# Progress in Understanding the Fluid Dynamics of Bubble Column Reactors - II

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High pressure operations are common in industrial applications of gas-liquid-solid fluidized beds and slurry bubble column reactors for resid hydrotreating, Fischer-Tropsch synthesis, coal methanation, methanol synthesis, polymerization and other reactions (Fan, 1989). The design and scale-up of these reactors require the knowledge of the hydrodynamics and heat and mass transfer characteristics in the reactor at high pressures. Works in the literature indicated that elevated pressures generally lead to an increased gas holdup in bubble column, slurry bubble column, and three-phase fluidized bed systems (Tarmy and Chang, 1984; Jiang et al., 1992; Luo et al., 1997). This phenomenon has been attributed to the small bubble size and bubble rise velocity at high pressures. The increased gas momentum in the bubble-formation process has been suggested as one factor behind the bubble size reduction. The mechanism through which the pressure affects the bubble formation and bubble rise velocity in slurry bubble column and three-phase fluidized bed systems is not currently understood.

The sizes of bubbles generated from gas distributors have a significant effect on the hydrodynamics and mass transfer in bubbling systems. Over the past decades, studies have been conducted on the bubble formation from a single orifice submerged in liquids. Mostly the investigations were under the ambient condition (Azbel, 1981); however, little is reported under elevated pressure conditions (Idogawa et al., 1987; LaNauze and Harris, 1974). An increase gas density was found to reduce the size of bubbles. Discrete bubbles are formed at low gas velocities. At a high gas velocity, jetting occurs and bubbles are formed from the top of the jet. In the discrete bubble regime, the bubble size is relatively uniform; in contrast, the bubbles formed from a jet are of a wide size distribution (Massimilla et al, 1961). The empirical correlation provided by Idogowa et al. (1987) indicated that the bubbling-jetting transition velocity in a liquid is proportional to the gas density to the power of -0.8. Few studies have been conducted in the presence of particles. The experimental data of Massimilla et al. (1961) revealed a significant effect of particles on the bubble formation under the ambient pressure condition. However, the effect of particles on the bubbling-jetting transition is still not clearly understood.

The objective of this work is to investigate the effects of particles and pressure on the bubble formation and the bubbling-jetting transition phenomena. Specifically, an optical fiber probe system is developed to study the bubbling and jetting behavior in liquid-solid media, along with visualization of the bubble formation and the jetting in a liquid. The initial bubble size is measured in shallow liquid-solid suspensions with various solids concentrations over a wide range of pressures. The bubbling-jetting transition velocity is identified from the probe signal, and the transition velocity is determined. In addition, the mechanisms underlying the particle and pressure effects on the bubble formation and the bubbling-jetting transition are discussed.

Further, the bubble rise characteristics in liquid-solid fluidized beds under frequently encountered industrial conditions, i.e. elevated pressure and temperature, and with a non-water based liquid medium, are investigated. The bubble rise velocity is studied in light of the apparent homogeneous (or effective) properties of liquid-solid fluidized beds under these conditions. A mechanistic model to predict the rise velocity is formulated based on a theoretical account of impact of particles onto the bubble. The effect of the variation in bubble rise velocity on the hydrodynamics of three-phase fluidized beds is also examined.

## Experimental

Figure 1 shows a schematic of the high pressure three-phase fluidized bed. The fluidized bed is a stainless steel column (1,372 mm in height and 101.6 mm ID) and can be operated at pressures up to 21 MPa and temperatures up to 180 °C. Three pairs of quartz windows are installed on the front and rear sides of the column. The liquid enters the column through a perforated plate distributor with 120 square-pitched holes of 1.0 mm diameter. A high-speed video camera (240 frames/second) is used to capture the images of bubbles emerging from the bed surface. The bubble size and frequency are obtained via image analysis. Nitrogen is injected into the liquid-solid medium through a stainless steel tubing of 3.175 mm OD and 1.585 mm ID. Further, a single-bubble generator is installed for the bubble rise velocity measurement. The bubble generator precisely controls the amount of gas flowing in. The liquid phase is Paratherm NF heat transfer fluid (viscosity = 0.028 Pa·s; surface tension = 0.029 N/m; and density = 869 kg/m<sup>3</sup> at 26.5 °C and 0.1 MPa).

An optical fiber probe system is developed for the detection of bubbles or jets at high pressures. The probe utilizes the difference in refractive index of gas and liquid to distinguish the gas phase from the liquid-solid suspension. The fiber cladding in the tip portion of the probe is partially ground in such a manner that it yields distinctive signals for gas void detection. The cross section of the tip perpendicular to the flow direction in this probe has a dimension of  $0.5 \times 4$  mm. The output of the photomultiplier is interfaced with a computer data acquisition system, which samples the signal for four seconds at a frequency of 5,000 Hz. The probe is movable in the radial direction under high pressure conditions, so that the tip and the orifice can be precisely aligned. This feature also permits observation of the influence of the tip on the bubble flow or jetting phenomena. Based on visualization, the probe is found to impose negligible disturbances on the bubble formation process and the bubbling-jetting transition, although it would alter the trajectories of bubbles.

