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A COLD FLOW MODEL FOR THE H-COAL REACTOR
PART II: DATA CORRELATIONS

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Experiments at ambient temperatures with extrudates of desulfurization catalyst fluidized with mixtures of nitrogen and coal char/kerosene slurries are used to simulate the fluid dynamics of the H-Coal reactor at actual operating conditions. Data from these studies presented in Part I are used to establish the validity of a model giving the volume fraction occupied by the gas, the slurry, and the catalyst phase. The importance of bubble terminal velocity and solids holdup in the gas wake volume in explaining bed contraction is discussed.

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SCOPE

In the H-Coal reactor, a fluidized bed is used to provide sufficient mixing between the slurried coal, hydrogen, and catalyst. A model to predict the holdups of the various phases in the reactor is necessary both for the design of equipment and for efficient operation. As discussed in Part I, experiments have been performed in a 15 cm ID cold flow pilot plant unit to measure catalyst bed expansions and phase holdups using model fluids. In some cases, small (approximately 3 μ m) coal char particles were slurried into a liquid phase to simulate the unreacted coal ash present in an actual reactor.

The data thus obtained provide the basis for testing and/or modifying existing models describing three-phase fluidized systems. Although previous investigators have reported experiments for air/water systems, they have developed empirical or semi-theoretical models for correlating their data. Ostergaard's (1965) work resulted in a model describing the effect of operating conditions on bed expansion. His model, although useful for the systems studied, cannot predict bed contraction because of the assumptions made. Bhatia and Epstein (1974) extended the previous model to allow prediction of bed contraction by allowing the solids concentration in the wake phase to vary. Darton and Harrison's (1975) approach emphasizes the importance of gas flow behavior in bed expansion. Using the Wallis (1969) drift flux method, he was able to identify two gas flow regimes (ideal bubbly and churn turbulent). Most investigators agree that bed contraction can occur when the gas flow is in the churn turbulent regime.

CONCLUSIONS AND SIGNIFICANCE

The holdup of the slurry and catalyst phases can be predicted with the application of the Richardson and Zaki (1954) correlation. The parameters used in this correlation were derived as a function of liquid or slurry viscosity.

Three-phase fluidization data were analyzed with two models. Through the use of a correlation developed by Darton and Harrison (1975), 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. The Darton-Harrison (1975) model is useful in identifying flow regimes, but cannot be used quantitatively for the range of operating conditions in this study. Therefore, a modified Bhatia-Epstein model (which uses explicit descriptions of the gas bubble and trailing wake as parameters) was selected as most promising for data analysis.

This model has been applied to data for various fluids, coal char concentrations, and temperatures. Model parameters include the gas bubble terminal velocity, and the solids concentration in the wake following the bubbles. It is shown that for the nitrogen/kerosene/catalyst system the bubble terminal velocity is small and the solids concentration in the wake

phase is zero. Addition of coal char fines results in a significantly higher bubble terminal velocity due to the increased viscosity of the slurry phase.

The solids concentration in the wake phase is also increased.

This work has established the significance of gas flow on the fluid dynamics of gas/slurry/catalyst systems. It has been shown that as the bubble terminal velocity (bubble size) increases, the gas flow changes from the ideal bubbly to the churn turbulent flow regime, and bed contraction is possible with increased gas velocity.

INTRODUCTION

In Part I of this study, data were presented with three-phase fluidized beds involving coal char/kerosene slurries, nitrogen, and cylindrical catalyst particles (1.8 mm in diameter, 5.1 mm in length. It was shown that the bed expansion obtained with such systems in a cold flow model simulates the bed expansion obtained in a process development unit operated at actual H-Coal operating conditions (700-755K, 13,340 to 21,378 KN/m²). Information presented in Part I is very useful in describing the volume fraction occupied by the gas, the slurry, and the catalyst phase. In Part II, models are developed to describe the fluid dynamics of the system as a function of the gas and liquid superficial velocities and the physical properties of the fluid and solid phase.

