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Engineering Development of Slurry Bubble Column Reactor (SBCR) Technology

**Quarterly Report
April 1 - June 30, 1997**

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ENGINEERING DEVELOPMENT OF SLURRY BUBBLE COLUMN REACTOR (SBCR) TECHNOLOGY

Quarterly Technical Progress Report No. 9 for the Period 1 April - 30 June 1997

Contract Objectives

The major technical objectives of this program are threefold: 1) to develop the design tools and a fundamental understanding of the fluid dynamics of a slurry bubble column reactor to maximize reactor productivity, 2) to develop the mathematical reactor design models and gain an understanding of the hydrodynamic fundamentals under industrially relevant process conditions, and 3) to develop an understanding of the hydrodynamics and their interaction with the chemistries occurring in the bubble column reactor. Successful completion of these objectives will permit more efficient usage of the reactor column and tighter design criteria, increase overall reactor efficiency, and ensure a design that leads to stable reactor behavior when scaling up to large diameter reactors.

Summary of Progress

Task 2: Technique Development

Computer Tomography (CT) scans of a large-diameter column with internals were completed and analysis was begun. These are the first CT scans in a large bubble-column with internals. Some problems (shadows) were experienced in the reconstructions. Work to understand if this is caused by the large diameter or the internals continues. In spite of the problems, some conclusions could be drawn (see Task 4).

(Washington University in St. Louis)

Bubble size distribution is a useful parameter for both computer modeling and understanding physical processes inside a bubble column. The dynamic gas disengagement technique offers potential as a way of measuring bubble size distribution. Additionally, dynamic gas disengagement can be measured in high-pressure, reacting columns using differential pressure measurements. A reliable correlation for bubble rise velocity is essential to the dynamic gas disengagement technique. A correlation has been developed to calculate the rise velocity of single bubbles in the high-pressure and high-temperature slurry bubble column. Predictions of the correlation are satisfactory.

(The Ohio State University)

Task 3: Model Development

The standard axial dispersion model (ADM) for the liquid phase in a bubble column was revisited, since it was realized that this model could describe the overshoots in tracer concentration that were seen during the LaPorte tracer trials. While the analysis shows that the model can fit the overshoot shape, the model still appears inadequate to model liquid flow. Consistent dispersion coefficients could not be found when the

region close to the injection points (i.e., the region where overshoots exist) was included in the analysis.

(Washington University in St. Louis)

One method of gaining insight into the effect of variables and of various closure methods for numerical modeling is to use the closure method in a simplified model. A one-dimensional gas phase model was used to test the effect of changing mixing length. It was shown that the equations can capture some of the essence of bubble-driven flow as found in bubble columns.

(Washington University in St. Louis)

Task 4: SBCR Experimental Program

CT measurements using Drakeoil in a 44-cm diameter bubble column fitted with internals to simulate a heat exchanger have shown that:

- a. Gas holdup increases with increasing superficial velocity.
- b. Gas holdup increases (slightly) axially up the column.
- c. Gas holdup is higher with internals than without.

Interpretation of the results continues.

(Washington University in St. Louis)

The differential pressure signals during bed collapse processes have been converted to the variation of gas holdup with time. From the variation of gas holdup with time, bubbles are divided into five groups based on bubble size. The bubble rise velocity and initial gas holdup in each group are obtained.

(The Ohio State University)

Washington University in St. Louis Research

The following report from Washington University for the period April - June 1997 is appended below. It contains

1. Objectives for the Second Budget Year
2. Gas Holdup Distribution Measurements by Gamma Ray Tomography (CT) for the Air-Drakeoil System in an 18-in. Column with Internals
3. Axial Dispersion Model for Batch Liquid-Responses with Overshoots
4. One-Dimensional Modeling of Gas-Liquid Flow in Bubble Columns
5. References

The Ohio State University Research

The following report from Ohio State University for the period April-June 1997 is provided below. It contains