### Gas Jetting and Bubble Formation

Injections of gas through an orifice are visualized to establish objective criteria for the bubblingjetting transition in liquid-solid suspensions over a wide range of the orifice Reynolds numbers,  $\text{Re}_{o,g}$ . Further, light intensity signals obtained by the probe are compared. At  $\text{Re}_{o,g} = 1,075$ , single bubbles are formed from the orifice. With increasing  $\text{Re}_{o,g}$  to 5,321, bubbles being formed at the orifice start to interact with the preceding ones. Bubble coalescence occurs between two bubbles and, sometimes, involving three and even four bubbles. At  $\text{Re}_{o,g} = 8,809$ , frequent coalescence of successive bubbles is observed, and a jet-like gas plume appears, which rarely breaks up near the orifice. This indicates the beginning of the bubbling-jetting transition. Under the jetting condition, the signals become irregular in shape and yield a wide range of dominant frequencies. At a higher  $\text{Re}_{o,g}$ , bubbles of various sizes break away from the top of the jets. Further, the jet penetration depth increases with an increase in  $\text{Re}_{o,g}$ . The behavior of void time fraction, another parameter calculated from the signal, can also be used to identify the regime transition. The void time fraction is defined as the ratio of the time occupied by the gas phase to the total sampling time. The void time fraction starts to level off around the regime transition.

The presence of particles in liquid has a significant effect on the motion of bubbles. Flow visualization of bubble formation in the liquid, with or without continuous liquid flow, reveals that all the bubbles follow the same trajectory. Once the probe is properly aligned with the orifice, the trajectory of the bubbles intercepts the tip of the probe, yielding consecutive steady peaks in the light intensity signals in the bubbling regime. However, the bubbles in liquid-solid suspensions have various trajectories. The probe can only detect bubbles periodically. Flow visualization also confirms that the bubbles escape the bed surface at different locations. The unsteady bubble trajectories in liquid-solid suspensions are due to the heterogeneous nature of the suspensions.

Figure 2 shows the effect of particles on the initial bubble size. The bubble size is obtained at the same distance above the orifice in the liquid and the liquid-solid suspensions. At both the ambient and 4.2 MPa pressures, the bubbles formed in the liquid-solid suspensions are larger than those formed in the liquid for a given  $\text{Re}_{o,g}$ . The bubble size increases with the solids holdup. The experimental data of Massimilla et al. (1961) showed a similar trend. This is because in addition to the buoyancy, surface tension, and liquid viscous forces that bubbles experience during the formation process in the liquid, bubbles also experience the drag force induced by particles in liquid-solid suspensions. Considering a pseudo-homogeneous medium, the apparent density and viscosity of a liquid-solid suspension are generally higher than those of the liquid. At a very low  $\text{Re}_{o,g}$ , the bubbles formed in these two systems are of the same size (0.23 cm). Above a certain  $\text{Re}_{o,g}$ , the initial bubble size for both systems starts to increase with  $\text{Re}_{o,g}$ .

It is found that the pressure has a significant effect on the bubble formation in the liquid-solid suspension as well as in the liquid (Fig. 3). In general, increasing pressure decreases the bubble sizes for a given solids holdup,  $\text{Re}_{o,g}$ , and temperature. The pressure, however, does not affect the initial bubble size at a low  $\text{Re}_{o,g}$ . Under this condition, the bubble size is dictated by the buoyancy and surface tension forces, and the effect of the gas momentum is negligible. In the range of high  $\text{Re}_{o,g}$ , the momentum of gas plays an important role in the force balance during the bubble formation process. The increased gas momentum at elevated pressures leads to earlier detachment of the bubble, and hence, a smaller bubble size. The pressure effect is relatively small when the pressure is above 4.2 MPa. This is because that the gas density is increased by 42 times when the pressure is elevated from 0.1 to 4.2 MPa; while at pressures above 4.2 MPa, the gas density is only increased by a maximum of 4.1 times.

The transition from the bubbling to the jetting is identified based on the characteristics of the signals from the optical fiber probe. The effects of particle and pressure on the transition are shown in Fig. 4. The presence of particles reduces the transition gas velocity. Particles affect the bubble-bubble interaction. The bubble wake is larger in the suspension than that in the liquid because of the larger bubble size in the suspension. The larger bubble wake results in a stronger bubble-bubble interaction, which induces the transition occurring at lower orifice gas velocities in the suspension than in the

liquid. It is noted that the particle size used in this study is 210  $\mu$ m. The particle effect on jetting phenomenon would be different in larger particle systems; for example, particles larger than 2.5 mm would enhance bubble breakup (Kim et al., 1977). It can be seen from Fig. 4 that increasing the pressure decreases the transition gas velocity in both the liquid and the liquid-solid suspensions due to the increased gas momentum.

