A New Approach for the Prediction of Gas Holdup in Bubble Columns Operated under Various Pressures in the Homogeneous Regime

Stoyan NEDELTCHEV* and Adrian SCHUMPE
Institute of Technical Chemistry, TU Braunschweig,
Hans-Sommer-Strasse 10, 38106 Braunschweig, Germany

Keywords: Bubble Columns, Gas Holdup, Bubble Shape, Hydrodynamics, Organic Liquids

A new semi-theoretical approach for prediction of gas holdups in the homogeneous flow regime is suggested. The model is based on two different expressions for the gas-liquid interfacial area. It is argued that in the case of oblate ellipsoidal bubbles (formed in the homogeneous regime) some correction factor should be introduced in order to render both expressions equivalent. The correction term is correlated to Eötvös number and a dimensionless gas density ratio. The method was capable of predicting 386 experimental gas holdups measured in 21 organic liquids, 17 liquid mixtures and tap water with an average relative error of 9.6%.

Introduction

Bubble columns are widely used in the chemical industry because of their simple construction and low energy requirement. A variety of absorption, oxidation, chlorination, oxychlorination, alkylation, carbonylation, hydroformylation, etc. reactions are carried out in these gas–liquid reactors. In the case of commercially important applications, bubble columns are usually operated at elevated pressures.

One of the keys to successful design and scale-up is the estimation of the gas holdup, which is strongly influenced by the behavior of gas bubbles and the liquid flow pattern in the column. The bubble characteristics (bubble size, bubble rise velocity and bubble wake phenomena) determine the gas holdup, the interfacial area and the gas residence time. The gas holdup also affects the flow pattern in the churn-turbulent regime, since liquid recirculation is driven by non-uniform radial gas holdup profiles.

The gas holdup \( \varepsilon_G \) is defined as the ratio of gas volume to total dispersion volume:

\[
\varepsilon_G = \frac{V_G}{V_G + V_L}
\]  

For constant cross-sectional area and batchwise operated liquid, the overall gas holdup can be expressed as follows:

\[
\varepsilon_G = \frac{H - H_0}{H}
\]

where \( H \) is the aerated liquid height and \( H_0 \) is the clear liquid height. The gas holdup \( \varepsilon_G \) can be estimated also as the superficial gas velocity \( u_G \) divided by the bubble rise velocity \( u_B \) (Guy et al., 1986). This illustrates that the \( \varepsilon_G \) value determines the gas residence time in the bubble bed. It is also a measure of the interfacial area available for phase interaction.

Smaller bubble size increases the overall \( \varepsilon_G \) by a lower rise velocity. The stability of small bubbles may vary from pure liquids to binary mixtures. In water, for example, salt and alcohol additives stabilize the bubbles by formation of an ionic or polar layer at the interface which depresses the coalescence rate (Schügerl et al., 1977; Oels et al., 1978; Hikita et al., 1980, Kelkar et al., 1983). In this case also the transition to heterogeneous flow is shifted to higher \( u_G \) values. Some of the major effects on the gas holdup \( \varepsilon_G \) will be discussed in the following subsections.

1. Influence of Various Factors on Gas Holdup

1.1 Effect of column design

Hughmark (1967) found the gas holdup \( \varepsilon_G \) to depend on column diameter \( D_c \) only when it was smaller than 0.1 m. Shah et al. (1982), Reilly et al. (1986) and Kumar et al. (1997) have concluded that for \( D_c \geq 0.15 \) m, column size has no effect on the overall gas holdup. Discussions about the nature of \( \varepsilon_G \) dependence on \( D_c \)
are given in Koide et al. (1979), Haque et al. (1986), Idogawa et al. (1987), Wilkinson and van Dierendonck (1990), Wilkinson et al. (1992) and Reilly et al. (1994).

It is generally accepted that for bubble columns with height-to-diameter ratios higher than 5, the gas holdup \( \varepsilon_G \) is independent of this ratio (Wilkinson et al., 1992). The authors concluded that the clear liquid height \( H_L \) had also no effect on radial holdup distribution.

The gas sparger design has a pronounced influence on initial bubble diameter, however, in the churn-turbulent regime attained at a higher \( u_G \), bubble coalescence and break-up play an important role and the bubble size will be insensitive to sparger design. At low \( u_G \) value, the gas holdup \( \varepsilon_G \) generated by the sintered plate is larger than that of the perforated plate (Kumar et al., 1997). Saxena and Chen (1994) reported a pronounced increase in \( \varepsilon_G \) values with a decrease in orifice diameter, however, the influence is small at hole diameters larger than \( 1 \times 10^{-3} \) m (Wilkinson et al., 1992). Schumpe and Deckwer (1982) argue that in the slug flow regime the gas holdup becomes independent of the sparger type.

