Gas Holdup in Slurry Bubble Columns: Effect of Column Diameter and Slurry Concentrations

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To study the influence of particle concentration on the hydrodynamics of bubble-column slurry reactors operating in the heterogeneous flow regime, experiments were carried out in 0.10, 0.19, and 0.38-m-dia. columns using paraffinic oil as the liquid phase and slurry concentrations of up to 36 vol. %. To interpret experimental results a generalization of the "two-phase" model for gas–solid fluid beds was used to describe bubble hydrodynamics. The two phases identified are: a dilute phase consisting of fast-rising large bubbles that traverse the column virtually in plug flow and a dense phase that is identified with the liquid phase along with solid particles and entrained small bubbles. The dense phase suffers backmixing considerably. Dynamic gas disengagement was experimented in the heterogeneous flow regime to determine the gas voidage in dilute and dense phases. Experimental data show that increasing the solid concentration decreases the total gas holdup significantly, but the influence on the dilute-phase gas holdup is small. The dense-phase gas voidage significantly decreases gas holdup due to enhanced coalescence of small bubbles resulting from introduction of particles. The dense-phase gas voidage is practically independent of the column diameter. The dilute-phase gas holdup, on the other hand, decreases with increasing column diameter, and this dependence could be described adequately with a slight modification of the correlation of Krishna and Ellenberger developed for gas–liquid systems.

Introduction

In processes for converting natural gas to liquid fuels, bubble-column reactors are finding increasing application. Bubble-column slurry reactors are attractive reactor configurations for Fischer-Tropsch and methanol syntheses (Eisenberg et al., 1994; Fox, 1990; Jager and Espinoza, 1995; De Swart and Krishna, 1995). There are considerable reactor design and scale-up problems associated with these synthesis technologies; these problems arise because of several special features of these processes. First, large gas throughputs are involved, necessitating the use of large-diameter reactors (typically 5–8 m), often in parallel. Second, the processes operate under high-pressure conditions (typically 10–80 bar). Third, in order to achieve economically high space-time yields, high slurry concentrations (typically 30–40 vol. %) need to be employed (Fox, 1990). Fourth, to obtain high conversion levels, large reactor heights, typically 20–30 m tall, are required. Finally, most of these processes are exothermic in nature, requiring heat removal by means of cooling tubes inserted in the reactor. Successful commercialization of the bubble-column reactor technology is crucially dependent on the proper understanding of the scaling up principles for these conditions, which fall outside the purview of most published theory and correlations, as can be ascertained by a careful examination of the published literature on bubble columns; see, for example, the recent comprehensive literature survey of Saxena (1995). The accurate estimation of the gas holdup under the conditions outlined in the foregoing is an essential factor in commercial reactor design for Fischer-Tropsch and methanol synthesis technologies.

The present experimental study distinguishes itself from earlier studies on the hydrodynamics of bubble-column slurry reactors (see, e.g., Bukur et al., 1987, 1992; Deckwer et al., 1980; Deckwer, 1992; Deckwer and Schumpe, 1993; Fukuma et al., 1987; Kara et al., 1992; Kelkar et al., 1984; Koide et al., 1984; O'Dowd et al., 1987; Saxena et al., 1992a,b; Saxena and Rao, 1993; Schumpe and Grund, 1986; Schumpe et al., 1987;

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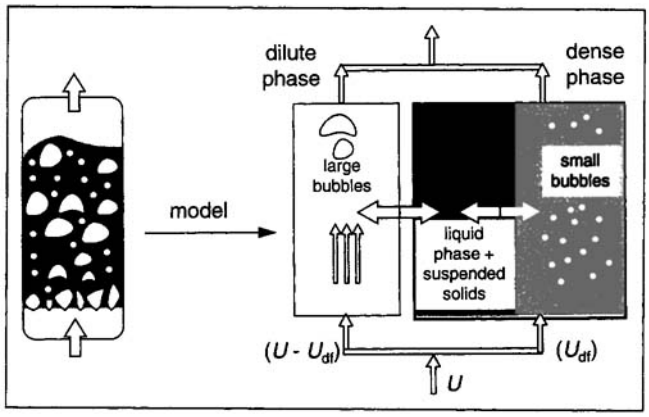

Figure 1. Generalized two-phase model applied to a bubble-column slurry reactor operating in the churn-turbulent regime.