Highlights

Dynamic Gas Disengagement (Bed Collapse) Technique

References

INTRINSIC FLOW BEHAVIOR IN A SLURRY BUBBLE COLUMN UNDER HIGH PRESSURE AND HIGH TEMPERATURE CONDITIONS

(Reporting Period: April 1 to June 30, 1997)

Highlights

1. The differential pressure signals during bed collapse processes have been converted to the variation of gas holdup with time. From the variation of gas holdup with time, bubbles are divided into five groups based on bubble size. The bubble rise velocity and initial gas holdup in each group are obtained.
2. A reliable correlation for bubble rise velocity is essential to the dynamic gas disengagement technique. A correlation has been developed to calculate the rise velocity of single bubbles in the high-pressure and high-temperature slurry bubble column. Predictions of the correlation are satisfactory.

Dynamic Gas Disengagement (Bed Collapse) Technique

Dynamic Gas Disengagement (DGD) or bed collapse technique offers a simple way to estimate the bubble size distribution in our high-pressure and high-temperature slurry bubble column. The experimental setup for the bed collapse technique and a typical response were described in the January-March 1997 quarterly report.

The differential pressure signal during a complete DGD process is divided into six stages, as described in the January-March 1997 report. (1) Immediately following the gas shutoff is a sudden jump in the differential pressure signal, which corresponds to the simultaneous escape of large and small bubbles. (2) In the second stage, the increase in the signal value is much more gradual, since only small bubbles escape from the bed. (3) The differential pressure remains at a relatively constant value for about 150 seconds because the particles are still fully suspended by the liquid motion induced by bubbles. The first three stages can be analyzed to evaluate the bubble size and bubble size distribution. In the last quarter, experiments were conducted to obtain differential pressure signals during bed collapse processes at zero liquid velocity and various gas velocities ranging from 5.2 to 32.4 cm/s. Figure 1 shows the differential pressure signals of the first three stages under five gas velocity conditions.

To evaluate the bubble size and bubble size distribution, the differential pressure signal should be converted into the variation of gas holdup with time. It can be seen from Figure 1 that the differential pressure signals in stage 3 have the same value of about 2950 Pa/m over the entire gas velocity range, which implies that the catalyst particles are uniformly distributed in the slurry bubble column under the conditions of this work. Based on this observation, it can be assumed that the particles are uniformly distributed in the

column, and therefore the ratio of liquid holdup to solids holdup is a constant, K , during the first three stages. K can be calculated from the signal at stage 3, i.e., the gas-free liquid-solid suspension. The differential pressure drop at this stage can be related to the gas-free liquid and solids holdups by the following equation:

$$\left(\frac{\Delta P}{\Delta z}\right)_d^0 = (\rho_l \epsilon_l^0 + \rho_s \epsilon_s^0 - \rho_l) g = \epsilon_s^0 (\rho_s - \rho_l) g \quad (1)$$

Solving Eq. (1) gives the solids holdup in the gas-free liquid-solid suspension:

$$\epsilon_s^0 = \frac{\left(\frac{\Delta P}{\Delta z}\right)_d^0}{(\rho_s - \rho_l) g} \quad (2)$$

K can then be written as

$$K = \frac{\epsilon_s^0}{\epsilon_l^0} = \frac{\epsilon_s^0}{1 - \epsilon_s^0} = \frac{(\Delta P / \Delta z)_d^0}{(\rho_s - \rho_l) g - (\Delta P / \Delta z)_d^0} \quad (3)$$

With K known, the differential pressure signal can be related to the variation of gas holdup with time during the dynamic gas disengagement process (stages 1 and 2), based on the following equation:

$$\begin{aligned} \left(\frac{\Delta P}{\Delta z}\right)_d &= (\rho_g \epsilon_g + \rho_l \epsilon_l + \rho_s \epsilon_s - \rho_l) g \\ &= \left\{ \frac{K(\rho_s - \rho_l)}{1 + K} + \left[\frac{(1 + K)\rho_g - (\rho_l + K\rho_s)}{1 + K} \right] \epsilon_g \right\} g \end{aligned} \quad (4)$$

