Gas Holdup and Solids Dispersion in a Three-Phase Slurry Bubble Column

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Studies with three-phase systems in bubble columns have shown that the effect of solids on gas holdup depends on a variety of factors. Important among these are: liquid properties and particle properties such as density, size and concentration in the slurry, and wettability. Most of these studies have been made with cold flow systems; and the data at high temperatures and pressures, i.e., with systems of potential industrial importance, are rare (Tarmy et al., 1984; Fan, 1989).

Several studies have been conducted on the system with molten wax as the liquid medium at elevated temperatures in bubble columns (e.g., Calderbank et al., 1963; Deckwer et al., 1980; Kuo, 1985; Bukur et al., 1987a,b; Patel et al., 1990), which is important for slurry-phase Fischer-Tropsch synthesis (FTS). Most of these studies were made with two-phase systems, in the absence of solids, and the emphasis was on measurements of holdups and bubble sizes. Deckwer et al. (1980) reported that the addition of solids (5.5 to 16 wt. % of 0–5 μm Al_2O_3) had very little effect on gas holdup.

The objective of this study was to obtain hydrodynamic data at elevated temperatures using a three-phase system of importance to FTS. In particular, the effects of solids type, size and concentration on gas holdup and axial solids dispersion were investigated. Two types of solids used, iron oxide and silica, simulate typical catalysts and supports, respectively, employed in FTS. A wider range of gas velocities (up to 0.12 m/s) than those used by Deckwer et al. (<0.04 m/s) was employed in this study. Additionally, data were obtained from experiments conducted with slurry circulation (continuous mode of operation), whereas all the previous studies with molten waxes were conducted in the batch mode of operation (without liquid flow).

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Experimental Equipment and Methods

Apparatus and procedure

The experiments were conducted in a 0.05 m ID by 3 m tall stainless steel column equipped with a single-hole 2 mm orifice plate distributor. The flow rate of nitrogen gas was monitored and controlled using a Brooks mass flowmeter. The column temperature was controlled using two temperature controllers, one for the bottom half and the other for the top half of the column. A variable-speed, positive displacement gear pump (Pulsafeeder, Model G12) was used to circulate the slurry. Five pressure transducers located along the column height at 0.1, 0.6, 1.2, 1.9 and 2.5 m above the distributor, respectively, were used to obtain differential pressure measurements. The column also had five slurry sampling ports at 0.3, 0.9, 1.6, 2.2 and 2.8 m above the distributor, respectively, which were staggered with respect to the pressure ports.

All experiments were conducted using prepurified nitrogen as the gas. Superficial gas velocities of 0.02, 0.04, 0.06, 0.09 and 0.12 m/s were employed in all runs. Pressure measurements were made three times for every gas velocity, with the first measurement made half an hour after the gas velocity was changed. After the last pressure measurement, slurry samples were withdrawn simultaneously from the column at five different locations using pneumatically actuated ball valves. Following slurry withdrawal, the samples were allowed to cool and solidify before being removed. The solids fraction of the slurry sample was determined using the Archimedean Principle by immersing the sample in acetone and measuring its apparent weight loss. For experiments conducted in the continuous mode of operation, the slurry flow rate was monitored using the calibration chamber.

FT-300 paraffin wax (average molecular weight of 730) from Dura Commodities Co., New York, was employed as the liquid medium in our study. Its physical properties were reported

elsewhere (Bukur et al., 1987b). Two types of solids were used (iron oxide and silica) in two size ranges (0–5 and 20–44 μm), with solids concentrations of 10, 20 and 30 wt. % in the slurry. The solids densities are 2,650 and 5,100 kg/m^3 for silica and iron oxide particles, respectively.

Measured quantities

The measured quantities were the pressure readings from the five pressure transducers and the weight fraction of solids for the slurry samples withdrawn from the five sampling ports. These quantities were used to determine the axial gas holdup profile, axial solids concentration profile, and the average gas holdup.