### 1.2 Effect of superficial gas velocity

Gas holdup \( \varepsilon_G \) generally increases with an increase in the superficial gas velocity \( u_G \), the effect being more pronounced in the dispersed bubble regime than in the churn-turbulent regime. The gas holdup \( \varepsilon_G \) values are usually correlated using the simple power relationship: \( \varepsilon_G \sim u_G^n \) (Reilly et al., 1994). The value of the exponent \( n \) provides an indication of the flow regime. As a rule, the \( n \) values corresponding to homogeneous bubbling conditions are higher than those for the turbulent bubbling regime. Shah et al. (1982) report that values of \( n \) in the range of 0.7–1.2 have been utilized to describe \( \varepsilon_G \) data falling into the homogeneous regime, and values of \( n \) within the range of 0.4–0.7 have been used to fit \( \varepsilon_G \) data belonging to the churn-turbulent regime.

### 1.3 Effect of gas properties and pressure

Wilkinson et al. (1992) and Lin et al. (1998) concluded that the gas holdup \( \varepsilon_G \) increases appreciably with an increase in pressure. This conclusion is valid for both low and high viscosity liquids. Idogawa et al. (1986, 1987) and Wilkinson and Van Dierendonck (1990) showed that gas holdup \( \varepsilon_G \) increased not only with an increase in pressure but also with an increase in gas density. The gas viscosity dependence is not that clear (Saxena and Chen, 1994).

### 1.4 Effect of Liquid Properties

Generally, gas holdup \( \varepsilon_G \) is strongly influenced by the liquid viscosity \( \mu_L \). The effect of this property is, however, rather controversial. The increase of \( \mu_L \) enhances bubble coalescence, however, bubble rise velocity \( u_G \) decreases with increasing \( \mu_L \). Measurements with methanol, ethanol and acetone revealed that \( \mu_L \) has a negligible influence on \( \varepsilon_G \) (Idogawa et al., 1986, 1987). Eissa and Schügerl (1975) examined also the effect of \( \mu_L \) on \( \varepsilon_G \). They observed an initial increase of \( \varepsilon_G \) with \( \mu_L \) followed by a decrease, and finally an increase.

The liquid surface tension \( \sigma_L \) has a profound influence on the gas holdup \( \varepsilon_G \) and its distribution (Kumar et al., 1997). It can be qualitatively concluded that \( \varepsilon_G \) increases with decreasing \( \sigma_L \) due to the lower \( u_G \). The effect of \( \sigma_L \) in systems containing pure liquids is, however, only slight. Idogawa et al. (1986, 1987) reported that the effect of \( \sigma_L \) was dependent on pressure. At atmospheric pressure, \( \varepsilon_G \) became constant and independent of \( \sigma_L \). Kelkar et al. (1987) found that \( \varepsilon_G \) remained unaltered when \( \sigma_L \) was varied using different concentrations of the same alcohol.

### 1.5 Gas holdup correlations

Most of the works in bubble columns dealing with gas holdup measurement and prediction are based on deep bubble beds (Hughmark, 1967; Akita and Yoshida, 1973; Kumar et al., 1976; Hikita et al., 1980; Kelkar et al., 1983; Behkish et al., 2007). A unique work concerned with gas holdup \( \varepsilon_G \) under homogeneous bubbling conditions was published by Hammer et al. (1984). The authors presented an empirical relation valid for pure organic liquids at \( u_G \leq 0.02 \text{ m·s}^{-1} \). Idogawa et al. (1987) proposed an empirical correlation for gas densities up to \( 121 \text{ kg·m}^{-3} \) and \( u_G \) values up to \( 0.05 \text{ m·s}^{-1} \). Kularkar et al. (1987) derived a relation to compute \( \varepsilon_G \) in the homogeneous flow regime in the presence of surface-active agents. By using a large experimental data set, Syeda et al. (2002) have developed a semi-empirical correlation for \( \varepsilon_G \) prediction in both pure liquids and binary mixtures. Pošarac and Tekić (1987) proposed a reliable empirical correlation which enables the estimation of gas holdup in bubble columns operated with dilute alcohol solutions. A number of gas holdup correlations were summarized by Hikita et al. (1980), Saxena and Chen (1994) and Kemoun et al. (2001). Recently, Gandhi et al. (2007) have proposed a support vector regression-based correlation for prediction of overall gas holdup in bubble columns. As many as 1810 experimental gas holdups measured in various gas–liquid systems were satisfactorily predicted (average absolute relative error: 12.1\%). The method is entirely empirical.

