

OHIO STATE UNIVERSITY

The report from Ohio State for the period follows.

INTRINSIC FLOW BEHAVIOR IN A SLURRY BUBBLE COLUMN UNDER HIGH PRESSURE AND HIGH TEMPERATURE CONDITIONS

Quarterly Report

(Reporting Period: October 1 to December 31, 2000)

Highlights

- The study of gas injection phenomena from a submerged single orifice in liquid-solid suspensions at high pressures has been completed. The effects of pressure and solids concentration on the bubbling-jetting transition velocity have been investigated.
- The bubbling-jetting transition velocity decreases with increasing pressure, and increases with the presence of particles.
- The study of flow fields and Reynolds stresses at high pressures using a two-dimensional laser Doppler velocimetry (LDV) system has been initiated. Experiments are currently being conducted in the 2-inch high-pressure vessel.
- The study of axial liquid-phase mixing at high pressures has been initiated. Previous studies on liquid-phase mixing at elevated pressures and measurement techniques were reviewed.

Work Conducted

1. Study of Bubbling-Jetting Transition

The effects of pressure and particle presence on the bubbling-jetting transition velocity from a single orifice connected to a gas chamber were studied. Figure 1 shows the effects of pressure and solids concentration on the transition velocity from the bubbling to jetting regimes. For both the liquid and the liquid-solid suspension, when pressure or gas density increases, the bubbling-jetting transition velocity decreases significantly, especially in the low-pressure range. At very high pressures, the decrease in the transition velocity with increasing pressure is relatively small. The acceleration of the transition to the jetting regime at high pressures is mainly due to an increase in the gas momentum. Based on the experimental data, it was found that the effect of pressure on the transition velocity can be expressed by the following relationship:

$$u_{jetting} \propto \rho_g^{-n} \quad (1)$$

where the index n is a function of the solids concentration. For the system used in this study, n varied from 0.3 for liquids to 0.4~0.5 for liquid-solid suspensions.

The effect of particle presence on transition velocity is also provided in Figure 1, which shows that the particle effect strongly depends on the pressure. In the low-pressure range ($P < 2.5$ MPa), the effect of particles on the transition velocity is significant. The presence of particles increases the transition gas velocity significantly. For example, at atmospheric pressure, the transition velocity in the liquid is about 7.0 m/s. When the solids concentration increases to 0.18 vol %, the transition velocity increases from 7.0 to 12.0 m/s (71% increase). In the high-pressure range ($P > 2.5$ MPa), the effect of solids concentration on the transition velocity is insignificant.

The effect of pressure on the critical mass flux of gas is shown in Figure 2. The critical mass flux of gas is defined as the mass flux of gas at the transition point, *i.e.*, equal to $\rho_g u_{jetting}$. It can be seen that the critical mass flux of gas is not constant and increases with an increase in pressure. At very high pressures ($P > 10$ MPa), the critical mass flux of gas tends to approach a constant value.

The effects of pressure and solids concentration on the critical Weber number, defined as the

Weber number at the transition point, $We_{cr} (= \frac{\rho_g u_{jetting}^2 D_0}{\sigma})$, are shown in Figure 3. For

bubbling-jetting transition in liquids, the critical Weber number increases significantly with an increase in pressure, which is possibly due to the increased liquid viscosity with pressure. Rabiger and Vogelpohl (1982) also found that the critical Weber number increases with an increase in liquid viscosity. At atmospheric pressure, the critical Weber number is about 3 for the present liquid system, while at high pressures (*e.g.*, $P = 8.3$ MPa), the critical Weber number is above 10. With an increase in solids concentration, the difference in the critical Weber number among various pressures becomes smaller. As shown in the figure, when the solids concentration reaches 30 vol %, the critical Weber number is almost constant over the pressure range studied. The effect of solids concentration on the critical Weber number is insignificant at high pressures.

2. Study of Flow Fields and Reynolds Stresses

In order to achieve better penetration of laser beams, new pairs of quartz windows and spacers were installed in the 2-inch, high-pressure column. The test and calibration of the LDV system were completed, and the measurements of liquid velocity profiles at high pressures are currently being conducted in the 2-inch vessel. Some preliminary results will be reported in the next quarter.