PREVIOUS WORK

The criteria which should be fulfilled by a three-phase fluidized model have been spelled out previously by Darton and Harrison (1975). One of these criteria stresses the importance of understanding liquid/solid fluidization. Although there are numerous correlations for predicting the bed height in liquid fluidized systems, a literature search has indicated that the most widely used method of correlating liquid/solid fluidization data is that of Richardson and Zaki (1954). The correlation relates the liquid volume fraction (ϵ_1) due to bed expansion to the ratio of the superficial liquid velocity (U_1) to the terminal velocity of a single particle (U_t):

$$\epsilon_1^n = U_1/U_t \quad (1)$$

For spherical particles:

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

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.

The addition of gas to a liquid fluidized bed increases the complexity of the system considerably. According to Stewart and Davidson (1964), addition of gas can cause bed contraction in some liquid fluidized beds because of the

formation of liquid wakes behind gas bubbles. The wakes move through the bed at the bubble velocity, and this results in a reduction in the interstitial liquid velocity in the rest of the bed, thus causing its contraction.

Although several empirical correlations have been reported in the literature, semi-theoretical models correlating three-phase fluidization data are of greater interest because they take into consideration details of the bed structure such as bubble rise velocity or bubble wake volume.

Ostergaard (1965) has proposed a model based on the assumption that the bed consists of three phases: a liquid fluidized bed (particulate phase), a bubble phase, and a wake phase. It is assumed that the wake phase moves through the bed at the bubble velocity, and the porosity of the wake phase is equal to that of the liquid fluidized bed. Because of the latter assumption, the model cannot predict a bed contraction.

Bhatia and Epstein (1974) extended Ostergaard's model by assuming that the solids concentration in the wake phase can be varied by means of an adjustable parameter, X_k , between zero ($X_k = 0$) and a solids content equal to that in the particulate phase ($X_k = 1$). Another parameter in the model is the bubble rise velocity, U_{tB} .

Darton and Harrison (1975) based their analysis on the drift flux approach, which is used to describe gas/liquid flow regimes (Wallis, p 69, 1969).

Physically the drift flux represents the volumetric flux of gas relative to a

surface moving at the average of gas plus liquid velocity. The drift flux is defined as follows:

$$V_{CD} = U_S \epsilon_g (1 - \epsilon_g) \quad (3)$$

where U_S is the gas/liquid slip velocity defined below:

$$U_S = U_G / \epsilon_g - U_L / (\epsilon_l + \epsilon_f) \quad (4)$$

By combining Equations 3 and 4, the following relationship is obtained:

$$V_{CD} = U_g (1 - \epsilon_g) - U_l \epsilon_g (1 - \epsilon_g) / (\epsilon_l + \epsilon_f) \quad (5)$$

Darton and Harrison (1975) analyzed the fluidization data of Michelsen and Ostergaard (1970) as shown in Figure 1. A plot of V_{CD} versus ϵ_g revealed two flow regimes: the ideal bubbly or bubble disintegration regime, in which the bubble rise as a uniform, steady cloud with little interaction; and the churn turbulent or bubble coalescing regime, which is the 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.

MODEL DEVELOPMENT

In this section, the data presented in Part I are used to test the models

described in the literature section. Modifications to these correlations will also be presented.

Liquid or Slurry Fluidization

For the liquid/solid systems, correlation of the liquid holdup with operating conditions and the physical properties of the system was carried out with the Richardson-Zaki model. Following Equation 1, plotting ϵ_l versus U_l on log-log paper or a plot of $\ln \epsilon_l$ on 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 2, 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 I. 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.

Three-Phase Fluidization

The three-phase fluidization data were first analyzed using the drift flux approach by Darton and Harrison (1975). As indicated in Figure 1, plotting the drift flux V_{CD} versus the 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 3. 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 3 is the effect of increased liquid flow rate on inhibiting the ideal bubbly to churn turbulent transition. Figure 4 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 3 and 4 indicate that a large amount of the data at high viscosity 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.

