the same. As expected, the small particle size results in greater bed expansion due to its lower terminal velocity.

Another factor which has also been considered in this study is the density of gas and the influence of gas type on the surface tension of the liquid. As reported in Table VIII, the surface tension of kerosene slightly

increases when it is in in contact with He instead of N_2. As shown in Figure 19, this small difference does not appear to have a significant effect on either bed expansion or the conditions under which bed contraction occurs. A similar conclusion is shown in Figure 20, where catalyst particles are fluidized with mineral oil and helium or nitrogen.

Bed expansions can be expressed in equivalent catalyst holdup (ϵ_c) with the equations listed in Appendix B. Plots of ϵ_c as a function of operating conditions and system physical properties are also reported in Appendix B. These plots will be very useful if it is desired to explore the application of various models in predicting the catalyst holdup ϵ_c.

In reviewing the gas/liquid (or slurry)/catalyst data, it has been shown that increasing the viscosity of the liquid phase results in increased bed expansion as the liquid rate increases, and in possible bed contraction as gas is added to the system. In explaining the bed contraction, the suggestions of other investigators were accepted stating that increased viscosities result in bubble coalescence and significant wake effects. Bubble coalescence should in addition result in a decrease in gas holdup because the latter is related to bubble velocity, U_b, and the average gas supeficial velocity, U_g, by the following equation (16):

$$\epsilon_g = \frac{U_g}{U_b} \qquad (14)$$

The gas holdup in the catalyst bed is plotted versus the gas velocity in Figure 21. It is shown that gas holdup increases with gas superficial velocity with kerosene at zero coal char fines concentration. However, when coal char fines are added to the system, the gas holdup in some cases is in fact reduced with increasing gas velocity. It appears that this reduction in gas holdup is a function of the relative liquid/gas flows and the viscosity of the continuous fluid phase. The low-viscosity kerosene has the highest holdup. Gas bubbles remain small and do not coalesce. The average bubble rise velocity (U_b) is correspondingly slow. In contrast to this, when coal fines are added to the system, gas coalesces to larger bubbles having a higher rise velocity. As a result of this, the gas holdup is reduced. Although various reasons can be invoked explaining the effect of coal fines on bubble coalescence, Figures 21 and 22 indicate that viscosity effects are important: the increased viscosity of the mineral oil decreases the holdup in a similar manner to the coal char kerosene slurries. The need to understand the changes in gas holdup discussed above necessitated the gas tracer tests discussed in the next section.

Three-Phase Fluidization--Tracer Data.--Residence time distribution curves obtained from radioactive gas tracer experiments can in principle be used to calculate the gas holdup (ϵ_g) inside the reactor using the linear velocity (V_g) and the superficial velocity (U_g) as follows:

$$\epsilon_g = \frac{U_g}{V_g} \qquad (15)$$

Typical residence time distribution data with (15.5 vol%) and without coal fines are shown in Figures 24 and 23, respectively.

The first and second moments for curves such as those shown in Figures 23 and 24 were calculated using the equations included in Appendix C. A comparison of the first and second moments calculated by procedures described in Appendix C for selected cases is shown in Table XVIII.

The gas linear velocity in the catalyst bed and above the catalyst (dilute phase) was calculated from the tracer results for several cases. The calculated velocities and test conditions are shown in Table XIX. Replicates for each case show generally good agreement with typically less than 10% error. However, experiments performed near the onset of churn turbulent behavior show wider variation.

Comparison of gas velocities with and without coal char at similar operating conditions shows that the average gas velocity in the catalyst bed is not significantly changed. However, addition of the fines causes a more rapid tracer response at the top of the bed, combined with increased dispersion and apparent backmixing. These results support earlier statements that the addition of coal fines enhances bubble coalescence, causing the formation of larger bubbles which rise quickly through the reactor.

As discussed earlier, this bubble coalescence is probably caused by the higher slurry viscosity due to coal fines. As reported by Calderbank, et al. (42), bubble coalescence is enhanced by more viscous fluids. The same author noted that larger bubbles tend to form and rise in the center of the column. This results in a downward motion of gas at the walls.

