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Gas hold-up in bubble columns: influence of alcohol addition versus operation at elevated pressures

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Abstract

Measurements were carried out in a 0.15 m diameter bubble column with air–water system to which ethanol was added in concentrations ranging from 0.03 to 1 vol.%. Alcohol addition results in a significant increase in the gas hold-up, ε. This increase in ε can be attributed to a delay in the point of transition from homogeneous to heterogeneous flow regime. The model of Krishna et al. (Chem. Eng. Sci. 54 (1999), 171) was found applicable, after accounting for the influence of alcohol addition on the regime transition parameters. Experimental data of Letzel et al. (Chem. Eng. Sci. 52 (1997), 3733), obtained in a 0.15 m diameter nitrogen–water bubble column, also showed that increased system pressures results in significantly increased gas hold-up. A careful comparison of the influence of alcohol addition and that of increased pressure leads to the conclusion that in the latter case, increased system pressures besides delaying the onset of the heterogeneous flow regime also promotes the break up of 'large' bubbles. One needs to modify the model to take account of the influence of increased pressure on the rise velocity of the swarm of 'large' bubbles. © 2000 Elsevier Science S.A. All rights reserved.

Keywords: Bubble columns; Alcohol additions; Regime transition parameters; Gas hold-up

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1. Introduction

There is considerable industrial interest in design and scale up of bubble column reactors operated at elevated pressures ranging to about 4–6 MPa, in view of the many practical applications in natural gas conversion technologies [1–3]. A bubble column reactor can be operated in either the homogeneous or heterogeneous flow regime. Though laboratory scale studies are often restricted to the homogeneous bubbly flow regime, prevailing at relatively low superficial gas velocities, industrial reactors are often operated at higher gas velocities in the heterogeneous flow regime. Examination of available literature shows that increased system pressure causes a delay in transition from the homogeneous to the heterogeneous flow regime [4–10]. The physical explanation for the delay in the regime transition with increased system pressure (i.e. increased gas density) is to be found in the reduced probability of propagation of instabilities leading to delayed flow regime transition [7–10].

Fig. 1 gives a schematic representation of the influence of increased pressure on the gas hold-up, an important design parameter. The influence of elevated pressure operation is very significant, as is evidenced by examining the recently published experimental results of Letzel et al. [4] for gas hold-up measured in a bubble column of 0.15 m diameter with the nitrogen–water system (see Fig. 2a). For example, for operation at a superficial gas velocity $U = 0.2$ m/s, the gas hold-up ε increases from a value of 0.29 at $p = 0.1$ MPa to a value which is twice as large for operation at $p = 1.2$ MPa (see Fig. 2b). In this paper, we attempt to simulate the operation at elevated pressures by deliberately delaying the regime transition point by adding small amounts of alcohol to an air–water bubble column. The objective of this exercise is to examine the extent to which it is possible to model the influence of elevated pressures by just taking account of the shift in the regime transition point.

Though there is published literature on the influence of alcohol addition on the gas hold-up in bubble

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columns [11–16], we decided to carry out our own experimental investigations for the following reasons: (1) in order to compare alcohol addition experiments with the high pressure experiments of Letzel [4], we needed to perform experiments in a 0.15 m diameter column with similar gas distribution device as that used by Letzel [4] and (2) our main focus is on the churn-turbulent regime of operation whereas most of the published works are restricted to the homogeneous bubbly flow regime.

2. Experimental

Experiments were carried out in a column of 0.15 m diameter, 4 m high, with the system air–water. The column configuration is shown in Fig. 3. In order to match the experimental results of Letzel et al. [4] we used a sieve plate distributor similar in configuration to that of Letzel. The sieve plate distributor had 625 holes of 0.5 mm diameter placed on a triangular pitch of 7 mm. The total gas hold-up was determined by measuring the hydrostatic pressure using a Validyne pressure sensor. The gas hold-up could be measured with a relative accuracy of ± 5%. Ethanol, added in various concentrations to demineralised water, was used as the liquid phase.