Bhatia-Epstein Model

Because data obtained in this study fell in both the churn turbulent and ideal bubbly regimes, the Bhatia-Epstein model was judged most appropriate for correlation of results. The major structure of the model will be highlighted. More details were reported by Bhatia and Epstein in 1974 and El-Temtamy and Epstein in 1978.

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 portions, as shown in Figure 5: gas, wake, and particulate. Following previous investigators, 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_{tB}), relative solids holdup in wake (X_k), and the wake volume behind the bubbles.

Following Henriksen and Ostergaard (1974), 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 6. 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 (Grace, 1970), the volume of the bubble wake increases with bed expansion. For a three-dimensional spherical cap bubble, the ratio of wake volume to bubble volume may be already computed, and is also displayed in Figure 6.

For a bed with a non-zero gas volume fraction, the wake volume ratio $K(=\epsilon_k/\epsilon_g)$ is incorporated following El-Temtamy and Epstein's (1978) correlation:

$$K = K_0 \exp(-5.08 \epsilon_g) \quad (6)$$

where $K_0 = \epsilon_k/\epsilon_g$ for a single bubble in a fluidized bed.

The particulate phase is described by the Richardson-Zaki correlation (Equation 1). The parameters n and U_t are derived from separate liquid/solid experimental runs.

The relative wake solids content, x_k , was calculated from an empirical correlation developed by El-Temtamy and Epstein (1978):

$$x_k = 1 - 0.877 U_t / [(U_g/\epsilon_g) - (U_l/\epsilon_l)] \quad (7)$$

In deriving the parameters U_{tB} and K_O , the experimental phase holdups were matched with model predictions by standard non-linear optimization techniques.

RESULTS

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 7 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 7 and 8 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 relative agreement of wake volumes for mineral oil and the coal char slurry is consistent with the corresponding similar bed expansions for these two fluids. Other investigators (Darton and Harrison, 1975; Rigby and Capes, 1970) have noted an increase in wake volume with increasing liquid velocity.

Correlation of Bubble Terminal Velocity, U_{tB}

It has been reported by Kim, et al., in 1977 that liquid rate has little — effect on the relative 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 3 and 4, 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 as $U_g - U_l$, as shown in Figures 9 and 10. Liquid kerosene alone results in low bubble terminal velocities. This is consistent with the drift flux analysis illustrated in Figure 3, which indicates that nitrogen/kerosene fluidization of catalyst particles involves gas flow in the ideal bubbly regime. On the other hand, Figure 9 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. Prior gas radiotracer tests (Vasalos, et al., 1979) also noted higher bubble rise velocities when coal char fines were added. A comparison of calculated bubble terminal velocities with coal char/kerosene slurries and mineral oil is shown in Figure 10.

The calculated bubble terminal velocities for mineral oil are lower than for the coal char/kerosene slurry. It is uncertain what causes this difference. The presence of the coal fines may promote rapid draining of the liquid film to adjacent bubbles, and thus accelerate coalescence of the gas. Alternatively, the viscosity of the coal char slurry may differ from that

of the mineral oil. Measurement by a variety of techniques has provided viscosities in the range of 0.004 to 0.008 Ns/m².

Correlation of Solids Holdup, X_k

The application of Equation 7 along with the previously described correlations for K_O and U_{tB} could be used to predict the holdup of all phases in three-phase systems discussed in this work. It was found, however, that at certain high velocities the solution for ϵ_g , ϵ_l , and 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 11. No correlation was necessary for liquid kerosene. For all other cases it is seen that the solids holdup ratio X_k is described by a correlation of the following form:

$$X_k = a/(U_{tB} + b) \quad (8)$$

Within experimental error, the curves for all three systems are identical.

The structure of this equation was chosen to be consistent with that reported by others (El-Temtamy and Epstein, 1974). 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

the ability of the Bhatia-Epstein model to predict holdups for a variety of operating conditions was then tested. Figures 12 and 13 show a comparison of holdups predicted using these correlations and the experimental data. In general the agreement is good, and it demonstrates that the application of this model for predicting holdups of the phases gives realistic values. 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.

