ENGINEERING DEVELOPMENT OF SLURRY BUBBLE COLUMN REACTOR (SBCR) TECHNOLOGY

**Quarterly Technical Progress Report No. 19
For the Period 1 October – 31 December 1999**

FINAL

Contractor
AIR PRODUCTS AND CHEMICALS, INC.
7201 Hamilton Blvd.
Allentown, PA 18195-1501

Bernard A. Toseland, Ph.D.
Program Manager and Principal Investigator

Robert M. Kornosky
Contracting Officer's Representative

Prepared for the United States Department of Energy
Under Cooperative Agreement No. DE-FC22-95PC95051
Contract Period 3 April 1995 – 2 April 2000

**NOTE: AIR PRODUCTS DOES NOT CONSIDER ANYTHING IN THIS
REPORT TO BE CONFIDENTIAL OR PATENTABLE.**

ENGINEERING DEVELOPMENT OF SLURRY BUBBLE COLUMN REACTOR (SBCR) TECHNOLOGY

Quarterly Technical Progress Report No. 19 For the Period 1 October – 31 December 1999

Contract Objectives

The major technical objectives of this program are threefold: 1) to develop the design tools and a fundamental understanding of the fluid dynamics of a slurry bubble column reactor to maximize reactor productivity, 2) to develop the mathematical reactor design models and gain an understanding of the hydrodynamic fundamentals under industrially relevant process conditions, and 3) to develop an understanding of the hydrodynamics and their interaction with the chemistries occurring in the bubble column reactor. Successful completion of these objectives will permit more efficient usage of the reactor column and tighter design criteria, increase overall reactor efficiency, and ensure a design that leads to stable reactor behavior when scaling up to large diameter reactors.

WASHINGTON UNIVERSITY IN ST. LOUIS

The report from Washington University for the period follows.

ENGINEERING DEVELOPMENT OF SLURRY BUBBLE COLUMN REACTOR (SCBR) TECHNOLOGY

**Nineteenth Quarterly Report
for
October 1 – December 31, 1999
(Budget Year 5 – 19th Quarter)**

**Chemical Reaction Engineering Laboratory (CREL)
Chemical Engineering Department
Washington University**

Objectives for the Fifth Budget Year

The objectives set for the Fifth Budget Year (October 1, 1999 to September 30, 2000) are listed below.

- Extension of CARPT database to high superficial gas velocity in bubble column
- Extension CARPT/CT database to gas-liquid-solid system at high superficial gas velocity
- Evaluation of the effect of sparger design on the fluid dynamics of bubble columns using the CARPT technique
- Interpretation of LaPorte tracer data
- Further improvement in Computational Fluid Dynamics (CFD) using CFDLIB and Fluent

HIGHLIGHTS FOR THE 19TH QUARTER

During this quarter we focused on utilizing the collected Computer Automated Radioactive Particle Tracking (CARPT) data in developing numerical means for assessing the state of liquid flow and mixing in bubble columns. These related efforts were pursued in parallel: 1) evaluation of the global liquid re-circulation profiles via a one-dimensional global model; 2) dynamic simulation of fluid dynamics in a 14-cm diameter column and comparison of simulation results with available CARPT data; and 3) dynamic simulation of tracer distribution in the 14-cm diameter column. All of these efforts are continuing, but the accomplishments of the past quarter are summarized below.

1. One-dimensional model for liquid velocity distribution in a bubble column

- The database collected by Computer Automated Radioactive Particle Tracking (CARPT) and Computer Tomography (CT) is being systematically analyzed in search of patterns needed for scaleup.
- Scaling the data for liquid re-circulation velocity in a column of a single diameter with superficial gas velocity yields two velocity profiles. One is characteristic of bubbly flow, and the other of churn turbulent flow.
- Liquid re-circulation velocities increase with column diameter, and the proper scaling factor is being sought.
- Reynolds shear stress increases with superficial gas velocity and column diameter.

2. Dynamic simulation of 14-cm column and comparison with CARPT measurement

- The numerically predicted global gas holdup, based on CFDLIB codes, i.e., the dynamic heights of the column, is in good agreement with the experimental measurements.
- The mean liquid axial velocity profiles, calculated by time- and azimuthal-averaging of the numerically simulated velocity field, agree well with the CARPT measurement in the case of the churn turbulent regime.
- Comparison of the computed Reynolds stress distribution with CARPT data indicates that such comparison is far from straightforward and sheds additional insight into the use of turbulence models.

3. Dynamic simulation of tracer distribution in bubble columns

- The dynamic numerical simulation of passive scalars, in both liquid and gas phases, in a fully developed bubble column flow has been successfully accomplished using the three-dimensional CFDLIB code.
- The procedure for evaluating the averaged axial turbulent eddy diffusivity of the liquid phase is outlined.

- The preliminary results for the estimated axial liquid eddy diffusivity in a 14-cm diameter column operated at 2.4 cm/s superficial gas velocity is within the same range as the experimental data measured by CARPT.
- The effect of the gas distributor on turbulent eddy diffusivity is observed experimentally.

