

Interfacial Area and Gas Holdup in a Bubble Column Reactor at Elevated Pressures

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The influence of pressure, liquid viscosity, and gas velocity on the gas holdup and specific gas–liquid interfacial area in a bubble column reactor has been studied. The 18.7 L reactor had an inner diameter of 15.6 cm with a dispersion height set equal to 3 times the diameter and was operated at pressures between 0.1 and 6.6 MPa. By means of the chemically enhanced absorption of CO₂ in diethanolamine, the gas–liquid interfacial area in the reactor has been determined. The viscosity has been changed in the range from 1 to 9.4 mPa·s by adding ethylene glycol to the mixture. It is determined that pressure has a small effect on the gas holdup in pure water, whereas it shows a pronounced effect for the more viscous liquids. This can be attributed to the influence of the increased pressure on the flow regime transition. For the most viscous liquid all interfacial area data were obtained in the fully heterogeneous regime. Here the interfacial area increased with increasing pressure and was moderately affected by the gas velocity. For the less viscous liquids both pressure and gas velocity affect the interfacial area; this influence depends on the flow regime. Therefore, the state of the flow regime has an important impact on the mode in which the operating parameters affect the interfacial area.

Introduction

Bubble column reactors, with and without suspended solids, are often used for chemical processes (Shah et al. (1982), Deckwer (1992)). For proper design and scaleup it is essential to have sufficient knowledge about the hydrodynamic behavior in the reactor. In particular, it is important to understand the influence of process parameters on among others the gas holdup ϵ , the liquid- and gas-side mass-transfer coefficients k_L and k_G , and the specific gas–liquid interfacial area a . Although industrial reactors ordinarily are operated at extreme conditions, i.e., high pressures and temperatures, most research has been carried out at ambient conditions. Only in the last years has research on the influence of pressure on the hydrodynamics increased. Of these works, most of it is concerning the influence of pressure on the gas holdup. The present study investigates the influence of pressure, liquid viscosity, and gas velocity on the gas holdup and specific interfacial area in a bubble column reactor. Variation in liquid viscosity was achieved by using aqueous mixtures with different amounts of ethylene glycol.

Literature Survey

In bubble column reactors the gas phase exists as a dispersed phase in a continuous liquid phase. Depending on the nature of the dispersion, three different types of flow regime can be distinguished (see Deckwer (1992)). At relatively low gas velocities the gas bubbles are almost uniformly distributed in size and rise quiescently to the top of the dispersion, the bubble flow regime. At larger gas velocities bubbles interact severely and a much more turbulent, heterogeneous system is present, the churn-turbulent regime. In small columns also a slug flow regime is feasible. It, however, is insignificant in large-scale industrial reactors.

A typical reported value for the superficial gas velocity at transition from bubble flow to churn-turbulent flow

in a water/air system at atmospheric conditions is 5 cm/s (see, e.g., Shah et al. (1982)). Depending on the gas and liquid properties, this value may change. Especially at elevated pressures the gas velocity may be as large as 10 cm/s before the transition occurs (Wilkinson et al. (1994), Reilly et al. (1994)).

The major part of the published research works about the pressure influence shows an increase in gas holdup with increasing pressure. This influence is smaller if a porous plate gas distributor is used, compared to a perforated plate (see among others Oyevaar et al. (1991)). Also the influence is smaller or absent for low gas velocities. Compared to low pressures, the number of large bubbles is reduced at high pressures and the bubble size distribution becomes narrower (among others Oyevaar et al. (1989), Wilkinson and van Dierendonck (1990)). More extensive literature surveys about the influence of pressure on the gas holdup are given by Oyevaar and Westerterp (1989) and Wilkinson (1991). Wilkinson too summarizes published research about the influence of gas density on the gas holdup.

Several investigators have used different liquids or liquid mixtures to investigate the influence of liquid properties on the gas holdup and mass transport parameters in bubble column reactors. Most of these investigations have been performed at atmospheric conditions. See among others Yoshida and Akita (1988) and Deckwer and Schumpe (1993) for extended reviews on this matter. In general, we refer to the book of Deckwer (1992), in which a very extensive review is given of all available data in literature. As we will correlate our data according to the interpretation of Krishna et al. (1991), we will briefly discuss his approach.