Rigby, et al. (46), also reported the tendency of gas bubbles to preferentially rise in the center of a gas/liquid fluidized bed. This results in a downward motion of gas at the walls which may cause the spreading of the tracer concentration, as indicated by the comparison of the second moments for Tests 1 and 2. This comparison indicates that coal fines cause the tracer to spread out in the reactor, possibly as a result of the downflow of gas near the walls.

A comparison of gas holdup calculated from gamma-ray scans with tracer results is shown in Table XX. In all cases, the values calculated from tracer results are significantly higher than those determined from gamma-ray scans. As discussed earlier, results from the tracer test reflect the gas holdup in the entire reactor cross-section, whereas the gas holdup calculated from gamma-ray scans reflects a value corresponding to a measurement taken across the diameter of the reactor.

To determine if a radial gas distribution could account for the difference between holdups calculated by the two methods, gamma-ray scans through three chords of the reactor were obtained. Gas holdups were determined through the reactor center and on two chords at different flow conditions. The test results are given in Table XXI.

Except for one value, the results are all within experimental error. This implies there is not a significant radial distribution of gas which could account for the large differences between holdups calculated by the two techniques.

It is believed that the discrepancy in the calculated holdups can be explained by the gas flow pattern described earlier. Flow of large gas bubbles in the center of the reactor followed by downward flow of gas near the walls will result in long residence time distributions. This in turn will give apparently low linear gas velocities and high gas holdups. On the other hand, the gas holdup calculated from other techniques is independent of the direction in the gas flow.

The qualitative picture of large bubbles traveling up in the center of the reactor causing downflow at the walls is consistent with the differences in holdups calculated by the two methods. Visual observation of the reactor also supports this model. A significant downflow of small gas bubbles can be seen at the reactor walls.

Results from the gamma-ray scans through different chords of a horizontal section are also consistent with this model for gas flow in the reactor. The gas holdup in the two different flow regimes could be the same, although the gas is traveling in different directions.

The ratio between the gas holdups calculated from gas tracer and gamma-ray tests can be related to the amount of gas in each flow regime. However, it must be assumed that the detector views the reactor impartially; tracer in the center of the reactor contributes equally to the total signal to the tracer at the walls. As the ratio of tracer to gamma-ray scan calculated gas holdups increases, the fraction of gas traveling down in the reactor or the cross-sectional area occupied by gas moving down increases. This ratio increases with the addition of coal fines to the reactor, supporting previous indications that coal fines enhance bubble coalescence to form large bubbles which would rise through the center of the reactor.

To quantify the extent of gas mixing inside the reactor, equations described in Appendix C were used to calculate the gas dispersion coefficients inside the bed (Detectors 1 to 2) and the region above the catalyst bed (Detectors 3 to 4). These results are reported in Table XXII. From the magnitude of the dispersion coefficients, it is confirmed that there is apparent gas backmixing throughout the reactor. Work describing a mixing model quantifying the observed results is discussed in a later section.

Three-Phase Fluidization--Data Analysis.--Three-phase fluidization data were analyzed using two different correlations: Darton and Harrison's drift flux approach, and Bhatia and Epstein's generalized wake model. Results of the drift flux analysis will be reviewed first. The drift flux V_{CD} is defined in Equation 9. Calculated values of drift fluxes for all runs are reported in Appendix B. As indicated in Figure 2, plotting V_{CD} vs. gas holdup ϵ_g could identify two flow regimes: churn turbulent and ideal bubbly.

This type of plot for the nitrogen/kerosene/catalyst system with 0 vol% and 17.8 vol% fines is shown in Figure 25. For kerosene with 0 vol% fines, the data fall in the ideal bubbly flow regime. On the other hand, addition of coal fines enhances the transition from ideal bubbly to churn turbulent flow by increasing the slurry viscosity. Also shown in Figure 25 is the effect of increased liquid flow rate on inhibiting the ideal bubbly to churn turbulent transition. Figure 26 confirms the fact that viscosity increase is the main reason for bubble coalescence. Mineral oil of about the same viscosity as coal char/kerosene slurry enhances the transition to churn turbulent. Figures 25 and 26 indicate that a large amount of the data at high viscosities lies in the transition region, suggesting that bubble size continually changes as the gas rate increases. For this reason, it is very difficult to use the drift flux model as a quantitative tool in correlating holdups for the experiments conducted in this study. Instead, an effort was made to test the Bhatia and Epstein model. The equations describing this model were previously presented in Table VI.

