Design Parameters Estimation for Scale-Up of High-Pressure Bubble Columns

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Experiments were carried out in bubble columns for a number of liquids at pressures between 0.1 and 2.0 MPa for two column sizes. Based on the experimental results as well as extensive literature data, the extent of the effect column dimensions have on gas holdup were determined, both at low and high pressures (which is of importance to scale-up). It was also demonstrated that none of the published empirical gas holdup equations incorporate the influence of gas density accurately. Therefore, a new improved gas hold-up equation is developed that incorporates the influence of gas and liquid properties with an average error of approximately 10%. Finally, it is also discussed to what extent the influence of pressure on other important design parameters such as the interfacial area, the liquid volumetric mass transfer coefficient, and gas and liquid mixing, can be estimated on the basis of empirical equations.

Introduction

Bubble column reactors, with and without suspended solids and in many configurations, are frequently applied in industry for various chemical processes (Mashelkar, 1970; Shah et al., 1982). In spite of the fact that bubble columns are used so frequently and have been studied extensively, an accurate design and scale-up of these bubble columns can still present a formidable task. Especially important design parameters, such as the gas holdup, the interfacial area, and the liquid volumetric mass transfer coefficient often cannot be estimated accurately enough. Furthermore, most industrial bubble columns are operated at extreme conditions with high temperatures and pressures, while most research reports have been focused on more ambient conditions. Recently, however, research on high pressures (for example, Idogawa et al., 1986, 1987a, b; Oyevaar, 1989; Wilkinson and van Dierendonck, 1990a) or a combination of high pressure and temperature (for example, de Bruijn et al., 1988; Clark, 1990; Deckwer et al., 1980; Mochida, 1985) has been increasing. Most of these works reported that gas holdup is higher, while the average bubble size is smaller at high pressure.

The main purpose of this article is to determine the influence of column dimensions on gas holdup (which is of importance to scale-up) and to establish whether the experimental influence of pressure (or gas density) and liquid properties on gas holdup can be correlated with empirical equations. It also discusses to what extent the influence of gas density on other important design parameters (mass transfer, gas and liquid mixing) can be estimated with empirical equations and to what extent experiments are still required for the estimation of these parameters.

Scale-Up Experiments

For the gas holdup experiments, two high-pressure bubble columns were used: bubble column with 0.158 m ID and a liquid height of 1.5 m (for pressures up to 2.0 MPa); bubble column consisting of a metal vessel equipped with three pairs of 0.15-m-dia. high-pressure sight glasses, in which either a 0.15-m- or a 0.23-m-ID transparent pipe could be placed with a liquid height of 1.2 m. The maximal pressure in the latter bubble column was 1.5 MPa and the maximal nitrogen supply rate was 400 Nm^3/h. Three pure liquids were used for the experiments (Table 1). In addition gas holdup experiments were

Table 1. Physical Properties of Liquids Used for Gas Holdup Experiments

Liquid (20°C)	ρ_l (kg/m^3)	σ (N/m)	η_l (Pa·s)
n-Heptane	684	0.020	0.00041
Mono-ethylene glycol	1,113	0.048	0.021
Water	998	0.072	0.001

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also carried out at 20°C and 40°C in liquid of the Hyam, phosphate oxime (HPO) preparation process (van Goolen, 1976) for comparison with the gas holdup obtained in an industrial (DSM) HPO bubble-column reactor ($U_l \simeq 0.11$ m/s, $U_g \simeq 0.14$ m/s, $V_r \simeq 60$ m^3, $H/D > 5$, $T \simeq 60$°C). The superficial gas velocity (based on the column midpoint pressure) was determined (for all experiments) with a calibrated gas turbine, and the gas holdup was measured with an overflow technique.

Experimental Gas Holdup Results and Discussion

Influence of column diameter on gas holdup

A number of authors have determined that gas holdup (under atmospheric conditions) is virtually independent of the column diameter provided its value is larger than 0.15 m (for example, Yoshida and Akita, 1965; van Dierendonck, 1970; Kaštánek et al., 1984). For smaller column diameters, wall effects become important because they cause an increase in gas holdup. Furthermore, bubbles with the size of column diameter can occur in small-diameter columns, resulting in slugging, but not in large-diameter columns.

Knowledge of the size of column diameter, above which no (or little) influence on gas holdup (and mass transfer) is expected, is critical to scale-up, because it determines the minimum scale at which pilot-plant experiments can be implemented to estimate the gas holdup (and mass transfer) in a large industrial bubble column. No published data, however, have dealt with the effect of column diameter on gas holdup above atmospheric pressures to confirm this value of 0.15 m for higher pressures. Therefore, we carried out experiments at various pressures to compare the results obtained in bubble columns with different column diameters.

Figures 1, 2 and 3 show the experimental results for deionized water, *n*-heptane, and mono-ethylene glycol in a 0.15- and 0.23-m-ID column for pressures from atmospheric to 1.5 MPa. Gas holdup increases markedly (especially for the liquids of low viscosity: *n*-heptane and water) with increasing pressure. The influence of column diameter also is found to be very small for all three liquids at both low and high pressures for these two column diameters. As Figure 4 shows, when our *n*-heptane results were compared with similar gas holdup experiments in *n*-heptane of Tarmy et al. (1984) in a 0.61-m-dia., 8.5-m-high bubble column, a slightly larger difference was found. These differences too are not very significant, however, and may very well be within the limits of experimental accuracy.

Gas holdup experiments by other researchers could also be used for comparison. Experiments of Idogawa et al. (1987a) in a 0.05-m-dia. column were between 30 and 50% higher than similar experimental data by Wilkinson and van Dierendonck (1990a) in a 0.15-m-dia. column (Figure 5) for both low and high pressures, which indicates that wall effects are also significant in a 0.05-m-dia. bubble column at high pressure. In Figure 6, comparisons are made of atmospheric gas holdup data in the nitrogen/mono-ethylene glycol system with column diameters between 0.16 and 0.44 m. Even for liquids that are as viscous as mono-ethylene glycol ($\eta_l = 0.021$ Pa·s), a column diameter, which is larger than 0.15 m, has little or no effect on gas holdup. It is concluded that column diameter has very little influence on the gas holdup for both low and high pressures and in different liquids provided that its diameter is in excess of 0.15 m.