The gas holdup in section *i-j* (the section between any two pressure transducers) is given by

$$\langle \epsilon_g \rangle_{ij} = 1 - \frac{\Delta P_{ij}/(S_{sl})_{ij}}{\Delta h_{ij}} \quad i = 1 \text{ to } 5, j = i + 1 \tag{1}$$

where

$$(S_{sl})_{ij} = \frac{1/\rho_{\text{water}}}{\langle W_s \rangle_{ij}/\rho_p + (1 - \langle W_s \rangle_{ij})/\rho_l} \tag{2}$$

The average gas holdup for the entire dispersion is estimated using a weighted average of the gas holdups in the individual sections

$$\epsilon_g = \frac{\sum_{i=1}^{5} \langle \epsilon_g \rangle_{ij} \Delta h_{ij}}{\sum_{i=1}^{5} \Delta h_{ij}} \quad j = i + 1 \tag{3}$$

Axial solids dispersion coefficients

The axial solids dispersion coefficients were estimated using the semiinfinite dispersion model (Kato et al., 1972; Smith and Ruether, 1985; O'Dowd et al., 1987). For a batch mode of operation, the solids concentration profile is given by

$$C_s = C_s^B \exp\left[-L\bar{\Phi}_l \frac{U_p}{E_s} x\right] \tag{4}$$

Solid concentration vs. axial position data can be used to obtain the estimates of U_p/E_s and the initial feed concentration, C_s^B by regression analysis. For batch experiments, U_p and E_s are not separable; and to obtain axial dispersion coefficients, one must assume values for the hindered settling velocity of the solids. There are various correlations available in the literature for estimating hindered settling velocities (e.g., Kato et al., 1972; Smith and Ruether, 1985; O'Dowd et al., 1987). The correlations proposed by Kato et al., Smith and Ruether, and O'Dowd et al., are all of the form:

$$U_p = aU_T^b U_g^c \bar{\Phi}_l^d \tag{5}$$

where U_T is the terminal settling velocity of a single particle in an infinite liquid medium and is calculated using Stokes' law. The mean particle diameters were used to calculate U_T (2.5 μm for particles in the range 0-5 μm, and 32 μm for particles in the range 20–44 μm). The constants in Eq. 5 are: (1.33, 0.75, 0.25, 2.5) for Kato et al.; (1.91, 0.8, 0.26, 3.5) for Smith and Ruether; and (1.69, 0.8, 0.23, 1.28) for O'Dowd et al.

Various correlations have been presented in the literature for predicting axial dispersion coefficients directly. The correlation proposed by Kato et al. is:

$$Pe_p = \frac{13Fr_g(1 + 0.009Re_pFr_g^{-0.8})}{1 + 8Fr_g^{0.85}} \tag{6}$$

This correlation includes a correction for the effect of particle size; however, this effect was not important for particles used in our study, and the numerator in Eq. 6 can be reduced to $13Fr_g$ as suggested by Kato et al. The equation proposed by Smith and Ruether is:

$$Pe_p = 9.6\left[\frac{Fr_g^6}{Re_g}\right]^{0.114} + 0.019Re_p^{1.1} \tag{7}$$

and the equation proposed by O'Dowd et al. for an unbaffled bubble column is:

$$Pe_p = 7.7\left[\frac{Fr_g^6}{Re_g}\right]^{0.098} + 0.019Re_p^{1.1} \tag{8}$$

Gas Holdup Results

Effect of liquid circulation velocity

Figure 1 shows the results from experiments conducted without solids. Foam was produced for the batch case ($U_l = 0$) with a gas holdup value of 28% at a superficial gas velocity of 0.04 m/s. However, gas holdups decreased significantly when the experiment was conducted in the continuous mode of operation using a superficial slurry velocity of 0.005 m/s. When the slurry velocity was increased to 0.02 m/s, gas holdup values were slightly lower than those observed at the slurry velocity of 0.005 m/s. The same trend (decrease in holdup with introduction of slurry circulation) was observed in three-phase experiments with either iron-oxide or silica particles.

The above results indicate that for FT-300 wax even a small upward liquid flow (0.005 m/s) lowers gas holdups markedly; however, a further increase in liquid flow has only a marginal

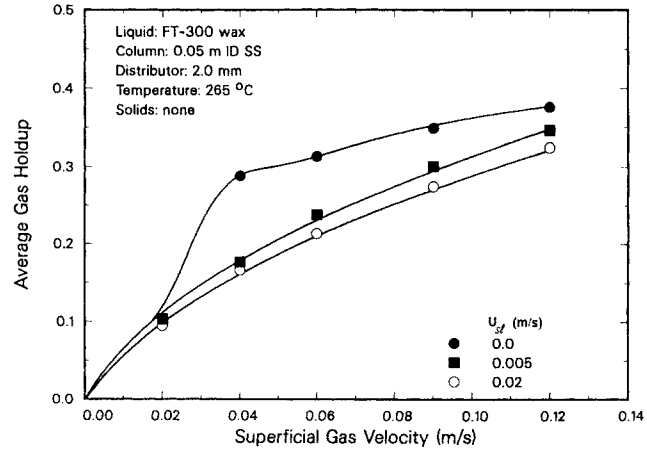

Figure 1. Effect of liquid circulation on average gas holdup in the absence of solids.