Shetty et al., 1992) in that (1) we concentrate our attention on the industrially important churn-turbulent regime of operation, (2) high slurry concentrations, ranging up to 36 vol. %, are used in the experiments, and (3) the influence of the column diameter has been specifically investigated by use of three columns with diameters 0.10, 0.19, and 0.38 m.

The interpretation of the experimental results in the churn-turbulent regime is on the basis of the generalized "two-phase" model developed by Krishna and coworkers (Ellenberger and Krishna, 1994; Krishna and Ellenberger, 1996; De Swart and Krishna, 1995) and pictured in Figure 1. The "dilute" phase is identified with the fast-rising "large" bubbles, which traverse the column virtually in plug flow. The "dense" phase is identified with the liquid phase along with the solid particles and the entrained "small" bubbles. The dense phase suffers a considerable degree of backmixing. The entering superficial gas velocity U is split in two parts: a portion of the gas U_{df} rises through the column in the form of "small" bubbles; the remainder $(U - U_{df})$ rises through the column in the form of "large" bubbles. The influence of column diameter and particle concentration has been studied on the gas holdup of both the dilute and dense phases.

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 plates with a pore diameter 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. To minimize the influence of gas disengaging from the plenum chamber the quick-closing valves were placed as near to the gas distributor as possible. Pressure taps were installed along the height of the columns. Validyne DP15 pressure sensors, mounted 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 2 for the 0.38-m column. This setup was typical.

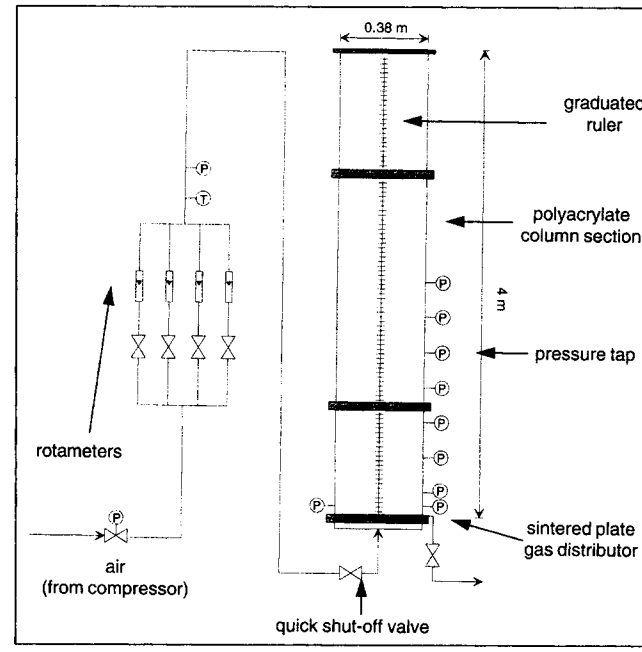

Figure 2. Experimental setup for the 0.38-m-diameter column.

Systems studied

All experiments were performed with paraffin oil (density, $\rho_L = 790$ kg·m^{-3}; viscosity, $\mu_L = 0.029$ Pa·s; surface tension, $\sigma = 0.028$ N·m^{-1}) as liquid phase. Air was used as the gas phase in all experiments. The solid phase used consisted of porous silica particles (skeletal density = 2100 kg·m^{-3}; pore volume = 1.05 mL·g^{-1}; particle-size distribution: 10% < 27 μm; 50% < 38 μm; 90% < 47 μm). An overview of the performed experiments is given in Table 1. Note that the solids concentration is expressed throughout this article in volume fraction of solids in gas-free slurry. In the definition of volume fractions of solids, the pore volume of the particles (liquid filled during operation) is counted as being part of the solid phase.

To investigate the gas holdup characteristics in the churn-turbulent regime, dynamic gas-disengagement experiments were performed. At the beginning of each experiment the gas

Table 1. Experimental Systems Studied

	Systems Studied		
D_T/[m]	Liquid	Solids Conc., ϵ_s/[−]	H_0/[m]
0.10	Paraffin oil	0	1.6
0.10	Paraffin oil	0.05	1.6
0.10	Paraffin oil	0.10	1.6
0.10	Paraffin oil	0.16	1.6
0.10	Paraffin oil	0.25	1.6
0.10	Paraffin oil	0.35	1.6
0.19	Paraffin oil	0	1.80
0.19	Paraffin oil	0.33	2.01
0.38	Paraffin oil	0	1.75
0.38	Paraffin oil	0.16	1.79
0.38	Paraffin oil	0.29	1.87
0.38	Paraffin oil	0.36	1.92