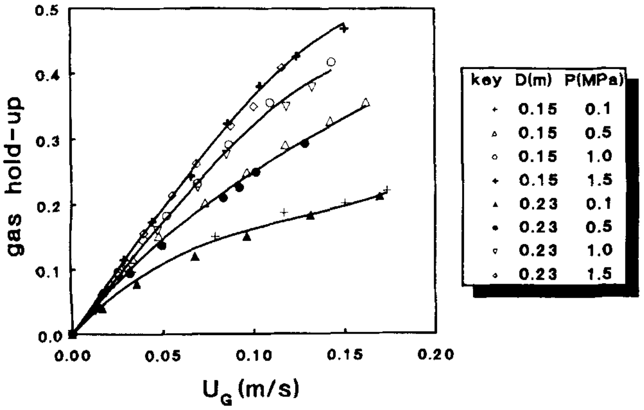

Figure 1. Gas holdup vs. superficial (nitrogen) gas velocity in deionized water for two column diameters: $D = 0.15$ m, $H = 1.5$ m, sparger ring with 37 holes of 2 mm; $D = 0.23$ m, $H = 1.2$ m, sparger ring with 4 holes of 7 mm.

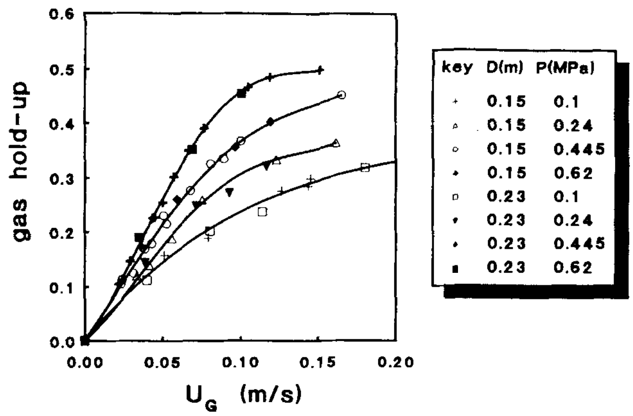

Figure 2. Gas holdup vs. superficial (nitrogen) gas velocity in *n*-heptane for two column diameters: $D = 0.15$ m and $D = 0.23$ m, both $H = 1.2$ m and a sparger ring with 4 holes of 7 mm.

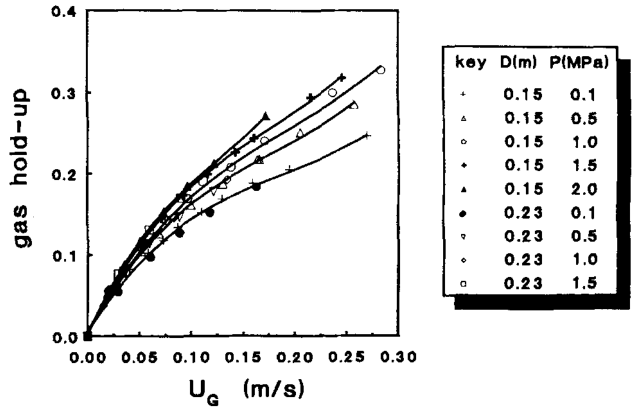

Figure 3. Gas holdup vs. superficial (nitrogen) gas velocity in mono-ethylene glycol for two column diameters (same as Figure 1).

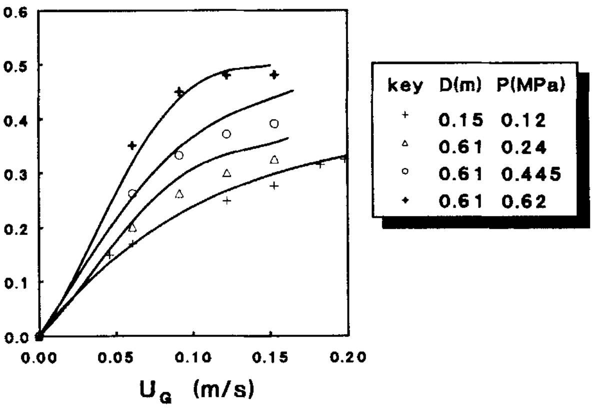

Figure 4. Gas holdup in *n*-heptane vs. superficial (nitrogen) gas velocity for four pressures: data of Tarmy et al., 1984 (+, ○, △, +) combined with data (——) of Figure 2.

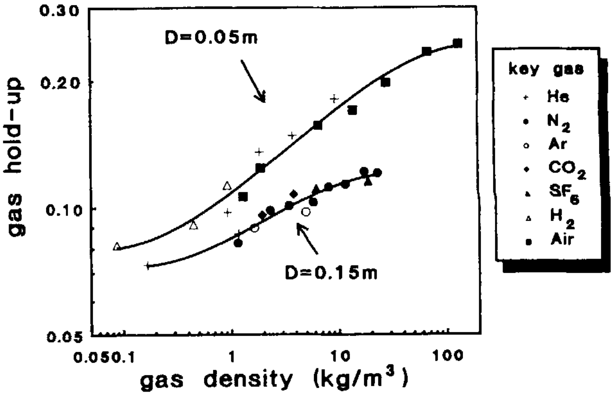

Figure 5. Gas holdup in water vs. gas density at a superficial gas velocity of 0.03 m/s: Data of Idogawa et al. (1987a) with $D=0.05$ m, $H=0.85$ m, combined with experimental data of Wilkinson and van Dierendonck (1990a) in a column with $D=0.15$ m, $H=1.5$ m.

Influence of column height on gas holdup

Gas holdup in a bubble column is usually not uniform. In general, three regions of different gas holdup are recognized. At the top of the column, there is often a kind of foam structure with a relatively high gas holdup, while the gas holdup near the sparger is sometimes measured to be higher (for porous-plate spargers) and sometimes lower (for single-nozzle spargers) than in the main central part of the column. Obviously the extent to which the gas holdup in the sparger region and in the foam region contribute to the overall average gas holdup depends on the colum height. In other words, if the bubble column is very high, then gas holdup near the sparger and in the foam region at the top of the column has little influence on the overall gas holdup, while the influence can be significant for low bubble columns. Furthermore, the column height can influence the value of the gas holdup due to the fact that liquid circulation patterns (that tend to decrease the gas holdup) are not fully developed in short bubble columns ($H/D<3$). The above-mentioned factors tend to cause a decrease in gas holdup with increasing column height. Most authors who studied this influence of column height on gas holdup, however, also claimed that this influence is negligible for column heights greater than 1-3 m and with height to diameter ratios above 5 (Kaštánek et al., 1984).