1. ONE-DIMENSIONAL MODEL FOR LIQUID VELOCITY DISTRIBUTION IN BUBBLE COLUMNS

1.1 Introduction

For design and scaleup purposes, it is important to have a reliable model to predict liquid recirculation. We used the CARPT/CT data base to develop a dimensionless model that should be useful for scaleup. The one-dimensional model equation for the time-averaged liquid velocity profile is

$$\frac{1}{r} \frac{d}{dr} \left[r(1 - \bar{\varepsilon})(\tau_t + \tau_m) \right] - \frac{dP}{dz} - \rho_l(1 - \bar{\varepsilon})g = 0 \quad (1.1)$$

where the turbulence shear stress, τ_t , and the molecular viscous shear stress, τ_m , are respectively defined as

$$\tau_t = -\rho_l \langle u'_r u'_z \rangle \quad \tau_m = \mu \frac{\partial \langle u_z \rangle}{\partial r}; \quad (1.2)$$

The azimuthal-averaged gas holdup, $\bar{\varepsilon}$, is a function of radial position. To validate a one-dimensional model, one needs information on the radial profile of the mean liquid velocity, $\langle u_z \rangle$, and Reynolds shear stress, $\langle u'_r u'_z \rangle$, which can be extracted from the CARPT database. In general, all these quantities are dependent on a number of process parameters such as superficial gas velocity, column size, physical properties of liquid and gas, and gas sparger type. Among these, the superficial gas velocity, U_g , and column diameter, D , are the two key parameters that dominate the fluid dynamics of a bubble column. In order to scale up and design bubble columns, a one-dimensional model in non-dimensional form is needed. Therefore, the column diameter and superficial gas velocity become the natural candidates for the characteristic integral length scale and velocity scale. To study the effects of D and U_g , we calculate and analyze $\langle u_z \rangle$ and $\langle u'_r u'_z \rangle$ for a 6 in. column operated at various superficial gas velocities and for 6, 8 and 18 in. columns operated at roughly the same superficial gas velocity of 12 cm/s.

1.2 Results and Discussion

Figure 1.1 shows the time-averaged liquid velocity profiles for the 14-cm diameter column at different operating conditions. As expected, $\langle u_z \rangle$ is proportional to the superficial gas velocity, U_g . The effect of U_g on the interstitial liquid velocities in the down-flow region is less than that in the up-flow region. This is due to the large area in the portion of the column where liquid flows downward and to the much higher liquid holdup in this region.

Scaling the liquid velocity by the superficial gas velocity, we reproduce the content of Figure 1.1 in Figure 1.2. It is immediately evident that the curves fall into two groups, one for U_g values of 2.4 and 4.8 cm/s, and the other for U_g values of 9.6 and 12 cm/s, which clearly represent the bubbly flow regime and churn turbulent flow regime, respectively. The fact that the non-dimensional profiles for the same flow regime overlap indicates that different similarity solutions may exist for different flow regimes. Notice that the non-dimensional liquid velocities of the turbulent flow group, i.e., U_g values of 9.6 and 12 cm/s, are smaller than those of the bubbly flow group, i.e., U_g values of 2.4 and 4.8 cm/s. This indicates that as flow transits from bubbly to churn turbulent, the dependence of liquid velocity on the superficial gas velocity, which is nearly linear in bubbly flow, changes.

Figure 1.3 shows the Reynolds shear stress, $\langle u'_r u'_z \rangle$. Again the difference between the bubbly and churn turbulent regime is clearly evident. Obviously the Reynolds shear stress is enhanced as the superficial gas velocity increases. However, at this time we are not sure about the proper scaling, which remains under investigation.

The effect of column size on liquid velocity is shown in Figure 1.4. It is not surprising that the non-dimensional liquid velocity profiles, $\langle u_z \rangle / U_g$, for 6 and 8 in. diameter columns are almost identical since the difference in column size is small. The results for the 18 in. column indicate that liquid velocity increases significantly as column size increases, which implies that the non-dimensional liquid velocity should be a function of a non-dimensional coefficient that contains both column diameter D and superficial gas velocity U_g . Finally, Figure 1.5 shows the effect of column size on the Reynolds shear stress. The trend is that $\langle u'_r u'_z \rangle$ increases as the column diameter becomes larger.

2. DYNAMIC SIMULATION OF 14-CM COLUMN AND COMPARISON WITH CARPT MEASUREMENT

2.1 Introduction

We have been working on the three-dimensional dynamic simulations of bubble columns for a period of time. One of the objectives is to perform the simulation for several superficial gas velocities for which we have CARPT data for liquid velocity. One important issue in gas-liquid Eulerian/Eulerian simulation is how to model the turbulence of the continuous phase (in this case, the liquid) properly. To study this issue, we present the results for two cases: one in the bubbly flow regime, the other in the churn turbulent regime.

