Effect of Gas Density on Large-Bubble Holdup in Bubble Column Reactors

Martin H. Letzel
Faculty of Applied Sciences, Delft University of Technology, 2628 BL Delft, The Netherlands
Dept. of Chemical Engineering, University of Amsterdam, 1018 WV Amsterdam, The Netherlands

Jaap C. Schouten and Cor M. van den Bleek
Faculty of Applied Sciences, Delft University of Technology, 2628 BL Delft, The Netherlands

Rajamani Krishna
Dept. of Chemical Engineering, University of Amsterdam, 1018 WV Amsterdam, The Netherlands

Increased system pressure or gas density changes the hydrodynamics in gas-liquid bubble columns. This has important consequences for the design of bubble column reactors, since the hydrodynamics at commercial operating conditions can differ considerably from the situation in cold flow experiments. Since empirical correlations have limited applicability, it is of importance to understand the physics of the effect of increased gas density, at least up to a point where still reasonable predictions about the gas fraction can be made.

In this article we explain the effect of gas density on large-bubble holdup by considering the effect of gas density on the rise velocity of large bubbles. We apply the Kelvin-Helmholtz theory to predict this influence on the large-bubble rise velocity. The theoretical dependence derived from this theory is checked by analyzing experimental gas-fraction data, acquired in a 0.15-m-ID column at system pressures up to 1.3 MPa.

Kelvin-Helmholtz Instability

The Kelvin-Helmholtz theory, as described by Lamb (1959), considers the propagation and growth of surface waves on the interface between two fluid phases, considered here without loss of generality to be gas and liquid phases. We consider the case where the upper fluid is the liquid phase with density ρ_l, and the lower fluid is the gas phase with density ρ_g. The gas and liquid phases have velocities ν_g and ν_l, relative to the interface. This configuration is representative for the situation near the roof of a bubble, as Figure 1 shows: close to the roof we have a near horizontal gas-liquid interface with liquid as the upper fluid, and, due to the velocity of the bubble, there will be a relative velocity between the gas and liquid phases. A complex disturbance η (m) with amplitude a (m), wave number k (m^{-1}) and velocity c ($m \cdot s^{-1}$) is superimposed on the gas-liquid interface

$$\eta = a e^{i(kct - kx)} \tag{1}$$

From conditions for continuity of pressure and velocity at the interface, Lamb (1959) derived that

$$c^2 = \frac{g}{k}\frac{\rho_g - \rho_l}{\rho_g + \rho_l} + \frac{k\sigma}{\rho_g + \rho_l} - \frac{\rho_g \rho_l}{(\rho_g + \rho_l)^2}(\nu_g - \nu_l)^2 \tag{2}$$

Equation 2 relates the square of the velocity of the disturbance c to the density of the two phases and to their velocity difference. If the square of the velocity c is negative, the so-

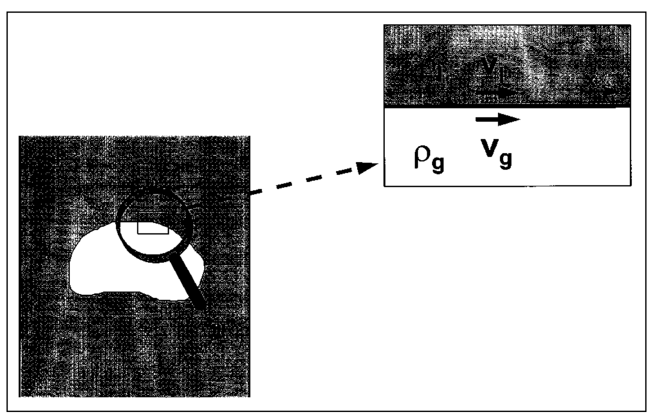

Figure 1. Interface between two fluids with different density and velocity.

Current address of J. C. Schouten: Eindhoven University of Technology, Laboratory of Chemical Reactor Engineering, P.O. Box 513, 5600 MB Eindhoven, The Netherlands.

lutions for c are complex and equal to $\pm iA$, where A is a positive real number and $A^2 = -c^2$. Substituting the solution $-iA$ in the original equation for the disturbance (Eq. 1), one obtains

$$\eta = ae^{i(-kiAt-kx)} = ae^{kAt-ikx} \tag{3}$$

A real number appears in the exponent of the disturbance, causing it to grow exponentially. In other words, when c^2 in Eq. 2 is negative, the interface is unstable for the disturbance given by Eq. 1.