3. Study of Axial Liquid-Phase Mixing

Literature Review

Liquid-phase mixing is an important parameter in the design of industrial reactors. The liquid-phase mixing strongly depends on the hydrodynamics and bubble characteristics. Previous studies have shown that pressure has significant effects on gas holdup, bubble size and bubble rising velocity. Therefore, the study of pressure effect on liquid-phase mixing is necessary to further understand flow behaviors in bubble columns and slurry bubble columns.

The studies regarding the axial liquid-phase mixing in gas-liquid systems at atmospheric conditions are extensive, especially for the air-water system; however, studies under high-pressure conditions are very scarce. Houzelot et al. (1985) measured the axial dispersion of the liquid phase in a bubble column with a diameter of 5 cm. They found that pressure did not affect axial dispersion, which was limited by the narrow experimental conditions in their study, *i.e.*, very low superficial gas velocity (<6 mm/s) and pressure (< 3 atm). Under very low gas velocities, the flow was always in the homogeneous bubbling regime, and a significant change in liquid-phase mixing was not expected. Sanchinnuan et al. (1984) experimentally investigated the extent of liquid-phase backmixing under coal hydroliquefaction conditions (*e.g.*, temperature between 164 and 384°C and pressure between 4.5 and 15 MPa) in a small bubble column reactor (1.9 cm in diameter). They did not describe the effect of pressure on axial mixing, and their study was limited by the small scale of the reactor.

Holcombe et al. (1983) determined the liquid axial-dispersion coefficient in a 7.8-cm diameter bubble column under pressures in the range of 3.0~7.1 atm. The superficial gas velocity varied up to 0.6 m/s. They used heat as a tracer to measure the thermal dispersion coefficient, which was found to be comparable to the mass dispersion coefficient. In their study, the effect of pressure on thermal dispersion coefficients was negligible. Wilkinson et al. (1993) measured the liquid axial-dispersion coefficient in a batch-type bubble column of 0.158-m diameter for the water-nitrogen system at pressures between 0.1 and 1.5 MPa using the electrical conductivity cell. They found that the liquid axial-dispersion coefficient actually increased with increasing pressure, especially under high gas velocity (> 0.10 m/s). They also found that the available theories in the literature describing liquid mixing under atmospheric pressure could not explain the pressure effect observed in their study. They proposed a procedure to estimate the liquid-phase dispersion coefficient at elevated pressures based on liquid holdup and the dispersion coefficient at ambient pressure, *i.e.*,

$$E_l(\text{high pressure}) = E_l(\text{atmospheric}) \cdot \frac{\varepsilon_l(\text{atmospheric})}{\varepsilon_l(\text{high pressure})} \quad (2)$$

Wilkinson et al.'s 1993 study was also limited by the narrow experimental conditions (low pressure) and limited batch system (air-water), and their conclusion on the pressure effect and the proposed correlation need to be further verified. Tarmy et al. (1984) used radioactive tracers to study the liquid back-mixing in pilot coal liquefaction reactors. They found that the measured dispersion coefficients at high pressures (17 MPa) were up to 2.5 times smaller than the predictions by literature correlations, which usually were proposed for ambient conditions in air-water systems. However, they did not present their mixing data in the paper. The detailed

information of high-pressure studies available in the literature regarding liquid-phase mixing in bubble columns is shown in Table 1.

Objective of Study

Although some research has studied the effect of pressure on liquid mixing in bubble columns, those studies were confined to low gas velocities or low pressures (less than 1.5 MPa), small column sizes and limited systems (air-water). Furthermore, the effect of pressure effect was still not conclusive. Further systematic studies are needed to cover a wide range of operating conditions and should be conducted in systems close to industrial applications. The objective of this study is to develop the suitable measurement technique to investigate axial liquid-phase mixing in a hydrocarbon liquid at high pressures.