In the empirical correlations, different dependencies on the physicochemical properties and operating conditions are implicit. This is primarily because of the limited number of liquids studied and different combinations of dimensionless groups used. For example, the gas holdup correlation proposed by Akita and Yoshida (1973) can be safely employed only within the set of systems used in the authors’ experiments (water, methanol and glycerol solutions). The effect of column diameter \( D \), was not verified and the presence of this parameter in the dimensionless groups is thus only formal. In general, empirical correlations can
describe ε₆ data only within limited ranges of system properties and working conditions. In this article a new semi-theoretical approach for ε₆ prediction is suggested which is expected to be more generally valid.

2. Mathematical Model for Gas Holdup Prediction

The specific gas–liquid interfacial area, defined as the surface area available per unit volume of the dispersion, is related to gas holdup ε₆ and the Sauter-mean bubble diameter dₛ by the following simple relation:

$$a = \frac{6\varepsilon₆}{dₛ} \tag{3}$$

Strictly speaking, Eq. (3) (especially the numerical coefficient 6) is valid only for spherical bubbles (Schügerl et al., 1977).

The formula for calculation of the interfacial area depends on the bubble shape. Excellent diagrams for bubble shape determination are available in the books of Clift et al. (1978) and Fan and Tsuchiya (1990) in the form of log–log plots of the bubble Reynolds number Reₖ vs. the Eötvös number E₀ with due consideration of the Morton number Mo. A comparison among the experimental conditions used in this work and the above-mentioned standard plots reveals that the formed bubbles are no longer spherical but oblate ellipsoidals that follow a zigzag upward path as they rise. Vortex formation in the wake of the bubbles is also observed. The specific interfacial area a of such ellipsoidal bubbles is a function of the number of bubbles Nₖ, the bubble surface Sₖ and the total dispersion volume Vₜotal (Painmanakul et al., 2005; Nedeltchev et al., 2006a, 2006b, 2007):

$$a = \frac{NₖSₖ}{Vₜotal} = \frac{NₖSₖ}{AH} \tag{4}$$

where A denotes the column cross-sectional area. The number of bubbles Nₖ can be deduced from the bubble formation frequency fₖ and bubble residence time (Painmanakul et al., 2005):

$$Nₖ = fₖ \frac{H}{uₖ} = \frac{Qₙ}{Vₙ} \frac{H}{uₖ} \tag{5}$$

where Qₙ is the volumetric gas flow rate, uₖ is the bubble rise velocity (see Eq. (16)) and Vₙ is the bubble volume (see Eq. (17)). The substitution of Eq. (5) into Eq. (4) yields:

$$a = \frac{fₖSₖ}{Auₖ} \tag{6}$$

If some dimensionless correction factor f_c due to the bubble shape differences is introduced, then Eqs. (3) and (6) might be considered equivalent:

$$\frac{6\varepsilon₆}{dₛ} = f_c \frac{Sₖ}{Auₖ} \tag{7}$$

Rearrangement of Eq. (7) yields:

$$\varepsilon₆ = f_c \frac{dₛ}{6Auₖ} \tag{8}$$

The surface Sₖ of an ellipsoidal bubble can be calculated as follows (Nedeltchev et al., 2006a, 2006b, 2007):

$$Sₖ = \pi \frac{1}{2} \left[ 1 + \left( \frac{h}{l} \right)^2 - \frac{1}{2e} \ln \left( \frac{1+e}{1-e} \right) \right] \tag{9}$$

where e is the bubble eccentricity:

$$e = \sqrt{1 - \left( \frac{h}{l} \right)^2} \tag{9a}$$

An oblate ellipsoidal bubble is characterized by its length l (major axis of the ellipsoid) and its height h (minor axis of the ellipsoid). The ellipsoidal bubble length l and height h can be estimated by the formulas derived by Tadaki and Maeda (1961) and Terasaka et al. (2004):

For 2 < Ta < 6:

$$l = \frac{dₛ}{1.14Ta^{-0.186}} \tag{10a}$$

$$h = 1.3dₛTa^{-0.352} \tag{10b}$$

For 6 < Ta < 16.5:

$$l = \frac{dₛ}{1.36Ta^{-0.28}} \tag{11a}$$

$$h = 1.85dₛTa^{-0.56} \tag{11b}$$

where

$$Ta = ReₖMo^{0.23} \tag{12}$$

$$Reₖ = \frac{dₖuₖDₖ}{\mu_L} \tag{13}$$

$$Mo = \frac{gH^4}{\rho_L \sigma_L} \tag{14}$$
It is worth noting that the major axis of a rising oblate ellipsoidal bubble is not always horizontally oriented (Yamashita et al., 1979). The same holds for the minor axis of a rising oblate ellipsoidal bubble, i.e., it is not necessarily vertically oriented (Akita and Yoshida, 1974).