Influence of sparger design on gas holdup

Recently, Wilkinson (1991) has shown that the influence of the sparger design on gas holdup is negligible (for various liquids and at various pressures) provided the sparger hole diameters are larger than approximately 1-2 mm (and care is taken to prevent maldistribution of gas at the sparger). Spargers with small hole diameters (less than 1 mm), however, lead to the formation of smaller bubbles and thus to a higher gas holdup and a higher interfacial area, and appear to be used most frequently for academic research on bubble columns. In spite of the advantageous characteristics of these spargers, in industry usually less effective spargers are used with larger hole diameters that are less sensitive to fouling. In high bubble columns, the influence of the sparger usually diminishes due to the ongoing process of bubble coalescence. Consequently, the relatively high gas holdup and mass transfer rate that can occur in small bubble columns as a result of the use of small sparger holes will not occur in general as noticeably in a high (industrial) bubble column.

So far it has been argued that the gas holdup is virtually independent of column dimensions and sparger layout (for low as well as high pressures) provided the following three criteria are fulfilled:

1. The column diameter has to be larger than 0.15 m.
2. The column height to diameter ratio has to be in excess of 5.
3. The hole diameter of the sparger has to be larger than 1-2 mm.

In other words, a scale-up procedure, in which the gas holdup, the volumetric mass transfer coefficient, and the interfacial area for a large-scale industrial bubble column are estimated on the basis of experimental data obtained in a pilot-plant

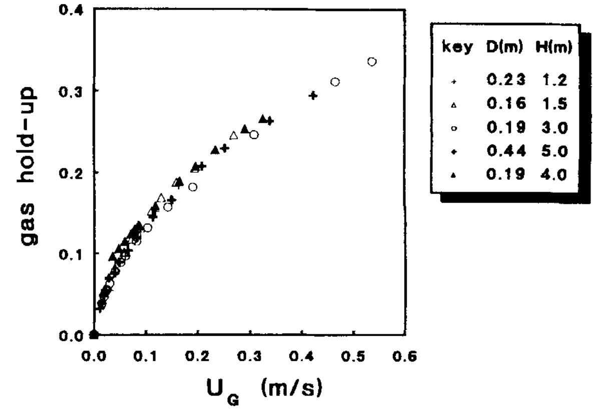

Figure 6. Gas holdup in mono-ethylene glycol vs. superficial (nitrogen) gas velocity at atmospheric pressure: data of Wezorke (1986); $H=3$ and 5 m and Krishna et al. (1991); $H=4$ m, combined with data of Figure 3.

Table 2. Gas-Density-Dependent Gas Holdup Equations

$\epsilon_g = 0.672 \left[\frac{U_g \eta_l}{\sigma}\right]^{0.578} \left[\frac{\eta_l^4 g}{\rho_l \sigma^3}\right]^{-0.131} \left[\frac{\rho_g}{\rho_l}\right]^{0.062} \left[\frac{\eta_g}{\eta_l}\right]^{0.107}$ or $\epsilon_g = 0.672\ U_g^{0.578} \cdot \rho_l^{0.069} \cdot \rho_g^{0.062} \cdot \sigma^{-0.185} \cdot \eta_l^{-0.053} \cdot \eta_g^{0.107} \cdot g^{-0.131}$	Hikita et al. (1980)
$\frac{\epsilon_g}{(1-\epsilon_g)} = 0.4 \left[\frac{U_g \eta_l}{\sigma}\right]^{0.87} \left[\frac{\eta_l^4 g}{\rho_l \sigma^3}\right]^{-0.27} \left[\frac{\rho_g}{\rho_l}\right]^{0.17}$ or $\frac{\epsilon_g}{(1-\epsilon_g)} = 0.4\ U_g^{0.87} \cdot \rho_l^{0.1} \cdot \rho_g^{0.17} \cdot \sigma^{-0.06} \cdot \eta_l^{-0.21} \cdot g^{-0.27}$	Hammer et al. (1984)
$\frac{\epsilon_g}{(1-\epsilon_g)} = 1.44\ U_g^{0.58} \cdot \rho_g^{0.12} \cdot \sigma^{-0.16\,\exp(-P)}$ P in MPa σ in mN/m	Idogawa et al. (1985)
$\epsilon_g = 296\ U_g^{0.44} \cdot \rho_l^{-0.98} \cdot \sigma^{-0.16} \cdot \rho_g^{0.19} + 0.009$	Reilly et al. (1986)
$\frac{\epsilon_g}{(1-\epsilon_g)} = 0.059\ U_g^{0.8} \cdot \rho_g^{0.17} \cdot (\sigma/72)^{-0.22\,\exp(-P)}$ P in MPa σ in mN/m U_g in cm/s	Idogawa et al. (1987a)

bubble column with small dimensions ($H/D<5$, $D<0.15$ m) or with porous-plate-type spargers, will in general lead to a considerable overestimation of these parameters.

Gas Holdup Estimation on the Basis of Empirical Equations

Among many gas holdup equations reported by Shah et al. (1982), most were developed on the basis of atmospheric data, usually only for one gas, thus they do not incorporate any influence of gas density. Recently, however, some equations were developed that are claimed to incorporate the influence of gas density (Table 2). All these equations account for the influence of gas velocity on gas holdup by assuming that gas holdup increases less than linear proportionally [$\epsilon_g \approx U_g^{0.4\text{–}0.6}$ or $\epsilon_g/(1-\epsilon_g) \approx U_g^{0.6\text{–}0.9}$] to the superficial gas velocity (U_g), which is a characteristic of the churn turbulent regime. From Figures 1 and 2, however, it appears that gas holdup in water and n-heptane first increases almost linearly with increasing gas velocity (which is a characteristic of the homogeneous bubble flow regime) until above a (pressure- or gas-density-dependent) gas velocity the slope of ϵ_g vs. U_g changes and the gas holdup is no longer linearly proportional to U_g. These results indicate that the flow regime transition is a function of gas density (as will be discussed in the next section). Obviously these trends do not follow from the equations in Table 2, and consequently none of these equations are expected to be successful for predicting gas holdup at both low and high pressures. This assumption is confirmed by a comparison (Table 4) between the gas holdup predicted from the above mentioned empirical equations and our gas holdup results (in water, mono-ethylene glycol, and n-heptane) combined with a selection (Table 3) of the published experimental gas holdup results. The selected experimental data were such that the three previously mentioned criteria were all fulfilled to prevent specific influences of the sparger design or column dimensions on the gas holdup.