3. Experimental results with alcohol addition

Fig. 4 shows the gas hold-up ε as a function of the superficial gas velocity U obtained with three typical ethanol concentrations used in our experiments. We note that while at low values of U, the values of ε are the same, the gas hold-up is significantly increased by

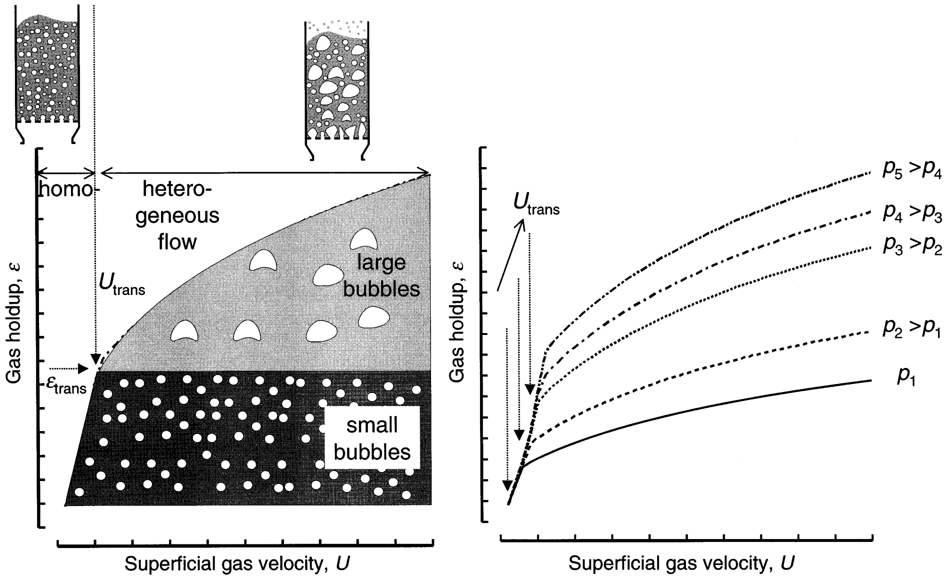

Fig. 1. Schematic representation of the influence of elevated pressure on the gas hold-up in a bubble column.

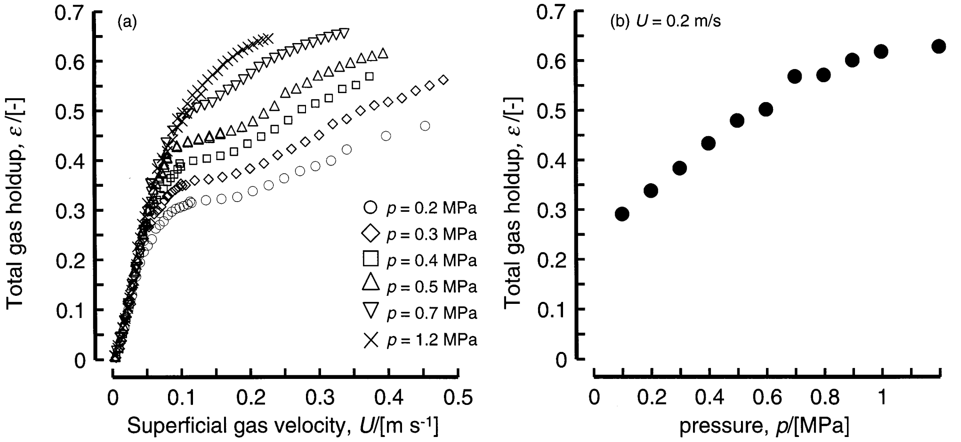

Fig. 2. Experimental data of Letzel et al. [4] describing the influence of elevated pressure on gas hold-up.