Solving Eq.(4) yields the gas holdup, ϵ_g :

$$\epsilon_g = \frac{(1 + K) \left(\frac{\Delta P}{\Delta z}\right)_d / g - K(\rho_s - \rho_l)}{(1 + K)\rho_g - (\rho_l + K\rho_s)} \quad (5)$$

Figure 2 shows the variations in gas holdup with time through the first three stages at various gas velocities.

The escape of bubbles from the slurry bubble column leads to the gas holdup variation during a bed collapse process. At any moment, bubbles of different sizes emerge simultaneously from the bed surface. Smaller bubbles have lower rise velocities and thus, stay in the bed for a longer period. During the bed collapse process, the slope of an ϵ_g vs.

t curve keeps decreasing because the bubble size inside the bed becomes smaller, as does the volumetric flow rate of bubbles escaping from the bed surface.

Assume that the bubbles are divided into N size groups, i.e., $d_{B,1}, \dots, d_{B,N-1}, d_{B,N}$, $d_{B,N}$ corresponding to the smallest bubbles. The ϵ_g vs. t signal can be approximated with N linear segments, designated as $(t_0, \epsilon_{g,0}), (t_1, \epsilon_{g,1}), \dots, (t_N, \epsilon_{g,N})$, shown in Figure 3. The change in slope implies the depletion of a group of bubbles. The decrease in gas holdup between t_{i-1} and t_i is due to the escape of the bubbles smaller than $d_{B,i}$. Therefore, the rise velocity ($U_{b,i}$) and holdup of bubbles of size $d_{B,i}$ ($\epsilon_{g0,i}$) can be determined by the following equations (Daly *et al.*, 1992):

$$\begin{cases} U_{b,i} = \frac{H}{t_i - t_0} \\ \epsilon_{g0,i} = \frac{-\left(HS_i + \sum_{j=i+1}^N U_{b,j} \epsilon_{g0,j}\right)}{U_{b,i}} \end{cases} \quad i = N, N-1, \dots, 1 \quad (6)$$

where H is the distance between the gas distributor and the top pressure port and S_i is the slope of the i th segment of curve. Note that calculations by Eq. (6) start from the smallest bubbles and end with the largest bubbles. Table 1 shows the results. With the initial gas holdup of each bubble group and the size of the bubbles, the number of bubbles in each group can be expressed as

$$n_{b,i} = \frac{AH\epsilon_{g0,i}}{\frac{\pi}{6} d_{b,i}^3} \quad (7)$$

A reliable and accurate correlation for the bubble rise velocity is the key to evaluating bubble size using the dynamic gas disengagement technique. The bubble rise velocity in a slurry bubble column is different from that of single bubbles, due to the interaction between bubbles and between bubbles and their surrounding medium. Correlations for bubble swarm rise velocity have been proposed in the literature (Peebles and Garber, 1953; Marrucci, 1965; Clift *et al.*, 1978; Abou-el-Hassan, 1983). Most of the correlations were established on the basis of single-bubble rise velocity. Unfortunately, no reliable correlation is yet available to predict the rise velocity of single bubbles under high-pressure conditions. In this quarter, experiments were conducted to develop a correlation for single-bubble rise velocity that could be applicable under high-pressure and high-temperature conditions. The following correlation (Fan and Tsuchiya, 1990) has been extended to such conditions:

$$U_b = \left\{ \left[\frac{Mo^{-1/4}}{K_b} \left(\frac{\Delta p}{\rho_m} \right)^{5/4} d_e'^2 \right]^{-n} + \left[\frac{2c}{d_e'} + \left(\frac{\Delta p}{\rho_m} \right) \frac{d_e'}{2} \right]^{-n/2} \right\}^{-1/n} \quad (8)$$

