Gas Hold-Up in Bubble Columns: Operation with Concentrated Slurries versus High Viscosity Liquid

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The hydrodynamics of bubble columns with concentrated slurries of paraffin oil (density, ρ_L = 790 kg/m^3; viscosity, μ_L = 0.0029 Pa·s; surface tension, σ = 0.028 N·m^{1}) containing silica particles (mean particle diameter d_p = 38 μm) has been studied in columns of three different diameters, 0.1, 0.19 and 0.38 m. With increasing particle concentration, the total gas hold-up decreases significantly. This decrease is primarily caused by the destruction of the small bubble population. The hold-up of large bubbles is practically independent of the slurry concentration. The measured gas hold-up with the 36% v paraffin oil slurry shows remarkable agreement with the corresponding data obtained with Tellus oil (ρ_L = 862 kg/m^3; μ_L = 0.075 Pa·s; σ = 0.028 N·m^{-1}) as the liquid phase. Dynamic gas disengagement experiments confirm that the gas dispersion in Tellus oil also consists predominantly of large bubbles. The large bubble hold-up is found to decrease significantly with increasing column diameter. A model is developed for estimation of the large bubble gas hold-up by introduction of an wake-acceleration factor into the Davies–Taylor–Collins relation (Collins, 1967), describing the influence of the column diameter on the rise velocity of an isolated spherical cap bubble.

On a étudié dans des colonnes de trois diamètres différents, soient 0,1, 0,19 et 0,38 m, l'hydrodynamique de colonnes à bulles avec des suspensions concentrées d'huile de paraffine (masse volumique, ρ_L = 790 kg/m^3; viscosité, μ_L = 0,0029 Pa·s; tension de surface, σ = 0,028 N·m^{-1}) contenant des particules de silice (diamètre moyen des particules d_p = 38 μm). Lorsque la concentration de particules augmente, la rétention de gaz totale diminue considérablement. Cette diminution est principalement due à la destruction de la population de petites bulles. La rétention de grosses bulles est pratiquement indépendante de la concentration des suspensions. La rétention de gaz mesurée avec la suspension d'huile paraffine à 36% volumique concorde remarquablement bien avec les données correspondantes obtenues avec de l'huile de Tellus (ρ_L = 86 kg/m^{-3}; μ_L = 0,075 Pa·s; σ = 0,028 N·m^{-1}) comme phase liquide. Des expériences de dégagement de gaz dynamiques confirment que la dispersion dans l'huile de Tellus se compose essentiellement de grosses bulles. On a trouvé que la rétention de grosses bulles diminuait de manière significative avec l'augmentation du diamètre de la colonne. On a mis au point un modèle pour l'estimation de la rétention de grosses bulles de gaz par l'introduction d'un facteur d'accélération dans le sillage dans la relation de Davies–Taylor–Collins (Collins, 1967), décrivant l'influence du diamètre de colonne sur la vitesse de montée d'une bulle à tête sphérique isolée.

Keywords: bubble columns, large bubbles, small bubbles, churn-turbulent flow regime, concentrated slurry, viscous liquid, bubble rise velocity, wall effect, scale effect.

Bubble column reactors are finding increasing use in industrial practice; this reactor technology figures prominently in processes for converting natural gas to liquid fuels and light olefins using Fischer Tropsch synthesis (Krishna et al., 1996; Maretto and Krishna, 1999; Sie and Krishna, 1999). There are considerable reactor design and scale-up problems associated with the Fischer Tropsch bubble column slurry reactor. Firstly, large gas throughputs are involved, necessitating the use of large diameter reactors, typically 5 to 8 m, often in parallel. Secondly, the process operates under high pressure conditions, typically 4 MPa. Thirdly, in order to obtain high conversion levels, large reactor heights, typically 30 to 40 m tall, are required along with the use of highly concentrated slurries, approaching 40 vol%. Finally, the process is exothermic in nature, requiring heat removal by means of cooling tubes inserted in the reactor. Successful commercialisation of this technology is crucially dependent on the proper understanding of the scaling up principles of bubble columns for the above mentioned conditions fall outside the purview of most published theories and correlations (Deckwer, 1992; Fan, 1989).

The objective of the present communication is to demonstrate that the hydrodynamics of bubble columns with concentrated slurries can be mimicked using a highly viscous liquid. The advantage of this approach is that experimentation with a viscous liquid is much simpler than with concentrated slurries, and parameters such as liquid velocity profiles and liquid phase backmixing can be determined more easily with liquids rather than with slurries. A further objective of this work is to present a more fundamentally based model for the estimation of the gas hold-up in slurry bubble columns than presented in our earlier work (Krishna et al., 1997).