Krishna et al. (1991) correlated the gas holdup by using separate equations for the homogeneous regime and the churn-turbulent regime. In the homogeneous regime the gas holdup is directly proportional to the gas velocity, while in the heterogeneous regime the extra gas holdup is proportional to $(u_G - u_{trans})^{0.8}$. Wilkinson et al. (1992) used the same assumptions used by Krishna et al. They, however, somewhat modified the

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model to estimate the gas holdup. They derived a set of correlations for holdup in pure liquids from experimental and literature data. In the homogeneous flow regime only small bubbles exist and the gas holdup depends linearly on the superficial gas velocity:

$$\epsilon = u_G/u_{sb} \quad \text{for } u_G \leq u_{trans} \quad (1)$$

In the heterogeneous flow regime the gas holdup from small bubbles remains constant, while the gas holdup from large bubbles increases with increasing gas velocity:

$$\epsilon = \frac{u_{trans}}{u_{sb}} + \frac{u_G - u_{trans}}{u_{lb}} \quad \text{for } u_G > u_{trans} \quad (2)$$

In Table 1 the relations are summarized as suggested by Wilkinson et al. (1992) to estimate the rise velocities u_{sb} and u_{lb} of the small and the large bubbles, respectively, and the velocity u_{trans} at transition from bubble to churn-turbulent flow.

Table 1. Correlations Given by Wilkinson et al. (1992) for the Determination of the Gas Velocities of Large and Small Bubbles and the Gas Holdup at Transition from the Homogeneous to the Heterogeneous Flow Regime

$$\frac{u_{sb}\eta_L}{\sigma} = 2.25 \left(\frac{\sigma^3 \rho_L}{g\eta_L^4} \right)^{-0.273} \left(\frac{\rho_L}{\rho_G} \right)^{0.03} \quad (3)$$

$$\frac{u_{lb}\eta_L}{\sigma} = \frac{u_{sb}\eta_L}{\sigma} + 2.4 \left(\frac{\eta_L(u_G - u_{trans})}{\sigma} \right)^{0.757} \left(\frac{\sigma^3 \rho_L}{g\eta_L^4} \right)^{-0.077} \left(\frac{\rho_L}{\rho_G} \right)^{0.077} \quad (4)$$

$$\frac{u_{trans}}{u_{sb}} = \epsilon_{trans} = 0.5 \exp(-193\rho_G^{-0.61}\eta_L^{0.5}\sigma^{0.11}) \quad (5)$$

Experimental Section

Experimental Method. For the determination of the specific gas-liquid interfacial area the chemical method of the enhanced absorption of CO₂, followed by a pseudo-first-order reaction with diethanolamine (DEA) has been used. The interfacial area can be deduced from the overall rate of absorption. This system has been reported elsewhere; see Stegeman et al. (1995) for a more thorough description.

For a carefully chosen DEA concentration the reaction is fast enough to convert all absorbed CO₂ in the liquid boundary. The reaction should not be too fast; otherwise, diffusion of DEA to the reaction zone reduces or limits the absorption rate. Further, in most gas-liquid systems with moderate reaction rates and low gas solubilities the resistance to mass transfer is located in the liquid phase.

The interfacial area can be determined directly from absorption experiments, provided the physicochemical parameters are known using eq 6.

$$a = \frac{\phi_{\text{mol,CO}_2}}{V_r m \overline{c_{\text{CO}_2,G}} \sqrt{k_{1,p} c_{\text{DEA}}^p D_{\text{CO}_2}}} \quad (6)$$

where the average $\overline{c_{\text{CO}_2,G}}$ is the mean driving force for mass transfer from the gas to the liquid phase. The liquid phase is assumed to be well mixed.