Equations (10a)–(14) were used to calculate both \( l \) and \( h \) values under the operating conditions examined. The Morton number \( Mo \) is the ratio of viscosity force to the surface tension force. The Tamaki number \( Ta \) characterizes the extent of bubble deformation; the \( Ta \) values fell always in one of the ranges specified above. This fact can be regarded as additional evidence that the bubbles formed under the operating conditions examined are really ellipsoidal.

Equations (10a)–(14) imply that one needs to know \( a \text{ priori} \) the bubble equivalent diameter \( d_e \). Literature frequently assumes that \( d_e \) can be approximated by the Sauter-mean bubble diameter \( d_s \). The latter was estimated by means of the correlation of Wilkinson et al. (1994):

\[
\frac{g \rho_g d_s^2}{\sigma_L} = 8.8 \left( \frac{\rho_L \mu_L}{\sigma_L} \right)^{-0.04} \left( \frac{\sigma_L}{g \mu_L^2} \right)^{-0.12} = \left( \frac{\rho_L}{\rho_g} \right)^{0.22} \\
(15)
\]

Equation (15) implies that the bubble size decreases as the superficial gas velocity \( u_g \) or the gas density \( \rho_g \) (operating pressure \( P \)) increases. The calculated \( d_s \) values for all liquids examined imply an ellipsoidal shape.

The bubble rise velocity \( u_e \) also takes part in the calculation of both \( l \) and \( h \) values. It was estimated from Mendelson’s (1967) correlation:

\[
u_e = \sqrt{\frac{2 \sigma_L}{\rho_L d_e} + \frac{gd_e}{2}} \\
(16)
\]

This equation is particularly suitable for the case of ellipsoidal bubbles. Equation (16) along with Eq. (15) (for \( d_s \) estimation) was used also to calculate the bubble Reynolds number \( Re_b \) (Eq. (13)) needed for the estimation of both \( l \) and \( h \) values.

The bubble equivalent diameter \( d_e \) of an ellipsoidal bubble can be also calculated by assuming a sphere of equal volume as the ellipsoidal bubble:

\[
V_b = \frac{\pi d_e^3}{6} = \frac{4}{3} \pi \left( \frac{l}{2} \right)^3 h \\
(17)
\]

or

\[
d_e = \left( \frac{l^2 h}{2} \right)^{1/3} \\
(17a)
\]

Estimating the characteristic length of ellipsoidal bubbles with the same surface-to-volume ratio (the same \( d_e \) value as calculated from Eq. (15)) required an iterative procedure but led to only insignificantly different values than simply identifying the equivalent diameter \( d_e \), when applying Eqs. (10a)–(10b) or (11a)–(11b). In other words, the differences between bubble diameters estimated by Eq. (15) and Eq. (17a) are negligibly small.

The main objective of this paper is to test the applicability of this semi-theoretical approach and especially to derive a correlation for the correction term \( f_e \) introduced in Eq. (8). Many liquids covering a large spectrum of physicochemical properties, different gas distributor layouts, and different gases at operating pressures up to 4 MPa are considered.

3. Experimental Results Considered

Numerous experimental gas holdups (386 values) were collected from two bubble columns. The first stainless steel column (\( D_o = 0.102 \) m, \( H_o = 1.3 \) m) was equipped with three different gas distributors: perforated plate, \( 19 \times \sigma 1 \times 10^{-3} \) m (D1), single hole, \( 1 \times \sigma 4.3 \times 10^{-3} \) m (D2) and single hole, \( 1 \times \sigma 1 \times 10^{-3} \) m (D3) (Jordan and Schumpe, 2001). In the second plexiglass column (\( D_o = 0.095 \) m, \( H_o = 0.85 \) m) the gas was always introduced through a single tube of \( 3 \times 10^{-3} \) m in ID (D4) (Öztürk et al., 1987). The \( \varepsilon_g \) values were measured in 21 organic liquids, 17 liquid mixtures and tap water (see Tables 1 and 2).

In Tables 1 and 2 the different combinations of liquids, gases, gas distributors and operating pressures are listed. In the 0.095 m in ID bubble column equipped with sparger D4 every liquid or liquid mixture was aerated with air. Table 1 shows that in the case of few liquids (carbon tetrachloride, tetralin, toluene and xylene) some other gases have been used. In the case of 0.102 m in ID bubble column no air was used, only nitrogen and helium.