Experimental

Experiments were performed in polyacrylate columns with inner diameters of 0.1, 0.19 and 0.38 m. The gas distributors used in the three columns were all made of sintered bronze plate (with a mean pore size of 50 μm). All columns were equipped with quick closing valves in the gas inlet pipe in order to perform dynamic gas disengagement, or bed collapse experiments. Pressure taps were installed along the height of the columns. Validyne DP15 pressure sensors, connected to the pressure taps and coupled to a PC, allowed the transient pressure signals to be recorded during dynamic gas disengagement experiments. The gas flow rates entering the column were measured with the use of a set of rotameters, placed in parallel, as shown in Figure 1 for the 0.38 m column. This setup was typical. Air was used as the gas phase in all experiments. Firstly,

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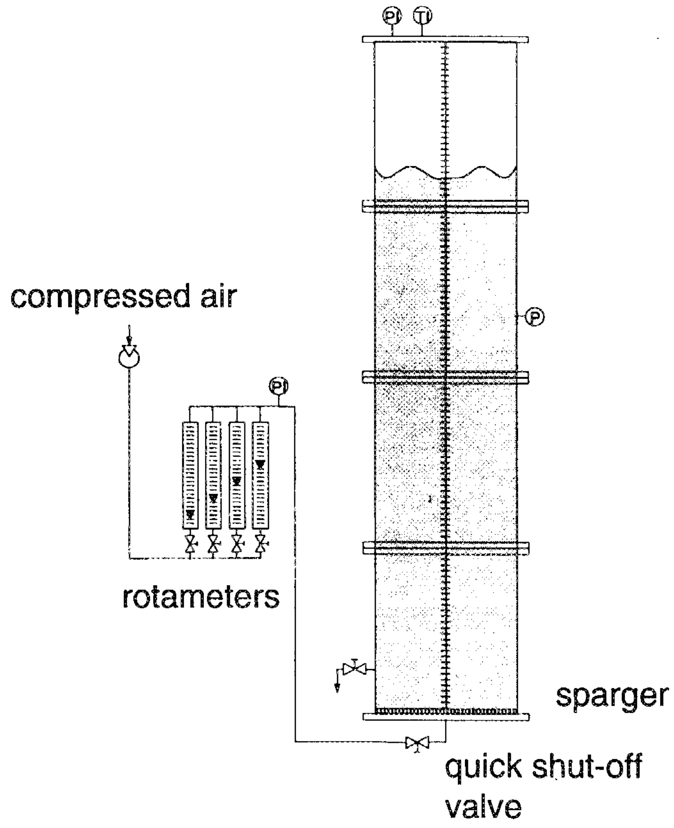

Figure 1. Experimental setup for the 0.38 m diameter column.

experiments were performed with paraffin oil (density, ρ_L = 790 kg/m^3; viscosity, μ_L = 0.0029 Pa·s; surface tension, σ = 0.028 N/m) as liquid phase to which solid particles in varying concentrations were added. The solid phase used consisted of porous silica particles whose properties were determined to be as follows: skeleton density = 2100 kg/m^3; pore volume = 1.05 mL/g; particle size distribution, d_p: 10% < 27 μm; 50% < 38 μm; 90% < 47 μm. The solids concentration ε_s, is expressed as the volume fraction of solids in gas free slurry. The pore volume of the particles (liquid filled during operation) is counted as being part of the solid phase.

To investigate the gas hold-up characteristics in the churn-turbulent regime, dynamic gas disengagement experiments were performed. For a set gas flow rate, the system was given time to reach steady state. At this moment the experimental run was commenced. During the experimental run the pressure at a certain height in the column was measured using a pressure transducer. A few seconds after the start of the run, the gas flow rate was instantaneously shut off using the quick closing valve. The measured pressure signals were interpreted to obtain information on the gas hold-ups (Krishna and Ellenberger, 1996).

Analogous experiments to determine the total gas hold-up and distribution of hold-ups of the large bubbles and dense phase were also carried out with Tellus oil (ρ_L = 862 kg/m^3; μ_L = 0.075 Pa·s; σ = 0.028 N/m) as the liquid.

Results and Discussion

The influence of the solids concentration on the total gas hold-up ε for varying superficial gas velocities are shown in Figure 2 for the 0.10 m and 0.38 m diameter columns. It is observed that increased particles concentration tends to decrease the total gas hold-up, ε, to a significant extent. This decrease in the total gas hold-up is due to the decrease in the hold-up of the small bubbles due to enhanced coalescence caused due to the presence of the small particles. At low solid concentrations there is a pronounced maximum in the gas hold-up which is typical of the transition region. With increased solids concentration the transition occurs at a lower superficial gas velocity and the transition "window" reduces in size. It is interesting to note that the transition region is wider for the 0.1 m diameter column than for the 0.38 m diameter column. Furthermore, we note from Figure 2 that the maximum in the gas hold-up seems to persist to higher solids concentration for the 0.38 m diameter column. Though the precise reasons for this is unclear, this phenomena could be linked to the stronger liquid circulations which are present in the larger diameter column.