Experimental System. In Figure 1 a simplified flow diagram of the experimental setup is given. The system, constructed from stainless steel 316, can be

Table 2. Physical Properties of the Used Liquid Phases

liquid mixture	water	DEA/water/ 20% ETG	DEA/water/ 40% ETG	DEA/water/ 60% ETG
DEA concn (mol L ⁻¹)	0	1.36	1.33	1.4
viscosity (Pa·s)	1.0 × 10 ⁻³	2.6 × 10 ⁻³	4.7 × 10 ⁻³	9.4 × 10 ⁻³
density (kg m ⁻³)	998	1045	1068	1090

operated at pressures ranging from 0.1 to 7.0 MPa. The chemical absorption measurements are performed by means of carbon dioxide absorption in diethanolamine solutions. There is a recycle loop for the gas system, while the liquid is regenerated after usage. Interfacial areas and gas holdup values are determined in three liquid mixtures with viscosities ranging from 2.6 to 9.4 × 10⁻³ Pa·s; see Table 2. While changing the viscosity over a wide range, the density and surface tension have been kept almost constant at values of 1050 kg m⁻³ and 65 mPa·s, respectively. The gas holdup too has been determined in pure demineralized water.

(1) Reactor. The geometry of the bubble column reactor is shown in Figure 2. The reactor is thermostated at 298 K by means of a heated jacket. A gas mixture of nitrogen and carbon dioxide is introduced via a perforated plate gas distributor with a diameter of 0.125 m and 284 holes of 0.4 mm in diameter. It has been mounted 0.062 m above the bottom plate and has an outer diameter of 0.152 m. The liquid phase is introduced at the top of the reactor about 5 cm below the dispersion level. It is withdrawn at the bottom of the reactor. The reactor is equipped with six glass windows on opposite sides of the reactor mounted at 45, 54, and 70 cm from the bottom of the reactor, respectively. Through these round windows of 3 cm diameter, the height as well as the type of dispersion can be observed. The diameter of the reactor is 156 mm, and the dispersion height is set to 0.64 m above the gas distributor. The dispersion height can be kept constant by using an overflow.

(2) Gas System. To reduce the gas consumption at high pressures, the gas is recycled in the miniplant by an air-driven Haskel gas booster (10, in Figure 1), type 8 AGD-5 which has an internal heat exchanger. The gas velocity is controlled by means of a bypass over the compressor. The gas mass flow is measured with an orifice meter.

The fresh gas feed consists of two streams: a mixture of 3 vol % CO₂ in 99.9% pure nitrogen and further pure nitrogen. Both flows, controlled by Hitec F-200 mass flow controllers, are introduced into the recycled gas phase in the static mixer (12). The setpoints of these mass flow controllers are adjusted so that the CO₂ concentration in the reactor inlet is approximately 1500 ppm. The pressure in the system is controlled with a Hitec P-500 pressure controller coupled to an electronic valve.

A demister (8) and a gas-liquid separator (9) are placed in the reactor outlet line to separate entrained liquid from the gas phase. These vessels also level out pulsations in the compressor inlet and outlet together with two additional buffer volumes of 10 L. At the reactor inlet and at the top of the reactor continuously small sample streams of about 33 mL/s are withdrawn and analyzed on-line for their CO₂ content by a Maihak UNOR6N infrared analyzer.

(3) Liquid System. Three different mixtures of DEA, ETG, and demineralized water have been used for the absorption measurements and gas holdup determinations; see Table 2. The gas holdup is deter-