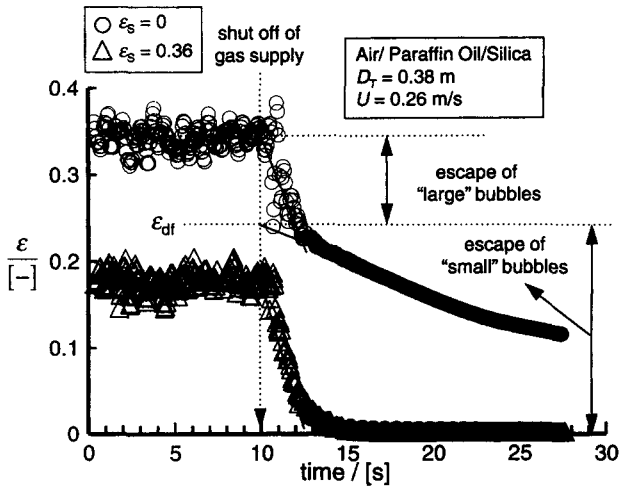

Figure 3. Dynamic gas-disengagement experiments for air/paraffin oil and air/36 vol. % paraffin oil slurry in the 0.38-m-dia. column.

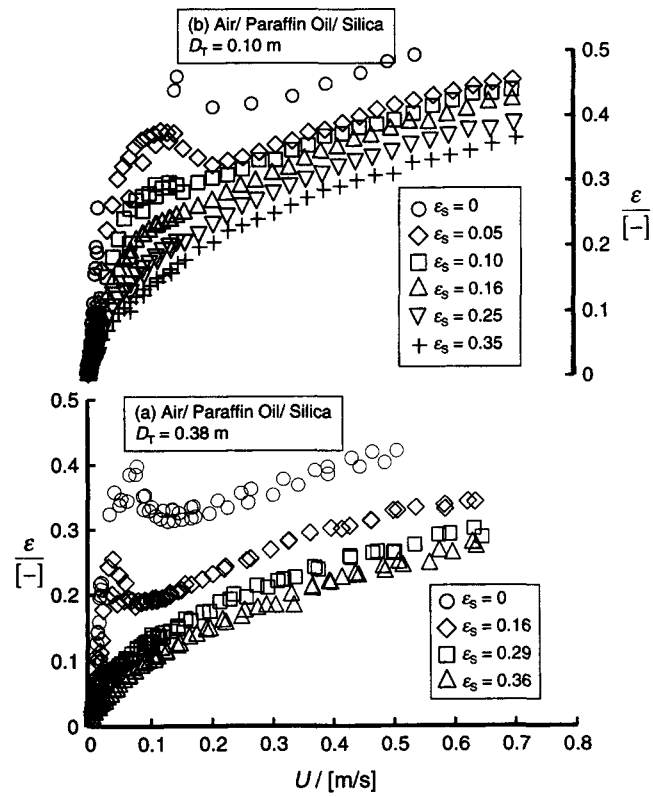

Figure 4. Influence of increased solids concentration on the total gas holdup for air/paraffin oil/silica in (a) 0.38-m-dia. column and (b) 0.1-m-dia. column.

flow rate was adjusted using one of four rotameters. The rotameters were initially calibrated to obtain precise values for the gas flow rate. 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. Ten seconds after the start of the run the gas flow was instantaneously shut off using the quick-closing valve. The measured pressure signals were interpreted to obtain information on the gas holdups (Daly et al., 1992).

Typical dynamic gas-disengagement profiles for air/paraffin oil and air/36 vol. % oil slurry in the 0.38-m column for the churn-turbulent flow regime of operation are shown in Figure 3. The total gas holdup ϵ is determined from the 10-s steady-state operation. After the shut off of the gas supply, the holdup 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 gas voidage in the "dense" phase, ϵ_{df}, is obtained as indicated in Figure 3. The gas holdup of the large bubbles, that is, the gas voidage of the dilute phase is obtained from

$$\epsilon_b = \frac{\epsilon - \epsilon_{df}}{1 - \epsilon_{df}}. \tag{1}$$

It is important to note that our definition of the dense-phase gas holdup, ϵ_{df}, differs from the small-bubble gas holdup used in the literature (Deckwer and Schumpe, 1993). The superficial gas velocity through the dense phase U_{df} is determined from the slope of the disengagement curve for the dense-phase gas, that is, the second linear portion of the disengagement process.