effect on the average gas holdup values. The significant differences in holdups between the batch and continuous modes of operation is due to changes in the foaming characteristics of the medium. For the batch case the foam accumulates at the top of the dispersion and increases the gas holdup, whereas in the continuous mode of operation the foam is removed from the column by the circulating slurry.

Results obtained in the continuous mode of operation by other researchers indicate that liquid velocity either has no effect on the average gas holdup (Shah et al., 1982) or decreases holdup slightly (Kara et al., 1982; Kelkar et al., 1984). However, it should be noted that most previous studies were conducted with liquids that do not have the tendency to foam. Shah et al. (1985) studied the effect of liquid velocity on holdup for a foaming system (aqueous ethanol solution). In the absence of liquid velocity, they obtained holdup values as high as 80% at a gas velocity of 0.20 m/s; however, when a liquid velocity of 0.0077 m/s was used, the holdup decreased to about 20%. Results from this study are in qualitative agreement with these findings.

Effect of solids concentration

Figure 2 illustrates the effect of solids concentration on gas holdup for experiments conducted in the batch mode of operation with small iron oxide particles. For superficial gas velocities greater than 0.04 m/s, the average gas holdups in the presence of solids are higher than those obtained without solids. Gas holdups increase as solids concentration is increased from 0 to 10 wt. %. A further increase in gas holdups is observed with the 20 wt. % slurry. For the slurry with 30 wt. % solids, gas holdups decrease relative to the 20 wt. % slurry, but they are still higher than those obtained in the experiment without solids. In experiments with 0–5 μm silica and with 20–44 μm iron oxide particles, the trends were similar to those shown in Figure 2: i.e., the addition of solids increased gas holdup in the batch mode of operation. As mentioned earlier, Deckwer et al. (1980) observed a small reduction in gas holdup values when solids were added to the nitrogen-paraffin wax system ($\Delta\epsilon_g < 0.02$ absolute) for gas velocities less than 0.04 m/s. In our study the addition of solids also had only a small effect on gas holdup values at gas velocities 0.02 and 0.04 m/s.

Figure 3 shows results from experiments conducted with

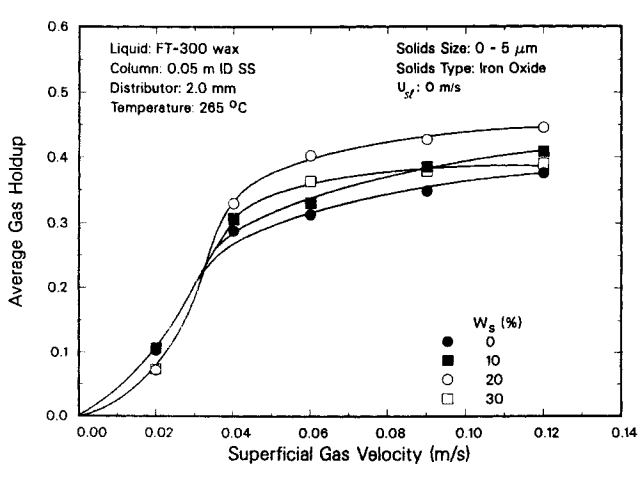

Figure 2. Effect of solids concentration on average gas holdup during batch mode of operation.

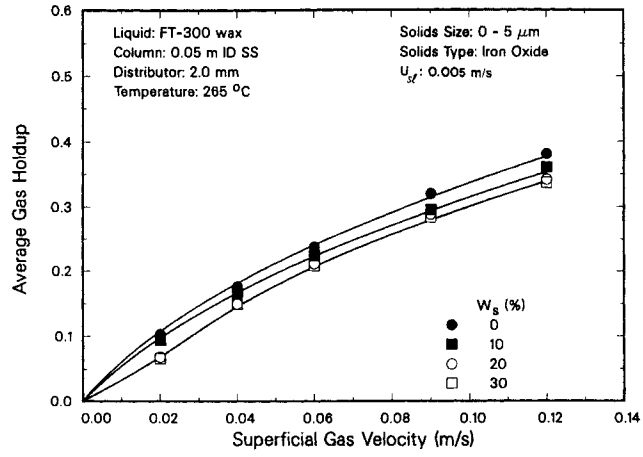

Figure 3. Effect of solids concentration on average gas holdup during continuous mode of operation.