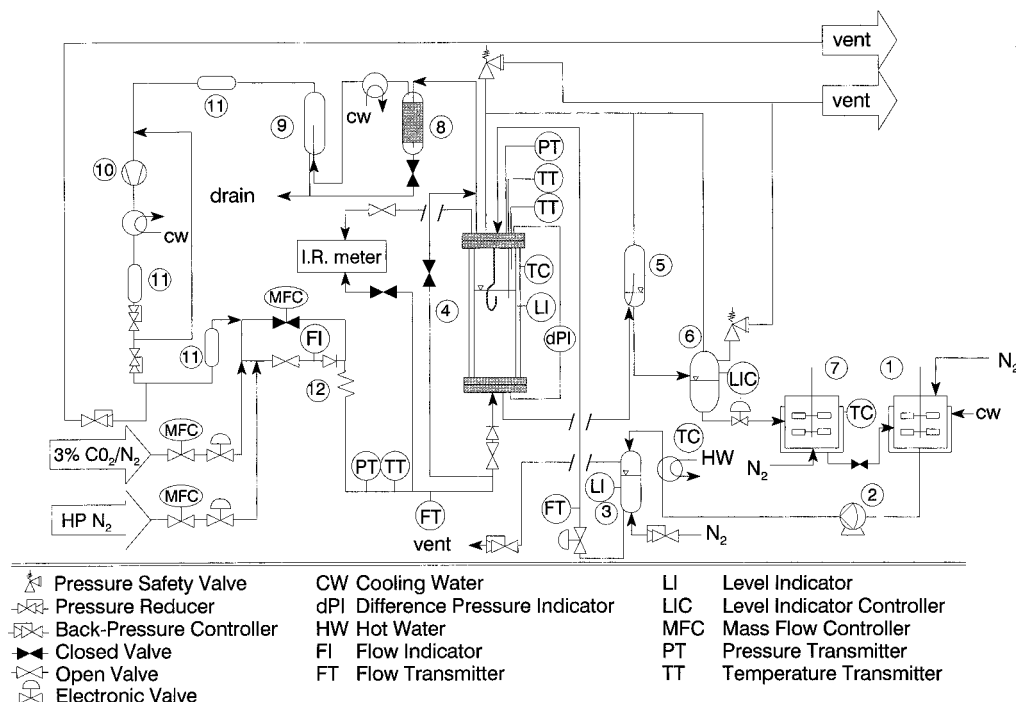


Figure 1. Experimental setup: 1, supply vessel; 2, liquid pump; 3, N₂-saturator; 4, reactor; 5, overflow vessel; 6, liquid buffer vessel; 7, regeneration vessel; 8, demister; 9, gas-liquid separator; 10, compressor; 11, gas buffer vessels; 12, gas mixer.

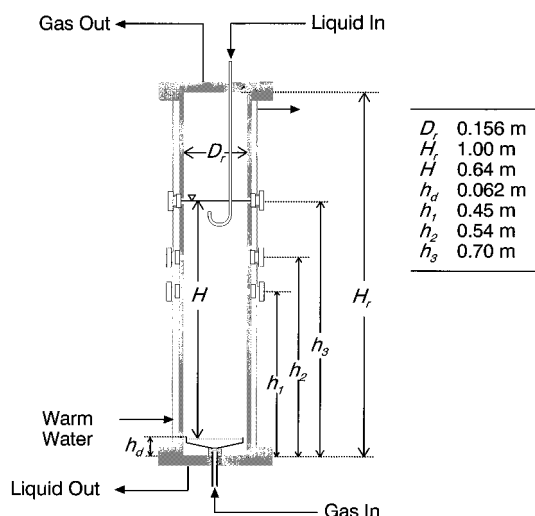


Figure 2. Experimental bubble column reactor and its dimensions.

mined in pure demineralized water as well. The fresh liquid mixtures are stored in two stirred, 60-L, supply vessels, placed under a constant nitrogen pressure of about 0.05 MPa. The jackets of these vessels are cooled with cooling water of about 290 K. The liquid phase is pumped to the reactor by a LEWA ELM-60 membrane pump (2, in Figure 1) with an adjustable stroke length. The liquid flow is heated up to 298 K in a heat exchanger. A vessel (3) is placed in the inlet line to dampen pulsations; it is also used to presaturate the liquid phase with nitrogen. The mass flow rate is measured on-line by means of a Brooks micromotion flowmeter, type D25.

After leaving the reactor, the liquid flow is directed to a 30-L buffer vessel (6), from which the liquid is discharged via a level controller to an atmospheric regeneration section.

Liquid samples are taken from the reactor and the regenerated liquid. They are analyzed for their amine concentration and CO₂ content by acidimetric titration.

Evaluation of the Interfacial Area

The interfacial area can be calculated directly from absorption measurements using eq 6. The absorption rate constant for our system has been determined by Oyevaar et al. (1990).

The spread in the residence time distribution in the gas phase in bubble column reactors can be assumed as negligible (see Deckwer (1992)). For the gas in plug flow, the mean CO₂ concentration is given by the logarithmic mean:

$$\overline{c_{\text{CO}_2, \text{G}}} = \frac{c_{\text{CO}_2, \text{in}} - c_{\text{CO}_2, \text{out}}}{\ln\left(\frac{c_{\text{CO}_2, \text{in}}}{c_{\text{CO}_2, \text{out}}}\right)} \quad (7)$$

Because conversions in the liquid phase are kept under 10% and because the turbulence caused by the gas phase will promote good mixing of the liquid, the liquid phase concentration is assumed to be constant over the reactor.