The gas holdups \( \varepsilon_g \) in 1-butanol, ethanol (96%), decalin, toluene, gasoline, ethylene glycol and tap water were recorded by means of differential pressure transducers in the 0.102 m stainless steel bubble column operated at pressures up to 4 MPa. The following relationship was used:

\[
\varepsilon_g = \frac{\Delta P_{no \ gas} - \Delta P_{gas}}{\Delta P_{no \ gas}} \\
(18)
\]

where \( \Delta P \) is the pressure difference between the readings of both lower (at 0 m) and upper (at 1.2 m) pressure transducers. The subscript “no gas” denotes the pressure difference for the clear liquid, whereas the subscript “gas” denotes the pressure difference for the dispersion.
The gas holdups $\varepsilon_G$ in all other liquids and liquid mixtures were estimated by visually observing the dispersion height under ambient pressure in the 0.095 m in ID bubble column and then applying Eq. (2). The upper limit of the homogeneous regime (transition gas velocity $u_{trans}$) was estimated by the formulas of Reilly et al. (1994).

### Table 1 Properties of the organic liquids and tap water (293.2 K)

<table>
<thead>
<tr>
<th>Liquid</th>
<th>$D_c$ [m]</th>
<th>Gas Sparger</th>
<th>Gases Used</th>
<th>$P$ [MPa]</th>
<th>$\rho_L$ [kg m$^{-3}$]</th>
<th>$\mu_L \times 10^{-3}$ [Pa s]</th>
<th>$\sigma_L \times 10^{-3}$ [N m$^{-1}$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Acetone</td>
<td>0.095</td>
<td>D4</td>
<td>air</td>
<td>0.1</td>
<td>790</td>
<td>0.327</td>
<td>23.1</td>
</tr>
<tr>
<td>Anilin</td>
<td>0.095</td>
<td>D4</td>
<td>air</td>
<td>0.1</td>
<td>1022</td>
<td>4.4</td>
<td>43.5</td>
</tr>
<tr>
<td>Benzene</td>
<td>0.095</td>
<td>D4</td>
<td>air</td>
<td>0.1</td>
<td>879</td>
<td>0.653</td>
<td>28.7</td>
</tr>
<tr>
<td>1-Butanol</td>
<td>0.095 0.102</td>
<td>D1, D2 D4</td>
<td>N$_2$, air, He</td>
<td>0.1–4.0</td>
<td>809</td>
<td>2.94</td>
<td>24.6</td>
</tr>
<tr>
<td>Carbon tetrachloride</td>
<td>0.095 D4</td>
<td>air, He, H$_2$, $\text{CO}_2$</td>
<td>0.1</td>
<td>1593</td>
<td>0.984</td>
<td>26.1</td>
<td></td>
</tr>
<tr>
<td>Cyclohexane</td>
<td>0.095</td>
<td>D4</td>
<td>air</td>
<td>0.1</td>
<td>778</td>
<td>0.977</td>
<td>24.8</td>
</tr>
<tr>
<td>Decalin</td>
<td>0.102</td>
<td>D1, D2 D4</td>
<td>N$_2$, He</td>
<td>0.1–4.0</td>
<td>884</td>
<td>2.66</td>
<td>32.5</td>
</tr>
<tr>
<td>1,2-Dichloroethane</td>
<td>0.095</td>
<td>D4</td>
<td>air</td>
<td>0.1</td>
<td>1234</td>
<td>0.82</td>
<td>29.7</td>
</tr>
<tr>
<td>1,4-Dioxane</td>
<td>0.095</td>
<td>D4</td>
<td>air</td>
<td>0.1</td>
<td>1033</td>
<td>1.303</td>
<td>32.2</td>
</tr>
<tr>
<td>Ethanol (96%)</td>
<td>0.102 D1, D2, D3</td>
<td>N$_2$, He</td>
<td>0.1–4.0</td>
<td>793</td>
<td>1.24</td>
<td>22.1</td>
<td></td>
</tr>
<tr>
<td>Ethanol (99%)</td>
<td>0.095</td>
<td>D4</td>
<td>air</td>
<td>0.1</td>
<td>791</td>
<td>1.19</td>
<td>22.1</td>
</tr>
<tr>
<td>Ethyl acetate</td>
<td>0.095</td>
<td>D4</td>
<td>air</td>
<td>0.1</td>
<td>900</td>
<td>0.461</td>
<td>23.5</td>
</tr>
<tr>
<td>Ethylbenzene</td>
<td>0.095</td>
<td>D4</td>
<td>air</td>
<td>0.1</td>
<td>867</td>
<td>0.669</td>
<td>28.6</td>
</tr>
<tr>
<td>Ethylene glycol</td>
<td>0.095 0.102</td>
<td>D1, D2 D4</td>
<td>N$_2$, air, He</td>
<td>0.1–4.0</td>
<td>1112</td>
<td>19.9</td>
<td>47.7</td>
</tr>
<tr>
<td>Gasoline</td>
<td>0.102</td>
<td>D1</td>
<td>N$_2$</td>
<td>0.1–4.0</td>
<td>692</td>
<td>0.464</td>
<td>21.6</td>
</tr>
<tr>
<td>Ligroin A (b.p. 90–110°C)</td>
<td>0.095</td>
<td>D4</td>
<td>air</td>
<td>0.1</td>
<td>714</td>
<td>0.470</td>
<td>20.4</td>
</tr>
<tr>
<td>Ligroin B (b.p. 100–140°C)</td>
<td>0.095</td>
<td>D4</td>
<td>air</td>
<td>0.1</td>
<td>729</td>
<td>0.538</td>
<td>21.4</td>
</tr>
<tr>
<td>Methanol</td>
<td>0.095</td>
<td>D4</td>
<td>air</td>
<td>0.1</td>
<td>790</td>
<td>0.586</td>
<td>22.2</td>
</tr>
<tr>
<td>Nitrobenzene</td>
<td>0.095</td>
<td>D4</td>
<td>air</td>
<td>0.1</td>
<td>1203</td>
<td>2.02</td>
<td>38.1</td>
</tr>
<tr>
<td>2-Propanol</td>
<td>0.095</td>
<td>D4</td>
<td>air</td>
<td>0.1</td>
<td>785</td>
<td>2.42</td>
<td>21.1</td>
</tr>
<tr>
<td>Tap water</td>
<td>0.095 0.102</td>
<td>D1, D2 D3, D4</td>
<td>N$_2$, air, He</td>
<td>0.1–4.0</td>
<td>1000</td>
<td>1.01</td>
<td>72.7</td>
</tr>
<tr>
<td>Tetralin</td>
<td>0.095</td>
<td>D4</td>
<td>N$_2$, air, $\text{CO}_2$</td>
<td>0.1</td>
<td>968</td>
<td>2.18</td>
<td>34.9</td>
</tr>
<tr>
<td>Toluene</td>
<td>0.095 0.102</td>
<td>D1, D2 D4</td>
<td>N$_2$, air, He</td>
<td>0.1–4.0</td>
<td>866</td>
<td>0.58</td>
<td>28.5</td>
</tr>
<tr>
<td>Xylene</td>
<td>0.095</td>
<td>D4</td>
<td>N$_2$, air, He, H$_2$, $\text{CO}_2$</td>
<td>0.1</td>
<td>863</td>
<td>0.63</td>
<td>28.4</td>
</tr>
</tbody>
</table>
4. Results and Discussion