Table 3. Selected Gas Holdup Data

Liquid	Reference
n-Butanol	Krishna et al. (1991); Zahradník et al. (1982)
Cyclohexane	van Dierendonck (1970); Küsters (1976)
n-Heptane	Coulon (1970); Tarmy et al. (1984); Wilkinson (1991)
Methanol	Küsters (1976)
Mono-ethylene glycol	Bach (1977); Krishna et al. (1991); Wezorke (1986); Wilkinson (1991)
n-Octanol	Bach (1977); Küsters (1976)
1,2 Propylene glycol	Wezorke (1986)
Tetrabromo ethane	Bach (1977)
Water	Akita and Yoshida (1973); Argo and Cova (1965); Hikita et al. (1980); Krishna et al. (1991); Petukhov and Kolokol'tsev (1965); Reilly et al. (1986); Wilkinson (1991); Zahradník et al. (1982)

Regime Transition

Influence of gas density on gas holdup and regime transition

From gas disengagement experiments (for example, Wezorke, 1986; Krishna et al., 1991) and visual observations in bubble columns it is known that the transition from the homogeneous bubble flow regime (which is characterized by small slow rising bubbles) to the churn turbulent regime is caused by the formation of large fast-rising bubbles. As a consequence, all factors that influence the occurrence (coalescence and bubble breakup) and formation (sparger design) of these large bubbles can in principle also influence the transition velocity (and thus gas holdup). It is well known, for instance, that the formation of large bubbles can be delayed to a higher value of U_{trans} (and gas holdup) when the coalescence rate is reduced by the addition of an electrolyte. Wilkinson and van Dierendonck (1990a) have demonstrated that a higher gas density increases the rate of bubble breakup especially for large bubbles. This higher bubble breakup counteracts the formation of large bubbles due to coalescence. As a result, at high pressure mainly small bubbles occur in homogeneous bubble flow, until for very high gas holdup the transition to the churn turbulent regime occurs because coalescence then becomes so important that larger bubbles are formed. In other words, the fact that both the gas holdup and the transition velocity in a bubble column are affected by pressure can be attributed to the influence of gas density on bubble breakup.

Influence of liquid properties on gas holdup and regime transition

Similarly, the influence of liquid properties such as viscosity and surface tension on the regime transition (and gas holdup) will also be qualitatively discussed. From articles on bubble breakup and bubble coalescence it is known that a higher liquid viscosity promotes coalescence of large bubbles (Crabtree and Bridgwater, 1971), whereas bubble breakup rates decrease for a high liquid viscosity. Consequently, on the basis of both these trends it can be argued that many (very) large bubbles occur especially in bubble columns with high-viscosity liquids. Furthermore, because of the high rise velocity of these large bubbles, the gas holdup in viscous liquids is expected to be low (which is confirmed by our experiments in mono-ethylene glycol), whereas the transition to the churn turbulent regime (due to the formation of large bubbles) already occurs at very low gas velocity. Apart from the liquid viscosity, the value of the surface tension also has a pronounced influence on bubble breakup (and thus on gas holdup). When the surface tension is lower, fewer large bubbles occur due to the fact that surface tension forces oppose deformation and bubble breakup (for example, Otake et al., 1977; Walter and Blanch, 1986). Consequently, the occurrence of large bubbles is minimal due to bubble breakup especially in those liquids that combine a low surface tension with a low liquid viscosity. As a result, relatively high gas holdup values are to be expected for such liquids (for example, *n*-heptane, Figure 2), whereas the transition to the churn turbulent regime due to the formation of large bubbles is delayed to relatively high gas holdup values.

Correlating Gas Holdup Incorporating Regime Transition

Recently, the influence of gas density (and thus pressure) on the transition velocity (U_{trans}) from homogeneous to churn turbulent flow was also reported by Krishna et al. (1991). Furthermore, Krishna et al. (1991) correlated the gas holdup for low and high gas densities, by using a separate equation for the homogeneous regime (below U_{trans}) and the churn turbulent regime. For superficial gas velocities below, the transition velocity (U_{trans}, the value of which depends on gas density), the gas holdup increases proportionally to the superficial gas velocity that leads to the following simple equation for the homogeneous bubble flow regime

For $U_g < U_{\mathrm{trans}}$,

$$\epsilon_g = A\,U_g \text{ (homogeneous bubble flow regime),} \qquad (1)$$

whereas for higher superficial gas velocities (in the churn turbulent regime) it was assumed (on the basis of gas disengagement experiments) that the gas velocity in excess of the transition velocity ($U_g - U_{\mathrm{trans}}$) flows through the bubble column in the form of large bubbles:

For $U_g > U_{\mathrm{trans}}$,

$$\epsilon_g = A\,U_{\mathrm{trans}} + C(U_g - U_{\mathrm{trans}})^{0.8} \text{ (churn turbulent regime),} \qquad (2)$$

The values of A, C and U_{trans} necessary to calculate the gas holdup by these equations were estimated on the basis of experimental data by Krishna et al. (1991), but no empirical equation was proposed to predict these values. The results of this equation, however, indicate that this approach is more likely to lead to a gas holdup equation that incorporates the influence of gas density accurately than equations in Table 2. Therefore, to develop a generalized gas holdup equation (incorporating both gas and liquid properties) a similar, but slightly different, approach was followed from Krishna et al. (1991). First of all, Eq. 1 can be rewritten to:

For $U_g < U_{\mathrm{trans}}$,

$$\epsilon_g = U_g / U_{s.b.} \text{ (homogeneous bubble flow regime),} \qquad (3)$$

where $U_{s.b.}$ is the rise velocity of small bubbles. On the basis of dimension analysis, the following equation for the value of $U_{s.b.}$ is proposed:

$$\frac{We_l}{Re_l} = \frac{U_{s.b.}\eta_l}{\sigma} = C \cdot \left[\frac{\sigma^3 \rho_l}{g\,\eta_l^4}\right]^{n1} \cdot \left[\frac{\rho_l}{\rho_g}\right]^{n2} \qquad (4)$$

For $U_g > U_{\mathrm{trans}}$,

$$\epsilon_g = U_{\mathrm{trans}}/U_{s.b.} + \frac{U_g - U_{\mathrm{trans}}}{U_{l.b.}}$$

$$\text{(transition and heterogeneous regime).} \qquad (5)$$

This equation shows that the total gas holdup is the sum of the contribution of the gas holdup before and after the regime transition where $U_{l.b.}$ is the contribution due to the rise velocity of large bubbles, for which $U_{l.b.} > U_{s.b.}$ must always hold. To comply with this requirement the following dimensionless equation was chosen:

$$\frac{\eta_l U_{l.b.}}{\sigma} = \frac{\eta_l U_{s.b.}}{\sigma} + C\left[\frac{\eta_l (U_g - U_{\mathrm{trans}})}{\sigma}\right]^{n3} \cdot \left[\frac{\sigma^3 \rho_l}{g\,\eta_l^4}\right]^{n4} \cdot \left[\frac{\rho_l}{\rho_g}\right]^{n5} \qquad (6)$$