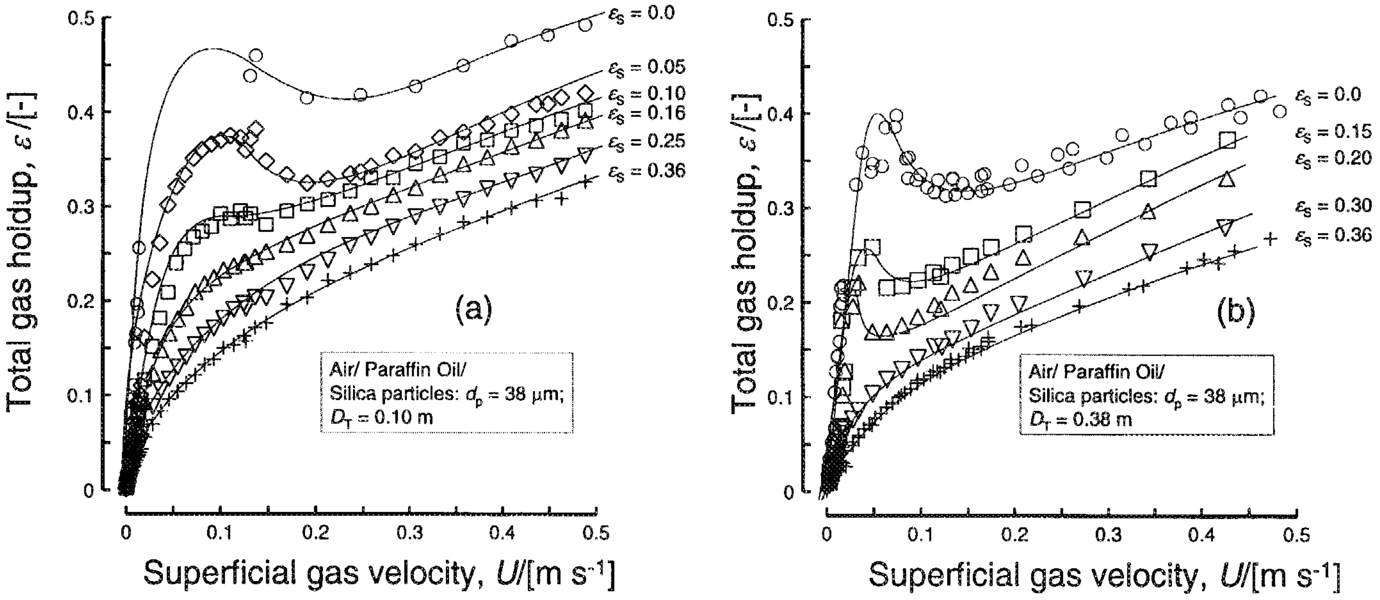

Figure 2. Influence of increased particles concentration on the total gas hold-up in columns of (a) 0.1 m, and (b) 0.38 m diameter.

Typical dynamic gas disengagement profiles for air–paraffin oil and air–36 vol% paraffin oil slurry in the 0.38 m column for the churn turbulent flow regime of operation are shown in Figure 3. After the shut-off of the gas supply, the hold-up decreases due to the escape of fast rising large bubbles (dilute phase). When the large bubbles have escaped the small bubbles leave the column. The voidage of gas in the dense phase, ε_{df}, is determined as indicated in Figure 3. The gas hold-up of the large bubbles, i.e., the dilute phase is obtained from $\varepsilon_b = (\varepsilon - \varepsilon_{df})/(1 - \varepsilon_{df})$. The terminology of dilute and dense phases is based on the two-phase model adopted earlier to describe the hydrodynamics of bubble columns in the churn-turbulent flow regime experiments (Krishna and Ellenberger, 1996); this model is adapted for slurry bubble columns in Figure 4.

Data on the gas hold-up in the dense (small bubbles) and dilute (large bubble) phases are shown in Figures 5 and 6 for the 0.38 m diameter column. Figure 5 shows the dense phase gas hold-up is approximately constant for churn-turbulent operation at superficial gas velocities exceeding about 0.1 m/s. This is a useful conclusion for scale up purposes. Furthermore, we note from the data in Figure 6 that the large bubble gas hold-up ε_b is practically independent of slurry concentration in the range $0.16 < \varepsilon_s < 0.36$. Figure 7(a) shows the collection of data on the gas hold-up in the dense phase, ε_{df}, for all column diameters and slurry concentrations. We see that the dense phase gas hold-up ε_{df} is virtually independent of the column diameter and is a significant decreasing function of the particle concentration ε_s. The unique dependence of the decrease in the dense phase gas voidage ε_{df} with increasing solids volume fraction ε_s is useful for scale-up purposes because this parameter can be determined in a relatively small diameter column under actual reaction conditions of temperature and pressure. It is clear that addition of silica particles has the effect of reducing the small bubble population virtually to zero when the slurry concentration approaches 40 vol%. The addition of solid particles tends to promote coalescence of small bubbles and the rise velocity of the small bubbles, V_{small}, increases with increasing ε_s; see Figure 7(b). The paraffin-oil slurry data on the dense phase