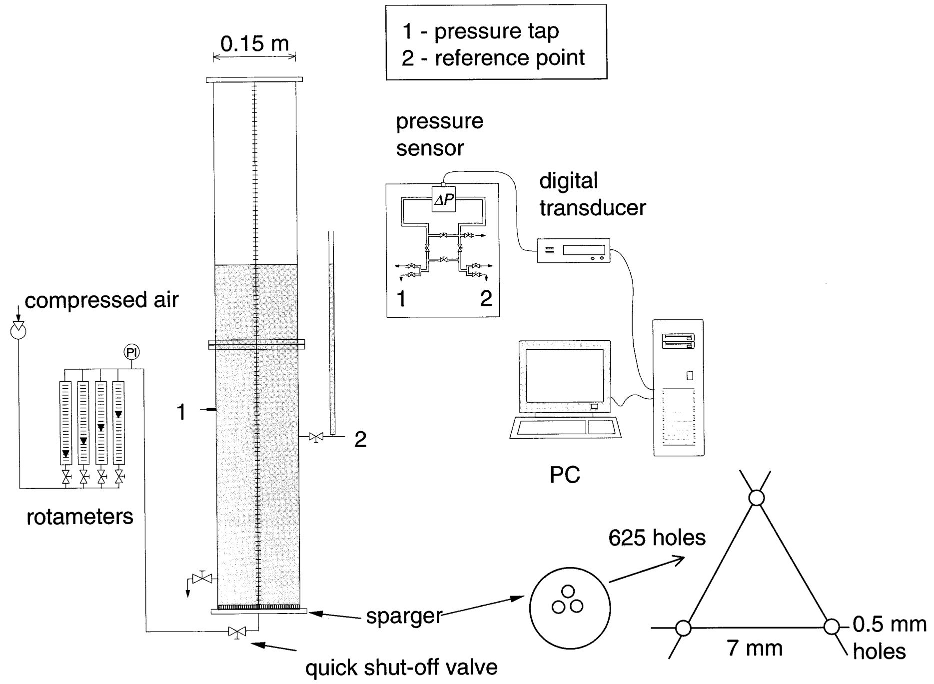

Fig. 3. Experimental set-up used in this work to study the influence of ethanol addition on gas hold-up.

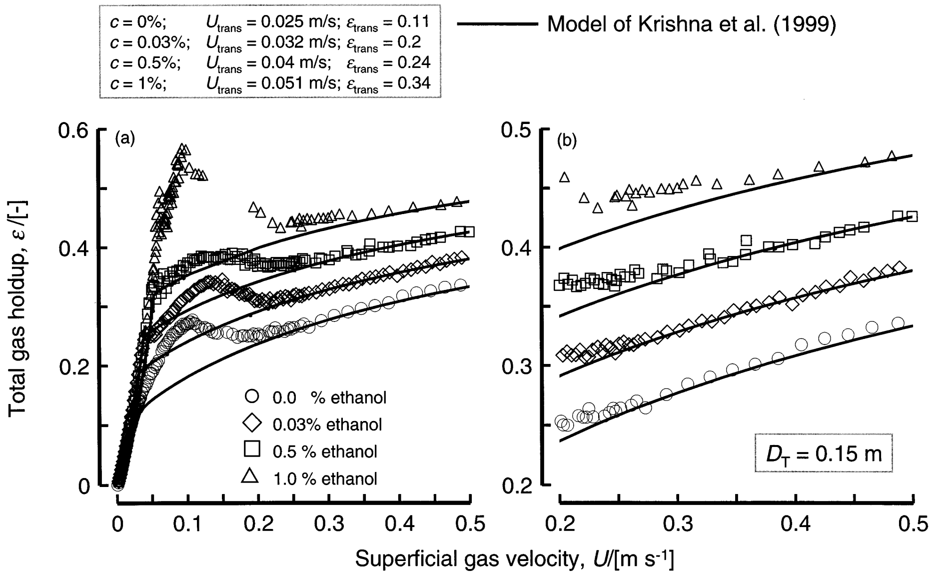

Fig. 4. Experimental data showing the influence of ethanol addition on gas hold-up: (a) shows both homogeneous and heterogeneous flow regimes; (b) focus on heterogeneous flow regime.

addition of ethanol for operation at high values of U. Clearly the influence of ethanol addition is more significant in the heterogeneous flow regime. In order to interpret the results, the first task is to find out the values of the regime transition parameters. The regime transition parameters $\varepsilon_{\mathrm{trans}}$ and U_{trans} can be determined from a Wallis plot [17]. In the Wallis plot the 'drift flux', $U\,(1-\varepsilon)$, is plotted against the gas hold-up, ε, as shown in Fig. 5 for the experimental data with 0% ethanol and 0.5% ethanol concentrations. The smooth curve in Fig. 5 are drawn using the Richardson–Zaki formulation [18], i.e. drift flux $= v_\infty\,\varepsilon(1-\varepsilon)^n$ where n is the Richardson–Zaki index and v_∞ is the rise velocity of a single gas bubble. For air–water system in the homogeneous regime, $n = 2$ and the rise velocity of a single bubble v_∞ can be obtained from the Wallis plot by data fitting (see Fig. 5). The Richardson–Zaki formulation is valid for the homogeneous bubbly flow regime. The point of deviation of the experimental values from the Richardson–Zaki curve is taken to indicate the regime transition point (see Fig. 5). The influence of ethanol concentration on the values of the transition parameters $\varepsilon_{\text{trans}}$ and U_{trans} is shown in Fig. 6 for the whole range of concentrations studied in our experiments. It is clear that the addition of small amounts of alcohol has the effect of increasing the values of $\varepsilon_{\text{trans}}$ and U_{trans}. In other words, the addition of alcohol has the effect of stabilizing the homogeneous bubbly flow regime. This stabilisation is caused by suppression of the coalescence tendency of small bubbles. The mechanism of the coalescence-preventing action of the alcohols was discussed and explained in detail by some authors [11–16]. The alcohols, when dissolved in water, are strongly adsorbed at the interface. They behave as hydrophobic materials and tend to be rejected from the bulk of the solution to the interface. They accumulate around the bubbles forming a 'protective' monolayer, and consequently the coalescence between the bubbles will be hindered. When a bubble moves through a liquid, adsorbed surface active material is pushed to the back of the bubble. This causes a surface tension gradient which opposes the tangential shear stress. This phenomenon increases the drag on the bubble and consequently the rise velocity is reduced.