The iterative method used to solve the Bhatia-Epstein equations given in Table VI is shown in Table XXIII. Prior to correlating the data obtained in this study, the sensitivity of this model was tested for changes in X_k (ratio of solids holdup in the wake to the solids holdup in the particulate phase), the Richardson-Zaki index n, terminal catalyst particle velocity (U_t), and gas bubble velocity (U_{t_B}). It was found that the model was most sensitive to terminal bubble velocity. Other variables have secondary effect. For example, a 10% change in either the Richardson-Zaki index or the catalyst terminal velocity (the error in determining these values) does not have a significant effect. Therefore, use of the Bhatia and Epstein correlation requires a good estimate of the bubble terminal velocity (i.e., bubble diameter).

A preliminary analysis of the data in this study with the Bhatia-Epstein model was performed. the variables which must be known to solve the set of equations are: U_l, U_g, X_k, U_t, U_{t_B}, and n. The gas and liquid velocities are known. The Richardson-Zaki index and catalyst terminal velocity were determined experimentally. The relative wake solids content, X_k, was calculated from an empirical correlation developed by El-Temtamy and Epstein (38):

$$X_k = 1 - 0.877 \frac{U_t}{U_s} \qquad (16)$$

The value of the terminal bubble velocity was then varied to give the best fit to experimental data.

Comparisons of experimental catalyst holdups with results from the Bhatia-Epstein model for tests with 0 vol% fines and 17.8 vol% showed that agreement between calculated and experimental holdups is generally good except when bed contraction occurred for the 17.8 vol% fines tests. In this case, the Bhatia and Epstein correlation did not predict bed contraction.

For the 0 vol% fines case, the calculated value for X_k was always about zero. Best agreement with the model was obtained with low terminal bubble velocities, corresponding to relatively small bubbles. With 17.8 vol% coal fines, X_k ranged from 0.1 to 0.8. For this case, best agreement with the model resulted in terminal bubble velocities significantly higher than the 0 vol% fines case. This indicates that with coal fines the gas coalesces to large bubbles which rapidly move through the reactor. Radioactive gas (argon-41) residence time distribution tests discussed earlier confirmed that these phenomena are actually occurring.

From the preceding discussion it appears that a Bhatia-Epstein type model offers the best promise in determining the phase holdups once operating conditions are specified. However, in fitting this model, it was found that in certain cases it failed to match some experiments. For this reason, a modified model describing the experimental results of this study was developed. This will be discussed in a later section.

Catalyst and Coal Fines Settling--Data Analysis

Examination of the data summarized in Table XXIV suggests that the bed settling rate after loss of flow is appreciably slower than the corresponding particle terminal velocity. The ratio of terminal velocity to settling rate appears to decrease as bed expansion increases. It is therefore likely that one factor controlling bed settling rate is the dynamics of displacing the excess liquid and gas held up in the expanded bed.

The settling rate of the coal fines in the reactor (as shown in Figure 7) was significantly slower. Particles of this low average diameter and low excess density above the fluid phase are probably forming a colloidal suspension. Although these results support the earlier claims that the fines are uniformly dispersed in the liquid phase, further settling tests should be performed using the higher-density reactor fines.

Comparison of Experimental Results with HRI PDU Data

The most important criterion for the correlations developed from these experimental data is to be able to predict the effect of operating conditions in an H-Coal reactor. Therefore, to test the validity of these correlations, data from cold flow studies should be compared with similar information from the H-Coal reactor at actual operating conditions.

