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Novel gas holdup and regime transition correlation for twophase bubble columns

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Abstract. The gas holdup is dimensionless parameter of fundamental and practical importance in the operation, design and scale-up of bubble columns. Unfortunately, the many relationships between the bubble column fluid dynamic parameters and the various variables characterizing the system make it difficult to find general correlations for the precise estimation of the gas holdup. Wilkinson et al. (1992), in their pioneering paper, proposed a correlation to predict the gas holdup in industrial-scale bubble columns, based on the physical properties of the phases and the operating conditions. However, this correlation lacks in generality, as it does not take in account the bubble column design. In this paper, we propose a generalization of the Wilkinson et al. (1992) gas holdup correlation to take into also the bubble column design parameters. Starting from considerations concerning the flow regime transition, corrective parameters are included to account for the effects introduced by the gas sparger openings, the bubble column aspect ratio and the bubble column diameter. The proposed correlation has been found to predict fairly well previously published gas holdup and flow regime transition data.

1. Introduction

Two-phase bubble columns are multiphase reactors where a gas phase is dispersed into a liquid phase in the form of dispersed bubbles or of "coalescence-induced" bubbles. The simplest bubble column configuration consists in a vertical cylinder, in which the gas enters at the bottom-through a gas sparger—and the liquid phase is supplied in batch mode or it may be led in either co-currently or counter-currently to the upward gas stream. Despite the simple bubble column design, complex fluid dynamics interactions and coupling between the phases—manifesting in the prevailing flow regimes [1]-exist. Therefore, the correct design and operation of these reactors rely on the proper prediction of the "reactor-scale" and the "bubble-scale" fluid dynamic properties. Among the many fluid dynamic properties, the gas holdup (ε_G)—a dimensionless parameter defined as the volume of the gas phase divided by the total volume of the system-is a "reactor-scale" fluid dynamic property of fundamental and practical importance. The gas holdup determines the mean residence time of the dispersed phase and, in combination with the size distribution of the dispersed phase, the interfacial area for the rate of interfacial heat and mass transfer. The many relations between the bubble column fluid-dynamic parameters and the variables characterizing the system make it difficult to find general correlations for the precise estimation of the gas holdup curve, which is the analytical relation between the gas superficial velocity U_G (viz. the ratio between the volumetric flow rate and the cross-sectional area) and $\varepsilon_G (\varepsilon_G = f(U_G))$. Numerous correlations are available in literature, although due to the fluid dynamic complexity of the problem none of them can be either considered of general validity or

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applied to a wide range of geometrical parameters and operating conditions. Wilkinson et al. [2], in their pioneering paper, proposed a correlation to predict the gas holdup in industrial bubble columns, based on the physical properties of the phases and the operating conditions (i.e., high pressure operation). The main idea is that the total gas holdup is the sum of the contribution of the gas holdup before and after the flow regime transition between the homogeneous and the transition flow regime (see ref. [3] for a discussion concerning the flow regime transitions). The homogeneous flow regime—associated with small superficial gas velocities—is referred as the flow regime where only "non-coalescence-induced" bubbles exist (e.g. as detected by the gas disengagement technique); the transition flow regime, is identified by the appearance of the "coalescence-induced" bubbles [4] and is characterized by large flow macro-structures with large eddies and a widened bubble size distribution. Thus, accordingly with Wilkinson et al. [2], the gas holdup can be estimated based on:

$$\varepsilon_{G} = \begin{cases} \frac{U_{G}}{U_{non-coalescence induced "bubbles}} & U_{G} < U_{G,trans} \\ \frac{U_{G,trans}}{U_{non-coalescence induced "bubbles}} + \frac{U_{G} - U_{G,trans}}{U_{ncoalescence induced "bubbles}} & U_{G} > U_{G,trans} \end{cases}$$
(1)