In a turbulent flow, the the velocity field can be expressed as

$$u_i = \langle u_i \rangle + u'_i \quad (2.1)$$

The well-known, one-dimensional model equation for the time-averaged liquid velocity profile is

$$\frac{1}{r} \frac{d}{dr} \left[r (1 - \bar{\varepsilon}) (\tau_t + \tau_m) \right] - \frac{dP}{dz} - \rho_l (1 - \bar{\varepsilon}) g = 0 \quad (2.2)$$

where the turbulence shear stress, τ_t , and the molecular viscous shear stress, τ_m , are respectively defined as

$$\tau_t = -\rho_l \langle u'_r u'_z \rangle \quad (2.3)$$

and

$$\tau_m = \mu \frac{\partial \langle u_z \rangle}{\partial r}; \quad (2.4)$$

From a CARPT measurement, one can directly evaluate the total Reynolds stress, i.e., Equation (2.3). Since the turbulence contains large scales, u_i'' , and small scales, u_i''' , the velocity field, (1.1), can be further decomposed into

$$u_i = \langle u_i \rangle + u_i'' + u_i''' \quad (2.5)$$

In a numerical simulation, it is very difficult to resolve the small turbulence scales, u_i''' . However, the effect of this part can be accounted for by a properly chosen turbulence model. The velocity field generated from such simulation would then be given by

$$u_i = \langle u_i \rangle + u_i'' \quad (2.6)$$

where the average $\langle u_i \rangle$ now incorporates the small fluctuations. Therefore, the Reynolds shear stress evaluated from this velocity field is $-\rho_l \langle u_r'' u_z'' \rangle$. In terms of numerically predicted values, the total Reynolds stress, τ_t , can then be expressed as

$$\tau_t = \mu_t \frac{\partial \langle u_z \rangle}{\partial r} - \rho_l \langle u_r'' u_z'' \rangle + f \quad (2.7)$$

Here μ_t is the modeled eddy viscosity, which accounts for the effect of small-scale turbulence, and f represents the possible interaction between large and small scales due to the non-linear dependence of the Reynolds stress on the velocity fluctuations. (It should be noted that no information on f can be obtained from a simulation using turbulence models.) In our current simulations, a model for bubble-induced turbulence is used to evaluate μ_t :

$$\mu_t \propto \varepsilon w_s d_b \quad (2.8)$$

where ε , w_s and d_b denote the local gas holdup, the slip velocity between the bubble and liquid (again a local value) and the bubble diameter, respectively. It is apparent that this model is like a localized mixing length turbulence model. The turbulence scales of bubble size or smaller would be modeled by Equation (2.8), and any scale larger than this would be solved by simulation, the results of which would then give the second term of the right-hand side of Equation (2.7).

2.2 Results and Discussion

For the 14-cm diameter column, the two cases with superficial gas velocities of 2.4 and 9.6 cm/s, which fall into bubbly flow and churn turbulent flow regimes, respectively, were simulated.

Figures 2.1 and 2.2 show the instantaneous gas volume fraction contours of the 14-cm diameter column in a fully developed state. The typical three-dimensional spiral structures are clearly observed. The purpose of showing these plots is to ensure that, qualitatively, this approach is not unreasonable.

Figure 2.3 shows the variation of the column's dynamic height with time. For both cases, i.e., $U_g=2.4$ and 9.6 cm/s, the columns are pre-filled with gas for numerical stability. The simulations start from $t=0$. The column with $U_g=2.4$ cm/s quickly reaches the steady state, while the one with $U_g=9.6$ cm/s approaches a stable level after about 120 seconds. As a measure of the global gas holdup, the numerically simulated dynamic heights are compared with the experimentally measured ones. Reasonable agreement, as shown by Figure 2.3, is obtained for both cases.

Figure 2.4 shows the instantaneous liquid velocity component in the axial direction at a central point of the column. For both cases, the nature of turbulence, i.e., multi-scale (refer here to the time scales), is observed. Particularly for the 2.4 cm/s case, the large-scale motion, i.e., the slow motion, is discerned. Further analysis requires energy spectrum.

In Figure 2.5 the numerically calculated time-averaged liquid axial velocity profiles are compared with those measured by the CARPT technique. Better comparison is found for the $U_g=9.6$ cm/s case than for the $U_g=2.4$ cm/s. It can be seen that the velocity inversion point for $U_g=2.4$ cm/s for the numerically computed profile is significantly different from the experimentally observed value. On the other hand, the comparison for the gas superficial velocity of 9.6 cm/s is good, except for the near-wall region. However, such discrepancy close to the walls could be due to the inaccuracy of CARPT measurement in the near-wall region. In fact, the studies of CARPT data have shown that the occurrence of the radioactive particle in the near-wall region is much lower than the average occurrence at all other positions of the column. When the particle visits the wall region, it is mainly carried by the strong downward flow of the liquid streams. Therefore, the

averaged values are likely to over-estimate reality. As the superficial gas velocity increases, the situation becomes more severe, as evident by comparison of Figures 2.5(a) and (b). This suggests the use of LDA for better measurements in the near-wall region.