A comparison of bed expansion in the HRI PDU with experimental bed expansion is shown in Figure 27. Results from PDU Run 7 (40), indicated by triangles, are compared with cold flow unit tests with 17.8 vol% coal char in kerosene at 75°F. Reactor slurry velocity was 0.09 ft/sec in all cases. PDU bed expansions are predicted well by these cold flow tests. The differences between bed expansions are within experimental error. The results indicate that a correlation developed from experimental data should closely model H-Coal PDU reactor bed expansions.

MODEL DEVELOPMENT

The objective of the model development is the description of the hydrodynamics of the H-Coal reactor. It is desired to obtain bed height and volume fraction occupied by gas, liquid slurry, and catalyst as a function of gas and liquid superficial velocity. Although it is not known with certainty that a model developed from cold flow studies will be applicable for the H-Coal reactor at actual operating conditions, the agreement of catalyst bed expansions at equivalent operating conditions suggests that the coal char/kerosene slurries are suitable model fluids. Additional recommendations for model validation will be made with the conclusions.

Since from the data analysis reported in the previous section it becomes evident that gas flow dynamics control the holdup of the various phases and the gas mixing inside the reactor, Bhatia and Epstein's approach will be used to take into account the changing bubble diameter with viscosity and operating conditions. Subsequent to the development of the model for predicting volume holdups, a gas mixing model will be presented.

Three-Phase Holdup Model

In developing this model it will be assumed that fines are distributed uniformly throughout the liquid phase. The ebullated bed is then assumed to be composed of three phases: gas, wake, and particulate. Following previous investigators (5,6,16), the assumption will be made that the particulate phase is composed of liquid and catalyst only, and that bubble wakes contain liquid and solids. Crucial parameters of the model are: bubble rise velocity (U_{t_B}), relative solids holdup in wake (X_k), and the wake volume behind the bubbles.

Following Henriksen and Ostergaard (48), it is assumed that bubbles in a three-phase fluidized bed behave qualitatively in the same manner as bubbles in pure liquids. The included angle θ of spherical cap bubbles depends on the viscosity of the fluidized bed as shown in Figure 28 (same as Figure 3 in Reference 48). As the bed expands, the apparent viscosity decreases, and this will result in a decrease in the bubble-included angle. Because the wake volume is that which completes the sphere defined by the bubble cap (49), the volume of the bubble wake increases with bed expansion.

The wake volume ratio K ($=\epsilon_k/\epsilon_g$) was incorporated into the model following El-Temtamy and Epstein's (38) correlation:

$$K = K_o \exp(-5.08\ \epsilon_g) \qquad (17)$$

where $K_o = \epsilon_k/\epsilon_g$ for a single bubble in a fluidized bed.

In deriving the parameters U_{t_B}, K_o, X_k for the model, the experimental phase holdups were matched with model predictions by standard non-linear optimization techniques. The following procedure was adopted:

1) Match experimental runs with U_{t_B} (the bubble rise velocity) and K_o as the independent variables with an optimization technique.

2) Correlate K_o and recalculate U_{t_B} values.

3) Correlate U_{t_B}.

4) Using the correlated U_{t_B}, K_o, Equations 16 and 17, and the mathematics of Table XXIII, predict phase holdups.

5) Compare predictions with experimental data. If substantial deviations exist, calculate X_k and make predictions using correlated U_{t_B}, K_o, and X_k.

Correlation of Wake Volume Ratio, K_o.--As discussed previously, the wake volume increases as the bed viscosity is reduced. It therefore follows that high bed expansions will result in an increased wake volume. Because with coal char/kerosene slurries bed expansion is mainly a function of liquid velocity, the calculated ratio K_o was correlated with liquid velocity. Figure 29 indicates that with kerosene and zero fines the wake volume shows little change with liquid velocity. In this case the gas flows in uniform, small bubbles and wake effects become unimportant. When coal fines are added to the system or when the viscosity of the liquid increases, Figures 29 and 30 indicate that bubble wake volume increases with liquid velocity. In this case bubble coalescence is predominant, the bubble size increases significantly, and wake effects are important. The correlations describing K_o as a function of U_l are given in Appendix G.