slurries containing different concentrations of 0–5 μm iron oxide particles using a superficial slurry velocity of 0.005 m/s. These results show a small change in holdup values with increasing solids concentration; however, holdup values in the presence of solids are consistently lower than those obtained without solids. The lowest holdup values were obtained with the slurry containing the highest concentration of solids. The same behavior was observed with other solids types and sizes when experiments were conducted in the continuous mode of operation. Also, in this mode of operation there was no significant effect of liquid circulation velocity (0.005 vs. 0.02 m/s) on gas holdup values.

In general, one would expect that the "pseudoviscosity" of the slurry will increase with increasing solids concentration, which in turn promotes the bubble coalescence resulting in an increase in bubble size and a decrease in holdup. However, results from this study and from studies with other systems indicate that the gas holdup either decreases (Kato et al., 1972; Smith and Ruether, 1985) or increases (Pandit and Joshi, 1984; Sada et al., 1986) with addition of solids particles. Pandit and Joshi (1984), in their study on the effect of particle size (d_p = 70–2,000 μm; ρ_p = 2,260–2,500 kg/m^3) in the air-water system, found that holdup increased with either small ($Re_p < 2$) or large particles ($Re_p > 500$), whereas the holdup decreased in experiments with particles having intermediate range of Reynolds number ($2 \le Re_p \le 300$). They reported that in the presence of fine particles ($d_p < 100$ μm) or relatively large particles ($d_p > 850$ μm), the average bubble diameter is smaller than that obtained in the absence of solids. In the intermediate range of particle size, however, larger bubbles are produced as compared to those in the absence of solids. Therefore, the holdup increased in experiments with small and large particles (smaller bubbles and lower bubble rise velocities), whereas it decreased in experiments with intermediate size particles (larger bubbles and higher bubble rise velocities). However, the physical reasons for decrease in bubble size with addition of solids particles are not completely understood. It has been postulated that large particles may cause bubble breakage due to their high kinetic energy (Fan, 1989). In the case of small particles a possible explanation for increase in holdup (i.e., decrease in the average bubble size) with addition of solids particles, may be given in terms of reduced particle wettability (Bhatia et al., 1972; Fan, 1989) and flotation phenomena (Adamson, 1982).

In the batch mode of operation, the iron oxide and silica particles, due to their poor wettability, adhere to the gas bubbles and stabilize them. The small bubbles, characteristic of the molten wax used in this study (Patel et al., 1990), are thus prevented from coalescing and are responsible for the higher holdup values (Figure 2). Furthermore, these bubbles are likely to form a stable layer of foam at the top of the column. When the column is operated in the continuous mode of operation (Figure 3), the decrease in gas holdup with the addition of solids could be attributed to two reasons. First, the circulating liquid entrains any foam (or froth) that forms at the top of the dispersion and prevents the formation of a stable foam layer. Second, based on the discussion by Fan (1989), the liquid circulation is likely to increase the relative velocity between the solids and the liquid, decreasing the contact angle and increasing the wettability of the particles. This would strip off the solids from the bubble surface and disperse them into the liquid. The gas holdups under these conditions would therefore be lower than those for the batch mode of operation.

Axial Solids Distribution

Axial solids concentration profiles for all runs conducted using the 0–5 μm iron oxide and silica particles were uniform in the continuous mode of operation. In the batch experiments with these solids, the profiles showed a slight gradient, with higher concentrations toward the bottom of the column. For experiments conducted in the batch mode of operation with 20–44 μm iron oxide and silica particles, significant gradients in axial solids distribution profiles were observed. However, when liquid circulation was introduced, the profile became uniform and suspension of solids improved significantly. These results indicate that solids suspension improves significantly with the introduction of a small upward liquid flow. This is not completely unexpected since the terminal settling velocity of the largest iron oxide particles (0.002 m/s) is lower than the upward liquid velocity (>0.005 m/s) used in our study.