Results

Influence of the Process Parameters on the Gas Holdup. The overall gas holdup in the reactor has been determined from the difference in the actual liquid height and dispersion height in the reactor. This method allows us to determine the gas holdup for ϵ values larger than 0.05 within $\pm 10\%$ accuracy.

(1) Gas Holdup in Water. In Figure 3 the gas holdup is given as a function of the gas velocity for pressures of $0.1 \leq P \leq 5.6$ MPa. In the plot also the gas holdup is indicated as calculated using the relations given by Akita and Yoshida (1973) and Idogawa et al. (1987). The relation of Akita and Yoshida is determined at atmospheric conditions, while the last authors correlate the gas holdup as a function of the gas density. Therefore, lines are given for N₂ at $P = 0.1$ and 5.6 MPa for the correlation of Idogawa et al. The values for the gas holdup are in the range between the calculated

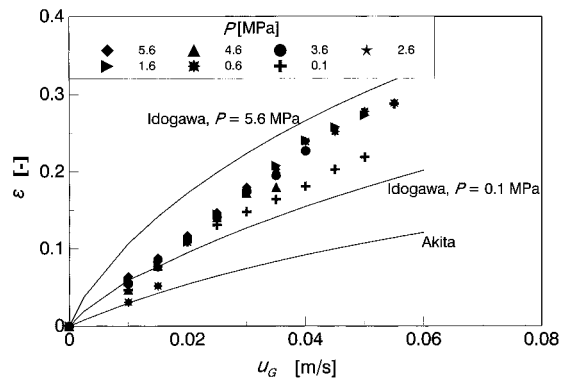


Figure 3. Gas holdup in water as a function of the gas superficial velocity compared to literature relations.

values using the relation of Idogawa et al., although our experiments only show a pressure influence for the lowest pressure. The values determined at the lowest pressure seem to follow the relation of Akita and Yoshida.

The gas holdup is also compared to the values calculated by the relations given by Wilkinson et al. (1992). Their correlations predict gas holdup values which are consistently 0.08–0.10 higher than our measured data. If we determine the bubble rise velocity in our system—from plots where u_G/ϵ is plotted versus P at the limiting value of $u_G \rightarrow 0$ at high pressures—it remains at a constant value of about 0.17–0.18 m/s in the homogeneous bubble flow regime. This value is lower than the one predicted by Wilkinson et al. (1992). Consequently, the gas holdup is larger. The possible reason for the difference may be the difference in the experimental setup. They have a gas inlet with larger hole diameters and a taller column than in our investigations.

(2) Influence of the Liquid Viscosity. With increasing liquid viscosities, it was visually observed that the bubble size distribution became much wider. The gas holdup decreases with increasing liquid viscosity for large gas velocities and small system pressures. At small gas velocities the gas holdup tends to increase with increasing liquid viscosity. The wider bubble size distribution indicates the dispersion already becomes heterogeneous at lower gas velocities, while in pure water almost all our experiments were performed in the homogeneous flow regime.

In Figure 4a–d plots of the gas holdup as a function of the gas velocity are given for the different liquid compositions. The plots show a remarkable difference between the gas holdup values in water, on the one hand, and those in viscous liquids, on the other hand. Whereas ϵ in water is only affected by pressure at low pressures, a large influence of the system pressure is found for the more viscous liquids. We have correlated the gas holdup using the more simple model of Krishna et al. (1991) because the relations given by Wilkinson et al. (1992) could not predict the location of the values of u_{trans} for our data. In Krishna's model the gas holdup is proportional to the gas velocity in the homogeneous regime:

$$\epsilon \propto u_G \quad \epsilon < \epsilon_{trans} \quad (8)$$

In the heterogeneous regime the gas holdup increases less than proportionally with the gas velocity:

$$\epsilon - \epsilon_{trans} \propto (u_G - u_{trans})^{0.8}$$

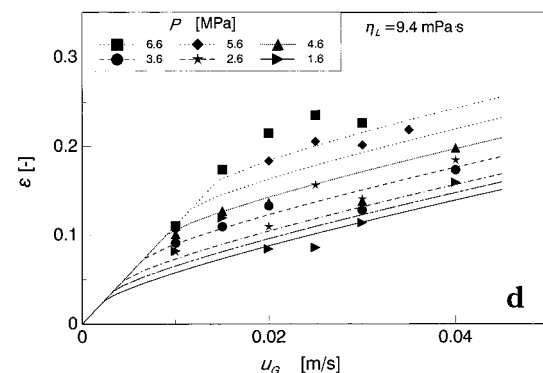
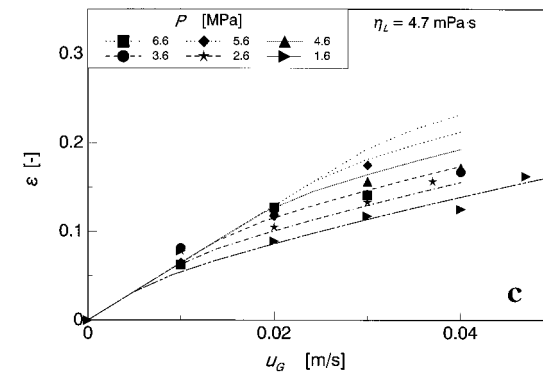
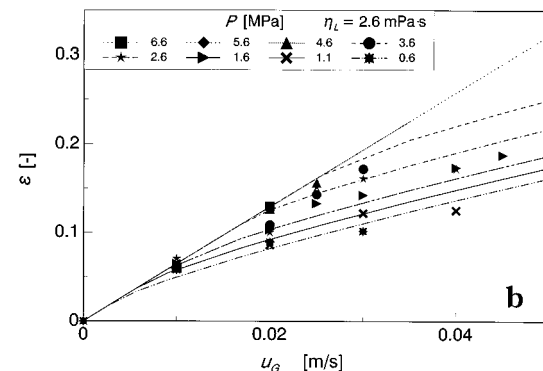
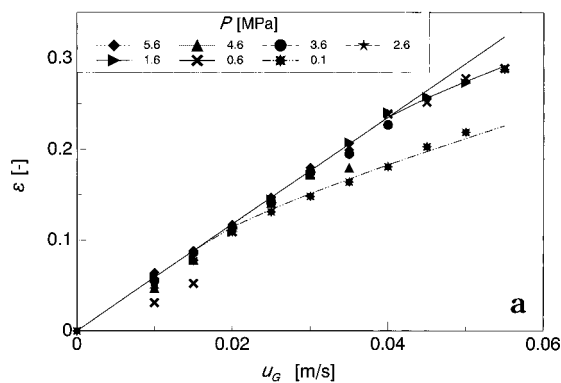


Figure 4. (a) Gas holdup in water as a function of the gas velocity for $0.1 \leq P \leq 5.6$ MPa. (b) Gas holdup as a function of the superficial gas velocity and the system pressure for $\eta_L = 2.6$ mPa·s. (c) Gas holdup as a function of the superficial gas velocity and system pressure for $\eta_L = 4.7$ mPa·s. (d) Gas holdup as a function of superficial gas velocity and the system pressure for $\eta_L = 9.4$ mPa·s.

For the viscous mixtures the values for u_{trans} follow a simple power law function: $u_{trans} = Const. \times \eta_L^{\alpha} P^{\beta}$, with α equal to 1.05, β equal to 1.28, and the constant depending on the liquid phase. The values of u_{trans} for water are determined separately because the gas holdup for water as a function of the gas velocity shows a

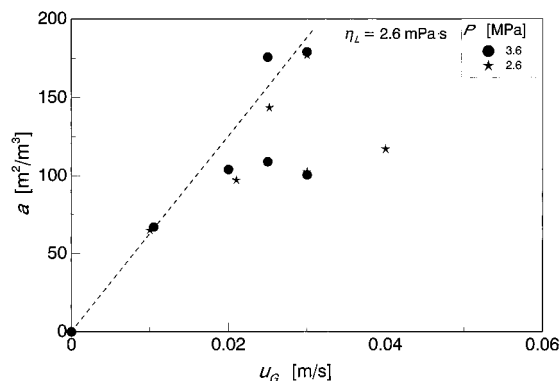


Figure 5. Interfacial area as a function of the superficial gas velocity and system pressure.

behavior different from those of the viscous mixtures. In accordance with Krishna et al., we observe that the liquid properties hardly affect the gas holdup in the heterogeneous flow regime. Although the gas holdup at equal small gas velocities is larger for higher liquid viscosities, it also follows that the transition from the homogeneous to the heterogeneous regime shifts to lower gas velocities with increasing liquid viscosity and with decreasing system pressures.