It is important to first determine whether the shift in the regime transition point with alcohol addition is able to explain the increased hold-up observed experimentally. To answer this question we apply the model of Krishna et al. [19] to calculate the gas hold-up.

4. Model of Krishna et al. [19] for gas hold-up

For superficial gas velocities $U \leq U_{\text{trans}}$, the gas hold-up in the homogeneous flow regime is given by:

$$\varepsilon = U/V_{\text{slip}}; \quad V_{\text{slip}} = v_{\infty}(1-\varepsilon) \tag{1}$$

In the heterogeneous flow regime, i.e. for $U \geq U_{\text{trans}}$, the total gas hold-up is given by:

$$\varepsilon = \varepsilon_{\text{b,large}} + \varepsilon_{\text{trans}}(1-\varepsilon_{\text{b,large}}); \quad \varepsilon_{\text{b,large}} = (U - U_{\text{trans}})/V_{\text{b,large}} \tag{2}$$

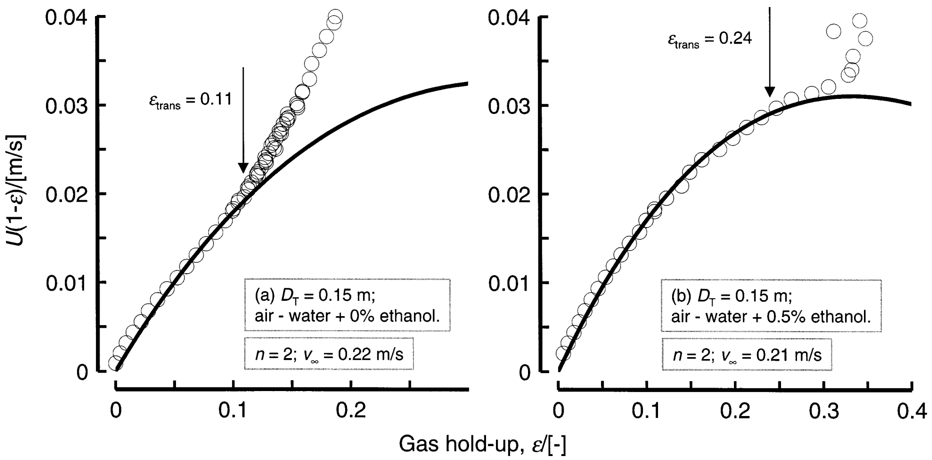

Fig. 5. Wallis plot to determine the regime transition parameters.

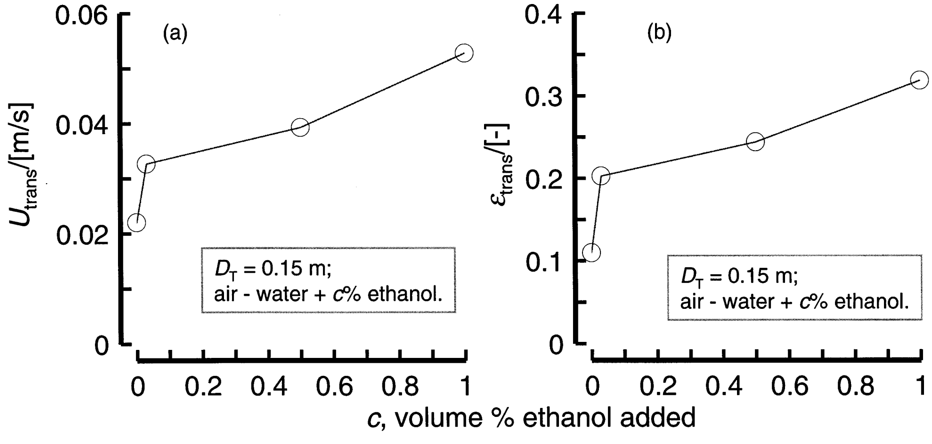

Fig. 6. Influence of ethanol addition on regime transition parameters.