The square of the growth factor of the disturbance is $k^2A^2 = -k^2c^2$, where

$$k^2c^2 = gk\frac{\rho_g - \rho_l}{\rho_g + \rho_l} + \frac{k^3\sigma}{\rho_g + \rho_l} - k^2\frac{\rho_g\,\rho_l}{(\rho_g + \rho_l)^2}(\nu_g - \nu_l)^2 \approx$$

$$gk + \frac{k^3\sigma}{\rho_l} - k^2\frac{\rho_g}{\rho_l}\nu_r^2 \tag{4}$$

and ν_r (m·s^{-1}) is the relative velocity of the phases with respect to each other. The approximation is valid for $\rho_g \ll \rho_l$. Figure 2 shows k^2c^2 as a function of the wavelength $\lambda (= 2\pi/k)$ of the disturbance for a water-nitrogen system at pressures of 0.1 MPa, 1.0 MPa, and 2.0 MPa, respectively. The wavelengths where $k^2c^2 < 0$ are unstable. At these wavelengths, the square root of $|k^2c^2|$ is the growth factor of the disturbance. In case of gas bubbles rising through a stagnant liquid, the relative velocity between the interfaces ν_r is obviously close to the bubble rise velocity. As a first approximation, we therefore assume that the relative velocity in Eq. 4 is equal to the bubble rise velocity V_b (m·s^{-1}). In Figure 2 we took $\nu_r = 1$ m·s^{-1}, which is a typical value for the large-bubble rise velocity (Krishna and Ellenberger, 1996).

One can observe that an increase in gas density increases the range of unstable wavelengths as well as the magnitude of k^2c^2 and thus the growth factor $\sqrt{-k^2c^2}$. The change of the growth factors with changing gas density was also observed by Wilkinson (1991). It is interesting to note that the effect of gas density is especially large in the range 0.01 m·s^{-1} to 0.05 m·s^{-1}, which is the order of the size of the large bubbles (de Swart et al., 1996).

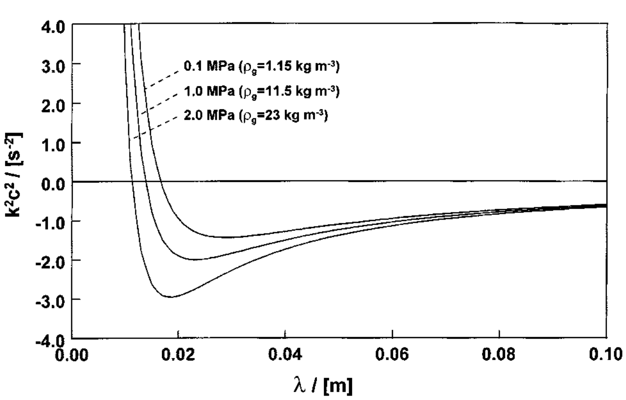

Figure 2. Equation 4 for the nitrogen-water system.

One can also observe from Eq. 4 that the relative velocity between the phases ν_r influences the growth factors. We will now illustrate what happens with a gas bubble with velocity $V_b = 1$ m·s^{-1} and gas density $\rho_g = 1.15$ kg·m^{-3} (corresponding to 0.1-MPa system pressure in a nitrogen-water system), when the gas density changes to say 11.5 kg·m^{-3} (corresponding to 1.0-MPa system pressure). From Figure 2, one can observe that disturbances with wavelengths larger than 0.02 m are unstable. Therefore, we can expect that on the surface of bubbles, larger than 0.02 m in diameter, disturbances with unstable wavelengths can occur. Large bubbles are therefore not stable. They still exist, however, because besides continuous breakup, there is also a continuous coalescence of bubbles (de Swart et al., 1996). The net result of these two opposing mechanisms is a dynamic equilibrium with a corresponding equilibrium bubble size.

When the gas density increases to 11.5 kg·m^{-3}, one can observe from Figure 2 that the range of unstable wavelengths, as well as the magnitude of the growth factors, increases. Therefore, the rate of breakup will increase. Since increased system pressure has a negligible influence on bubble coalescence (Sagert and Quinn, 1976), this will make the dynamic equilibrium between breakup and coalescence shift to smaller equilibrium bubble sizes. Together with the bubble size, the bubble velocity will decrease. From Eq. 4, one observes that the decrease in bubble velocity will increase the stability. We may expect that a new dynamic equilibrium is reached when the bubble surface is marginally stable for the same set of wavelengths and when the disturbance has the same growth factors for the unstable wavelengths as in the case with lower gas density.

Inversely, when the gas density is decreased, the range of unstable wavelengths will decrease slightly, and the growth factors of unstable wavelengths will be smaller. Therefore, coalescence will be temporarily favored over breakup. This will result in a larger bubble size and, therefore, a larger bubble velocity, until a new equilibrium is reached.