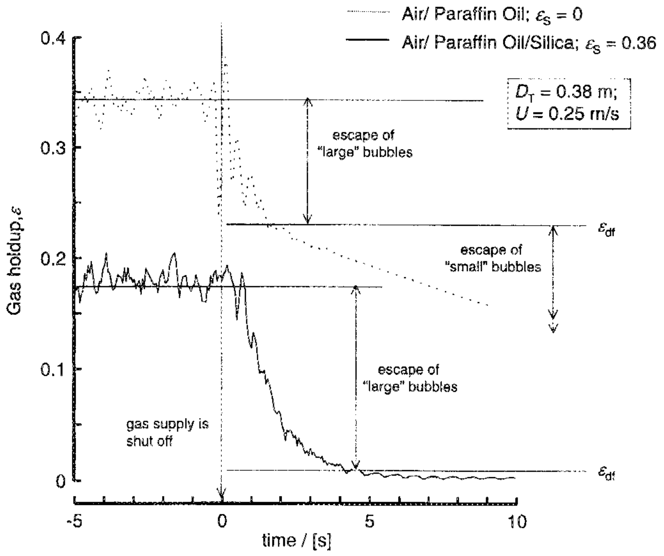

Figure 3. Dynamic gas disengagement experiments for air/paraffin oil and air/36 vol% paraffin oil slurry in the 0.38 m diameter column.

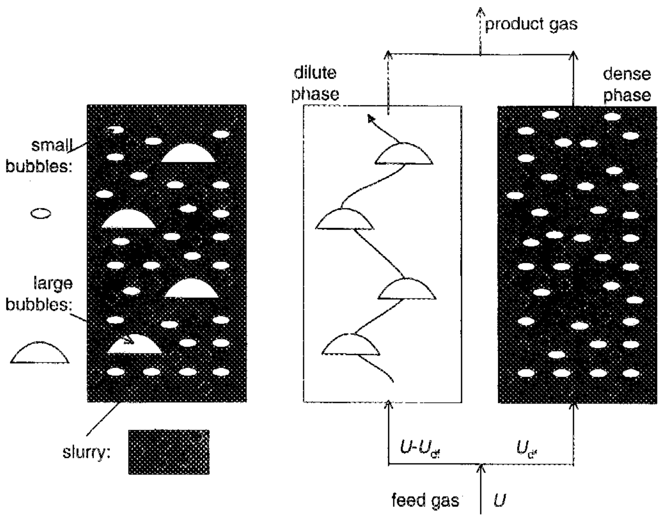

Figure 4. Generalized two-phase model applied to a bubble column slurry reactor operating in the churn-turbulent regime.

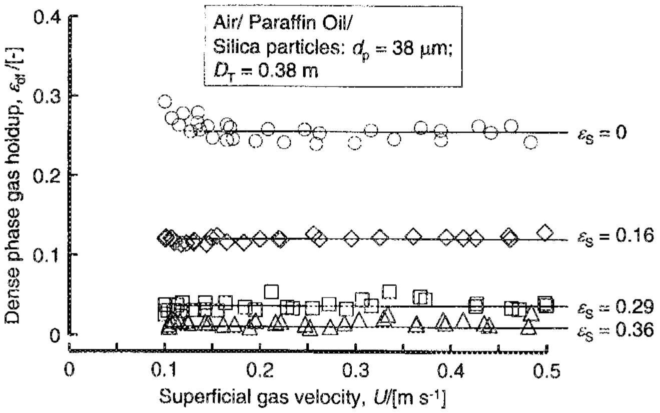

Figure 5. Influence of increased solids concentration on the dense phase gas hold-up for air/paraffin oil slurries in a 0.38 m diameter column.

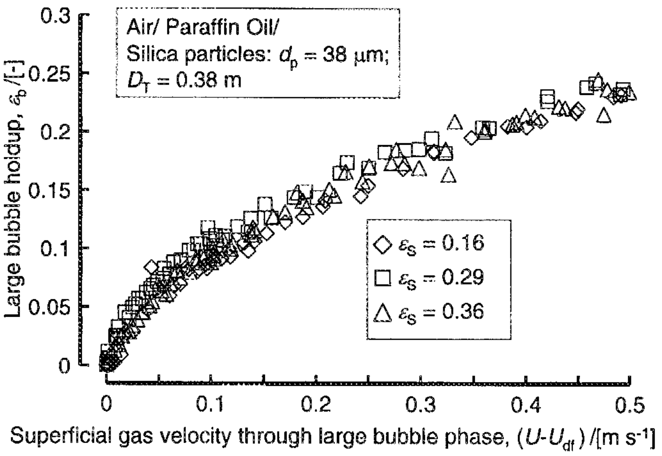

Figure 6. Large bubble gas hold-up in 0.38 m diameter column for various slurry concentrations.