Influence of the Process Parameters on the Interfacial Area. The major part of our measurements was conducted in the heterogeneous regime. During the experiments we observed a large scatter in the determined interfacial area, especially in the transition region. The value of the interfacial area seemed to depend on the history of the measurements (see Figure 5) where the interfacial area is given for $P = 2.6$ and 3.6 MPa and a liquid viscosity equal to 2.6 mPa·s. Here low and high values for the interfacial area are found at the same gas velocity. The highest values are found when the measurements were preceded with experiments performed at higher pressures and smaller gas velocities. Probably in that case the system still behaves as homogeneous, giving larger interfacial areas compared to the measurements in the fully heterogeneous regime.

In the homogeneous flow regime the interfacial area is almost proportional to the gas velocity; thus, bubble sizes are almost constant with increasing gas velocity. In the heterogeneous regime larger bubbles are formed which contribute only moderately to the interfacial area. We observe that the transition from the homogeneous to the heterogeneous regime cannot be given as a distinct transition point; between the two regimes there is a transition region. In this region several factors simultaneously determine the interfacial area; also the sequence of changes in operating conditions possibly may affect the interfacial area. In this transition region, although gas holdup still may increase, the interfacial area may change in any direction depending on the coalescence and bubble break-up behavior of the dispersion (see, e.g., Schumpe and Deckwer (1982)).

Because the hydrodynamics in the homogeneous and the heterogeneous regime are different, in Figure 6 only values found in the heterogeneous regime and some low values in the transition regime are given. In this figure the different pressures are represented by different symbols, while the shadings of the symbols indicate the liquid viscosity; black, gray, and white for respectively 9.6 , 4.7 , and 2.6 mPa·s. At the highest viscosity the heterogeneous flow prevailed over the whole experimental range and no data points are found in the homogeneous regime. Only a small influence of the gas

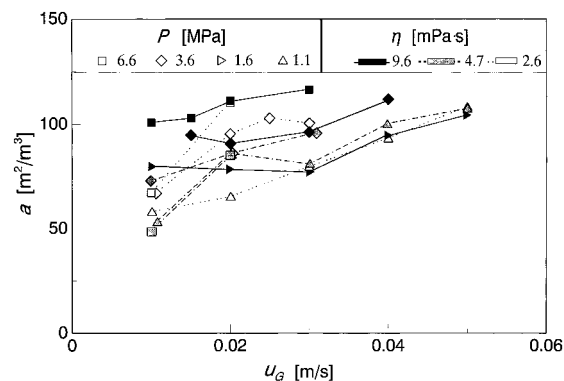


Figure 6. Interfacial area as a function of the superficial gas velocity and for $P = 2.6$ MPa and $P = 3.6$ MPa, $\eta_L = 2.6$ mPa·s.

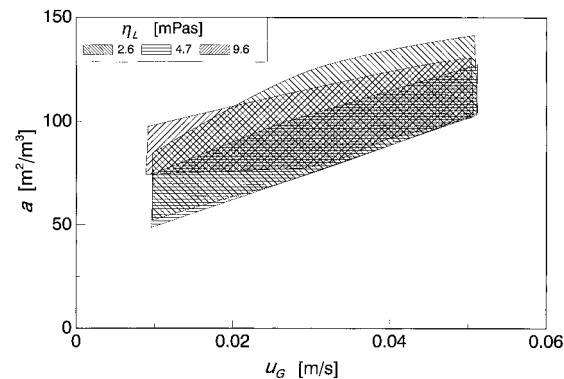


Figure 7. Experimentally determined ranges of the interfacial area for $1.6 \leq P \leq 6.6$ MPa.

velocity can be observed and a noticeable influence of the system pressure. Although the transition velocity u_{trans} has a lower value for large values of η_L , the gas holdup increases more rapidly in the more viscous liquid. This explains the larger initial value of a for the most viscous liquid.