In summary we can say that, although we do not know the details of the breakup process of bubbles, we can deduce the following from Eq. 4: two bubbles at densities ρ_{g1} and ρ_{g2} will have the same spectrum of growth factors, if their rise velocities, V_{b1} and V_{b2}, relate as

$$\rho_{g1}V_{b1}^2 = \rho_{g2}V_{b2}^2 \tag{5}$$

Therefore, using the Kelvin-Helmholtz theory we can predict that

$$V_b \propto \frac{1}{\sqrt{\rho_g}} \tag{6}$$

We shall test this conclusion by comparison with experiments.

Experimental Studies

Figure 3 shows the experimental setup. A glass bubble column, 0.15 m in diameter and 1.2 m high, is located in a steel vessel. The liquid phase is demineralized water, and nitrogen is sparged into the reactor through a 0.1-m-diameter perfo-

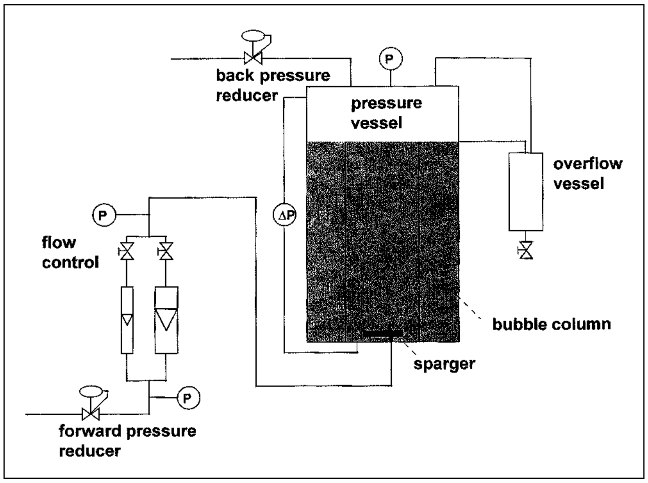

Figure 3. Experimental setup.

rated plate with 200 evenly distributed orifices, 0.5 mm in diameter. This ensures an equal distribution of the gas over the distributor area. The pressure in the vessel can be controlled with a back-pressure reducer. Gas fractions are measured by means of an overflow vessel, or by means of a Validyne DP15 pressure sensor. Both methods agree well.

Results

Figure 4 shows gas fractions measured at different superficial gas velocities at system pressures ranging from 0.1 to 1.3 MPa. The effect of elevated system pressure is clearly observable: the initial part of the gas holdup vs. gas velocity curve is linear. This linear relation breaks down in the heterogeneous regime. The departure from linearity is taken as the regime transition. As illustrated in Figure 4, this transition shifts to higher gas fractions and slightly higher gas velocities as the system pressure increases (Letzel et al., 1997).

From Figure 4, one can furthermore observe that the slope of the gas fraction curve in the heterogeneous regime increases with increasing gas density. At these higher gas velocities, the increase in gas fraction is caused only by the "large-bubble" phase, as was shown by Ellenberger and Krishna (1994).

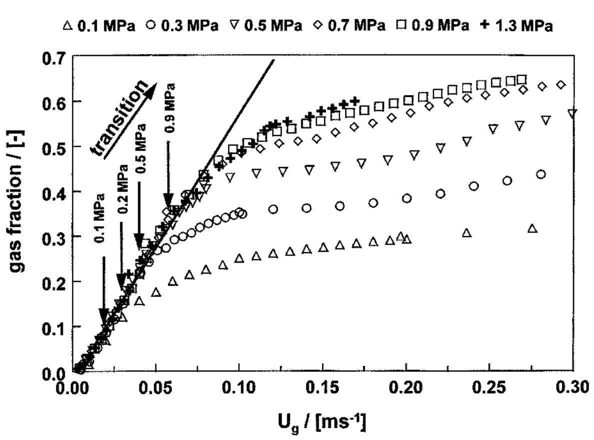

Figure 4. Gas fraction as a function of superficial gas velocity.

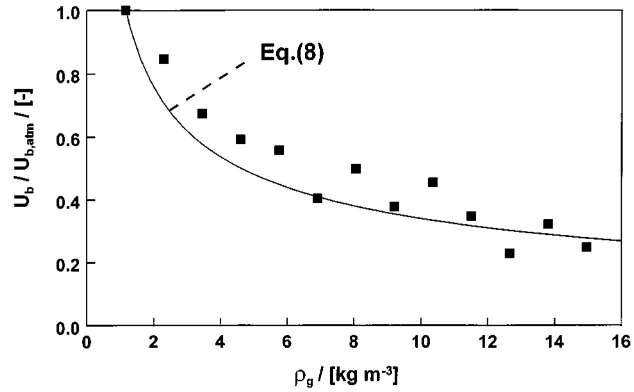

Figure 5. Bubble velocity at elevated vs. atmospheric pressure as a function of gas density, compared with the theoretical dependence (Eq. 8).