Results and Discussion

The influence of the solids concentration on the total gas holdup ϵ for varying superficial gas velocities is shown in Figure 4a for the 0.38-m- and 0.1-m-dia. columns. It is observed that increased particle concentration tends to decrease the total gas holdup, ϵ, to a significant extent. This conclusion was also reached by Koide et al. (1984), Kara et al. (1982), Kelkar et al. (1984), and Yasunishi et al. (1986), and can be attributed to an increase in the mean bubble diameter as the solids concentration is increased (Yasunishi et al., 1986). A close-up of the experimental $\epsilon - U$ data in the homogeneous bubbly flow regime, prevailing at low superficial gas velocities, emphasizes the significant influence of increased particle concentration on the rise velocity of the bubble swarm (see Figure 5). For the paraffin oil–silica slurries under study the rise velocity of the small bubbles was found to increase linearly with the particle volume fraction according to

$$V_{\text{small}} = V_{\text{small},0}\left(1 + \frac{0.8}{V_{\text{small},0}}\epsilon_s\right); \quad V_{\text{small},0} = 0.095 \text{ m/s}, \tag{2}$$

where $V_{\text{small},0}$ is the rise velocity of the small-bubble population in solids-free paraffin oil. Increased rise velocity of the small bubbles signifies larger bubble diameters caused by increased bubble coalescence. In the homogeneous regime, therefore, the total gas holdup decreases with increasing particle concentration due to increasing bubble diameters of the small-bubble population. The total gas holdup in the homogeneous flow regime can be estimated from

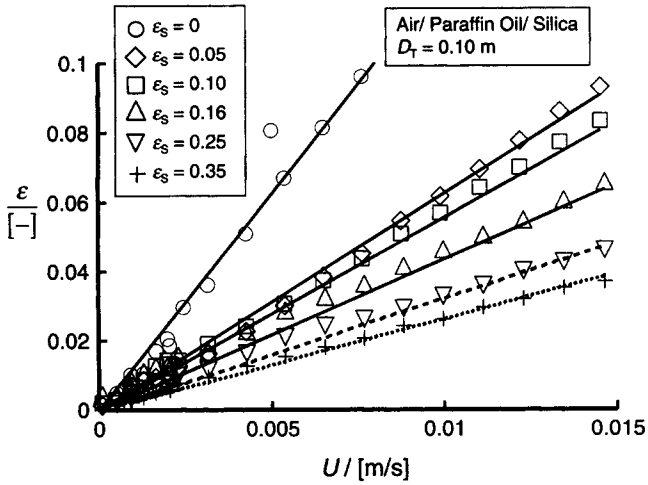

Figure 5. Influence of increased solids concentration on the total gas holdup for air/paraffin oil/silica in 0.1-m-dia. column, focusing on data at low superficial gas velocities.

$$\epsilon = \frac{U}{V_{\text{small}}} \quad \text{(homogeneous flow regime: } U \le U_{df}\text{)}. \quad (3)$$

This phenomenon also manifests itself in the heterogeneous flow regime, prevailing at superficial gas velocities $U > U_{df}$, where it is noted that the dense-phase gas holdup ϵ_{df} is significantly reduced with increasing solids concentration; Figure 6 shows the typical results for the 0.38-m-diameter column. Also seen in Figure 6 is that the dense-phase gas holdup is approximately constant for churn-turbulent operation at superficial gas velocities exceeding about 0.1 m/s. Figure 7 shows the collection of data on ϵ_{df} for all column diameters and slurry concentrations listed in Table 1. We see that the dense-phase gas holdup ϵ_{df} is virtually independent of the column diameter and is a significant decreasing function of the particles concentration, empirically fitted as

$$\epsilon_{df} = \epsilon_{df,0}\left(1 - \frac{0.7}{\epsilon_{df,0}}\epsilon_s\right); \quad \epsilon_{df,0} = 0.27. \quad (4)$$

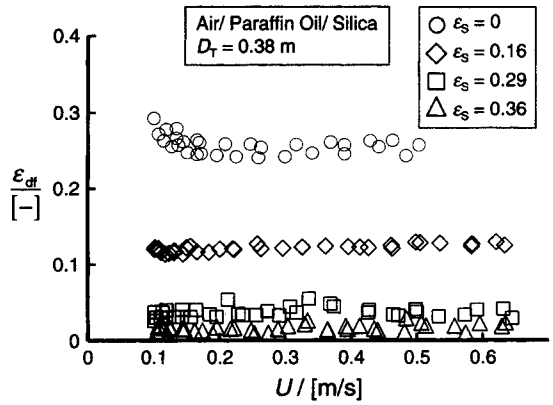

Figure 6. Influence of increased solids concentration on the dense-phase gas holdup for air/paraffin oil slurries in a 0.38-m-dia. column.