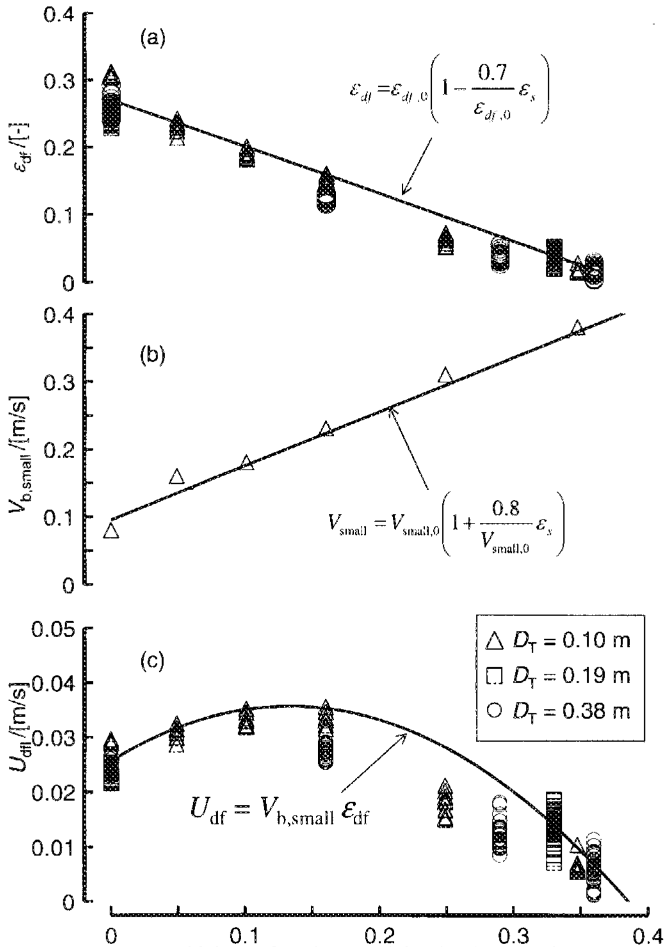

Figure 7. Influence of particles concentration ε_s on (a) dense phase gas voidage, ε_{df}, (b) rise velocity of the small bubbles, V_{small}, and (c) superficial gas velocity through the dense phase U_{df}.

voidage ε_{df} and the small bubble rise velocity V_{small} can be correlated as:

$$\varepsilon_{df}=\varepsilon_{df,0}\left(1-\frac{0.7}{\varepsilon_{df,0}}\varepsilon_s\right);\quad V_{small}=V_{small,0}\left(1+\frac{0.8}{V_{small,0}}\varepsilon_s\right) \tag{1}$$

where the paraffin-oil parameters $\varepsilon_{df,0}$ = 0.27 and $V_{small,0}$ = 0.095 m/s. The superficial gas velocity through the dense phase (see Figure 4) can be estimated from $U_{df} = V_{small}\,\varepsilon_{df}$.

Our earlier study on the modelling of the Fischer Tropsch slurry reactor has shown that slurry concentration of at least 35 vol% is desirable from the point of view of commercial viability (Maretto and Krishna, 1999). We therefore focus further attention on the influence of column diameter on the hydrodynamics of a 36 vol% paraffin oil slurry system. The total gas hold-up ε measured with this slurry concentration in the three columns is compared in Figure 8 with the corresponding data obtained with air–Tellus oil. It is interesting to note that the gas hold-ups are remarkably close to each other for all three columns studied.

Dynamic gas disengagement experiments were also performed in the three columns with air–Tellus oil. A typical experiment carried out in the 0.38 m diameter column operating at a superficial gas velocity U = 0.25 m/s is shown in Figure 9, in which comparison is made with the corresponding experiment with the air–36 vol% slurry. In the air–Tellus oil system, the dispersion consists predominantly of large bubbles and the values of the dense phase voidage $\varepsilon_{df} \approx 0.02$ and $U_{df} \approx 0.01$ m/s.

The large bubble hold-up ε_b in the air–Tellus oil system is found to decrease significantly with the column diameter; see Figure 10(a). From the experimental data on ε_b, the rise velocity of the swarm of large bubbles V_b can be calculated from the relation:

$$V_b = (U - U_{df})/\varepsilon_b \tag{2}$$

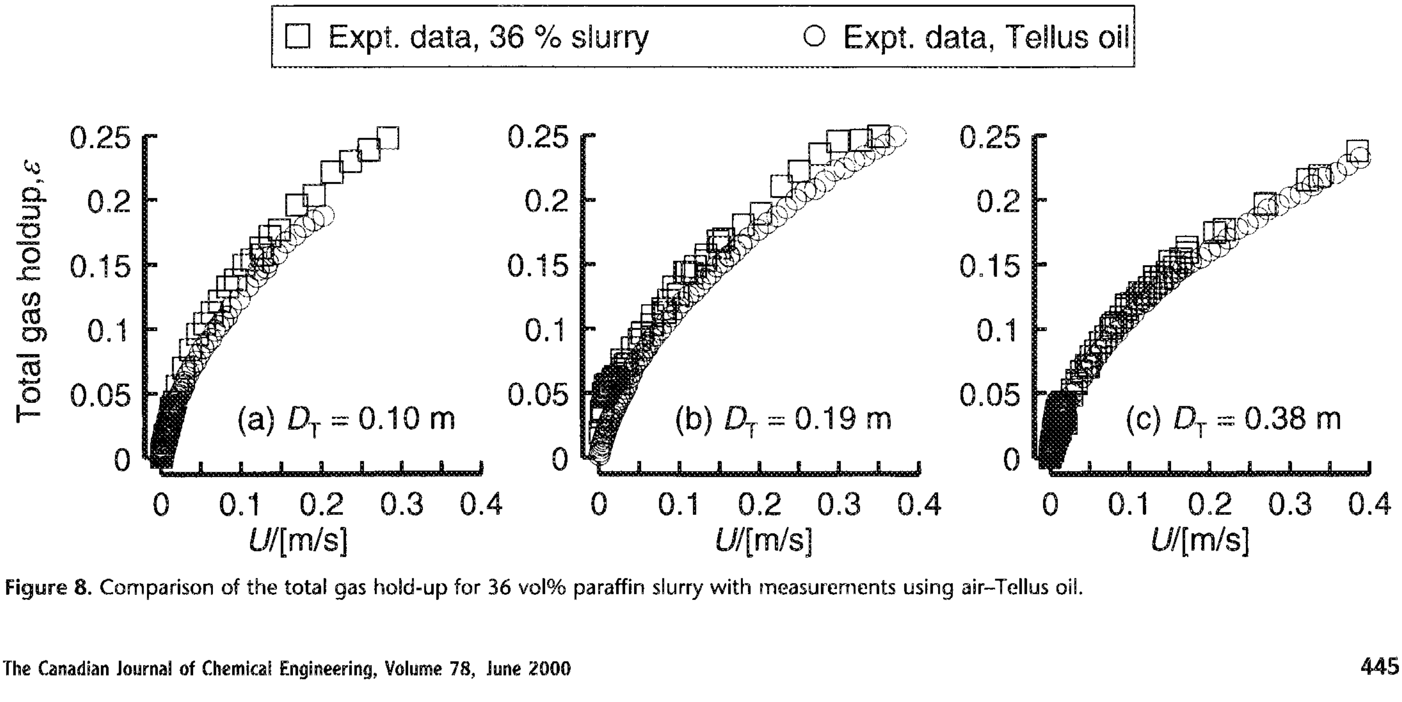

Figure 8. Comparison of the total gas hold-up for 36 vol% paraffin slurry with measurements using air–Tellus oil.

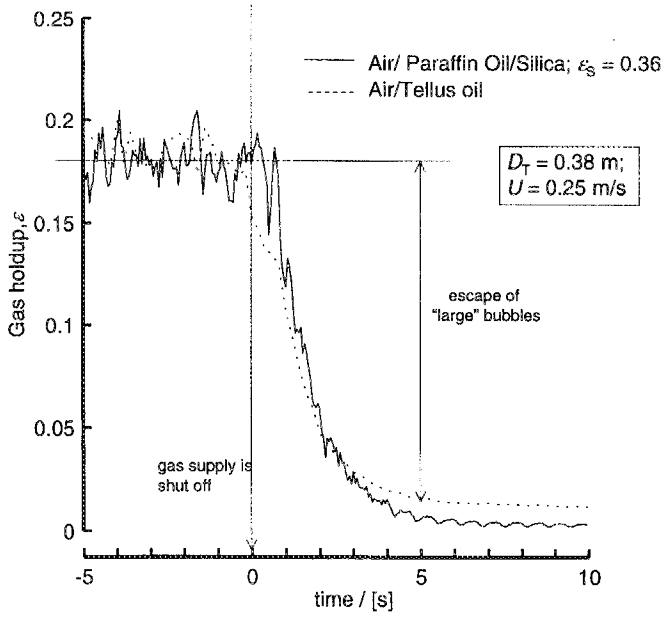

Figure 9. Comparison of dynamic gas disengagement curves for 36 vol% paraffin slurry with measurements using air–Tellus oil.