Correlation of Bubble Terminal Velocity, U_{t_B}.--It has been reported previously (50) that liquid rate has little effect on the rise velocity of bubbles in the moving liquid phase. For a given fluid system, it has been found that gas velocity is the major operating variable in determining bubble size and velocity. However, from the drift flux analysis shown in Figures 25 and 26, it was found that the transition from ideal bubbly to churn turbulent is delayed as the liquid velocity increases. For this reason, the bubble terminal velocity was correlated with $U_g - U_l$, as shown in Figures 31 and 32. Liquid kerosene alone results in low bubble terminal velocities. This is consistent with the drift flux analysis illustrated in Figure 25, which indicates that nitrogen/kerosene

fluidization of HDS-2A catalyst involves gas flow in the ideal bubbly regime. On the other hand, Figure 31 indicates that with coal fines the bubble terminal velocity increases very rapidly as $U_g - U_l$ increases, again consistent with the drift flux analysis. A comparison of calculated bubble terminal velocities with coal char kerosene slurries and mineral oil is shown in Figure 32. The calculated bubble terminal velocities for mineral oil are lower than for the coal char/kerosene slurry. It is uncertain what causes this difference. As shown in Table VII, the viscosity of mineral oil at 175°F is 4.2 cp. The viscosity of coal char/kerosene slurries at 80°F is about 8 cp according to data reported in Table IX. The authors of these reports do not have great confidence in these viscosity measurements because of possible settling of coal fines. The following explanations are therefore likely:

1) If it is assumed that the viscosity of the coal char/kerosene slurries is indeed twice as high as that of mineral oil, the higher bubble terminal velocity is consistent with previous findings (50), indicating that higher-viscosity liquids lead to increased bubble coalescence.

2) If, however, the viscosity of the coal char/kerosene slurry is assumed to be equal to that of mineral oil at 175°F (based on capillary viscometer measurements and matching bed expansions), the increased bubble coalescence with coal fines may be due to promotion of rapid draining of the liquid film between two adjacent bubbles.

The work in determining the viscosity of coal char/kerosene slurries will continue in the future. In the meantime, correlations have been derived to describe the functional relationship of U_{t_B} with $U_g - U_l$. These are also reported in Appendix G.

Correlation of Solids Holdup, X_k.--The application of Equation 16 along with the previously described correlations for K_o and U_{t_B} could be used to predict the holdup of all phases in three-phase systems discussed in this work. It was found, however, that in certain cases the solution for ϵ_g, ϵ_l, ϵ_c did not converge to realistic values. This failure resulted from the functional dependence of X_k on the slip velocity, which in turn depends on the gas and liquid holdups. It was therefore necessary to correlate the experimental values of X_k with bubble terminal velocity as shown in Figure 33. No correlation is shown for liquid kerosene because in this case the solids holdup ratio was found to be equal to zero. For all other cases it is seen that the solids holdup ratio X_k is described by a correlation of the following form:

$$X_k = \frac{a}{U_{t_B} + b} \qquad (18)$$

Coefficient values are given in Appendix G.

The structure of Equation 18 was chosen to be consistent with that reported by others (38). As the viscosity of the liquid or slurry phase increases,

X_k increases as the bubble terminal velocity becomes higher. With liquid kerosene alone, X_k is negligible because the terminal velocity of small bubbles is very low.

Model Predictions.--Figures 34 and 35 show a comparison of experimental data with model predictions. In general, the agreement is good and it demonstrates that the application of this model for predicting holdups of the phases gives realistic values. The flow diagrams and Fortran coding of this predictive program appear in Appendix E. Further work is planned in the future to establish the application of this model for predicting the gas and catalyst holdup in the PDU H-Coal reactor.