Measurement Technique

Axial mixing of the liquid phase is normally described by a one-dimensional dispersion model. The dispersion coefficient can be determined by unsteady and steady tracer injection methods. It has been verified that both measurement principles lead to the same results (Deckwer et al., 1974). In the steady injection method, a tracer is injected at the exit or some other convenient point, and the concentration profile is obtained by sampling upstream. With the unsteady injection method, a variable flow of tracer is injected, usually at the contactor inlet, and samples are normally taken at the exit. The most common form of injection is the single impulse, and the response of the tracer input is then obtained at the sampling point. Other forms of injection are the step change, oscillatory and random forms. The tracer for both the steady and unsteady tracer injection methods can be an electrolyte, a dye or heat. The mass dispersion method normally uses electrolyte as the tracer, and the change in electrical conductivity is measured. This method is only suitable for aqueous liquids. The thermal dispersion method uses heat as the tracer, and the temperature distribution within the column is measured. This method can also be applied to non-aqueous liquids, for example, hydrocarbon liquids. The thermal dispersion method is thought to give more accurate results than the mass dispersion method, because the experimental operation is easily achieved for the former.

In this study, because the liquid used was hydrocarbon liquid, the thermal dispersion method was chosen to study the axial liquid-phase mixing. The axial dispersion coefficients of the liquid phase were measured by the steady-state thermal dispersion method, *i.e.*, introducing heat close to the outlet of the liquid phase and measuring the upstream temperature profile in the liquid when the temperature distribution reached steady state. The schematic of typical axial temperature distribution across the column is shown in Figure 4. The dispersion coefficient can be determined from the temperature profile based on the one-dimensional dispersion model. Since the heat capacity of gas is much smaller than that of liquid, the temperature change in the column is mainly due to the backmixing of liquid. Considering that the heat losses through the column wall and the gas-liquid interface are negligible, the following differential energy balance equation applies (Aoyama et al., 1968; Wendt et al., 1984):

$$\frac{k_l}{\rho_l C_{pl}} \frac{d^2 T}{dz^2} + \frac{U_l}{1 - \varepsilon_g} \frac{dT}{dz} = 0 \quad \text{with } E_l \equiv \frac{k_l}{\rho_l C_{pl}} \quad (3)$$

where E_l is the effective thermal dispersion coefficient and is comparable to the mass dispersion coefficient, z is the axial position from the liquid outlet and $z=0$ represents the liquid outlet. This differential equation can be solved analytically after the boundary conditions for a semi-infinite reactor are introduced, *i.e.*,

$$\begin{aligned} T &= T_m \quad \text{at } z = 0 \\ T &= T_0 \quad \text{at } z = \infty \end{aligned} \quad (4)$$

where T_0 and T_m are the inlet and outlet liquid temperatures, respectively. T is the liquid temperature at distance z from the liquid outlet. The analytical solution of the differential equation is:

$$\ln \frac{T - T_0}{T_m - T_0} = -\frac{U_l}{E_l(1 - \varepsilon_g)} z \quad (5)$$

Equation (5) indicates that the relationship between $\ln[(T-T_0)/(T_m-T_0)]$ and z is linear, and the dispersion coefficient, E_l , can be calculated from the slope of the temperature distribution curve, provided that the dispersion coefficient is constant and the gas holdup and superficial liquid velocity are known. It was proven that dispersion coefficients measured by mass and thermal methods are comparable, as shown in Figure 5 (Wendt et al., 1984). The validity of the thermal dispersion technique was also verified by other researchers (Aoyama et al., 1968; Holcombe et al., 1983).

Future Work

The axial dispersion coefficients of the liquid phase will be measured by the steady-state thermal dispersion method. The study of flow fields and Reynolds stresses at high pressures using a two-dimensional laser Doppler velocimetry (LDV) system will be undertaken.

Notations

C_{pl}	heat capacity of liquid, J/(kg·°C)
D_0	orifice diameter, m
E_l	liquid-phase dispersion coefficient, m ² /s
k_l	thermal conductivity of liquid, W/(m·°C)
P	system pressure, Pa
T	temperature at axial position z , °C
T_0	liquid inlet temperature, °C
T_m	liquid outlet temperature, °C
u_0	superficial orifice gas velocity, m/s
$u_{jetting}$	bubbling-jetting transition velocity, m/s
U_l	superficial liquid velocity, m/s
V_c	gas chamber volume, m ³

We	Weber number, $\frac{\rho_g u_0^2 D_0}{\sigma}$, dimensionless
Z	axial height from the gas-liquid outlet, m
ε_g	gas holdup, dimensionless
ε_l	liquid holdup, dimensionless
ε_s	solids concentration, dimensionless
ρ_g	gas density, kg/m ³
ρ_l	liquid density, kg/m ³
σ	surface tension, N/m