CONCLUSIONS

A Bhatia-Epstein model has been applied to analyze data from a three-phase gas/solid/liquid slurry reactor. The experiments were conducted using kerosene/coal char slurries as a fluid to model the H-Coal reactor, and pure mineral oil and kerosene for comparison.

Model parameters were obtained over a range of gas and liquid velocities. The magnitude of these parameters and their variation with operating conditions are consistent with prior literature.

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NOMENCLATURE

a	Constant in X_k correlation	cm/sec
b	Constant in X_k correlation	cm/sec
d	Particle diameter	cm
D	Diameter of cylindrical vessel	cm
n	Exponent in Richardson-Zaki correlation	--
K	Wake volume/bubble volume in a fluidized bed	--
K_0	Wake volume/bubble volume for a single bubble	--
U_g	Gas superficial velocity	cm/sec
U_l	Liquid superficial velocity	cm/sec
U_t	Catalyst terminal velocity	cm/sec
U_{tB}	Bubble terminal velocity	cm/sec
V_{CD}	Drift flux	cm/sec
X_k	Ratio of solids holdup in bubble wake to solids holdup in particulate phase	--
ϵ_f	Fines volume fraction	--
ϵ_g	Gas volume fraction	--
ϵ_k	Wake volume fraction	--
ϵ_l	Liquid volume fraction	--
θ	Included angle of bubble	degrees

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TABLE I
VARIATION IN RICHARDSON-ZAKI INDEX

<u>Run No.</u>	<u>Test Nos.</u>	<u>Liquid</u>	<u>Fines, Vol%</u>	<u>Temp, K</u>	<u>Index n</u>
201	2-11	Kerosene	0	295	2.68
203	1-4	"	1	293	3.07
204	1-6	"	5.1	294	3.44
205	1-8, 35	"	10.4	292	3.57
206	1-9	"	15.5	302	3.27
208	1-6, 26-28	"	15.5	337	3.42
209	1-4, 24-25	"	15.5	309	3.22
210	1, 7, 13, 19	"	11.9	295	3.58
211	1, 7, 13	"	17.8	300	3.71
212	1, 3, 7, 10	"	0	295	3.58
214	1-11	"	15.5	338	4.98
310	1-5, 6, 13	Kerosene	0	295	2.46
320	1-5	"	0	297	3.84
427	3-6	Mineral Oil	0	311	3.8
428	3-5	"	0	325	3.5
426	4-7	"	0	338	3.4
423	2-4	"	0	352	3.5

Figure 1

**Drift flux vs. gas holdup:
Darton and Harrison (1975)**

Figure 2

Correlation of liquid-solid data

Figure 3

Drift flux--Effect of operating conditions

Figure 4

Drift flux--Effect of liquid viscosity

Figure 5

Bhatia-Epstein model

Figure 6

**Variation of bubble included angle
with liquid viscosity**

Figure 7

Wake volume--Effect of coal fines

Figure 8

Wake volume--Effect of liquid type

Figure 9

Bubble terminal velocity--Effect of coal fines

Figure 10

Bubble terminal velocity--Effect of liquid type

Figure 11

Relative solids holdup--Effect of liquid type

Figure 12

**Catalyst holdup-predicted vs. actual
kerosene and 17.8 vol % coal char**

Figure 13

**Gas holdup-predicted vs. actual
kerosene and 17.8 vol% coal char**

Figure 12

**Catalyst holdup-predicted vs. actual kerosene
and 17.8 vol.% coal char**

Figure 13

**Gas holdup-predicted vs. actual kerosene
and 17.8 vol% coal char**

- Ebullated bed assumes 3 phases

Gas

Wake

Particulate

Assumptions:

- Particulate phase—liquid and catalyst only
- Bubble wake—liquids and solids

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