Gas Mixing Model

Gas flow in gas/liquid systems has been shown (42,52) to be generally characterized by an upflow of gas bubbles in the center region of the bed and a downflow of gas bubbles near the wall region. An extension of these findings to the systems studied under this work could be used to describe the various degrees of gas mixing which are implied by the tracer data presented earlier. For this reason, a circulation model consisting of n and m completely mixed tanks is proposed. This is shown schematically in Figure 36, where n and m tanks (CSTR's) represent the gas mixing characteristics in the upflow and downflow regions, respectively. This model is in agreement with visual observation on the cold flow unit (small gas bubbles may be seen flowing down along the outer walls) and the literature (42). A general method for calculation of residence time distribution in such recycle systems has been published recently (53). A similar model was independently developed during the summer of 1979 as part of this project. This model is documented in Appendix F.

Figure 37 shows the application of this model to the residence time data for one tracer test with kerosene/no coal char. The definitions of $E(\theta)$ (the exit age distribution at the top of the catalyst bed) and θ (dimensionless time) may be found in Levenspiel (54).

The model parameters, found by trial and error, illustrate that the gas flow up through the catalyst bed is intermediate between plug flow and backmix. The 10% downflow through one CSTR illustrates the magnitude of the recycle flow. The volume ratio of 0.875 indicates that all tanks are of equal volume.

Model parameters for the 15.5 vol% coal char/kerosene slurries appear in Appendix F. This tracer work indicates the flow regimes in the ebullated bed are complex, and that a proper description of this flow is necessary to predict gas holdup and residence times.

CONCLUSIONS AND RECOMMENDATIONS

This report presents the results of work aimed at understanding the hydrodynamic behavior of the H-Coal reactor. A summary of the literature search related to the fluid dynamic behavior of gas/liquid/solid systems was presented. Design details of a cold flow unit were discussed. The process design of this cold flow model followed practices established by HRI in their process development unit.

The cold flow unit has been used to conduct experiments with nitrogen, kerosene, or kerosene/coal char slurries, and HDS catalyst, which at room temperature have properties similar to those existing in the H-Coal reactor. Mineral oil, a high-viscosity liquid, was also used. The volume fractions occupied by gas/liquid slurries and catalyst particles were determined by several experimental techniques. The use of a mini computer for data collection and calculation has greatly accelerated the analysis and reporting of data. Data on nitrogen/kerosene/HDS catalyst and coal char fines were presented in this paper.

Correlations identified in the literature search were utilized to analyze the data. From this analysis it became evident that the Richardson-Zaki correlation describes the effect of slurry flow rate on catalyst expansion.

Three-phase fluidization data were analyzed with two models. Through the use of a correlation developed by Darton and Harrison, the two bubble flow regimes were identified: ideal bubbly and churn turbulent. The effect of viscosity on the flow transition between regimes was described. Increased viscosity due to coal char addition in kerosene or use of a viscous fluid enhances bubble coalescence and the transition to churn turbulent flow. Although the Darton-Harrison model was useful in identifying flow regimes, it could not provide quantitative correlations (drift flux vs. gas holdup) because most of the data with viscous fluids fall in the churn turbulent regime. For this reason, the Bhatia and Epstein model was used to derive parameters describing the experimental data from this work. It was shown that the bed could be described by the gas bubbles, wakes following the bubbles, and the liquid/solid particulate phase. Correlations were derived describing bubble rise velocity, wake volume, and solids holdup in the wake as a function of operating parameters. Predictions with this model appear realistic.

One of the significant findings in this work is the observation by Battelle that H-Coal fluids are non-Newtonian. This should be further explored with additional measurements of H-Coal liquids.

It is recommended that an experimental program be carried out in the PDU reactor in which the ebullated bed height will be externally monitored with changes in operating conditions at low, medium, and high viscosities. These viscosities will be achieved via different slurry recycle rates. Additional studies at ambient temperatures are also needed to establish the bubble size along with direct measurements of the bubble rise velocity.

Use of optically clear solid/liquid systems will permit the use of laser techniques for this purpose. Finally, since this study established that coal char is uniformly distributed in the liquid phase, additional liquid tracer tests will be useful to derive mixing parameters for the liquid phase.