where

$$U'_b = U_b (\rho_m / \sigma g)^{1/4}, \quad d'_e = d_e (\rho_m g / \sigma)^{1/2} \quad (9)$$

and $Mo = g \Delta \rho \mu_m^4 / (\rho_m^2 \sigma^3)$ with $\Delta \rho = \rho_m - \rho_g$. ρ_m and μ_m are the effective density and viscosity of the liquid-solid medium, respectively. Three empirical constants in Eq. (8), n , c and K_b , reflect the separate factors governing the rate of bubble rise. In the practical use of Eq. (8), n ranges from 0.8 (for contaminated liquids) to 1.6 (for purified liquids); $c = 1.2$ and 1.4 for monocomponent and multicomponent liquids, respectively; and

$$K_b = \max(K_{b0} Mo^{-0.038}, 12) \quad (10)$$

where $K_{b0} = 14.7$ and 10.2 for aqueous solutions and organic solvents/mixtures, respectively. The effective viscosity of liquid-solid suspensions is calculated by the following equation:

$$\frac{\mu_m}{\mu_l} = \exp \left[\frac{K \epsilon_s}{1 - (\epsilon_s / \epsilon_{sc})} \right] \quad (11)$$

with two parameters correlated by

$$K = \{3.1 - 1.4 \tanh[0.3(10 - 10^2 U_t)]\} / \phi \quad (12a)$$

$$\epsilon_{sc} = \{1.3 - 0.1 \tanh[0.5(10 - 10^2 U_t)]\} \epsilon_s \quad (12b)$$

where U_t is the particle terminal velocity in liquid (m/s), ϵ_s is the solids holdup at the incipient fluidization/packed state, and ϕ is the shape factor of the particles. Figure 4 compares the experimental data with the predictions of Eqs. (8)-(12). The particles in the experiments were 210- μ m glass beads. It can be seen that the correlation proposed here can be used under the experimental conditions of this work. Equations (8)-(12) can then be used to convert the bubble rise velocity to bubble size for each group of bubbles.

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Notations

P	Pressure
z	Distance from the distributor
g	Gravitational constant
K	Constant defined in Eq. (3)
H	Bed height
t	Time
t_0	Time of gas shut-off
U_b	Bubble rise velocity
d_b	Bubble size
A	Cross-sectional area of the column
U_t	Particle terminal velocity
Mo	Morton number of liquids
d_e	Volume equivalent diameter of bubbles

Greek

ρ	Density
ε	Holdup
μ	Viscosity
ϕ	Shape factor of particles

Subscripts

g	Gas phase
l	Liquid phase
s	Solid phase
m	Liquid-solid medium

Superscript

0	Gas-free liquid-solid suspension
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Table 1. Bubble Rise Velocities and Holdups of Various Bubbles Groups ($U_g = 32.4$ cm/s)

i	t_i (s)	$\epsilon_{g0}(t_i)$	S_i	U_{bi} (m/s)	$\epsilon_{g0,i}$
0	1.64	0.43			
1	2.05	0.245	-0.451	0.84	0.068
2	2.54	0.105	-0.286	0.38	0.181
3	3.25	0.045	-0.0845	0.21	0.120
4	6.03	0.018	-0.00971	0.079	0.022
5	10.02	0	-0.00451	0.041	0.038

Figure 1. Dynamic Pressure Drop Signals in Bed Collapse Processes at Various Gas Velocities

N₂-Paratherm NF-GB210

FT (1990) correlation with
 $\mu_m = 3.8 \text{ (mPa s)}$

(mPa s)
 15
 16
 17

(mPa s)
 210
 220
 230

(b) 87.5°C

Pressure (MPa)

0.1
 10.4
 17.3

d_e (mm)

Figure 4. Comparison between the predictions and the experimental data, under high pressure conditions (Solids holdups for +, open, and filled symbols are 0, 0.381 and 0.555, respectively; lines: predictions).