Finally, Figure 2.6 compares the profiles of Reynolds stresses. As discussed earlier, what is really compared here is the $\langle u_r'' u_z'' \rangle$ calculated from the dynamic, numerically simulated liquid velocity field, with the $\langle u_r' u_z' \rangle$ calculated from the CARPT measurement. The former contains only the contribution from the large-scale fluctuations, while the latter represents contributions from all the scales measured by CARPT. In fact, due to the limitation of the CARPT technique, the Reynolds stress calculated based on measurement does not contain the contribution from the smallest turbulent fluctuations, which cannot be captured by the currently used large ($d_p=2.3\text{mm}$), neutrally boyant radioactive tracking particle. However, if the turbulence model used manages to cover a wider range of small-scale fluctuations than those missed by CARPT, then one would expect $\langle u_r' u_z' \rangle$ observed by CARPT to still be larger than $\langle u_r'' u_z'' \rangle$ computed by the model. Therefore, the numerically calculated Reynolds stress profile should be lower than the one from CARPT measurement.

For the case of churn turbulent flow, i.e., $U_g=9.6\text{ cm/s}$, as shown in Figure 2.6(b), the comparison of computed and measured velocity cross correlation is consistent with the above discussion. If the portion that has already been modeled is added, as indicated by the first term on the right-hand side of Equation (2.7) to the $\langle u_r'' u_z'' \rangle$, the summation should be very close to measured $\langle u_r' u_z' \rangle$, provided that the large/small interaction term, f , is small.

However, for bubbly flow, $U_g=2.4\text{ cm/s}$, this is not the case, as shown by Figure 2.6(a). The numerically computed profile for the velocity cross correlation is higher than the experimental one. Actually the intensity of turbulence is very low for this case. In addition to the unsatisfactory comparison of the mean velocity, as shown in Figure 2.5(a), this indicates that the turbulence model, such as given by Equation (2.8), may not be needed for the cases within the bubbly flow regime. In other words, for bubbly flow, a simulation without any turbulence model should be able to resolve all the scales that can be detected by the CARPT regime. This assertion will be tested in future work.

The flow condition and parameters used in the simulation are summarized in Table 2.1.

Table 2.1. Flow Condition Computational Parameters

	Column diameter (cm)	Superficial gas velocity U_g (cm/s)	Static liquid height (cm)	Bubble diameter used in drag law (cm)
Case 1	14	2.4	120	0.5
Case 2	14	9.6	98	1.5

3. Dynamic Simulation of Tracer Dispersion in Bubble Columns

3.1 Introduction

One of the major objectives of our ongoing modeling and dynamic simulation of bubble column flow is to study the liquid/gas dispersion and mixing. The widely used one-dimensional axial dispersion model requires the effective axial dispersion coefficient for which no suitable accurate correlation has been found to date. On the other hand, the two-dimensional convection-diffusion model requires axial and radial eddy diffusivities and the liquid velocity profile. It was demonstrated that such a model, with the velocity profile and eddy diffusivity values provided from the CARPT studies, can predict well the independently measured liquid tracer exit age density function (Degaleesan *et al.*, 1996, 1997). This implies that such a model can accurately predict bubble column reactor performance for the first-order reaction schemes and is a model of choice for mildly nonlinear reactions. Moreover, the parameters of the model need to be supplied, which, if based on experimentation, is a very expensive and tedious proposition. Therefore, we attempt here to obtain the modeled parameters by three-dimensional numerical simulation of the flow field and tracer spreading. In the current report, we verify the capability of the dynamic tracer simulation to evaluate the averaged liquid turbulent diffusivity.

Before a tracer simulation is started, one needs to generate a fully developed turbulent liquid/gas velocity field inside the bubble column. The Eulerian/Eulerian two-fluid model is selected to dynamically simulate the bubble driven flow. Following Drew (1983), below are the governing equations for the motion of gas-liquid flow in bubble columns, consisting of the continuity equation for each phase,

$$-\frac{\partial \varepsilon}{\partial t} + \nabla \cdot [(1 - \varepsilon)\mathbf{u}_l] = 0 \quad (3.1)$$

$$\frac{\partial \varepsilon}{\partial t} + \nabla \cdot (\varepsilon \mathbf{u}_g) = 0 \quad (3.2)$$

as well as the momentum equations for each phase,

$$\rho_l(1-\varepsilon)\left(\frac{\partial \mathbf{u}_l}{\partial t} + \mathbf{u}_l \cdot \nabla \mathbf{u}_l\right) = \rho_l(1-\varepsilon)\mathbf{g} - (1-\varepsilon)\nabla p - (\mathbf{M}_d + \mathbf{M}_{vm}) + \nabla \cdot [(1-\varepsilon)\boldsymbol{\sigma}_l] + \nabla \cdot [(1-\varepsilon)\boldsymbol{\sigma}_l^b] \quad (3.3)$$

$$\rho_g \varepsilon \left(\frac{\partial \mathbf{u}_g}{\partial t} + \mathbf{u}_g \cdot \nabla \mathbf{u}_g \right) = \rho_g \varepsilon \mathbf{g} - \varepsilon \nabla p + (\mathbf{M}_d + \mathbf{M}_{vm}) \quad (3.4)$$