To complete this set of equations an equation for U_{trans} is also required. As explained before, this transition velocity decreases for an increase in liquid viscosity and surface tension, whereas an increase in gas density has the opposite effect. Therefore, an empirical equation was sought that depends on the liquid properties as well as on the gas density:

$$U_{\mathrm{trans}} = F(\sigma, \rho_g, \rho_l, \eta_l) \qquad (7)$$

Optimal values for the parameters in Eqs. 4, 6 and 7 were then determined with the aid of nonlinear regression analysis. For this regression analysis, the experimental gas holdup data in Table 3 in combination with the gas holdup results in deionized water, *n*-heptane and mono-ethylene glycol were used. The regression analysis was repeated with different equations for U_{trans}. The best result with the smallest average error is obtained with:

$$\frac{U_{s.b.}\eta_l}{\sigma} = 2.25\left[\frac{\sigma^3 \rho_l}{g\,\eta_l^4}\right]^{-0.273} \cdot \left[\frac{\rho_l}{\rho_g}\right]^{0.03} \qquad (8)$$

$$\frac{\eta_l U_{l.b.}}{\sigma} = \frac{\eta_l U_{s.b.}}{\sigma} + 2.4\left[\frac{\eta_l(U_g - U_{\text{trans}})}{\sigma}\right]^{0.757} \cdot \left[\frac{\sigma^3 \rho_l}{g\,\eta_l^4}\right]^{-0.077} \cdot \left[\frac{\rho_l}{\rho_g}\right]^{0.077} \qquad (9)$$

$$\frac{U_{\text{trans}}}{U_{s.b.}} = \epsilon_{g.\text{trans}} = 0.5 \cdot \exp(-193\,\rho_g^{-0.61} \cdot \eta_l^{0.5} \cdot \sigma^{0.11}) \quad \text{(SI Units)} \qquad (10)$$

A parity plot for these equations is shown in Figure 7. These equations show an improved fit for the gas holdup data as compared to all other recent gas holdup equations (Table 2) that incorporate the influence of gas density in different ways. The average error of this equation was just under 10%, while the largest error was 40% (errors defined in Table 4). The range within which the physical constant has been varied for these equations is: $\sigma = 0.02\text{-}0.073$ N/m, $\eta_l = 0.0004\text{-}0.055$ Pa·s, $\rho_l = 683\text{-}2{,}960$ kg/m^3, $\rho_g = 0.09\text{-}38$ kg/m^3. Finally, although Eqs. 3 and 5 can be used to predict the gas holdup with a higher accuracy than previous equations, there are still a number of effects that can in practice complicate an accurate gas holdup estimation. Possible causes for such effects and possible limitations of Eqs. 3 and 5 will therefore be discussed in the next sections.

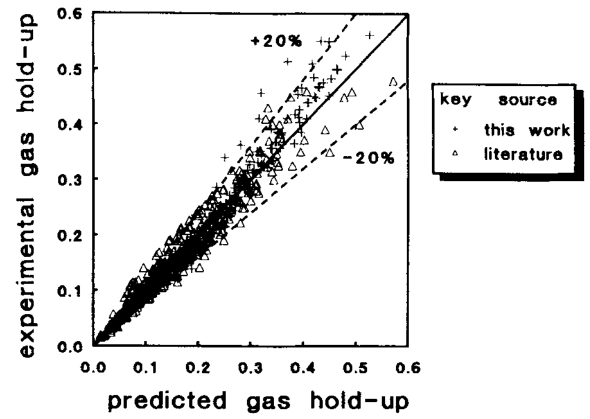

Figure 7. Parity plot of gas holdup predicted by Eqs. 3 and 5 vs. experimental data from this work and data in Table 3.

Miscellaneous Influences on Gas Holdup

Influence of liquid composition on gas holdup

The influence of liquid properties (especially liquid viscosity and surface tension) on gas holdup has been studied by using experimental gas holdup results obtained from different pure liquids. Other authors, however, have sometimes varied liquid properties for gas holdup experiments by mixing liquids in different proportions or by adding surfactants. Gas holdup results obtained by these two different methods often are not the same. In the first case when only pure liquids are used, gas holdup decreases with an increase in liquid viscosity (due to the formation of many large bubbles). In the case of liquid mixtures, however, Bach and Pilhofer (1978) (also Godbole et al., 1982; Khare and Joshi, 1990) determined that gas holdup does not decrease if the viscosity of water is increased by adding glycerol, CMC, or glucose, but passes through a maximum (Figure 8). Presumably this initial increase in gas holdup is due to the fact that the coalescence rate in mixtures is lower than pure liquids.

Futhermore, adding an electrolyte to water is also known to hinder coalescence with the result that smaller bubbles occur and a higher gas holdup than pure water (this is also confirmed by the relatively high gas holdup values as determined in the HPO process liquid, Figure 9). These influences are obviously not incorporated in Eqs. 3 and 5 (that are based on gas holdup in pure liquids). Consequently, the use of these equations for predicting the gas holdup in noncoalescing liquid mixtures and electrolyte solutions will lead to a conservative (low) estimate of the gas holdup for both low and high pressures.

Influence of liquid velocity and gas holdup

The liquid velocity in a bubble column is usually relatively low, and consequently its influence on gas holdup is often claimed to be negligible (for example, Akita and Yoshida, 1973, with $U_l < 0.04$ m/s) or small (Kelkar et al., 1983). In principle, however, liquid flowing cocurrently upward will lower gas holdup, while a countercurrent liquid flow will increase gas holdup (Otake et al., 1981). There are different methods of correlating gas holdup data as a function of the liquid velocity (Hills, 1976). However, for low liquid velocities, as in bubble columns, an approach based on the slip velocity is preferable (Hills, 1976). For this approach it is assumed that the velocity difference between the bubbles and the liquid (the slip velocity U_s) is only a function of the gas holdup:

Table 4. Statistical Analysis of Gas Holdup Equations

Equations	Hikita (1980)	Hammer (1984)	Idogawa (1985)	Reilly (1986)	Idogawa (1987a)	This Work
Avg. Rel. Error, %*	20	18	29	55	65	10
Max. Rel. Error, %**	97	81	223	445	210	40

$$^{*}\sum^{N}\frac{\sqrt{(\epsilon_{g.p} - \epsilon_{g.p})^2}}{\epsilon_{g.e}N} \cdot 100\%$$

$$^{**}\left[\frac{\epsilon_{g.p} - \epsilon_{g.e}}{\epsilon_{g.e}}\right]_{\text{max.}} \cdot 100\%$$