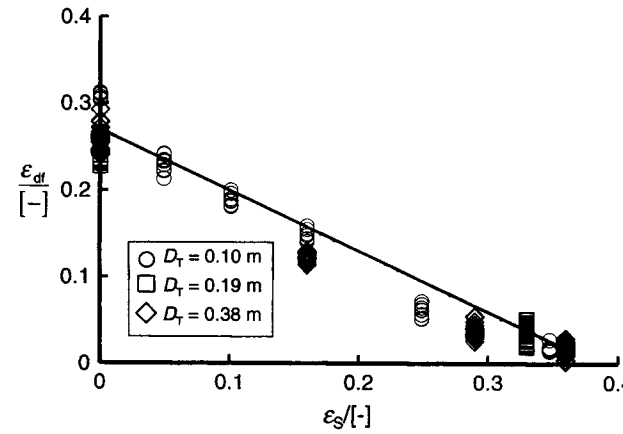

Figure 7. Reduction in the dense-phase gas holdup with increasing solids volume fraction.

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. The dense-phase gas holdup for the gas–liquid system, $\epsilon_{df,0}$, can be estimated using Reilly et al. (1994) correlation for the gas voidage at the regime transition point ϵ_{trans} as suggested by Krishna and Ellenberger (1996). Krishna and Ellenberger (1996) have verified that the Reilly correlation adequately reflects the influence of gas density on the regime transition point, and therefore when Eq. 3 is combined with the Reilly correlation, can be used to estimate the dense-phase gas holdup at increased system pressures and slurry concentrations. The superficial gas velocity at the regime transition point can be estimated from

$$U_{df} = V_{\text{small}}\epsilon_{df}, \quad (5)$$

where the small-bubble rise velocity and the dense-phase gas holdup at the regime transition point are estimated from Eqs. 3 and 4.

In an earlier study of hydrodynamics of gas–liquid bubble column reactors, Krishna and Ellenberger (1996) proposed the following correlation for the dilute-phase gas holdup

$$\epsilon_b = \alpha_2 \frac{1}{D_T^N}(U - U_{df})^{0.58}, \quad (6)$$

which is a dimensional equation in which SI units need to be inserted for $(U - U_{df})$ and D_T. For gas–liquid systems Krishna and Ellenberger (1996) determined the fit parameters to be $\alpha_2 = 0.268$ and $N = 0.18$. From Eq. 6 it follows that $\epsilon_b(D_T/D_{ref})^N$ should be independent of the column diameter where D_{ref} is an arbitrarily chosen reference diameter. Figure 8a and 8b show a plot of $\epsilon_b(D_T/D_{ref})^N$ vs. $(U - U_{df})$ for the complete data set in Table 1, taking $D_{ref} = 0.1$ m. The Krishna-Ellenberger model fit constant $\alpha_2 = 0.268$ is seen to hold quite well for slurry concentrations below 10 vol. %. For high slurry concentrations, in excess of 16 vol. % the experimental data could be fitted remarkably well taking α_2

