In addition to the viscosities of the pure liquids reported in Table VI, an effort was made to measure the viscosities of kerosene/coal char slurries. A capillary tube viscometer having an ID of 1.6 mm and a length of 100 cm was then constructed to measure the viscosity of the coal char slurries. The viscometer was connected to the unit as shown in Figure 10. The viscosities of 11.5 and 15 vol% coal char/kerosene slurries were determined to be 3.5 cp at 22°C and 3.7 cp at 25°C, respectively.

The particle size distribution of the catalyst and coal char is also important in determining the hydrodynamic properties of the H-Coal reactor. The cumulative size distribution of the coal char particles which was used to make slurries under this study was measured by IIT Research Institute using an optical microscope interfaced with a Quantimet 720 computerized image analyzer. The distribution is reported in Table VII. The geometric mean particle diameter is 3.4 microns. As shown in Table VIII, these fines supplied by HRI have density and particle size distribution similar to the fines found in the recycle stream of the H-Coal reactor (35).

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 IX.

Unit Data

Detailed experimental test conditions for the runs included in this paper are reported in References 34 and 36. A summary is shown below:

Run No.	Test No.	Temp, °C	Fines, Vol%	Gas	Range of Slurry Flow Velocities, cm/sec	Range of Gas Flow Velocities, cm/sec
201	9-44	24	0	N_2	1.5-6.0	0-7.6
203	1-11	24	1	N_2	1.5-6.0	0-6.1
204	1-28	24	5	N_2	1.5-5.5	0-4.6
205	1-34	24	10	N_2	1.5-6.0	0-7.6
205	37-64	24	11.5	N_2	1.5-6.0	0-7.6
206	1-56	24	15.0	N_2	2.1-6.0	0-8.8
206	57-74	24	16.5	N_2	2.6-4.6	0-7.6
208	1-31	65	16.5	N_2	2.6-6.0	0-7.6
209	1-30	38	16.5	N_2	2.6-6.0	0-7.6
212	1-15	24	0	N_2	2.6-6.0	0-7.6

For each test, a given amount of catalyst was loaded to the reactor filled with kerosene. The catalyst particles were allowed to soak with kerosene for a period of over 24 hours. 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 11. From plots of this type, a bed height was established for each test. Based on this bed height, the initial catalyst charge, the delta P 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 calculational procedures to establish the holdup of each phase are reported in Appendix A.

RESULTS AND DISCUSSION

The objective of this section is to present some preliminary data to describe the effect of coal char concentration, temperature, gas and slurry velocity on bed behavior. Based on these data, the validity of existing models described in the literature section will be tested. Finally, an attempt will be made to compare the results of this study with actual data from the HRI PDU. 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, 10, 11.5, 15, and 16.5 vol% coal char/kerosene slurries are given in Table X. 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 206-(2). These results are given in Table XI. This analysis was performed to determine if there was 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 206-2 and 206-67 in Table X 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 X 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).

Slurry Fluidization

The effect of coal fines slurry concentration on bed expansion at zero gas flow is shown in Figure 12. Increasing the slurry concentration from 0 to 16.5 vol% (viscosity increase from 1.4 cp to ~ 3.7 cp at 24°C) increased bed expansion by 20 to 30% over the range of slurry velocities studied. The effect of liquid temperature on bed expansion at a slurry concentration of 16.5 vol% is illustrated in Figure 13. As can be seen, bed expansion declines with increasing temperature.

Figures 12 and 13 suggest that bed expansion data can be correlated via a Richardson-Zaki format (Equation 1). 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 XII. The results indicate that the index changed from 2.9 to approximately 3.5 as the slurry concentration increased from 0 to 16.5 vol%. (The 4.8 reported for the 16.5 vol% slurry at room temperatures appears to be incorrect.) The values are within experimental error of each other. This is expected because

the index is insensitive to changes in viscosity in this Reynolds number range. For example, according to the correlation for n given in Table I ($1 < Re < 200$), to obtain a 10% change in n, the viscosity must change by a factor of three.

Whereas there is no significant change in n, the directional changes are correct. In general, the index increases with increasing viscosity as expected. Future tests with a much higher viscosity liquid, mineral oil, will be used to evaluate the effect of viscosity on n.

Three-Phase Fluidization

Three-phase fluidization data for both kerosene and kerosene slurries are presented in Figures 15 and 16. It has been previously discussed that the addition of gas in a liquid fluidized bed will generally result in an increased bed expansion. Figure 15 indicates that although this is usually true for the nitrogen/kerosene/catalyst system, bed expansion is in fact reduced in some cases when 16.5 vol% coal fines are present. The effect of temperature on bed expansion with the 16.5 vol% coal char slurry system is shown in Figure 16. Figures 15 and 16 illustrate the conditions under which bed contraction occurs. As Ostergaard has reported previously (30), the bed contraction is a function of the relative liquid/gas flows and the viscosity of the particulate phase. Bed contraction occurs as a result of liquid traveling through the reactor in gas bubble wakes. This liquid passes through the reactor quickly at the gas linear velocity. The rest of the bed is depleted of liquid, causing the catalyst bed to contract.

The data in Figure 15 and 16, replotted in terms of catalyst volume fraction versus gas velocity with liquid velocity as a parameter, are

shown in Figures 17 and 18. The same effect of liquid viscosity on bed contraction can also be seen in these figures. The increased viscosity results in greater bed expansion. The results also indicate that gas bubbles are smaller with smaller liquid wakes at lower viscosity because higher gas velocities are required to cause bed contraction for the high-temperature tests.