Most of the gas–liquid systems given in Tables 1 and 2 were characterized with Tadaki numbers $Ta$ lower than 6 and thus Eqs. (10a)–(10b) for the estimation of both bubble length $l$ and bubble height $h$ were applied. Only in the case of ethylene glycol-(helium, air and nitrogen), 1-butanol-(helium and air) and decalin-helium the $Ta$ values exceeded 6 and then Eqs. (11a)–(11b) were used.

It was found that the dimensionless correction factor $f_c$ can be correlated successfully to both the $Eo$ number and a dimensionless gas density ratio:

$$f_c = 0.78 Eo^{-0.22} \left( \frac{\rho_G}{\rho_G^\text{ref}} \right)^{0.07}$$

$$= 0.78 \left( \frac{g (\rho_L - \rho_G) d_l^2}{\sigma_L} \right)^{-0.22} \frac{\rho_G}{1.2}^{0.07}$$

(19)

where $\rho_G^{\text{ref}}$ is the reference gas density (1.2 kg·m$^{-3}$ for air at ambient conditions: 293.2 K and 0.1 MPa). All experimental gas holdup data (386 points) were fitted with an average error of 9.6%.

The dimensionless gas density ratio is probably needed because the correlation of Wilkinson et al. (1994) was derived for pressures up to 1.5 MPa only, whereas the present data extend up to the pressure of 4 MPa. It is worth noting that Krishna (2000) also used such a dimensionless gas density ratio for correcting his correlations for large bubble rise velocity and dense-phase gas holdup.

**Figure 1** illustrates the decrease of the product $f_c (\rho_G / 1.2)^{-0.07}$ with increasing $Eo$. At smaller bubble sizes (with shapes approaching spheres), $Eo$ will be lower and thus $f_c$ higher (gradually approaching unity). It is worth noting that most of the liquids are characterized with $Eo$ values in a narrow range between 2 and 8.