The subscript ‘ l ’ and ‘ g ’ indicate the properties for the liquid phase and the gas phase, respectively, and ε denotes the volume fraction of the gas phase, i.e., local gas holdup. The inter-phase momentum exchange term, $\mathbf{M}_d$, which is caused by drag force acting on bubbles, is given by

$$\mathbf{M}_d = \frac{6\varepsilon(1-\varepsilon)}{\pi d_b^3} \mathbf{F}_d \quad (3.5)$$

in which the drag force, $\mathbf{F}_d$, is expressed as

$$\mathbf{F}_d = \frac{1}{8} \rho_l \pi d_b^2 C_D |\mathbf{u}_l - \mathbf{u}_g| (\mathbf{u}_l - \mathbf{u}_g) \quad (3.6)$$

where d_b represents the bubble diameter. For the drag coefficient, C_D , we use the following expression (Drew 1983):

$$C_D = \max \left[\frac{24}{Re} (1 + 0.15 Re^{0.687}), f \frac{8}{3} \frac{Eo}{Eo + 4} \right] \quad (3.7)$$

in which

$$f = \left\{ \frac{1 + 17.67(1-\varepsilon)^{9/7}}{18.67(1-\varepsilon)^{3/2}} \right\}^2 \quad (3.8)$$

The Eotvos number, Eo , and bubble Reynolds number, Re , are defined as

$$Eo \equiv E g \rho_c d_p^2 / \gamma \quad (3.9)$$

and

$$Re \equiv d_b |\mathbf{u}_l - \mathbf{u}_g| / \nu_l \quad (3.10)$$

respectively. γ and ν_l denote the surface tension and dynamic viscosity of the liquid phase, respectively. Additional inter-phase momentum exchange, $\mathbf{M}_{vm}$, caused by the added-mass force, is given by

$$\mathbf{M}_{vm} = \frac{1}{2} (1-\varepsilon) \varepsilon C_{vm} \left(\frac{D\mathbf{u}_l}{Dt} - \frac{D\mathbf{u}_g}{Dt} \right) \quad (3.11)$$

where the added-mass coefficient, C_{vm} , is taken to be

$$C_{vm} = 1 + 3.32\varepsilon + O(\varepsilon^2) \quad (3.12)$$

In the momentum equation for the liquid phase, Equation (3.3), we adopted a model for the bubble-induced stress, as proposed by Sato *et al.* (1981)

$$\sigma_c^b = \rho_c \nu_b^t (\nabla \mathbf{u}_l + \nabla \mathbf{u}_l^T) \quad (3.13)$$

in which the bubble-induced additional viscosity is calculated by

$$\nu_b^t = k_b \varepsilon d_b |\mathbf{u}_l - \mathbf{u}_g| \quad (3.14)$$

The empirical constant k_b takes a value from 0.2 to 0.6; in this simulation, the value is 0.4.

The Equations (3.1) to (3.14) consist of a complete two-fluid model for describing the turbulent motion in gas/liquid systems. The three-dimensional dynamic simulations for the cylindrical bubble columns operated at various conditions can then be performed by using CFDLIB, in which the two-fluid model above is implemented. Such simulations generate fully developed flow fields, i.e., $\mathbf{u}_l(\mathbf{x}, t)$, $\mathbf{u}_g(\mathbf{x}, t)$ and $\varepsilon(\mathbf{x}, t)$. The tracer simulations can then be started. If we use some type of passive scalar, for example C , as the local tracer concentration for the liquid phase, the convection-diffusion equation for C is given by

$$\frac{\partial(1-\varepsilon)C}{\partial t} + \nabla \cdot [(1-\varepsilon)C\mathbf{u}_l] = D_m^l \nabla^2 C \quad (3.15)$$

in which D_m^l is the molecular diffusivity. Similarly the governing equation for the gas tracer is written as

$$\frac{\partial \varepsilon S}{\partial t} + \nabla \cdot [\varepsilon C \mathbf{u}_g] = D_m^g \nabla^2 S \quad (3.16)$$

where S is chosen to denote the local gas phase tracer concentration. Equations (3.15) and (3.16), which are also numerically implemented in CFDLIB, are then solved, together with the fluid dynamic equations, (3.1) to (3.4), to generate the tracer concentration as a function of time and location in the column, $C(\mathbf{x}, t)$ and $S(\mathbf{x}, t)$, for the liquid and gas phase, respectively. Since in bubble columns the dispersion of passive scalars is dominantly controlled by turbulence, the molecular diffusivities, D_m^l and D_m^g , can be taken as negligible and thus are given zero value during the simulations. It should be emphasized that during the tracer simulation, the fluid dynamic equations are still being solved continuously. Since the tracers are passive scalars, their existence in the column, either in liquid phase or in gas phase, do not change the fluid dynamics at all.