Where $U_{non-coalescence induced"}$ bubbles and $U_{coalescence induced"}$ bubbles refer to the rising velocity of the "non-coalescence induced" bubbles and the "coalescence induced" bubbles, respectively. In particular, the terms in Eq. (1) have been evaluated by Wilkinson et al. [2], by using a non-linear regression of the experimental data obtained in two bubble columns having different inner diameters ($d_c = 0.15$ and $d_c = 0.23$ m), operated at different operating pressures and filled by different liquid phases (n-heptane, monoethylene glycol, and water):

$$U_{G,trans} = 0.5U_{non-coalescence induced "bubbles} \exp\left(-193\rho_{G}^{-0.61}\mu_{L}^{0.5}\sigma^{0.11}\right)$$
(2)

$$U_{non-coalescence induced "bubbles} = 2.25 \frac{\sigma}{\mu_L} \left(\frac{\sigma^3 \rho_L}{g \mu_L^4}\right)^{-0.273} \left(\frac{\rho_L}{\rho_G}\right)^{0.03}$$
(3)

$$U_{\text{"coalescence induced "bubbles}} = \frac{\sigma}{\mu_{l}} \begin{cases} \frac{\mu_{l}U_{\text{"non-coalescence induced "bubbles}}}{\sigma} + \frac{\sigma}{\rho} \\ + 2.4 \left[\frac{\mu_{l}(U_{g} - U_{g,trans})}{\sigma} \right]^{0.757} \left(\frac{\sigma^{3}\rho_{l}}{g\mu_{l}^{4}} \right)^{-0.077} \left(\frac{\rho_{l}}{\rho_{s}} \right)^{0.077} \end{cases}$$
(4)

In Eqs. (2-4) μ_L is the liquid phase viscosity, ρ_L is the liquid phase density and σ is the surface tension. Unfortunately, the above correlation, despite is considered the state-of-the-art in bubble columns, it lacks in generality, as it does not take in account the bubble column design parameters (i.e., sparger openings, aspect ratio and bubble column inner diameter). In this respect, the reader should refer to our recent paper concerning the importance of bubble column design parameters in gas holdup correlations as well as scaling up criteria [4].

Therefore, in this paper, we propose a generalization of the Wilkinson et al. [2] gas holdup correlation to take in account both the physical properties and the operating conditions, as well as the bubble column design. The proposed correlation aims to provide an engineering practical tool able to estimate the global bubble column parameters. Starting from considerations concerning the flow regime transition, corrective parameters are included in the gas holdup correlation to account for the effects introduced by the gas sparger openings, the bubble column aspect ratio and the bubble column diameter.

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2. The proposed scheme of correlation

A general and precise correlation for the gas holdup depends on the many parameters characterizing the system: (a) the pressure and the temperature (p_c and T_c); (b) the gas and liquid superficial velocities (U_G and U_L); (c) the physical and the interface properties; (d) the bubble column design parameters (i.e., bubble column inner diameter, d_c , bubble column height, H_c , initial liquid height, H_0 , and diameter of the gas sparger openings, d_o , see Figure 1).



Bubble column inner diameter: d_c

Figure 1. Bubble column design parameters.

The "*reactor-scale*" and the "*bubble-scale*" fluid dynamic properties are also related to the prevailing flow regime. Therefore, a gas holdup correlation should take into account the flow regime transition point between the homogeneous flow regime and the transition flow regime ($U_{G,trans}$). This concept has the underlying assumption that systems having similar flow regime transition points show similar "*reactor-scale*" behaviour. Hence, a correlation for the gas holdup would be as follows:

$$\varepsilon_{g} = f\left(U_{g}, U_{L}, U_{g,trans}, p_{c}, T_{c}, \mu_{L}, \mu_{g}, \rho_{L}, \rho_{g}, n, n^{*}, \sigma, d_{c}, H_{0}, d_{o}, g, ...\right)$$
(5)

In Eq. (5), *n* and n^* are the actual and the critical concentration of active compounds [4]. Indeed, the fluid dynamics in bubble columns filled with binary liquid phases cannot be entirely modelled by the bulk physical properties and the properties of the interface should be considered [5, 6]. This issue, as well as the influence of U_L , has been addressed in ref. [4] and, to keep the discussion as simple as possible, their influence has been neglected in the following (viz. pure liquid phase in the batch mode). Thus, Eq. (5) simplifies to Eq. (6):

$$\varepsilon_{g} = f\left(U_{g}, U_{g,trans}, p_{c}, T_{c}, \mu_{L}, \mu_{g}, \rho_{L}, \rho_{g}, \sigma, d_{c}, H_{0}, d_{o}, g\right)$$
(6)