Figure 4 shows results for axial dispersion coefficients based on the three point (closed symbols) and four point (open symbols) least square fit of the data to the semiinfinite dispersion model for both iron oxide (top) and silica particles (bottom). The dispersion coefficients (E_s) were calculated from experimentally determined U_p/E_s values using Kato et al.'s correlation for the settling velocity (Eq. 5). Values predicted by Eqs. 6–8 are also shown in the figure. For iron oxide particles, the dispersion coefficients obtained using all the data points (four points) for solids concentrations were lower than the dispersion coefficients obtained when the solids concentrations corresponding to the uppermost sample port were omitted (three data points), whereas for silica particles the two sets of results are similar. This is due to the differences in the amount of foam produced, in experiments with the two types of solids. In the absence of foam, the results from our study are described well by the Kato et al. (1972) correlation for the hindered settling velocity and the Smith and Ruether's (1985) correlation for the axial dispersion coefficient.

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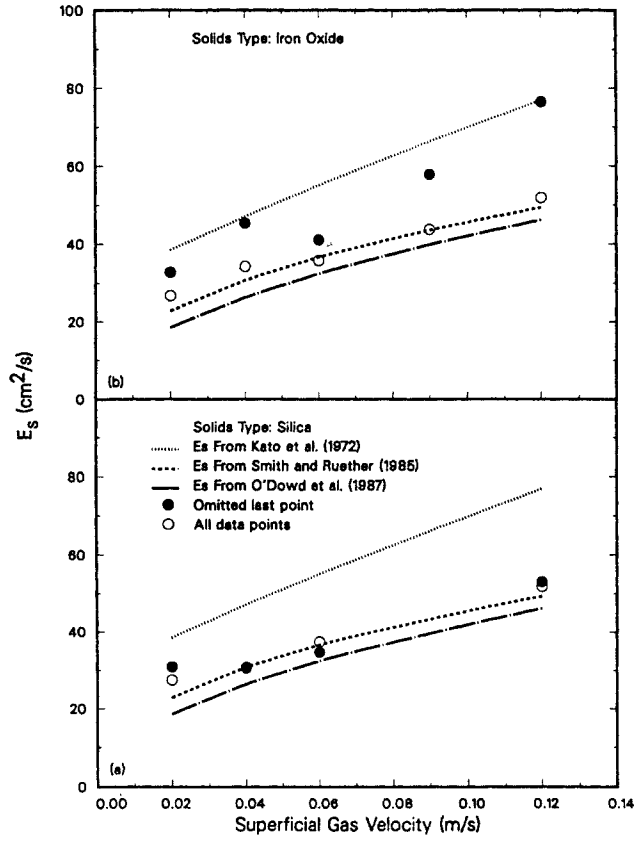

Figure 4. Effect of superficial gas velocity on axial solids dispersion coefficient using Kato et al. correlation for hindered settling velocity (FT-300, 265°C, batch mode, 20 wt. % of 20–44 μm particles).

Notation

a,b,c,d = constants in Eq. 5
C_s = solids concentration, kg/m^3
C_s^B = solids concentration at the bottom of the dispersion, kg/m^3
d_c = column diameter, m
d_p = particle diameter, m
E_s = axial dispersion coefficient, m^2/s (cm^2/s in Figure 4)
Fr_g = Froude number $= U_g/\sqrt{gd_c}$
g = acceleration due to gravity, 9.81 m/s^2
L = expanded bed height, m
Pe_p = particle Peclet number $= (U_g d_p)/E_s$
Re_g = Reynolds number $= (U_g d_c \rho_l)/\mu_l$
Re_p = particle Reynolds number $= (U_T d_p \rho_l)/\mu_l$
S_{sl} = specific gravity of the slurry
U_g = superficial gas velocity, m/s
U_p = hindered settling velocity of particles, m/s
U_T = terminal rise velocity of a single particle in an infinite medium $= [d_p^2 g(\rho_p - \rho_l)]/18\,\mu_l$
$\langle W_s \rangle$ = average weight fraction of solids in the slurry
x = dimensionless height above the distributor

Greek Letters

ϵ_g = gas phase holdup
Δh_{ij} = distance between pressure ports i and j, m
ΔP_{ij} = pressure differential between transducers i and j, m
μ_l = liquid viscosity, N · s/m
ρ_g = gas density, kg/m^3
ρ_l = liquid density, kg/m^3

ρ_p = solids density, kg/m^3
ρ_{water} = water density, kg/m^3
$\bar{\Phi}_l$ = average volume fraction of liquid in the slurry

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