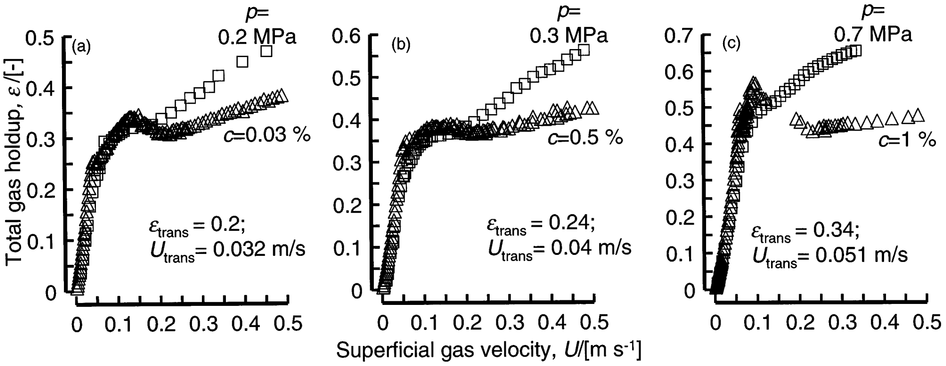

Fig. 7. Comparison of the influences of elevated pressure with alcohol addition: (a) elevated pressure results of Letzel at 0.2 MPa compared with results obtained with 0.03% ethanol solution; (b) elevated pressure results at 0.3 MPa compared with results obtained with 0.5% ethanol solution; (c) elevated pressure results at 0.7 MPa compared with results obtained with 1.0% ethanol solution.

where $V_{\mathrm{b,large}}$ is the rise velocity of the swarm of large bubbles. Krishna et al. [19] obtained a correlation for the large bubble velocity in the form:

$$V_{\mathrm{b,large}} = 0.71\sqrt{g\, d_{\mathrm{b,large}}}\ (\mathrm{SF})(\mathrm{AF}) \tag{3}$$

where two correction factors are introduced into the classical Davies–Taylor [20] relation for the rise of a single spherical cap bubble in an infinite volume of liquid. Bubbles in a liquid assume a spherical cap shape when the criterion Eötvös number, $E\ddot{o} > 40$, is met; see Clift et al. [21]. For the air–water system, the criterion $E\ddot{o} > 40$ is met for bubbles larger than 17 mm in diameter. The scale correction factor (SF), accounts for the influence of the column diameter and is taken from the work of Collins [22] to be:

$$\begin{aligned} &\mathrm{SF} = 1 \quad \text{for} \quad d_{\mathrm{b,large}}/D_{\mathrm{T}} < 0.125 \\ &\mathrm{SF} = 1.13\exp(-d_{\mathrm{b,large}}/D_{\mathrm{T}}) \quad \text{for} \\ &\quad 0.125 < d_{\mathrm{b,large}}/D_{\mathrm{T}} < 0.6 \\ &\mathrm{SF} = 0.496\sqrt{D_{\mathrm{T}}/d_{\mathrm{b,large}}} \quad \text{for} \quad d_{\mathrm{b,large}}/D_{\mathrm{T}} > 0.6 \end{aligned} \tag{4}$$

The acceleration factor AF accounts for the increase in the large bubble velocity over that of a single, isolated, bubble due to wake interactions. This factor increases as the distance between the large bubbles decreases [19]. Since the average distance between large bubbles will decrease as the superficial gas velocity through the large bubble phase increases, a linear relation is postulated for AF:

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

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

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

The model parameters α, β, γ and δ were determined by Krishna et al. [19] by multiple regression of the measured data on the rise velocity of large bubbles in low viscosity liquids. The fitted values are:

$$\alpha = 2.73; \quad \beta = 4.505; \quad \gamma = 0.069; \quad \delta = 0.376 \tag{7}$$

The continuous curves in Fig. 4 have been drawn with Eqs. (1)–(7) using the parameters $\varepsilon_{\mathrm{trans}}$ and U_{trans} determined from the Wallis plots and reported in Fig. 6. Except for the transition regime, characterised by a maximum in the gas hold-up, the model of Krishna et al. [19] works very well.