Now we assume that (a) the physical properties and the operating conditions and (b) the bubble column design parameters provide two different effects. Thus, from Eq. (6) follows Eq. (7):

$$\varepsilon_{g} = f\left(U_{g}, U_{g,trans}, p_{c}, T_{c}, \mu_{L}, \mu_{g}, \rho_{L}, \rho_{g}, \sigma, g, \ldots\right) \cdot f\left(d_{c}, H_{0}, d_{o}\right)$$
(7)

In the right side of Eq. (7) the geometrical parameters are the same as the ones listed in the three Wilkinson et al. [2] scale-up criteria. At this point, a scheme of correlation is selected: the dependence on the physical properties have been accounted by using the Wilkinson et al. [2] correlation:

$$\varepsilon_{G} = f_{Wilkison \ et \ al. (1992)} \left(U_{G}, p_{c}, T_{c}, \mu_{L}, \mu_{G}, \rho_{L}, \rho_{G}, \sigma, g, \ldots \right) \cdot f\left(d_{c}, H_{0}, d_{o} \right)$$
(8)

To generalise Eq. (8), the geometrical parameters are modelled by using the corresponding nondimensional groups, which are related to the fluid dynamics in the bubble column:

$$\varepsilon_{G} = f_{Wilkison \ et \ al. (1992)} \left(U_{G}, p_{c}, T_{c}, \mu_{L}, \mu_{G}, \rho_{L}, \rho_{G}, \sigma, g, \ldots \right) \cdot f \left(D_{H}^{*}, d_{o}^{*}, AR \right)$$
(9)

The non-dimensional groups in Eq. (9) are defined as follows:

• **Dimensionless bubble column height** (*AR*, *aspect ratio*). The aspect ratio is defined as the ratio between the initial liquid height, H_0 , and the diameter of the column, d_c (see ref. [4]):

$$AR = \frac{H_0}{d_c} \tag{10}$$

In the systems where the bubble sizes are not at their maximum equilibrium size (and where coalescence may occur), the liquid height will influence the extent of the coalescence. Consequently, the gas holdup would decrease with H_0 , because the higher the column the longer the time the bubbles have to coalesce and, thus, the lower the mean residence time of the gas phase. Furthermore, in shorter bubble columns, the liquid circulation patterns (that tends to decrease the gas holdup) are not fully developed and the end-effects are more evident.

• **Dimensionless sparger opening** (d_o^*) . Depending on the gas sparger opening, a bubble size (d_b) is imposed as boundary condition at the lower level of the bubble column. Hence, the diameters of the sparger openings can be related to the bubble size produced by the bubble nucleation at the sparger, estimated by the well-known Gaddis and Vogelpohl correlation [7]:

$$d_{o}^{*} = \frac{d_{b}}{d_{o}} = \left(\frac{6d_{o}\sigma}{\rho_{L}g}\right)^{1/3} / d_{o}$$
(11)

• **Dimensionless diameter** (D_{H}^{*}). The dimensionless diameter represents the quantification of the Rayleigh–Taylor instabilities [8] at the "*reactor-scale*". In particular, the dimensionless diameter is related to the Eötvös number of the slug bubbles as follows:

$$D_{H}^{*} = \frac{D_{H}}{\sqrt{\sigma / g(\rho_{L} - \rho_{G})}} \xrightarrow{Circular bubble column}} \frac{d_{c}}{\sqrt{\sigma / g(\rho_{L} - \rho_{G})}} = \frac{1}{\sqrt{\sigma / d_{c}^{2}g(\rho_{L} - \rho_{G})}} = \frac{1}{\sqrt{1 / Eo_{c}}} = \sqrt{Eo_{c}} = \sqrt{Eo_{slug-bubble}}$$
(12)

Where D_H is the hydraulic diameter, d_c is the bubble column inner diameter, σ is the surface tension, g is the acceleration due to gravity, $\rho_L - \rho_G$ is the density difference between the two phases, and $Eo_c = Eo_{slug-bubble}$ is the Eötvös number computed using the bubble column diameter, which is also the characteristic length of the slug bubbles. Bubble columns with D_H^* greater than the critical value $D_{H_cr}^* = 52$ [9] are considered to be large-diameter bubble columns. When the $d_c > D_{H_cr}^*$, the stabilizing effect of the channel wall on the interface of the Taylor bubbles decreases, and slug flow cannot be sustained anymore because of the Rayleigh-Taylor instabilities.