**Figure 2** illustrates that the correction factor $f_c$ increases with gas density $\rho_G$ (operating pressure) leading to bubble shrinkage. For example, the correction term $f_c$ decreases in the following sequence: toluene > ethanol > decalin > 1-butanol > ethylene glycol. The smallest bubble size is formed in toluene, whereas the largest bubble size is formed in the case of ethylene glycol. When very small (spherical) bubbles are formed, the correction factor $f_c$ should be equal to unity, and both expressions for the interfacial area should become identical (see Eq. (7)).
Figures 3 and 4 exhibit that the experimental gas holdups $\varepsilon_G$ measured in 1-butanol, decalin and toluene at pressures up to 4 MPa can be predicted reasonably well irrespective of the gas distributor type.

The same result holds for ethylene glycol and tap water (see Figure 5). The successful prediction of gas holdups in ethylene glycol should be regarded as one of the most important merits of the presented method since the viscosity is much higher than that of the other liquids.

Figure 6 shows, for ethanol (96%) as an example, that the gas distributor type is not so important. The same holds for 1-butanol, decalin and toluene (Figures 3–5) and tap water. This fact is in agreement with the work of Wilkinson et al. (1992), who stated that once the hole size of the gas distributor is greater than 1–2 $\times$ 10$^{-3}$ m, then it has no significant effect on the gas holdup and the mass transfer coefficient.

Eight organic liquids and tap water have been aerated not only with air or nitrogen but also with other gases (helium, hydrogen and carbon dioxide). Figure 7 shows that the developed model is capable of predicting satisfactorily the experimental gas holdups at these operating conditions.

Figure 8 shows that the model predicts reasonably well the experimental gas holdups measured in 15 organic liquids at ambient pressure. This fact should be regarded as further evidence that by the introduction of a correction term, the presented method becomes generally applicable.

Figure 9 exhibits that the proposed method for gas holdup prediction along with the new correction factor (Eq. (19)) is also applicable to various liquid mixtures.

Tables 1 and 2 and Figures 3–9 reveal that our approach is applicable to 21 organic liquids, 17 liquid mixtures and tap water covering the following ranges of the main physicochemical properties:

$692 \leq \rho_L \ [\text{kg} \cdot \text{m}^{-3}] \leq 1593$

$0.327 \times 10^{-3} \leq \mu_L \ [\text{Pa} \cdot \text{s}] \leq 19.9 \times 10^{-3}$

$20.4 \times 10^{-3} \leq \sigma_L \ [\text{N} \cdot \text{m}^{-1}] \leq 47.7 \times 10^{-3}$

4.1 Statistical comparison among the gas holdup predictions based on the current approach and some well-known empirical correlations

A statistical comparison among the gas holdup predictions based on our approach, and the ones based on the empirical correlation of Hammer et al. (1984) (which is considered the most reliable one in the
literature thus far) has been performed. This dimensionless correlation reads as follows:

\[
\frac{\varepsilon_G}{1 - \varepsilon_G} = 0.4 \left( \frac{\mu_G \mu_L}{\sigma_L} \right)^{0.87} \left( \frac{\mu_L^2 \rho_L}{\rho_l \sigma_L^2} \right)^{-0.27} \left( \frac{\rho_G}{\rho_l} \right)^{0.37}
\]

(20)

It is worth noting that the gas holdup predictions based on Hammer et al.’s (1984) correlation are closer to the experimental values as compared with the other correlations summarized by Wilkinson et al. (1992) and Kemoun et al. (2001).

In addition, our gas holdup correlation was compared with the well-known empirical relationship derived by Akita and Yoshida (1974) for \( \varepsilon_G < 0.14 \):

\[
8.887 \left( \frac{gD_i^3}{V_L} \right) ^{-0.083} \left( \frac{gD_i^3 \rho_l}{\sigma_L} \right) ^{-0.125} \varepsilon_G^{1.11} = \frac{\mu_G}{\sqrt{gD_i}}
\]

(21)

In Table 3 comparisons of all three approaches are summarized in terms of both the average relative errors (ARE) and the maximum relative errors (MRE) based on 21 organic liquids, 17 liquid mixtures and tap water.

Fig. 3 Parity plot for gas holdups in 1-butanol and decalin sparged with nitrogen through gas distributors D1 and D2 at various pressures

Fig. 4 Parity plot for gas holdups in toluene and decalin sparged with nitrogen through gas distributors D1 and D2 at various pressures
In both cases (organic liquids/tap water and liquid mixtures) our approach yields better predictions than the ones produced by means of the empirical correlations of Akita and Yoshida (1974) and Hammer et al. (1984). Especially in the case of 21 organic liquids and tap water, the average relative error (ARE) based on our predictions is over two times lower as compared to AREs in the other two cases.