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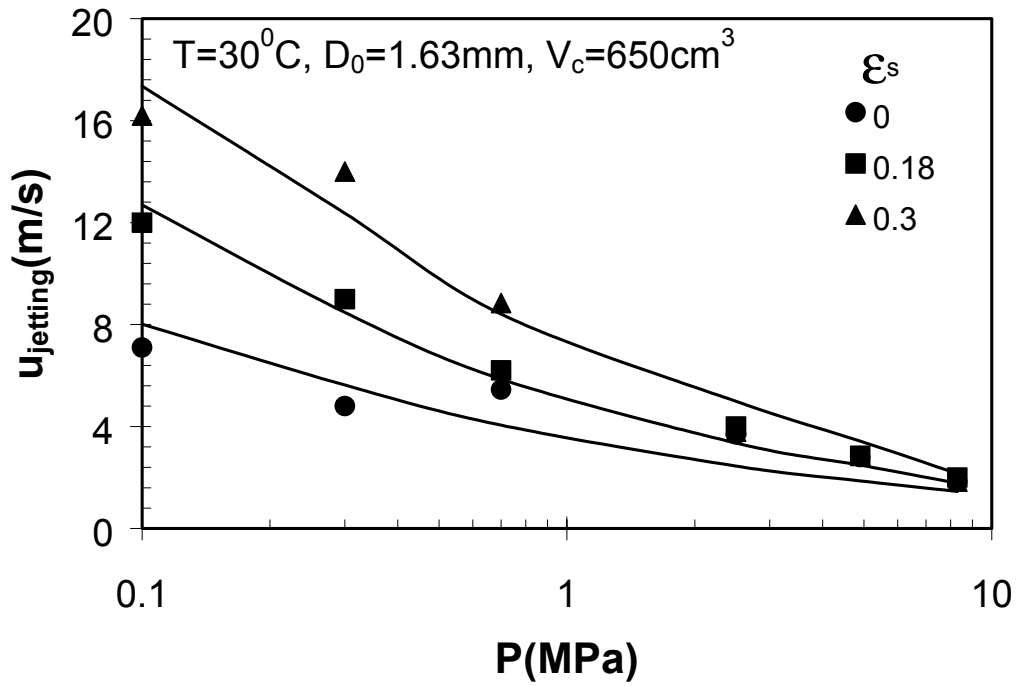


Figure 1 Effects of Pressure and Solids Concentration on Bubbling-Jetting Transition Velocity