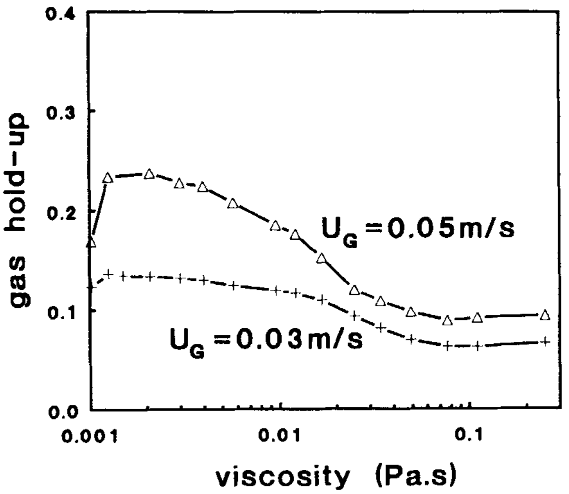

Figure 8. Gas holdup in glycerol/water mixtures vs. viscosity: experimental data of Bach and Pilhofer, 1978.

$$U_s=\frac{U_g}{\epsilon_g}\pm\frac{U_l}{1-\epsilon_g}=F(\epsilon_g)\qquad \begin{array}{l} -\text{ for cocurrent flow}\\ +\text{ for countercurrent flow}\end{array} \tag{11}$$

In other words, it is assumed that gas holdup data for each gas/liquid system with and without liquid flow, all lie on single curves when this slip velocity is plotted vs. gas holdup. If the liquid velocity remains below 0.3 m/s, Hills (1976) demonstrated that this assumption is relatively accurate. In Figure 10, our experimental gas holdup results for the HPO process liquid (in the 0.15-m-dia. column, for which $U_l=0$) are recalculated to the so-called slip velocity and plotted vs. the gas holdup in combination with the slip velocity calculated from experiment in the large-scale ($V_r \simeq 60$ m^3) industrial HPO reactor (with $U_l \simeq 0.11$ m/s, $U_g \simeq 0.14$ m/s, $\rho_g \simeq 12$ kg/m^3). Apart from indicating the usefulness of the slip velocity approach, this comparison (in Figure 10) again shows that the influence of gas density in large-scale ($D \gg 0.15$ m, $H/D>5$), industrial, high-pressure bubble columns is comparable (within the limits of accuracy) to results obtained in 0.15-m-dia. (and $H/D>5$) bubble columns. Furthermore, although the gas holdup equations (Eqs. 3 and 5) have been determined for $U_l=0$, it is also possible to estimate the gas holdup when the liquid velocity is not negligible by a simple (graphical) procedure that is based on the above mentioned slip velocity approach. First, calculate the function $F(\epsilon_g)$ in Eq. 11 from the present gas holdup equations with $U_s=F(\epsilon_g)=U_g/\epsilon_g$ (because $U_l=0$). Then, draw the result vs. ϵ_g in a figure together with Eq. 11: $U_g/\epsilon_g \pm U_l/(1-\epsilon_g)$ for the value of U_g and U_l at which the gas holdup is to be determined. The gas holdup for this specific combination of U_g and U_l is then obtained by determining (graphically or numerically) the gas holdup value at which the two lines intersect.

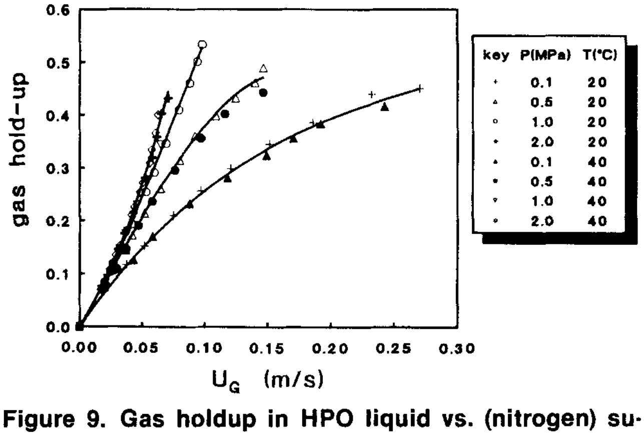

Figure 9. Gas holdup in HPO liquid vs. (nitrogen) superficial gas velocity at 4 pressures and two temperatures: $D=0.15$ m, $H=1.5$ m, sparger ring with 19 holes of 10 mm; composition of HPO liquid, 0.48 mol/L H_3PO_4, 0.71 mol/L $NH_3OH.\ H_2PO_4$, 0.92 mol/L $NH_4.\ H_2PO_4$, 1.14 mol/L NH_4NO_3.

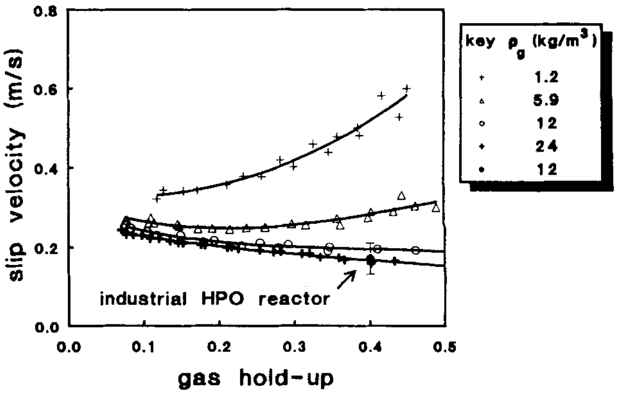

Figure 10. Slip velocity vs. gas holdup in HPO process liquid as a function of gas density, combined with the slip velocity calculated from data of the industrial HPO bubble-column reactor.