For $\eta_L = 2.6$ mPa·s the interfacial area shows a gradual increase with an increase in gas velocity. For $\eta_L = 4.7$ mPa·s we observed a more pronounced influence of the gas velocity at $P \geq 4.6$ MPa.

In Figure 7 we have given the ranges in which interfacial areas experimentally are found for pressures between 1.6 and 6.6 MPa for the three liquid phase viscosities. A direct influence of the liquid viscosity cannot be observed: At low gas velocities the interfacial areas are higher at larger viscosities, while at the larger gas velocities the opposite seems to be correct. The cause must be sought in bubble coalescence and breakup behavior.

Discussion and Conclusions

From this study it can be concluded that the influence of pressure, gas velocity, and liquid viscosity on the gas holdup and gas-liquid interfacial area in bubble columns cannot be ignored. The gas holdup can be evaluated from the holdups in the homogeneous and the heterogeneous regimes. In the homogeneous regime this holdup is directly proportional to the gas velocity, while in the heterogeneous regime the increase in gas holdup is less pronounced. In the homogeneous regime it is hardly affected by the system pressure. The transition point from homogeneous to heterogeneous flow for increasing pressures shifts to higher gas velocities and thus to larger gas holdups. An increase in liquid viscosity has an opposite effect: the gas velocity at transition decreases rapidly with increasing viscosi-

ties. Compared to literature data for work at atmospheric pressure, larger gas holdup values are found in our work, especially for the more viscous mixtures. This is probably due to a broader bubble size distribution in the more viscous mixtures. Throughout the whole range of gas velocities in the viscous liquid with $\eta_L = 9.4$ mPa·s the heterogeneous flow regime prevailed.

The interfacial area measurements performed in the least viscous liquids exhibited a transition zone rather than a distinct transition point. Whether the system behaves like homogeneous or heterogeneous in this region depends on the operating history. No firm conclusions can be drawn in these regimes, the experiments should also be conducted at higher gas velocities to get a more complete picture of the influence of gas velocity and pressure in the heterogeneous regime.

We assume the unexpected behavior in the transition range is caused by the coalescence and bubble break-up behavior which still is not well understood and cannot be described yet by physical properties like density, surface tension, and viscosity solely. Much research is still required.

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Notation

a = specific interfacial area, $\text{m}^2 \cdot \text{m}^{-3}$ dispersion
 c = concentration, $\text{mol} \cdot \text{m}^{-3}$
 D_A = diffusivity of component A in the liquid phase, $\text{m}^2 \cdot \text{s}^{-1}$
 D_r = reactor diameter, m
 DEA = diethanolamine
 H = height of the dispersion, m
 H_r = reactor height, m
 k_L = liquid-phase mass-transfer coefficient, $\text{m} \cdot \text{s}^{-1}$
 k_G = gas-phase mass-transfer coefficient, $\text{m} \cdot \text{s}^{-1}$
 $k_{n,p}$ = reaction rate constant for a reaction of order (n, p),
 $\text{m}^{3(p+n-1)} \cdot \text{mol}^{-(p+n-1)} \cdot \text{s}^{-1}$
 m = c_L/c_G , distribution coefficient
 P = absolute pressure in the reactor, Pa
 T = absolute temperature, K
 u = superficial gas velocity at the prevailing pressure and based on the empty cross-section area of the vessel, $\text{m} \cdot \text{s}^{-1}$
 u_{swarm} = rise velocity of gas bubble swarm, $\text{m} \cdot \text{s}^{-1}$
 V_r = effective reactor volume, m^3

Greek Symbols

ϵ = gas holdup
 ϵ_{trans} = gas holdup at transition point from homogeneous to heterogeneous flow
 ϕ_{mol} = molar flow rate, $\text{mol} \cdot \text{s}^{-1}$
 ρ = density, $\text{kg} \cdot \text{m}^{-3}$
 σ = surface tension, $\text{N} \cdot \text{m}^{-1}$
 η = kinematic viscosity, $\text{Pa} \cdot \text{s}$

Superscripts and Subscripts

G = gas
 in = inlet
 L = liquid
 lb = large bubble
 n = reaction rate order for component A in the liquid phase
 out = outlet
 p = reaction rate order of component B in the liquid phase
 r = reactor
 sb = small bubble

trans = transition from homogeneous to heterogeneous flow regime

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