It is assumed here that in the heterogeneous flow regime the dense-phase gas fraction ϵ_{df} and the superficial gas velocity through the dense phase U_{df} ($\mathrm{m \cdot s^{-1}}$) are constant. The increase in gas velocity beyond U_{df} then results exclusively in an increase in the large-bubble fraction, $\epsilon_b = \epsilon - \epsilon_{df}$. With this assumption, we can estimate the average large-bubble velocity from the slope of the gas fraction curve, without the need of estimating ϵ_{df}. Since the slope is fairly constant at gas velocities above 0.1 $\mathrm{m \cdot s^{-1}}$, we estimate one average large-bubble velocity at each system pressure. If α is the slope of the curve in the heterogeneous regime, one can write for the average large-bubble velocity V_b.

$$V_b \approx \frac{\Delta U_g}{\Delta \epsilon} = \frac{1}{\alpha} \tag{7}$$

The average large-bubble velocity is thus inversely proportional to the slope of the gas fraction curve.

Figure 5 shows the relative change of the large-bubble velocity with increasing system pressure. $V_{b,\mathrm{atm}}$ is the average large-bubble velocity at atmospheric pressure ($\rho_g = 1.15\ \mathrm{kg \cdot m^{-3}}$). Also drawn in Figure 5 is the relation (cf. Eq. 6)

$$\frac{V_b}{V_{b,\mathrm{atm}}} = \frac{\sqrt{\rho_{\mathrm{atm}}}}{\sqrt{\rho_g}} \tag{8}$$

Since for $\rho_g = \rho_{\mathrm{atm}}$, we have $V_b/V_{b,\mathrm{atm}} = 1$. A good agreement between the experimentally determined bubble velocities and Eq. 8 is found.

Conclusions

From the Kelvin-Helmholtz theory, it can be concluded that the effect of increased gas density is that the surface of large bubbles becomes unstable for a wider range of wavelengths, and that the growth factors of the unstable surface waves increase. In a dynamic equilibrium between bubble coalescence and bubble breakup, the effect will be a shift of this equilibrium towards smaller bubbles. If we assume that for a new equilibrium the bubbles must be unstable for the same set of wavelengths and have the same growth factors at unstable wavelengths, we find that the velocity of the large bubbles should be inversely proportional with the square root of the gas density.

Large-bubble velocities were determined experimentally from gas fraction measurements. The observed change of bubble velocity with gas density agrees very well with the prediction from Kelvin-Helmholtz theory (Eqs. 6 and 8).

This result provides strong evidence that the physical explanation of the effect of pressure is correct; it will, therefore, prove helpful in predicting gas fractions in bubble column reactors operated at elevated pressures.

Notation

A = square root of $-c^2$, $m \cdot s^{-1}$
g = gravity acceleration constant, $m \cdot s^{-2}$
t = time, s
x = horizontal position on interface, m
λ = wavelength of disturbance, m
σ = surface tension, $N \cdot m^{-1}$

Literature Cited

de Swart, J. W. A., R. E. Van Vliet, and R. Krishna, "Size, Structure and Dynamics of 'Large' Bubbles in a Two-Dimensional Slurry Bubble Column," *Chem. Eng. Sci.*, **51**, 4619 (1996).
Ellenberger, J., and R. Krishna, "A Unified Approach to the Scale-Up of Gas-Solid Fluidized Bed and Gas-Liquid Bubble Column Reactors," *Chem. Eng. Sci.*, **49**, 5391 (1994).
Krishna, R., and J. Ellenberger, "Gas Holdup in Bubble Column Reactors Operating in the Churn-Turbulent Flow Regime," *AIChE J.*, **42**, 2627 (1996).
Krishna, R., P. M. Wilkinson, and L. L. Van Dierendonck, "A Model for Gas Holdup in Bubble Columns Incorporating the Influence of Gas Density on Flow Regime Transitions," *Chem. Eng. Sci.*, **46**, 2491 (1991).
Lamb, H., *Hydrodynamics*, 6th ed., Cambridge Univ. Press, Cambridge, U.K. (1959).
Letzel, H. M., J. C. Schouten, C. M. Van den Bleek, and R. Krishna, "Influence of Elevated Pressure on the Stability of Bubbly Flows," *Chem. Eng. Sci.*, **52**, 3733 (1997).
Sagert, N. H., and M. H. Quinn, "The Coalescence of H_2S and CO_2 Bubbles in Water," *Can. J. Chem. Eng.*, **51**, 392 (1976).
Wilkinson, P., "Physical Aspects and Scale-Up of High Pressure Bubble Columns," PhD Thesis, Univ. of Groningen, The Netherlands (1991).

Manuscript received Feb. 4, 1998, and revision received June 16, 1998.