### Pressure and Temperature Effects on Bubble Rise Velocity

The bubble rise velocity decreases with an increase in pressure for a given solids holdup. The extent of reduction is as high as by 50% from 0.1 to 17.3 MPa. More drastic reduction in U<sub>b</sub>, however, arises from the addition of solid particles. While the particle effect is small at low solids holdup ( $\varepsilon_s < 0.4$ ), the effect is appreciable at high  $\varepsilon_s$  (= 0.545), especially for high liquid viscosity. A comparison of the data at 26.5°C and 87.5°C, for the same  $\varepsilon_s$  of 0.545, indicates that the viscosity effect appears to be significant. The reduction of the bubble rise velocity with an increase in pressure can lead to a significant increase in the gas holdup of three-phase fluidized beds. The extent of the increase in gas holdup was reported to be around 100% at all gas velocities when the pressure is increased from 0.1 to 15.6 MPa (Luo et al., 1997). By comparing the pressure effect on the gas holdup with that on the bubble rise velocity, it can be stated that the increase in gas holdup with pressure is a consequence of the decreases in both the bubble size and the bubble rise velocity, i.e. larger bubbles broken into smaller ones and their rise velocities further reduced by pressure.

The decrease in bubble rise velocity occurs due to corresponding variations of gas and liquid properties with pressure. In the presence of solid particles, it is assumed that the particles modify only homogeneous properties of the surrounding medium. The calculated results based on the Fan-Tsuchiya correlation (Fan and Tsuchiya, 1990) are extended to liquid-solid suspensions (Tsuchiya et al., 1997) and generalized for high-pressure conditions.

Jean and Fan (1990) developed a theoretical/mechanistic model that accounts for impact forces on a rising bubble due to particles. The model can predict the bubble rise velocity for small particles ( $d_p < 500 \text{ mm}$ ), low-to-intermediate solids holdups ( $\varepsilon_s < 0.45$ ) and large spherical-cap bubbles ( $d_e > 15 \text{ mm}$ ). Their model is extended to cover a smaller bubble size range. The model is based on a force balance on a rising bubble involving the net gravity, liquid drag and particle-bubble collision forces.

## Conclusion

The bubble formation from a single nozzle is experimentally studied in both the liquid and the liquid-solid suspensions. The initial bubble size in the suspension is generally larger than that in the liquid. The initial bubble size in the liquid is determined through the balance of the buoyancy, the surface tension, and the liquid viscous forces. An additional drag force on the bubble in liquid-solid suspensions, induced by bubble-particle interactions, contributes to the larger initial bubble size in the suspension. As the pressure increases for a given Re<sub>o,g</sub>, the initial bubble size decreases due to the increased gas momentum. The transition from the bubbling to the jetting is identified via gas void signals obtained from the optical fiber probe. It is found that the bubbling-jetting transition in the liquid-solid suspensions occurs at a lower gas velocity than that in the liquid, due to inherent bubble

coalescence characteristics in the suspension. The nozzle gas velocity at the transition decreases with an increase in pressure, in both the liquid and the liquid-solid suspensions.

The rise velocity of single bubbles in liquid-solid fluidized beds decreases with an increase in pressure and with a decrease in temperature. This decrease, combined with the pressure (and temperature) effect on reducing the bubble size, contributes to high gas holdups observed at high pressures. The bubble rise velocity in liquids and liquid-solid suspensions with low solids holdups can be reasonably estimated using the predictive equation available for ambient conditions, if the *in-situ* physical properties of the gas and liquid are used. Significant reduction in the rise velocity occurs at high solids holdups, especially for high liquid viscosity. The extent of reduction can be examined in terms of an increase in the apparent suspension viscosity by applying the homogeneous, Newtonian analogy. A mechanistic analysis indicates that the heterogeneous characteristics of liquid-solid suspensions can be satisfactorily accounted for by considering the particle-bubble collision behavior.

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Figure 1. Schematic of the experimental setup. (1. Liquid inlet; 2. Perforated plate distributor; 3. Quartz windows; 4. High Speed Video camera; 5. TV and VCR; 6. Gas and liquid outlet; 7. Lighting; 8. Optical fiber probe; 9. Light source; 10. Photomultiplier; 11. Data acquisition system; 12. Nitrogen cylinder.)



Figure 2. Effect of particles on the initial bubble size at various  $Re_{o,g}$ : (a) P = 0.1 MPa; (b) P = 4.24 MPa.



Figure 3. Pressure effects on the bubble formation (a) in liquid; (b) in liquid-solid suspensions,  $\varepsilon_s = 0.54$ .



Figure 4. Effects of pressure and particle on the bubbling-jetting transition.