It should be noted that the statistical comparison presented in Table 3 is not based on all 386 data points but on a representative sample of 250 gas holdup values (213 points for randomly selected organic liquids/tap water and 37 points for all liquid mixtures).

Table 3 reveals that in the homogeneous flow regime our semi-theoretical approach (see Eqs. (3)–(17)) improves the gas holdup predictions and turns out to be the most reliable one. It is worth underlying that our approach is applicable at $u_G \leq u_{trans}$, where the transition gas velocity $u_{trans}$ can be predicted by the correlations of Reilly et al. (1994). In this work, the $u_{trans}$ values were always less than 0.04 m·s$^{-1}$.

**Conclusions**

A new semi-theoretical approach for gas holdup $\varepsilon_G$ calculation in the homogeneous regime of bubble
column operation is suggested. In this simple model, the geometrical characteristics (length and height) of the ellipsoidal bubbles play a central role. It is found that the introduction of some correction factor is mandatory in order to render equivalent two expressions for the interfacial area: the classical one strictly valid for the rigid spheres, and the one for oblate ellipsoidal bubbles. The model presented implies that \( \varepsilon_G \) is directly proportional to the correction factor, bubble formation frequency, bubble surface and Sauter-mean bubble diameter, and inversely proportional to both the bubble rise velocity and column cross-section (Eq. (8)). The correction factor \( f_c \) was correlated to both the \( \text{Eötvös} \) number and a dimensionless gas density ratio. The method is capable of predicting experimental gas holdups \( \varepsilon_G \) measured in 21 organic liquids, 17 liquid mixtures, and tap water with an average relative error of 9.6%.

### Acknowledgements

Dr. Stoyan Nedeltchev expresses his gratitude to the Alexander von Humboldt Foundation (Germany) for the postdoctoral...
fellowship (2005-2007) that made this research work possible.

Nomenclature

- $A$ = cross-sectional area of the reactor $[m^2]$
- $a$ = specific interfacial area (referred to dispersion volume) $[m^{-1}]$
- $D_i$ = column diameter $[m]$
- $d_i$ = bubble equivalent diameter $[m]$
- $d_0$ = Sauter-mean bubble diameter $[m]$
- $Eo$ = Eotvos number, Eq. (19) $[—]$
- $e$ = bubble eccentricity, Eq. (9a) $[—]$
- $f_b$ = bubble formation frequency $[s^{-1}]$
- $f_c$ = correction factor $[—]$
- $g$ = gravitational acceleration $[m \cdot s^{-2}]$
- $H$ = aerated liquid height $[m]$
- $h$ = height of an ellipsoidal bubble $[m]$
- $H_e$ = Morton number, Eq. (14) $[—]$
- $N_b$ = number of bubbles in the dispersion $[—]$
- $n$ = exponent $[—]$
- $P$ = operating pressure $[Pa]$
- $Q_{ce}$ = gas flow rate $[m^3 \cdot s^{-1}]$
- $R_{nb}$ = bubble Reynolds number, Eq. (13) $[—]$
- $S_p$ = bubble surface $[m^2]$
- $T_a$ = Tadaki number, Eq. (12) $[—]$
- $u_w$ = bubble rise velocity $[m \cdot s^{-1}]$
- $u_{sc}$ = superficial gas velocity $[m \cdot s^{-1}]$
- $u_{trans}$ = transition gas velocity $[m \cdot s^{-1}]$
- $V_b$ = bubble volume $[m^3]$
- $V_{ei}$ = gas volume $[m^3]$
- $V_l$ = liquid volume $[m^3]$
- $V_{disp}$ = dispersion volume $[m^3]$
- $\Delta P$ = pressure difference (0–1.2 m) $[Pa]$
- $\varepsilon_0$ = gas holdup $[—]$
- $\mu_l$ = liquid viscosity $[Pa \cdot s]$
- $\nu_l$ = liquid kinematic viscosity $[m^2 \cdot s^{-1}]$
- $\rho_0$ = gas density $[kg \cdot m^{-3}]$
- $\rho_{ref}$ = reference gas density (air at ambient conditions) $[kg \cdot m^{-3}]$
- $\rho_l$ = liquid density $[kg \cdot m^{-3}]$
- $\sigma_l$ = liquid surface tension $[N \cdot m^{-1}]$

Literature Cited


Oels, U., J. Lucke, R. Buchholz and K. Schügerl; “Influence of Gas
Distributor Type and Composition of Liquid on the Behavior of a Bubble Column Bioreactor,” *Germ. Chem. Eng.*, 1, 115–129 (1978)