Thus, the terms in Wilkinson et al. [2] correlation (Eq. (1)) have been modified as follows:

$$U_{G,trans} = A U_{G,trans,Wilkison\ et\ al.(1992)} D_{H}^{+m_{1}} A R^{m_{2}} d_{o}^{+m_{3}}$$
(13)

$$U_{non-coalescence induced "bubbles} = B U_{non-coalescence induced "bubbles, Wilkison et al. (1992)} D_{H}^{* n_{1}} A R_{0}^{* n_{2}} d_{o}^{* n_{3}}$$
(14)

It worth noting that the coalescence-induced term has not been modified, as the influence of the bubble column design in the heterogeneous flow regime is lower compared with the homogeneous flow regime. In Eqs. (13-14), A, B, (m_1, m_2, m_3) and (n_1, n_2, n_3) are calibration parameters; Eqs. (13-14) are related to the Wilkinson et al. [2] correlation, by the following terms:

$$U_{G,trans,Wilkison\ et\ al.(1992)} = 0.5U_{non-coalescence\ induced\ "bubbles,Wilkison\ et\ al.(1992)} \exp\left(-193\rho_{G}^{-0.61}\mu_{L}^{0.5}\sigma^{0.11}\right)$$
(15)

$$U_{\text{"non-coalescence induced "bubbles, Wilkison et al. (1992)} = 2.25 \frac{\sigma}{\mu_L} \left(\frac{\sigma^3 \rho_L}{g \mu_L^4}\right)^{-0.273} \left(\frac{\rho_L}{\rho_G}\right)^{0.03}$$
(16)

3. The experimental dataset and methods

To calibrate and validate the proposed scheme of correlation, gas holdup and flow regime transition point data are needed. To this end, a comprehensive experimental dataset of gas holdup data has been taken from the literature. These gas holdup data have been post-processed, by using a systematic approach, to estimate the regime transition point. In the following, firstly, the method to estimate the regime transition point has been discussed (Section 3.1) and, secondly, the whole dataset has been presented (Section 3.2).

3.1. Flow regime transition analysis

Although the flow transition from the homogeneous to the transition flow regime does not happen instantaneously [36], the definition of an approximate transition point is helpful for modelling the hydrodynamic of bubble columns. In this study, we employ two methods to invetsigate the flow regime transition (as discussed in ref. [4]): (a) the swarm velocity and (b) the drift flux method. In particular, the flow regime transition point is computed as the average of the values obtained by the two methods:

$$U_{G,trans} = \frac{U_{G,trans,swarm} + U_{G,trans,Wallis}}{2}$$
(17)

$$\varepsilon_{G,trans} = \frac{\varepsilon_{G,trans,swarm} + \varepsilon_{G,trans,Wallis}}{2}$$
(18)

3.1.1. Swarm velocity method

This method, proposed by Zuber and Findlay [37], is based on the evaluation of the swarm velocity:

$$U_{swarm} = U_{g} / \varepsilon_{g}$$
(19)

In this method, the swarm velocity is plotted against the superficial gas velocity: U_{swarm} is constant in the homogeneous flow regime and starts to increase as the system enters the transition/heterogeneous flow regime ($U_{G,trans}$). The appearance of the first large bubble is responsible for the increase in the swarm velocity and is an indication of flow regime transition.

3.1.2. Wallis plot method

The drift-flux method was proposed by Wallis [38]. This method is based on the drift flux (that represents the gas flux through a surface moving with the speed of the two-phase mixture) and it is experimentally obtained as follows:

$$J_{T} = U_{G} \left(1 - \varepsilon_{G} \right) \tag{20}$$

Theoretically, the drift flux is written in terms of a parameter, U_b , whose dependence upon ε_G varies with the prevailing flow regime:

$$J_{E} = U_{b} \left(1 - \varepsilon_{G} \right) \tag{21}$$

The idea is to employ a model for U_b valid for the homogeneous flow regime and to plot J_E and J_T in the same graph as a function of ε_G . In the homogeneous flow regime J_E is equal to J_T and the transition point is, thus, defined when:

$$J_{E} = U_{b} \left(1 - \varepsilon_{G} \right) \tag{22}$$

The evaluation of U_b is a matter of discussion in the literature, as different models were proposed and applied. In this study, we follow the approach of Krishna et al. [39], which is based on the empirical model of Richardson and Zaki [40]:

$$U_{b} = u_{\infty} \left(1 - \varepsilon_{G}\right)^{n-1}$$
(23)

where *n* is fluid-dependent ($n \approx 2$ for water) and u_{∞} —the terminal velocity of an isolated bubble—should be fitted with the aid of the experimental data in the determination of the flow regime transition point. Combining Eq. (21) and Eq. (23) results:

$$J_{E} = u_{\infty} \varepsilon_{G} \left(1 - \varepsilon_{G}\right)^{n}$$
(24)