Two models discussed in the literature review section were evaluated for correlation of the three-phase fluidization data (liquid and slurry). The data were first analyzed with the Darton-Harrison drift flux method given in Equation 9. As discussed in a previous section, if drift flux is plotted versus gas holdup, lines defining two flow regimes can be discerned: ideal bubbly and churn turbulent.

This type of plot for the nitrogen/kerosene/catalyst system is shown in Figure 19. At all flow rates, the data fall in the ideal bubbly flow regime. The effect of 16.5 vol% coal fines on the reactor flow regime can be seen by comparing Figures 19 and 20. Coal fines enhance the transition from ideal bubbly to churn turbulent flow by increasing the slurry viscosity. As reported by Ostergaard (17) and Massimila (18), higher viscosity favors bubble coalescence, causing the flow transition to churn turbulent. Also shown in Figure 20 is the effect of increased liquid flow rate on inhibiting the ideal bubbly to churn turbulent transition.

The effect of temperature on the flow transition with the 16.5 vol% coal char slurry system is shown in Figure 21. Lower temperature increases the slurry viscosity, enhancing the transition to churn turbulent flow. The drift flux plots shown in Figures 19, 20, and 21 give a qualitative description of the effects of gas and liquid flow rates and fluid

properties on reactor flow conditions. However, because a large amount of the data lie in the transition region, it is difficult to use this model as a quantitative tool in correlating holdups for three-phase systems. For this reason, the far more complex correlation of Bhatia and Epstein (33) was tested as a model for three-phase data.

The iterative method used to solve the Bhatia-Epstein equations given in Table V is shown in Table XIII. 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 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):

$$X_k = 1 - 0.877 \frac{V_l}{U_s} \qquad (12)$$

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 and Epstein model for tests with 0 vol% fines and 16.5 vol% fines are shown in Figures 22 and 23. Agreement between calculated and experimental holdups is generally good except when bed contraction occurred for the 16.5 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 16.5 vol% coal fines, X_k was found to be equal to about 0.8. For this case, best agreement with the model resulted in terminal bubble velocities ten to twenty times 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 are under way to establish whether these phenomena are actually taking place.

COMPARISON OF EXPERIMENT RESULTS WITH HRI PDU DATA

The most important criterion for the correlation developed from these experimental data is to model operating conditions of an H-Coal reactor. Therefore, data used to develop this correlation must also be consistent with reactor operating conditions.

A comparison of bed expansion in the HRI PDU with experimental bed expansion is shown in Figure 24. Results from PDU Run 7 (40), indicated by triangles, are compared with tests with 16.5 vol% coal char in kerosene at 24°C and 65°C. Reactor slurry velocity was 2.6 cm/sec in all cases.

PDU bed expansions are best predicted by tests at 24°C with 16.5 vol% coal char in kerosene. The differences between bed expansions are within experimental error. The results indicate that a correlation developed from experimental data should closely model H-Coal reactor bed expansions.

CONCLUSIONS

This paper reports 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 the conditions in the H-Coal reactor. 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 directionally 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 existence of two bubble flow regimes was identified: ideal bubbly and churn turbulent. The effect of viscosity on this flow transition was described. Increased viscosity due to coal char addition or decreased temperature enhances bubble coalescence and the transition to churn turbulent flow. Results of an initial analysis of three-phase data with the Bhatia-Epstein model have been encouraging. However, neither model is currently totally satisfactory. The Darton-Harrison model is useful in identifying flow regimes, but cannot be used quantitatively for the range of operating conditions in this paper. To use the Bhatia-Epstein model, a better correlation for calculation of the ratio of solids in the wake to solids in the particulate phase and more detailed information on bubble behavior are required. Future work should emphasize these two areas.

A comparison of bed expansion from the nitrogen/kerosene/HDS catalyst system with similar data obtained from the HRI PDU at similar gas and slurry flows indicates excellent agreement. It is therefore hoped that understanding the fluid dynamics behavior of the systems studied at ambient temperature will provide information useful in the H-Coal process.

ACKNOWLEDGMENTS

This work is conducted under a contract from the Department of Energy (Contract EF-77-C-01-2588). The permission to publish this work is gratefully acknowledged. The authors also appreciate the contribution provided by Hydrocarbon Research, Incorporated. Several people in the Amoco Research Center also have assisted directly or indirectly in this work: C. C. Wallin with the literature search; S. E. Shields and A. P. Vander Klay in systems design; T. D. Evans and A. W. Larson in

mechanical design; J. W. Jones and G. D. Cera in computer services; C. F. Washington and C. J. Fuhs for assistance in unit operation; and J. W. Gorman in data analysis.

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NOMENCLATURE

		Typical Units
A	Cross-sectional area of column	cm^2
C_D	Drag coefficient for single particle or bubble	--
$C_{D\,M}$	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
f	Frequency of formation of bubble cluster	sec^{-1}
Fr	Froude number	--
g	Acceleration of gravity	cm/sec^2
G	Dimensionless group	--
$H_{G\,L}$	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
l	Particle length	cm
M	Morton number	--
M'	Rheological parameter	$gm/cm\ sec^{n'-2}$

		Typical Units
n	Richardson-Zaki index or exponent in ideal bubbly flow regime models	--
n'	Rheological parameter	--
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	--
Utb	Bubble terminal velocity	cm/sec
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 bubble	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	--
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_e	Effective viscosity	poise
μ_l	Liquid viscosity	poise
σ	Surface tension	dynes/cm
λ	Wavelength of disturbance	cm
ϕ	Solids volume fraction	--

Subscripts

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

		Typical Units
g	Gas	
c	Catalyst	

Superscripts

''	Two-phase region	
'''	Three-phase region	