Assuming axisymmetric flow field in a bubble column and neglecting the molecular diffusivity, Equation (3.15) can be converted into the following form in terms of cylindrical coordinates,

$$\frac{\partial(1-\varepsilon)C}{\partial t} + \frac{1}{r} \frac{\partial(1-\varepsilon)ru_r C}{\partial r} + \frac{\partial(1-\varepsilon)u_z C}{\partial z} = \frac{1}{r} \frac{\partial}{\partial r} \left[D_{rr} (1-\varepsilon) r \frac{\partial C}{\partial r} \right] + \frac{\partial}{\partial z} \left[D_{zz} (1-\varepsilon) \frac{\partial C}{\partial z} \right] \quad (3.17)$$

where the coefficients D_{zz} and D_{rr} represent the axial and radial turbulent eddy diffusivities, respectively. They are related to the turbulent transport by the following definitions:

$$\langle u'_z C' \rangle \equiv -D_{zz} \frac{\partial C}{\partial z} \quad (3.18)$$

$$\langle u'_r C' \rangle \equiv -D_{rr} \frac{\partial C}{\partial r} \quad (3.19)$$

where $\langle \rangle$ represents the ensemble averaging. Equation (3.17) represents the averaged balance equation for the non-volatile (passive) species, and is a transient, two-dimensional convection-diffusion equation. The phasic averaging that has been performed to arrive at this equation (see Degaleesan 1997) refers to any time interval, which may be small or large.

Performing a volume integration of the above equation over a section of the column, as illustrated in Figure 3.1, one gets

$$\iint \frac{\partial \varepsilon C}{\partial t} ds dz + \iint \frac{\partial}{\partial z} (\varepsilon u_z C) ds dz = \iint \frac{\partial}{\partial z} \left(D_{zz} \varepsilon \frac{\partial C}{\partial z} \right) ds dz \quad (3.20)$$

in which $\int ds$ represents the area integration over the column's cross-section. The term involving the radial turbulent eddy diffusivity, D_{rr} , vanishes since the boundary condition of zero flux in radial direction at $r = R$ was imposed. The axial turbulent eddy diffusivity, D_{zz} , may be a function of r . However, the volume-averaged axial turbulent eddy diffusivity can be defined as

$$\bar{D}_{zz} = \frac{\iint \frac{\partial \varepsilon C}{\partial t} ds dz + \iint \frac{\partial}{\partial z} (\varepsilon u_z C) ds dz}{\iint \frac{\partial}{\partial z} \left(\varepsilon \frac{\partial C}{\partial z} \right) ds dz} \quad (3.21)$$

If the integration along the axial direction, z , extends from z_1 to z_2 , as indicated in Figure 1, Equation (3.21) can be further written as

$$\bar{D}_{zz} = \frac{\iint \frac{\partial \varepsilon C}{\partial t} ds dz + \int [(\varepsilon u_z C)_{z_2} - (\varepsilon u_z C)_{z_1}] ds}{\int \left[\left(\varepsilon \frac{\partial C}{\partial z} \right)_{z_2} - \left(\varepsilon \frac{\partial C}{\partial z} \right)_{z_1} \right] ds} \quad (3.22)$$

To calculate the right-hand side of Equation (3.22), the liquid velocity field, $\mathbf{u}_l(\mathbf{x}, t)$, the gas holdup distribution, $\varepsilon(\mathbf{x}, t)$, and the tracer concentration distribution, $C(\mathbf{x}, t)$ are needed. In fact, all of these quantities are obtained from a dynamic tracer simulation, in which Equations (3.1) to (3.4) and (3.15) are numerically solved. In such a way, the volume-averaged turbulent eddy diffusivity of the liquid phase in a cylindrical bubble column can be evaluated numerically. Notice that $\bar{D}_{zz}$ in Equation (3.22) is still a function of time. In fact $\bar{D}_{zz}$ is calculated from the liquid velocity, local gas holdup and local tracer concentration at time t , as one can see from Equation (3.22). As with an ensemble averaging, $\bar{D}_{zz}$, which is now a volume-averaged quantity, should be further averaged over time to obtain the averaged turbulent eddy diffusivity. In the dynamic simulations presented here, the CFDLIB code outputs the time-dependent results every Δt second. Usually the tracer simulations for a 14-cm diameter column operated at 2.4 cm/s superficial gas velocity last about 20 to 30 seconds. For this particular case, the results show that the liquid tracer concentration reaches a nearly uniform distribution at about 15 seconds after the tracer-injection. The time interval for output, Δt , is set to 0.1 second. The averaged turbulent eddy diffusivity, $\langle D_{zz} \rangle$, is therefore calculated as,

$$\langle D_{zz} \rangle = \frac{1}{N} \sum_{n=1}^N \bar{D}_{zz}(t_0 + n\Delta t) \quad (3.23)$$

where t_0 denotes the time when the tracer is injected into the column and $N\Delta t$ represents the total time of the tracer simulation. Both Δt and N are adjustable according to the flow conditions and column geometry.

In this report, we present a preliminary result on the tracer simulation and the calculation of axial turbulent eddy diffusivity, as outlined above, for the liquid phase in a three-dimensional bubble column. The bubble column being simulated is 14 cm in diameter and operated at a gas superficial velocity of 2.4 cm/s.