The large bubble swarm velocity therefore increases with increasing column diameter, as can be seen in Figure 10(a). Our objective now is to develop a model to describe the scale dependence on V_b. In our earlier work (Krishna et al., 1999), we had developed a model for rise velocity of a swarm of large bubbles in the form:

$$V_b = 0.71\sqrt{g d_b}(SF)(AF) \tag{3}$$

where we introduce two correction factors into the classical Davies–Taylor (1950) relation for the rise of a single spherical cap bubble in an infinite volume of liquid. Equation (2) is valid for bubble sizes and system properties for which the Eötvös number, $Eö > 40$ (see Clift et al., 1978). The scale correction factor (SF) accounts for the influence of the column diameter and is taken from the work of Collins (1967) to be:

$$\begin{aligned} SF &= 1 && \text{for } d_b / D_T < 0.125 \\ SF &= 1.13\exp(-d_b / D_T) && \text{for } 0.125 < d_b / D_T < 0.6 \\ SF &= 0.496\sqrt{D_T / d_b} && \text{for } d_b / D_T > 0.6 \end{aligned} \tag{4}$$

The acceleration factor AF accounts for the increase in the large bubble velocity over that of a single, isolated, bubbles; this acceleration is due to wake interactions. This factor increases as the distance between the large bubbles decreases. Since the average distance between large bubbles will decrease as the superficial gas velocity through the large bubble phase increases, we postulate a linear relation for AF:

$$AF = \alpha + \beta(U - U_{df}) \tag{5}$$

and a power-law dependence of the bubble size on $(U - U_{df})$:

$$d_{b,large} = \gamma(U - U_{df})^{\delta} \tag{6}$$

The model parameters α, β, γ and δ were determined by multiple regression and found for Tellus oil to be:

$$\alpha = 2.25; \quad \beta = 4.09; \quad \gamma = 0.069; \quad \delta = 0.376 \tag{7}$$

The model developed above for Tellus oil is compared with the large bubble hold-up for concentrated slurries in Figure 10(b).

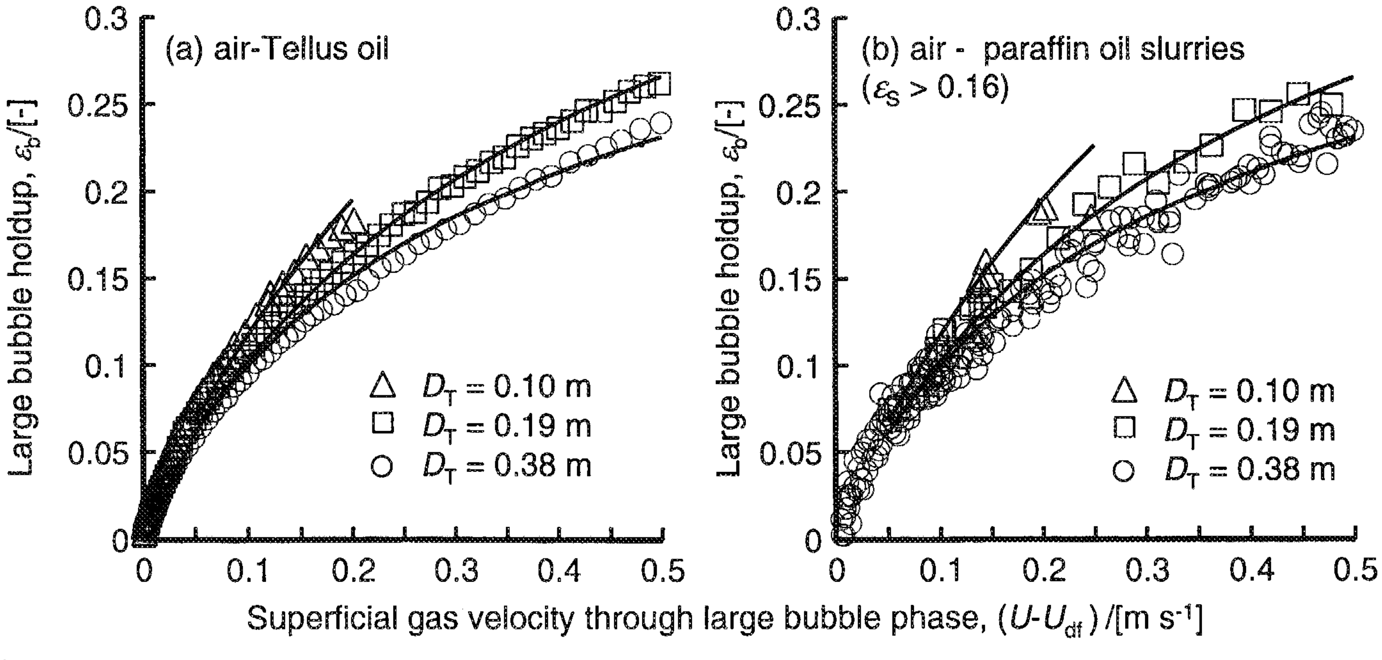

Figure 10. Influence of column diameter on the hold-up of large bubbles ε_b in (a) Tellus oil, and (b) paraffin oil slurry. Experimental data compared with predictions of model (solid lines) using Equations (2) to (7).