Influence of suspended solids on gas holdup

Bubble columns are used frequently for chemical reactions that require suspended catalyst particles. Numerous examples have been listed by Shah et al. (1982) and Mashelkar (1970) including biochemical reactions, hydrogenation of liquid petroleum fractions, and coal liquefaction, while a number of books and review articles have been published recently (Pandit and Joshi, 1984 and 1986; Fan, 1989; Beenackers and van Swaaij, 1986) that deal with the estimation of parameters necessary for the design of slurry bubble columns (and other three-phase reactors). From these publications it has become clear that the addition of solids to a bubble column will in general lead to a small decrease in gas holdup (Reilly et al., 1986) and the formation of larger bubbles; an exception occurs for very small particles (0–100 µm) at low weight fractions (usually below 4% by weight). For such conditions, Khare and Joshi (1990) have given numerous examples that can be explained only by assuming that coalescence of bubbles is hindered by small particles and that this leads to smaller bubbles and higher gas holdup values. No systematic research, however, has been done as to the influence of pressure on gas holdup in a three-phase environment. The scarce gas holdup values that have been reported in the literature for three-phase bubble columns under pressure, however, are generally relatively high (Blum and Toman, 1977; Kürten, 1982; Brown, 1984; Tarmy et al., 1984; Clark, 1990). Consequently, it appears that the significant increase in gas holdup that occurs in two-phase bubble columns (due to the higher gas density) will also occur in three-phase bubble columns. Therefore, we assume that the influence of gas density and liquid properties as predicted by Eqs. 3 and 5 is also correct for slurry bubble columns, although differences will occur especially for small (coalescence hindering) particles and when a high volume fraction of solids is applied.

Influence of temperature on gas holdup

In the literature it has been demonstrated that a temperature increase in general leads to a higher gas holdup (for example, Bach and Pilhofer, 1978; Küsters, 1976). A change in temperature can have an influence on gas holdup, for a number of reasons: due to the influence of temperature on the physical properties of the liquid, as well as the influence of temperature on the vapor pressure. Both the surface tension and the liquid viscosity decrease if the temperature is increased and, as follows from Eqs. 3 and 5, both a lower surface tension and a lower liquid viscosity will lead to a higher gas holdup, which can partly explain high gas holdup values measured at high temperatures by various researchers. The fact that a higher temperature also increases the vapor pressure of the liquid has two effects. First, if the feed gas to a bubble column is not presaturated, the evaporation of the liquid phase can lead to a substantial increase in the volumetric gas flow, which obviously also increases the gas holdup (for example, Zou et al., 1988; Wilkinson and van Dierendonck, 1990b). Second, a higher vapor pressure can influence the gas density. The latter effect is especially noticeable if the molecular weight of the liquid phase is very different from the molecular weight of the gas phase. In other words, if the gas holdup equations (Eqs. 3 and 5) are used for predicting gas holdup at high temperatures, the correct gas density must be determined, and the superficial gas velocity must be corrected for possible evaporation effects.

Mass Transfer

The interfacial area and the liquid volumetric mass transfer coefficient are considered the most important design parameters for bubble columns. Consequently, many authors have experimentally determined the values of these parameters and empirical equations for the estimation of the interfacial area (Akita and Yoshida, 1974) or the volumetric mass transfer coefficient (Akita and Yoshida, 1973; Hikita et al., 1981; Hammer et al., 1984) have been proposed. However, these published empirical equations do not account for any influence of pressure. Although it is obvious that both the increase in gas holdup and the decrease of the bubble size with increasing pressure lead to a higher interfacial area (and $k_l a$):

$$a = 6\epsilon_g / d_s \tag{12}$$

Therefore, the interfacial area (and $k_l a$) of a high-pressure bubble column will be underestimated by the published empirical equations. Thus, in principle, an accurate estimation of both the interfacial area and the volumetric mass transfer coefficient for high-pressure conditions will at present require experiments at high pressure, which are much more complicated than those at atmospheric pressure. Consequently, a procedure whereby the volumetric mass transfer coefficient or the interfacial area can be estimated on the basis of atmospheric results would be advantageous. Wilkinson (1991) proposed such a procedure (based on experiments) for the estimation of the liquid volumetric mass transfer coefficient at high pressure on the basis of atmospheric results:

$$\frac{k_l a\ (\text{high pressure})}{k_l a\ (\text{atmospheric})} = \left[\frac{\epsilon_g\ (\text{high pressure})}{\epsilon_g\ (\text{atmospheric})}\right]^n \quad \text{with } n = 1.0\text{–}1.2 \tag{13}$$

This equation shows that the liquid volumetric mass transfer coefficient increases with increasing pressure in spite of the fact that a (small) decrease of the liquid-side mass transfer coefficient (k_l) is expected at higher pressure. A (small) decrease in the liquid-side mass transfer coefficient for higher pressure (and thus smaller average bubble diameters) is expected mainly because the liquid-side mass transfer coefficient is known to decrease for smaller bubble size (Calderbank and Moo-Young, 1961). The magnitude of the liquid volumetric mass transfer coefficient increase (as predicted by Eq. 13) depends on the ratio of the gas holdup (which can be estimated on the basis of Eqs. 3 and 5). To calculate the volumetric mass transfer coefficient at high pressure from Eq. 13, a value for the volumetric mass transfer coefficient at atmospheric pressure is required. In principle, such data can be obtained from published empirical equations that are all based on atmospheric data (for example, Akita and Yoshida, 1973; Hammer et al., 1984) or from published experimental values of the volumetric mass transfer coefficient.

Similarly, the increase in interfacial area with increasing pressure obviously (Eq. 12) depends partly on the relative extent to which the gas holdup increases with increasing pressure (which can be calculated from Eqs. 3 and 5) and partly on the decrease in bubble size with increasing pressure. The extent to which the Sauter mean bubble diameter decreases with increasing pressure is estimated (Wilkinson, 1991) to be proportional to $\rho_g^{-0.11}$ on the basis of photos. Combining this result with Eq. 12 leads to:

$$\frac{a\ (\text{high pressure})}{a\ (\text{atmospheric})} = \frac{\epsilon_g\ (\text{high pressure})}{\epsilon_g\ (\text{atmospheric})} \cdot \left[\frac{\rho_g\ (\text{high pressure})}{\rho_g\ (\text{atmospheric})}\right]^{0.11} \tag{14}$$

In principle, Eq. 14 is the desired equation for estimating the interfacial area for high pressure on the basis of atmospheric data. If, however, no experimental data are available, then the interfacial area can be estimated by using an empirical equation (for example, Akita and Yoshida, 1974; van Dierendonck, 1970; Wilkinson, 1991) for the bubble diameter in combination with Eqs. 3, 5 and 14. However, there are considerable differences among various empirical equations for predicting d_s. Consequently, the value of the estimated Sauter mean bubble diameter (and interfacial area) depends on the choice of the empirical equations (differences can easily be as much as a factor of two for apparently equal conditions). Furthermore, in electrolyte solutions the bubble size is generally much smaller than in pure liquids.