3.2 Results and Discussion

To ensure an accurate evaluation of the dispersion coefficient, the numerical tracer simulation can only be started when the flow field of the column reaches a statistically quasi-steady state. Since the tracer dispersion in the axial direction is largely controlled by the mean axial momentum and its fluctuation, we calculated the time- and azimuthal-

averaged profiles of the liquid axial velocity, $\langle u_z \rangle$, and the axial turbulence intensity, $\langle u'_z u'_z \rangle$. For the 14-cm diameter column operated at $U_g=2.4$ cm/s, two experiments were conducted. One was for the column with perforated plate type **6A** as the gas distributor, and the other was for the column with distributor **6B**. Figure 3.2 depicts the gas distributors. In Figure 3.3, comparisons are made between the computed numerical values and the corresponding experimental data. As this figure shows, using different gas distributors does not induce large differences in the mean axial velocity, and the computed profile is satisfactorily close to the experimental ones. However, the effect of gas distributors on turbulence intensities is significant. As shown in Figure 3.3, the axial liquid turbulence intensity for the column with the **6B** distributor is about three times larger than that for the column with distributor **6A**. The numerical profile lies in between. Notice that in a numerical simulation, the type of gas distributor was not exactly matched due to the limited mesh resolution. As shown in Figure 3.2, the gas is introduced through the numerical cells in the shaded area only. With the current mesh system, this matches most closely the real gas distributors **6A** and **6B**.

Figure 3.4 shows the time evolution of liquid tracer concentration in the 14-cm diameter column operated at a superficial gas velocity of 2.4 cm/s. An impulse of tracer is injected at the center of the column ($r=0$) and close to the bottom ($z=3$ cm). The injection is made when the flow field reaches the fully developed state. Due to liquid recirculation, i.e., batch flow, the liquid tracer cannot escape the column. Therefore, the tracer concentration eventually approaches a uniform pattern. On the other hand, as shown in Figure 3.5, the gas tracer eventually exits the column. One can also observe the downward dispersion of the gas tracer due to the carrying of gas bubbles by liquid back flow near the wall region.

Figure 3.6 displays the liquid tracer concentrations observed at different axial locations as functions of time. C_0 is proportional to the cross-sectional average concentration and is defined as

$$C_0(z) \equiv \int C ds \quad (3.24)$$

where the area integral, $\int () ds$, is taken over the cross section at a axial location, z . It can be seen that at the time of 10 seconds after the tracer injection, there is still a spatial non-uniformity of tracer distribution. The complete simulation indicates that after about 15 seconds the tracer concentration field reaches uniform distribution along the z direction. From this simulation the liquid velocity, liquid holdup, tracer concentration and the spatial gradient of the tracer concentration are sampled every 0.1 second. The averaged axial turbulent eddy diffusivity is calculated by using Equations (3.22) and (3.23). This result is listed in Table 3.1, in which the experimentally measured values of eddy diffusivity obtained by CARPT for distributors **6A** and **6B** are also listed. The effect of the gas distributor on diffusivity is quite significant. However, the calculated turbulent eddy diffusivity is within the same range as the experimental data. It should be

pointed out that the calculated turbulent diffusivity should be independent of the location of tracer injection. A reliable value can only be obtained by performing a series of tracer simulations with different locations of the initial injection.

Table 3.1 Flow Condition Computational Parameters

D (cm)	U_g (cm/s)	H_s (cm)	d_b (cm)	Computed $\langle D_{zz} \rangle$ (cm ² /sec)	$\langle D_{zz} \rangle$ 6A (cm ² /sec)	$\langle D_{zz} \rangle$ 6B (cm ² /sec)
14	2.4	120	0.5	72	48	75

Reference

Degaleesan, S., S. Roy, S.B. Kumar, and M.P. Dudukovic, "Liquid mixing based on convection and turbulent dispersion in bubble columns," *Chem. Eng. Sci.* 51, 1976 (1996).

Degaleesan, S., "Fluid dynamic measurements and modeling of liquid mixing in bubble columns," D. Sci. Thesis, Washington University in St. Louis, (1997).

Degaleesan, S., M.P. Dudukovic, B.A. Toseland, and B.L. Bhatt, "A two compartment convective-diffusive model for slurry bubble column reactor," *Ind & Eng. Chem. Res.*, 36 (11), 4670 (1997).

Drew, D.A. "Mathematical modeling of two-phase flow," *Ann. Rev. Fluid Mech.*, 15, 261 (1983).

Sato, Y., M. Sadatomi and K. Sekoguchi, "Momentum and heat transfer in two-phase bubble flow I," *Int. J. Multiphase Flow*, 7, 167 (1981).

Figure 1.1 Mean Axial Liquid Velocity of 14-cm (D=6 in.) Column Operated at Different Superficial Gas Velocities, U_g .

Figure 1.2 Non-Dimensional Mean Axial Liquid Velocity for 14-cm (D=6 in.) Column Operated at Different Superficial Gas Velocities, U_g . (The velocities are scaled by their superficial gas velocities.).