5. Influence of ethanol addition versus influence of elevated pressure

The inescapable conclusion to be drawn from Fig. 4 is that the influence of ethanol addition on the gas hold-up is entirely describable by a model which takes account of delay in the regime transition. It is interesting to speculate whether the influence of increased pressure on the gas hold-up can also solely be ascribed to a shift in the regime transition point. In order to test this hypothesis we compare the gas hold-ups at 0.2, 0.3 and 0.7 MPa, measured by Letzel et al. [4] with data measured in this work with 0.03, 0.5 and 1% ethanol, respectively (see Fig. 7a–c). Consider Fig. 7(b); the transition parameters for the experiment of Letzel at $p = 0.3$ MPa and for $c = 0.5\%$ ethanol (this work) are nearly the same, viz. $\varepsilon_{\mathrm{trans}} = 0.24$; $U_{\mathrm{trans}} = 0.04$ m/s. The model of Krishna et al. [19] would anticipate that the gas hold-ups for these two experiments would be identical since the two experiments were carried out in columns of the same diameter D_{T} and with the similar distributor configuration. The experimental data however show that operation at elevated pressures leads to a significantly higher hold-up in the heterogeneous regime. The same observation holds when we compare the experiments of Letzel at $p = 0.2$ MPa with the data from this work for $c = 0.03\%$ ethanol and the 0.7 MPa results with the 1% ethanol results. The comparison of the data presented in Fig. 7 leads us to conclude that increased pressure has the additional effect of increasing the large bubble hold-up, $\varepsilon_{\mathrm{b,large}}$ because the differences in the gas hold-up values are in the heterogeneous flow regime. The reason for this increase is the enhanced break up of the large bubble population due to decreased bubble stability [23]. We can therefore modify the model of Krishna et al. [19] for the large bubble rise velocity by introducing a correction factor to account for the influence of elevated pressures:

$$V_{\mathrm{b,large}} = 0.71\sqrt{g\, d_{\mathrm{b,large}}}(\mathrm{SF})(\mathrm{AF})(\mathrm{DF}) \tag{8}$$

where DF is the density correction factor. Using the Kelvin–Helmholtz stability theory as basis, Letzel et al. [23] concluded that this correction factor is inversely proportional to the square root of the gas density. For air at atmospheric conditions used in the experiments, $\rho_G = 1.29$ kg/m^3 and the density correction factor is unity, i.e. DF = 1. For any gas at any system pressure, having a gas density ρ_G, the density correction factor can be calculated from:

$$\mathrm{DF} = \sqrt{1.29/\rho_G} \tag{9}$$

The Fischer–Tropsch synthesis of hydrocarbons in a bubble column slurry reactor using synthesis gas, a mixture of CO and H_2, is carried out at a pressure of about 3 MPa; the syngas density at this pressure is 7 kg/m^3 and the large bubble rise velocity at these conditions is only a fraction $\sqrt{1.29/7} = 0.43$ of the velocity it would have in cold-flow experiments carried out under atmospheric pressure conditions with air as the gas phase. This underlines the importance of the density correction factor developed above.

In order to apply the model of Krishna et al. [19] we need information on the regime transition parameters. For design and prediction purposes, the Reilly et al. correlation [6] may be used to estimate the gas hold-up at regime transition:

$$\varepsilon_{\mathrm{trans}} = 0.59\, B^{1.5}\sqrt{\rho_G^{0.96}\, \sigma^{0.12}/\rho_L} \tag{10}$$

where the parameter $B = 3.85$. The superficial gas velocity at regime transition is $U_{\mathrm{trans}} = v_\infty\, \varepsilon_{\mathrm{trans}}(1 - \varepsilon_{\mathrm{trans}})$ where the 'small' bubble rise velocity is given by Reilly et al. [6] as:

$$v_\infty = \sigma^{0.12}/(2.84\rho_G^{0.04}) \tag{11}$$

With the modification given by Eqs. (8) and (9), the Krishna et al. model can be combined with Eqs. (10) and (11) to predict the gas hold-up for various system pressures. These predictions are compared with the experimental data in Fig. 8 and we see that the agreement with the model with data is excellent. Also shown in Fig. 8 with broken lines are model calculations using Eqs. (1)–(11) taking the density correction factor as unity, i.e. DF = 1. It is clear that the experimental data of Letzel et al. [4] cannot be modelled without considering the density correction factor.

6. Conclusions

The following major conclusions can be drawn from the present investigation.