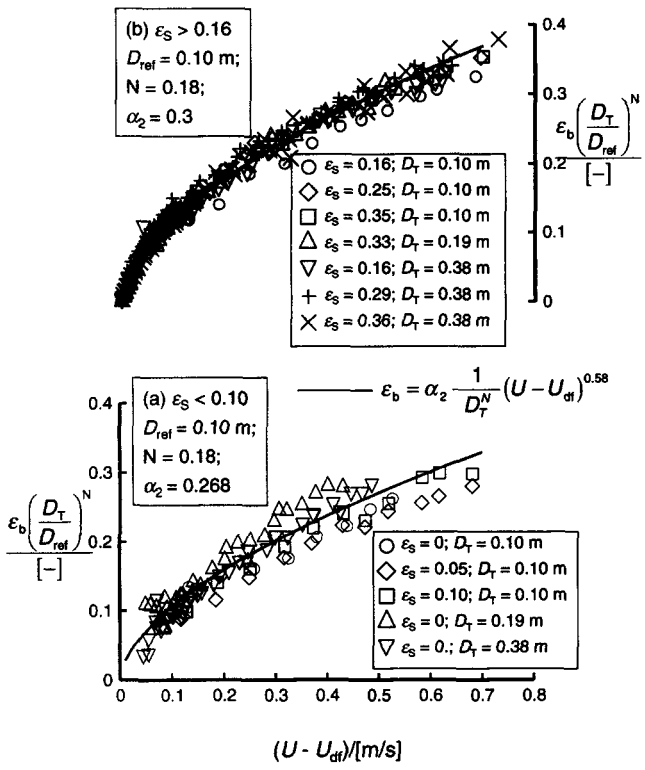

Figure 8. Influence of increased solids concentration and column diameter on the dilute-phase gas holdup.

(a) Data for low slurry concentrations ($\epsilon_s < 0.10$), and for (b) for high slurry concentrations ($\epsilon_s > 0.16$). Also drawn are the Krishna-Ellenberger correlation, Eq. 6 taking (a) $\alpha_2 = 0.268$ and (b) $\alpha_2 = 0.3$.

$= 0.3$. This increase in the value of the parameter α_2 is to be attributed to the vastly enhanced slurry viscosity at high slurry concentrations, which tends to lower the rise velocity of the large bubble population.

On the basis of the insights gained in the foregoing we may set up the following model for calculation of the total gas holdup in the heterogeneous flow regime

$$\epsilon = \epsilon_b + \epsilon_{df}(1 - \epsilon_b) \quad \text{(heterogeneous flow regime } U > U_{df}\text{)}. \tag{7}$$

The applicability of Eq. 7 for estimation of the total gas holdup is demonstrated in Figure 9 for three sets of data with varying column diameters and slurry concentrations.

Conclusions

The major conclusions emerging from this work are listed below.

- The total gas holdup ϵ is significantly decreased with increasing slurry concentration
- The dense-phase gas holdup ϵ_{df} (= small-bubble holdup) in the heterogeneous regime of operation is practically independent of the operating gas velocity above 0.1 m/s. Further, ϵ_{df} is practically independent of the column diameter. The dense-phase gas holdup is significantly reduced with increasing solids concentration; for estimation purposes we recommend the use of Eq. 4 where the value of $\epsilon_{df,0}$ can be estimated from the Reilly et al. (1994) correlation for the gas voidage at the regime transition point.
- The dilute-phase gas holdup ϵ_b is virtually independent of slurry concentration, but is a significant function of the column diameter. The fit constant is to be taken as $\alpha_2 = 0.268$ for low slurry concentrations ($\epsilon_s < 0.10$), and for high slurry concentrations ($\epsilon_s > 0.16$) we take $\alpha_2 = 0.3$.

The splitting of the total gas holdup ϵ into two parts: (1) a scale-independent, system- and slurry-concentration-dependent dense-phase gas holdup ϵ_{df}, and (2) a scale-dependent, system-and-slurry-concentration independent dilute-phase gas holdup ϵ_b is an important clue to the estimation of the gas holdup in slurry bubble columns.

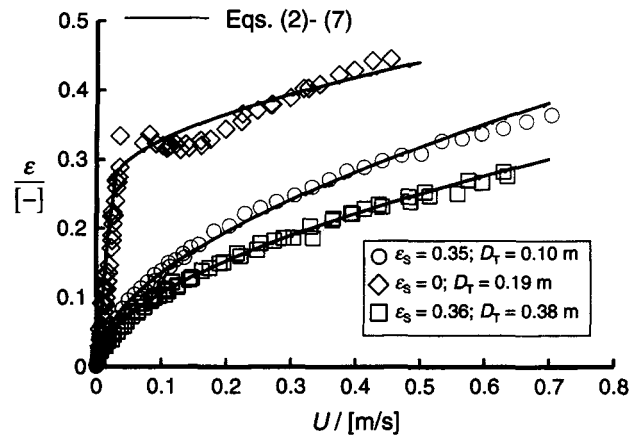

Figure 9. Test of experimental data for total gas holdup against the model predictions using Eqs. 2–7.

Notation

d_p = particle diameter, m
D_T = column diameter, m
H_0 = height of ungassed bed, m
N = power in the rise velocity correlation

Subscripts

G = referring to gas phase
0 = pure liquid property

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