So far it is argued that at higher pressure a favorable increase of both the liquid volumetric mass transfer coefficient ($k_l a$) and the interfacial area are to be expected. The gas-phase mass transfer coefficient (k_g), however, decreases with increasing pressure (see Eq. 15) due to the fact that the gas diffusion coefficient is inversely proportional to pressure ($D_g \approx 1/P$, provided the pressure is not too high, Reid et al., 1988).

$$k_g \approx \mathbb{D}_g^n \approx P^{-n} \tag{15}$$

The value of n in this equation is usually found to be close to 0.5 (Cho and Wakao, 1988). As a consequence, the favorable increase in the interfacial area (and $k_l a$) due to a higher pressure will in some cases (especially for soluble gases) be limited by the decrease in the gas-phase mass transfer coefficient at higher pressures. For such cases, however, reactors other than bubble columns are presumably preferable. Finally, it is also possible that the performance of a high-pressure bubble column is limited for relatively slow reactions, because for slow reactions a decrease of the liquid-phase reaction volume (as a result of the higher gas holdup at high pressure) can lead to a lower overall reaction rate.

Liquid Dispersion

In many articles, liquid dispersion in bubble columns has been described on the basis of the one-dimensional axial dispersion model. One of the main results of these articles is that the value of the one-dimensional dispersion coefficient increases considerably with increasing column diameter, whereas the column height has no influence on its value. Consequently, for the scaleup of experimental results obtained in a small pilot plant to a larger industrial bubble-column reactor, it is relevant to determine whether the axial dispersion coefficient influences the reactor performance. Obviously, this requires a procedure to estimate the axial dispersion coefficient. From the results of Wilkinson (1991), it appears that a reasonably accurate estimate can be made of the liquid dispersion coefficient on the basis of published empirical equations for both low and high pressures if the following correction factor is used:

$$E_l\ (\text{high pressure}) = E_l\ (\text{atmospheric}) \cdot \frac{\epsilon_l\ (\text{atmospheric})}{\epsilon_l\ (\text{high pressure})} \tag{16}$$

Gas-Phase Dispersion

Like the liquid-phase dispersion, the gas-phase dispersion is also usually described on the basis of a one-dimensional axial dispersion coefficient. The value of this gas-phase dispersion coefficient (that is also much influenced by the column diameter, Eqs. 17 and 18) can have a considerable influence on the performance of a bubble column, especially in the case of high gas-phase conversions (for example, Deckwer, 1976). At present, however, the value of this gas-phase dispersion coefficient has been determined only for a limited range of conditions (at atmospheric pressure), while no experiments have been carried out at high pressure. The (atmospheric) results obtained by a number of authors were correlated (Mangartz and Pilhofer, 1980) to the following equation that depends markedly on the bubble swarm velocity (U_g/ϵ_g) and thus on gas holdup:

$$E_g = 50 \cdot D^{1.5} \cdot \left[\frac{U_g}{\epsilon_g}\right]^3 \tag{17}$$

Joshi (1982) determined a different equation (based on the results of a number of authors, for air/water systems) for predicting E_g that also depends (to a lesser extent) on the value of the gas holdup:

$$E_g = 110 \frac{U_g^2}{\epsilon_g} D^2 \tag{18}$$

If these equations are used to estimate E_g at high pressure, a lower value is predicted as compared to atmospheric pressure (especially for Eq. 17) due to the fact that the gas holdup increases with increasing pressure. In principle, such a trend is not unexpected because at low pressure (in the churn turbulent regime) a much wider range of bubble sizes occurs in a bubble column (as compared to high pressure) with large differences in rise velocity which leads to a large residence time distribution (large E_g) of these bubbles. However, more research is required to determine whether this trend is predicted accurately by Eq. 17 or Eq. 18.

Conclusions

From the present investigation on the scaleup of pressurized bubble columns, the following conclusions can be drawn:

- Experimental gas holdup data obtained in a bubble column with a minimum diameter of 0.15 m, $H/D > 5$ or $H > 1$–3 m and for perforated-plate or single-nozzle spargers with hole diameters in excess of 1–2 mm can be used to estimate the gas holdup in a larger bubble column without correction for low and high pressures.
- The influence of gas density and liquid properties on gas holdup are correlated by Eqs. 3 and 5 with an average accuracy of ±10% provided pure coalescing liquids are used.
- For noncoalescing water/electrolyte solutions or noncoalescing liquid mixtures, gas holdup increases above the value predicted by Eqs. 3 and 5 for low and high pressures.
- The increase in the liquid volumetric mass transfer coefficient and the interfacial area as a result of an increase in gas density can be estimated with Eqs. 13 and 14.
- The advantageous influence of an increased liquid volumetric mass transfer coefficient and interfacial area due to an increase of the gas density can (especially for soluble gases) be limited partly by a decrease in the gas-phase mass transfer coefficient (Eq. 15) at higher pressures.
- The axial dispersion coefficient for the gas phase is likely to be influenced by pressure; no experimental data, however, are available at present for pressures above atmospheric. Therefore, whether Eq. 17 or Eq. 18 predict the gas-phase dispersion coefficient accurately at high pressure is still unknown.

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Notation

a = interfacial area, m^2/m^3
C = constant
d_s = Sauter mean bubble diameter, m
D = column diameter, m
$\mathbb{D}_g$ = gas-phase diffusion coefficient, m^2/s
E_g = gas-phase dispersion coefficient, m^2/s
E_l = liquid-phase dispersion coefficient, m^2/s
g = gravitational acceleration, m/s^2
H = column height, m
$k_l a$ = liquid volumetric mass transfer coefficient, L/s
k_g = gas-side mass transfer coefficient, m/s
P = pressure, Pa
U_g = superficial gas velocity, m/s
U_l = superficial liquid velocity, m/s
U_s = slip velocity, defined by Eq. 11, m/s
$U_{\rm trans}$ = velocity at regime transition, Eq. 10, m/s
V_r = volume of bubble column reactor, m^3

Greek letters

$\Delta\rho$ = density difference between gas and liquid, kg/m^3
ϵ_g = gas holdup
$\epsilon_{g,\rm trans}$ = gas holdup at regime transition
ϵ_l = liquid holdup
η_g = gas viscosity, Pa·s
η_l = liquid viscosity, Pa·s
ρ_g = gas density, kg/m^3
ρ_l = liquid density, kg/m^3
σ = surface tension, N/m

Subscripts/superscripts

e = experimental value
g = gas
l = liquid
l.b. = large bubbles
p = predicted value
s.b. = small bubbles

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