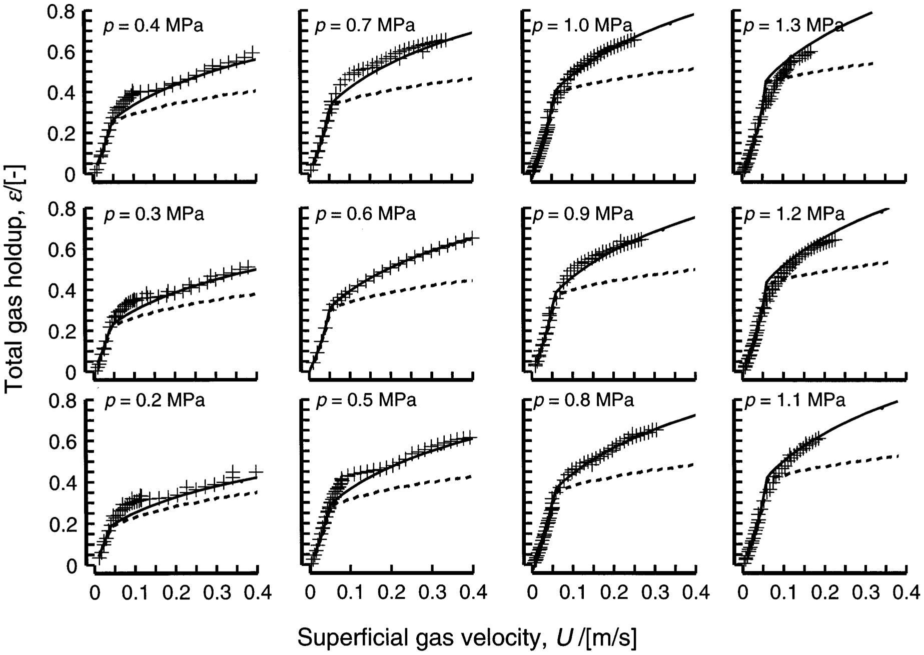

Fig. 8. Influence of increased system pressure on gas hold-up. Experimental data of Letzel et al. [4], indicated by (+). The continuous line represent the model of Krishna et al. [19]: (———) model using Eq. (8) with correction for large bubble break up; (---------) model using Eq. (3), taking DF = 1.

(1) The addition of small amounts of ethanol to water tends to result in a significant increase in the gas hold-up. This increase in the gas hold-up is entirely attributable to the delay in the regime transition point. By incorporating the appropriate values of $\varepsilon_{\mathrm{trans}}$ and U_{trans}, the model of Krishna et al. [19] provides a very good description of the measured data in the churn-turbulent flow regime.

(2) The reason behind the delay in regime transition due to alcohol addition is coalescence suppression. On the other hand, increased system pressure causes a delay in regime transition due to a reduction in the probability of propagation of instabilities which trigger transition to the heterogeneous flow regime.

(3) The influence of alcohol addition and the influence of increased system pressure on the gas hold-up are not equivalent. Increased system pressure has two separate effects: (1) the regime transition is delayed, i.e. the values $\varepsilon_{\mathrm{trans}}$ and U_{trans} are increased with increased p, and (2) the large bubble rise velocity is decreased according to an inverse square root dependence on p. The model of Krishna et al. [19], modified to take account of the system pressure on the large bubble swarm velocity, provides an excellent description of the measured data of Letzel et al. [4] at elevated pressures. For estimation of the gas hold-up at elevated pressures we therefore recommend the inclusion of the density correction factor given by Eqs. (8) and (9).

Appendix A. Nomenclature

AF	acceleration factor (dimensionless)
B	constant in Reilly correlation
c	concentration of ethanol in water (vol.%)
$d_{\mathrm{b,large}}$	large bubble diameter (m)
D_{T}	column diameter (m)
DF	density correction factor, defined by Eqs. (8) and (9) (dimensionless)
$E\ddot{o}$	Eötvös number, $g(\rho_{\mathrm{L}}-\rho_{\mathrm{G}})d_{\mathrm{b}}^2/\sigma$
g	gravitational acceleration (9.81 m/s^2)
n	Richardson–Zaki index (dimensionless)
p	system pressure (Pa)
SF	scale factor given by the Collins relations, Eq. (4) (dimensionless)
U	superficial gas velocity (m/s)
U_{trans}	superficial gas velocity at regime transition (m/s)
$(U-U_{\mathrm{trans}})$	superficial gas velocity through the large bubble phase (m/s)
v_{∞}	rise velocity of 'small' bubbles (m/s)
$V_{\mathrm{b,large}}$	rise velocity of 'large' bubbles in a swarm (m/s)
$\alpha, \beta, \gamma, \delta$	parameters defined by Eqs. (5)–(7)
ε	total gas hold-up (dimensionless)
$\varepsilon_{\mathrm{b,large}}$	hold-up of large bubbles (dimensionless)
$\varepsilon_{\mathrm{trans}}$	gas hold-up at the regime transition point (dimensionless)
μ_{L}	viscosity of liquid phase (Pa s)
ρ_{G}, ρ_{L}	density of gaseous and liquid phases (kg/m^3)
σ	surface tension of liquid phase (N/m)

Subscripts

G	referring to gas
L	referring to liquid
b,large	referring to 'large' bubbles
trans	referring to the transition point
T	tower or column

References

[1] R. Krishna, J. Ellenberger, S.T. Sie, Reactor development for conversion of natural gas to liquid fuels: a scale up strategy relying on hydrodynamic analogies, Chem. Eng. Sci. 51 (1996) 2041–2050.