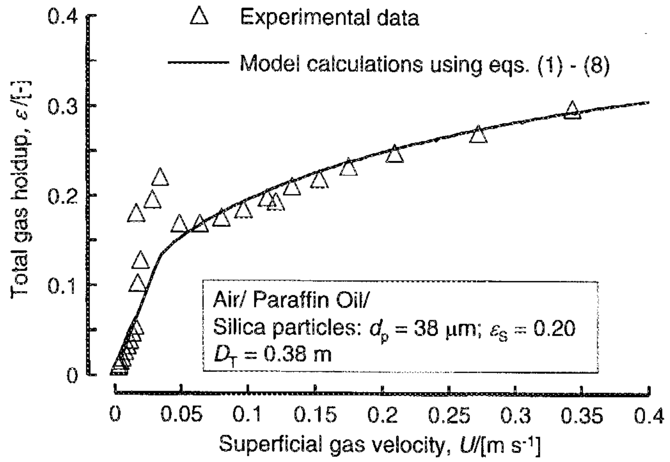

Figure 11. Estimation of the total gas hold-up for a 20 vol% slurry in 0.38 m diameter column compared with experimental data. Estimations using Equations (1) to (8).

The agreement between the model and experiment is remarkably good.

The total gas hold-up for bubble column slurry reactors can be calculated from the model of Krishna and Ellenberger (1996):

$$\varepsilon = \varepsilon_b + \varepsilon_{df}(1 - \varepsilon_b) \qquad (8)$$

where we use Equation (1) to estimate the dense phase gas hold-up as a function of the slurry concentration.

In order to illustrate how our developed model is to be used we estimate the gas hold-up in a 0.38 m diameter column operating with 20 vol% paraffin oil slurry at a superficial gas velocity U = 0.2 m/s. The values of the parameters $\varepsilon_{df,0}$ and $V_{small,0}$ for paraffin oil are, respectively, 0.27 m/s and 0.095 m/s. The rise velocity of small bubbles V_{small} is estimated from Equation (1) to be 0.255 m/s. The voidage of the dense phase ε_{df} is estimated from Equation (1) to be 0.13. The superficial gas velocity through the dense phase, $U_{df} = V_{small}\varepsilon_{df}$ = 0.033 m/s. This means that the superficial gas velocity through the large bubbles $(U - U_{df})$ = 0.167 m/s. The large bubble diameter can be calculated from Equations (6) and (7), d_b = 0.0352 m. The scale factor, using Equation (4), SF = 1. The acceleration factor, from Equations (5) and (7), AF = 2.91. The large bubble rise velocity V_b can be calculated using Equations (3); this yields V_b = 1.22 m/s. The large bubble hold-up is therefore $\varepsilon_b = (U - U_{df})/V_b$ = 0.137. From Equation (8) the total gas hold-up can be estimated ε_{df} = 0.249. Similar calculations, performed over a whole range of superficial gas velocities have been performed and compared in Figure 11 with experimental data. The agreement is very good for superficial gas velocities exceeding 0.1 m/s, which is the range of interest in industrial practice.

Conclusions

The major conclusions emerging from this work are listed below:

1. The total gas hold-up ε is significantly reduced with increasing slurry concentration.
2. The dense phase gas hold-up ε_{df} in the heterogeneous regime of operation is practically independent of the operating gas velocity above 0.1 m/s. Furthermore, ε_{df} is practically independent of the column diameter.
3. The dense phase gas hold-up ε_{df} is significantly reduced with increasing solids concentration; for estimation purposes we recommend the use of Equation (1).
4. The dilute phase gas hold-up ε_b is virtually independent of slurry concentration for slurry concentrations $\varepsilon_s > 0.16$.
5. The hydrodynamics of slurry bubble columns with concentrated slurries is analogous to that of a highly viscous liquid. The same model, based on a modification of the Davies–Taylor–Collins relations for describing the wall effect on the rise velocity of single spherical cap bubbles, can be used to describe the large bubble gas hold-up, ε_b, in both cases. This model is outlined in Equations (2) to (7).

Nomenclature

AF	acceleration factor
d_b	large bubble diameter, (m)
D_T	column diameter, (m)
$Eö$	Eötvös number, $g(\rho_L - \rho_G)d_b^2/s$
g	acceleration due to gravity, (9.81 m/s^2)
SF	scale correction factor
U	superficial gas velocity, (m/s)
$(U - U_{df})$	superficial gas velocity through the large bubble phase, (m/s)
U_{df}	superficial gas velocity through the dense phase, (m/s)
V_b	rise velocity of the large bubble swarm, (m/s)

Greek Symbols

$\alpha,\beta,\gamma,\delta$	parameters defined by Equations (5) and (6)
ε	total gas hold-up
ε_b	voidage of gas in the dilute phase (large bubbles)
ε_{df}	voidage of gas in the dense phase
ε_s	volume fraction of particles in slurry
μ	viscosity of phase, (Pa·s)
ρ	density of phase, (kg/m^3)
σ	surface tension of liquid phase, (N/m)

Subscripts

b	referring to large bubble population
df	referring to the dense phase
G	referring to gas phase
L	referring to liquid phase
T	tower or column

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