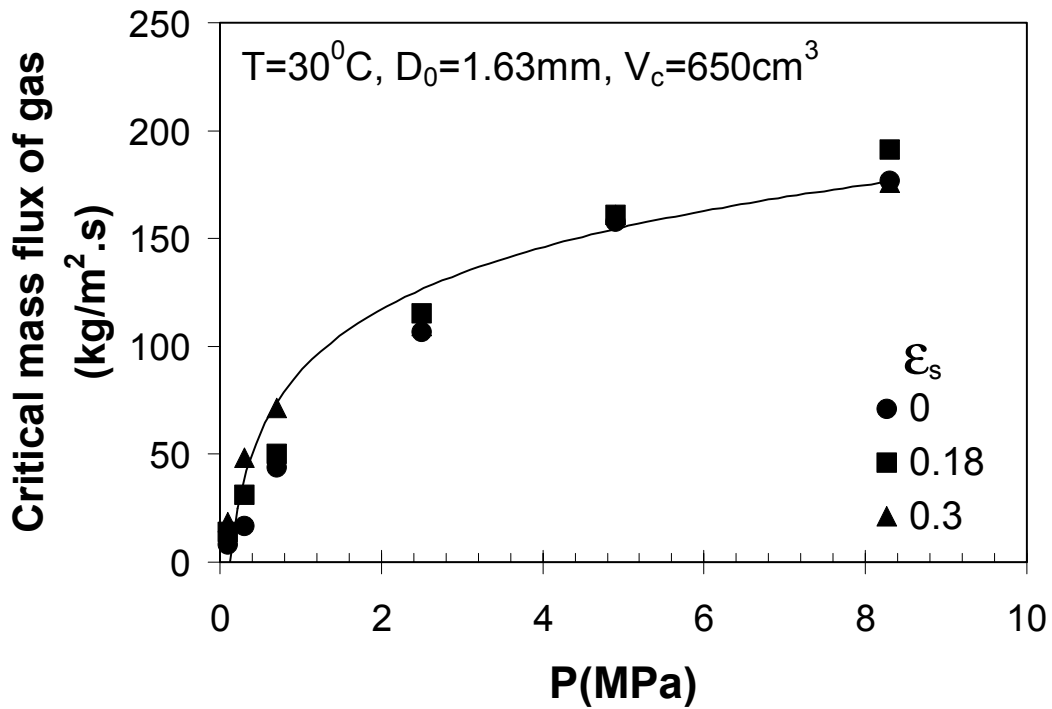


Figure 2 Effect of Pressure on the Critical Mass Flux of Gas Through the Orifice in Liquid and Liquid-Solid Suspensions

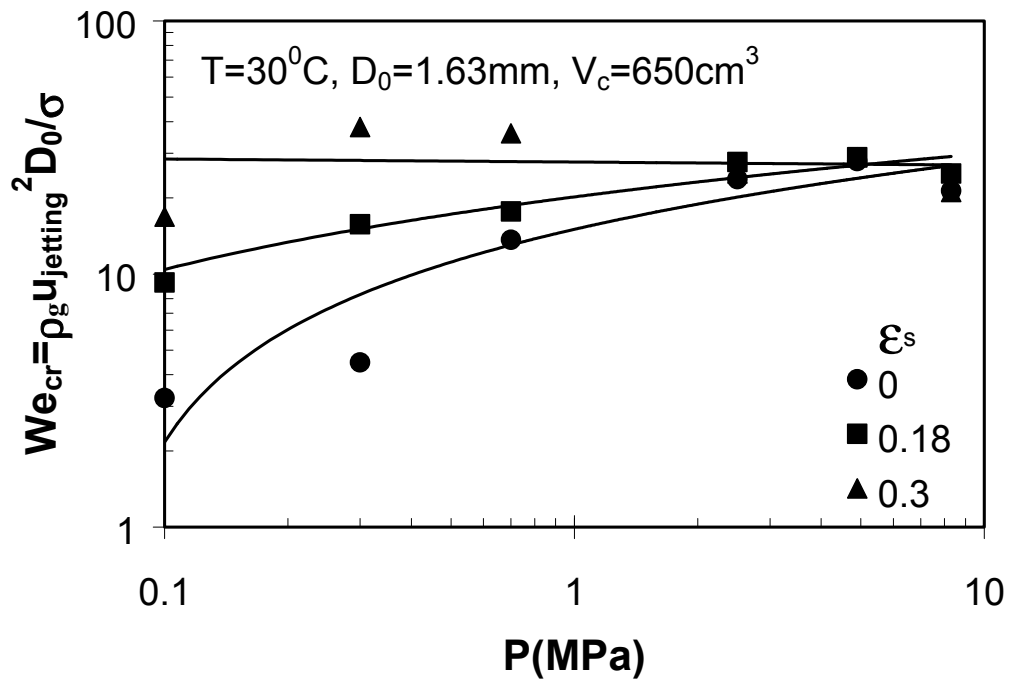


Figure 3 Critical Weber Number as a Function of Pressure and Solids Concentration

Table 1 Available High-Pressure Studies in the Literature Regarding Liquid-Phase Mixing in Bubble Columns

Reference	Technique	System	D (cm)	P (MPa)	U_g (cm/s)	U_l (cm/s)	Pressure effect
Holcombe et al. (1983)	thermal	N ₂ -water	7.8	0.3~0.7	0~60	0~2.0	insignificant
Sancinmuan et al. (1984)	mass	N ₂ -tetralin	1.9	4.5~15	0.2~1.1	0.1~0.3	insignificant
Houzelot et al. (1985)	mass	N ₂ -water	5.0	0.3	0.25~0.6	0.025~0.1	insignificant
Wilkinson et al. (1993)	mass	N ₂ -water	15.8	0.1~1.5	0~20	batch	increase
Tarmy et al. (1984)	mass	H ₂ -hydrocarbon-	2.4				
	(radioactive	coal	6.6	17	0.6~6.5	0.15~1.6	decrease
	tracer)		61				