3.2. Dataset employed

Table 1 displays the dataset employed as well as the corresponding transition points, computed by the methods described in Section 3.1. It is worth noting that *AR* has been computed by using H_0 and, when this parameters was not available, by an estimation based on H_c . In data post-processing, the mid-point conditions have been used to compute the physical properties (see ref [4]).

Table 1. Dataset employed for calibration and validation of the correlation with corresponding transition point (using the methods in Section 3.1)

Reference	d_c	d_o	Free area	H_{C}	H_{θ}	$U_{G,trans}$	$\mathcal{E}_{G,trans}$
	[m]	[mm]	[%]	[m]	[m]	[m/s]	[-]
Chaumant et al. [10]	0.200	1.0	0.80	1.60	-	0.0391	0.1224
Chilekar [11]	0.290	0.5	0.46	1.60	-	0.0439	0.1796
Han and Al-Dahhan [12]	0.162	0.5	0.16	-	1.81	0.0640	0.2193
Jin et al. [13]	0.162	1.0	0.21	2.50	0.45	0.0408	0.1411
Kemoun et al. [14]	0.162	0.4	0.04	2.50	1.80	0.0498	0.1882
Krishina et al. [15]	0.160	10.0	7.42	1.20	-	0.0569	0.1263
Krishina and Ellenberger [16]	0.630	2.5	0.10	4.00	-	0.0976	0.1583
Krishina and Ellenberger [16]	0.380	0.5	-	4.00	1.70	0.0694	0.1843
Krishina et al. [17]	0.150	1.0	2.78	-	1.00	0.0285	0.1289
Kumar et al. [18]	0.154	1.0	0.51	2.72	1.15	0.0339	0.1275
Letzel et al. [19]	0.150	0.5	0.22	1.20	-	0.0524	0.1814
Ohki and Inoue [20]	0.160	3.0	1.30	3.00	0.40	0.0594	0.1299
Veera and Jochi [21]	0.385	1.0	0.42	3.20	-	0.0500	0.1365
Reilly et al. [22]	0.300	1.5	0.75	5.00	-	0.0421	0.1754
Sal et al. [23]	0.330	2.0	1.08	3.00	0.66	0.0736	0.1128
Sal et al. [23]	0.330	1.0	0.98	3.00	0.66	0.0616	0.1066
Deckwer et al. [24]	0.200	1.0	0.14	4.44	-	0.0418	0.2078
Vandu and Krishna [25]	0.380	0.5	0.48	-	1.60	0.0443	0.1941
Akita and Yoshida [26]	0.152	5.0	0.11	4.00	-	0.0313	0.0777
Patil et al. [27]	0.380	3.7	-	-	0.684	0.0730	0.1068
Patil et al. [27]	0.380	3.7	-	-	1.026	0.0744	0.1052
Patil et al. [27]	0.380	3.7	-	-	1.406	0.0764	0.1019
Godbole et al [28]	0.305	1.66		2.44	0.60	0.0590	0.1261
Gopal and Sharma [29]	0.200	6.0	0.09	-	0.80	0.0449	0.0909
Gopal and Sharma [29]	0.600	2.0	0.07	-	3.60	0.0592	0.1411
Schumpe and Grund [30]	0.300	1.0	-	-	3.60	0.0400	0.2416
Schumpe and Grund [30]	0.300	1.0	-	-	3.60	0.0600	0.1237
Wilkinson et al. [2]	0.150	2.0	-	-	1.50	0.0461	0.0918
Wilkinson et al. [2]	0.230	7.0	-	-	1.20	0.0518	0.0961
Thorat et al. [31]	0.385	1.5	0.11	-	1.54	0.0612	0.1817
Sasaki et al. [32]	0.200	1.4	0.18	2.00	0.80	0.0500	0.1500
Passos et al. [33]	0.090	0.4	-	-	2.00	0.0180	0.1129
Pjontek et al. [34]	0.102	3.18	2.24	1.80	-	0.0300	0.1616
Ruzicka et al. [35]	0.140	0.5	0.20	-	0.40	0.0360	0.2753