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NOMENCLATURE

		Typical Units
A	Cross-sectional area of column	cm^2
C_D	Drag coefficient for single particle or bubble	--
C_{DM}	Drag coefficient for multi-particle system	--
d	Particle diameter	cm
$\bar{d}$	Average particle diameter	cm
d_e	Equivalent spherical diameter of bubble	cm
d_p	Diameter of a circle if the same area as the projected particle when lying in its most stable position	cm
d_s	Diameter of a sphere with the same volume as the particle	cm
d_{sm}	Sauter-mean bubble diameter	cm
D	Bed or tube diameter	cm
D_c	Diameter of capillary	cm
E_g	Dispersion coefficient	sec/cm^2
f	Frequency of formation of bubble cluster	sec^{-1}
Fr	Froude number	--
g	Acceleration of gravity	cm/sec^2
G	Dimensionless group	--
H_{GL}	Three-phase bed height	cm
H_L	Liquid/solid bed height	cm
j_l	Liquid flux	gm/cm^2 sec
j_l^*	Dimensionless liquid flux	--
$\bar{k}$	Average of wake volume to bubble volume ratio	--
K	Shape factor	--
K'	Effective hydrodynamic volume of particle	cm^3
K_o	Single bubble wake volume to bubble volume ratio	--
l	Particle length	cm
M	Morton number	--
M'	Rheological parameter	gm/cm sec

		Typical Units
n	Richardson-Zaki index or exponent in ideal bubbly flow regime models	--
n'	Rheological parameter	--
P_e	Peclet number	
Q_g	Gas volumetric flow rate	cm^3/sec
r	Particle or bubble radius	cm
r^*	Dimensionless particle radius	--
r_e	Equivalent spherical bubble radius	cm
r_o	Orifice radius	cm
R	Radius of curvature	cm
Re	Particle Reynolds number	--
Re_b	Bubble Reynolds number	--
Re_f	Minimum fluidization particle Reynolds number	--
Re_m	Reynolds number for multi-particle system	--
Re_t	Particle Reynolds number based on U_t	--
U_b	Bubble rise velocity	cm/sec
U_g	Superficial gas velocity	cm/sec
U_l	Superficial liquid velocity	cm/sec
U_m	Mean velocity	cm/sec
U_r	Relative velocity between particles and liquid	cm/sec
U_s	Gas/liquid slip velocity	cm/sec
U_t	Terminal velocity of an isolated particle or bubblr	cm/sec
U_{tb}	Bubble terminal velocity	cm/sec
U_{lo}	Superficial liquid velocity at incipient fluidization	cm/sec
U_{sl}	Velocity of gas slug	cm/sec
U_l'	Superficial liquid velocity in the particulate fluidized phase in a three-phase system	cm/sec
v_b	Bubble volume	cm^3

		Typical Units
V_g	Gas linear velocity	cm/sec
V_{CD}	Gas drift flux	cm/sec
We	Weber number	--
X_k	Ratio of solids holdup in wake to solids holdup in particulate phase	--

Greek

β	Number of small bubbles forming a cluster	--
ϵ	Bed voidage	--
ϵ_g	Volume fraction of gas	--
ϵ_l	Volume fraction of liquid	--
ϵ_s, ϵ_c	Volume fraction of solids (catalyst particles)	--
ϵ_w	Volume fraction of wake phase	--
δ	Pore diameter of gas distributor	cm
ρ_g	Gas density	gm/cm^3
ρ_s	Density of particles	gm/cm^3
ρ_l	Density of liquid	gm/cm^3
ρ_b	Density of fluidized bed	gm/cm^3
τ_w	Wall shear stress	$dynes/cm^2$
μ	First moment	sec
μ_e	Effective viscosity	poise
μ_l	Liquid viscosity	poise
σ	Surface tension	dynes/cm
σ^2	Second moment	sec^2
λ	Wavelength of disturbance	cm
$\emptyset$	Solids volume fraction	--

Subscripts

k	Wake phase
f	Liquid fluidized phase (particulate phase)
l	Liquid

Nomenclature

Subscripts (Continued)

g	Gas
c	Catalyst

Superscripts

11	Two-phase region
111	Three-phase region