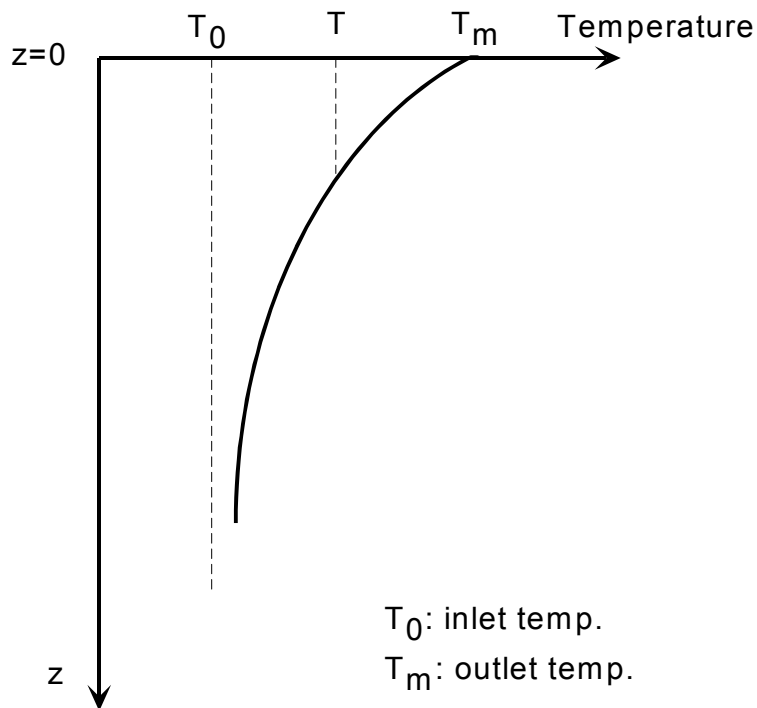


Figure 4 Typical Axial Temperature Distribution for Steady-State Thermal Dispersion Method

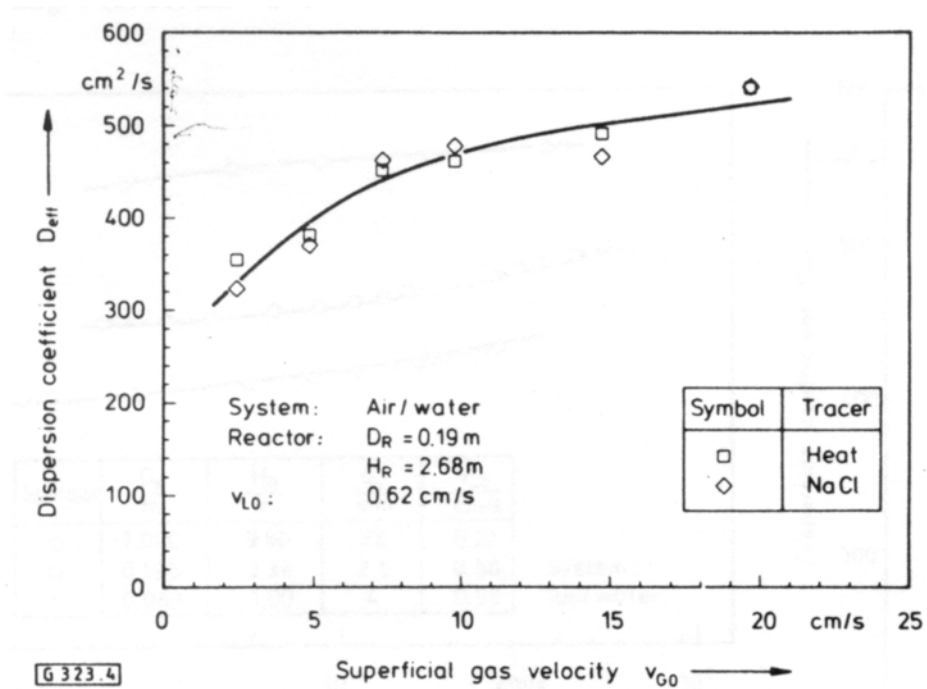


Figure 5 Comparison of Dispersion Coefficients Measured by Mass and Thermal Methods (Wendt et al., 1984)