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4. Novel correlation: calibration and validation procedures

4.1. Correlation calibration

Based on the experimental dataset employed (viz. Table 1 data and flow regime transition points), the coefficients A, B, (m_1, m_2, m_3) and (n_1, n_2, n_3) in Eqs. (13-14) have been obtained. These coefficients have been obtained by mean of regression analysis and the results are as follows:

$$U_{G,trans} = 2.1U_{G,trans, Wilkison et al. (1992)} D_{H}^{*0.5} A R^{-0.31} d_{a}^{*0.01}$$
(25)

$$U_{non-coalescence induced "bubbles} = 0.5U_{non-coalescence induced "bubbles, Wilkison et al. (1992)} D_{H}^{*0.4} A R_{0.3}^{-0.3} d_{o}^{*-0.39}$$
(26)

4.2. Correlation validation

In this section, the proposed scheme of correlation has been compared with the experimental dataset obtained (Table 1). Figure 2 compares the experimental and the predicted flow regime transition points (in terms of the gas superficial velocity and gas holdup). The proposed correlation has been able to predict the transition points, for the different experimental setups considered, within the boundaries of $\pm 20\%$ of error. This result is an advancement compared with the previous correlation to estimate the flow regime transition point in bubble columns. Indeed, at present, the Wilkinson et al [2] correlation was not able to take into account the bubble column design (as observed in our previous studies, see for example ref. [4]).



Figure 2. Comparison between predicted and experimental transition points.

Figure 3 compares the experimental and the predicted rising velocity of the "non-coalescence induced" bubbles. According to Wilkinson et al. [2], the rising velocity of the "non-coalescence induced" bubbles is related to the rising velocity of bubbles in the homogeneous regime, so it can be easily computed by using the definition of the swarm velocity. The proposed correlation is able to predict the experimental data within the boundaries of $\pm 10\%$ of error, for the different datasets considered. Finally, Figure 4 and Figure 5 compare the experimental and the predicted gas holdup curves for some of the studies listed in Table 1. It is shown that the proposed correlation has been found to predict fairly well previously published gas holdup data. Therefore, the proposed scheme of correlation can be used as a tool to estimate the main bubble column global parameters in the design of bubble columns as a function of the operating conditions and design parameters.

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Figure 3. Comparison between predicted and experimental rising velocity of the "non-coalescence induced" bubbles



Figure 4. Comparison between predicted and experimental gas holdup curves.

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Figure 5. Comparison between predicted and experimental gas holdup curves: parity plot.

5. Conclusions

We have proposed a generalised version of the well-known Wilkinson et al. [2] gas holdup correlation. Starting from considerations concerning the flow regime transition, corrective parameters are included in the gas holdup correlation to account for the effect of the changes introduced by the sparger openings, the aspect ratio and the diameter. The proposed correlation is able to predict the gas holdup and the flow regime transitions, by taking into account the physical properties of the phases, the bubble column operating conditions, as well as the bubble column design. The proposed scheme of correlation has been calibrated and validated over a wide range of literature studies. The main outcomes of this study are as follows:

- the proposed correlation has been found to predict the rising velocity of the "*non-coalescence induced*" bubbles within the boundaries of ±10% of error;
- the proposed correlation has been found to predict previously published gas holdup and flow regime transition data within the boundaries of $\pm 20\%$ of error;
- the proposed scheme of correlation can be used as a tool to estimate the main bubble column global parameters in the design of bubble columns as a function of the operating conditions and design parameters.

On the other hand, it should be noted that the proposed correlation is valid only for two-phase bubble columns in the batch mode without the contribution of active agents. Future studies should extend the proposed scheme of correlation by taking into account: (a) the influence of the solid load is not studied; (b) co-current and counter-current bubble column; (c) the influence of active agents. In this respect, the proposed scheme of correlation can be coupled with another gas holdup correlation recently proposed by the authors in ref. [4] (which takes into account the influence of the liquid superficial velocity and the concentration of the active agents).

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