[2] C. Maretto, R. Krishna, Modelling of a bubble column slurry reactor for Fischer–Tropsch synthesis, Catal. Today 52 (1999) 279–289.

[3] S.T. Sie, R. Krishna, Fundamentals and selection of advanced Fischer–Tropsch reactors, Appl. Catal. A 186 (1999) 55–70.

[4] H.M. Letzel, J.C. Schouten, C.M. van den Bleek, R. Krishna, Influence of elevated pressure on the stability of bubbly flows, Chem. Eng. Sci. 52 (1997) 3733–3739.

[5] P.M. Wilkinson, A.P. Spek, L.L. Van Dierendonck, Design parameters estimation for scale-up of high-pressure bubble columns, AIChE J. 38 (1992) 544–554.

[6] I.G. Reilly, D.S. Scott, T.J.W. De Bruijn, D. MacIntyre, The role of gas phase momentum in determining gas hold-up and hydrodynamic flow regimes in bubble column operations, Can. J. Chem. Eng. 72 (1994) 3–12.

[7] R. Krishna, J.W.A. De Swart, D.E. Hennephof, J. Ellenberger, H.C.J. Hoefsloot, Influence of increased gas density on the hydrodynamics of bubble column reactors, AIChE J. 40 (1994) 112–119.

[8] H.C.J. Hoefsloot, R. Krishna, Influence of gas density of flow regime transitions in homogenous flow in bubble columns, Ind. Eng. Chem. Res. 32 (1993) 747–750.

[9] R. Krishna, J. Ellenberger, D.E. Hennephof, Analogous description of gas–solid fluidized beds and bubble columns, Chem. Eng. J. 53 (1993) 89–101.

[10] R. Krishna, J. Ellenberger, C. Maretto, Flow regime transition in bubble columns, Int. Comm. Heat Mass Transfer 26 (1999) 467–475.

[11] M. Jamialahmadi, H. Müller-Steinhagen, Effect of alcohol, organic acid and potassium chloride concentration on bubble size, bubble rise velocity and gas hold-up in bubble columns, Chem. Eng. J. 50 (1992) 47–56.

[12] G. Keitel, U. Onken, Inhibition of bubble coalescence by solutes in air/water dispersions, Chem. Eng. Sci. 37 (1982) 1635–1638.

[13] B.G. Kelkar, S.P. Godbole, M.F. Honath, Y.T. Shah, N.L. Carr, W.D. Deckwer, Effect of addition of alcohols on gas hold-up and backmixing in bubble columns, AIChE J. 29 (1983) 361–369.

[14] F.L. Müller, J.F. Davidson, On the effects of surfactants on mass transfer to viscous liquids in bubble columns, Trans. Inst. Chem. Eng. 73 (1995) 291–296.

[15] K. Schügerl, U. Oels, J. Lücke, Bubble columns bioreactors, Adv. Biochem. Eng. 7 (1977) 1–84.

[16] J. Zahradnik, G. Kuncová, M. Fialová, The effect of surface active additives on bubble coalescence and gas hold-up in viscous aerated batches, Chem. Eng. Sci. 54 (1999) 2401–2408.

[17] G.B. Wallis, One-Dimensional Two-Phase Flow, McGraw-Hill, 1969.

[18] J.F. Richardson, W.N. Zaki, Sedimentation and fluidisation: part I, Trans. Inst. Chem. Eng. 32 (1954) 35–53.

[19] R. Krishna, M.I. Urseanu, J.M. Van Baten, J. Ellenberger, Rise velocity of a swarm of large gas bubbles in liquids, Chem. Eng. Sci. 54 (1999) 171–183.

[20] R.M. Davies, G.I. Taylor, The mechanics of large bubbles rising through extended liquids and through liquids in tubes, Proc. R. Soc. London A200 (1950) 375–390.

[21] R. Clift, J.R. Grace, M.E. Weber, Bubbles, drops and particles, Academic Press, San Diego, 1978.

[22] R. Collins, The effect of a containing cylindrical boundary on the velocity of a large gas bubble in a liquid, J. Fluid Mech. 28 (1967) 97–112.

[23] H.M. Letzel, J.C. Schouten, C.M. van den Bleek, R. Krishna, Influence of gas density on the large-bubble hold-up in bubble column reactors, AIChE J. 44 (1998) 2333–2336.