Figure 1.3 The Radial Distributions of Reynolds Shear Stress, $\langle u'_r u'_z \rangle$, in 14-cm (D=6 in.) Column Operated at Different Superficial Gas Velocities, U_g

Figure 1.4 Non-Dimensional Mean Axial Liquid Velocity for Different-Sized Columns Operated at Approximately the Same Superficial Gas Velocities, U_g

Figure 1.5 The Radial Distributions of Reynolds Shear Stress, $\langle u'_r u'_z \rangle$, in Different-Sized Columns Operated at Approximately the Same Superficial Gas Velocity, U_g

Figure 2.1 Instantaneous Gas Holdup Contours on a Plane Cutting Through the Center of the Column. Column Diameter, $D=14$ cm.

Figure 2.2 Instantaneous Gas Holdup Contours on Cross-Sectional Planes (3D view) Column. (Column diameter (D)=14 cm)

Figure 2.3 Variation of Column's Dynamic Height with Time. (Column diameter (D)=14 cm)

Figure 2.4 The Instantaneous Liquid Axial Velocity at the Center Point of the Column. (Column diameter (D)=14 cm)

(a) $U_g = 2.4$ cm/s

(b) $U_g = 9.6$ cm/s

Figure 2.5 Time-Averaged Liquid Axial Velocity Profiles - Numerical and Experimental. ($D=14$ cm)

Figure 2.6 Reynolds Stress, $\langle u'_r u'_z \rangle$, Profiles - Numerical and Experimental. (D=14 cm)

Figure 3.1 Computational Mesh System for Fluid Dynamic and Tracer Simulation in Bubble Columns

Figure 3.2 The Perforated Plate Gas Distributors used on the 14-cm Diameter Column: 6A (holes of 0.4 mm; 3 concentric circles; 0.05% porosity) and 6B (holes of 1 mm; 6 concentric circles; 0.62% porosity). (Computational mesh at column's bottom: the gas is injected through the shadowed area only.)

Figure 3.3 Time-Averaged Liquid Axial Velocity, $\langle u_z \rangle$, and Turbulence Intensity, $\langle u'_z u'_z \rangle$, for 14-cm Column Operated at Superficial Gas Velocity of $U_g = 2.4$ cm/s. (The experimental data, from CARPT measurement, are for two different perforated plate gas distributors: 6A and 6B.)

Figure 3.4 Time Evolution of the Liquid Tracer Concentration Inside a 14-cm Diameter Column at $U_g=2.4$ cm/s. [The tracer is released at the center of the column, near the bottom, at time $t=0$. There is no net flow of liquid (batch liquid).]

Figure 3.5 Time Evolution of the Gas Tracer Concentration Inside a 14-cm Diameter Column at $U_g=2.4$ cm/s. (The tracer is released at the center of the column, near the bottom, at time $t=0$.)

Figure 3.6 Numerical Detector Responses for Liquid Tracer Injection at $r=0$, $z/D=0.2$, in a 14-cm Column Operated at $U_g=2.4$ cm/s

THE OHIO STATE UNIVERSITY

The report from Ohio State University for the period follows.

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

Quarter Report

(Reporting Period: October 1 to December 31, 1999)

Simulation of Single Bubble Formation in Liquid-Solid Suspensions at High Pressures with Pressure Fluctuations in the Gas Chamber

Highlights

- The behavior of bubble formation from a single orifice connected to a gas chamber in a non-aqueous liquid and a liquid-solid suspension at high pressures (up to 8.3 MPa) was investigated. An optic fiber probe was used to measure the initial bubble size in liquids and liquid-solid suspensions.
- A two-stage spherical bubble formation model was extended to describe the bubble formation process in liquid-solid suspensions with pressure fluctuations in the gas chamber. The effects of particle and pressure on bubble motion and pressure balance at the gas-liquid interface were taken into consideration in the model through various forces.
- The model can reasonably predict the initial bubble sizes obtained in this study, as well those reported in the literature under high-pressure conditions. The predicted trends of pressure and particle effects on initial bubble size under various bubble formation conditions also agreed well with the experimental observations.
- The model simulation showed that pressure in the gas chamber and gas flow rate through the orifice change significantly with time during bubble formation. Even for bubble formation under constant flow conditions, there still exist fluctuations in pressure in the gas chamber and in gas flow rate through the orifice at the beginning of bubble formation.

Work Conducted

Model Equations

Bubble Motion Equation

The motion equation of a rising bubble in liquid-solid suspensions can be described based on a balance of all the forces acting on the bubble (Luo et al., 1998). The forces induced by the liquid include the upward forces [effective buoyancy (F_B) and gas momentum (F_M)], and the downward resistance [liquid drag (F_D), surface tension force (F_σ), bubble inertial force ($F_{I,g}$), and Basset force (F_{Basset})]. Two other downward forces on the bubble account for the particle effect on bubble motion, i.e., the particle-bubble collision force (F_C) and the suspension inertial force ($F_{I,m}$), due to acceleration of the liquid and particles surrounding the bubble (Luo et al., 1998). Therefore, the overall force balance on the bubble formed in liquid-solid suspensions can be written as

$$F_B + F_M = F_D + F_\sigma + F_{Basset} + F_{I,g} + F_C + F_{I,m}. \quad (1)$$

The expansion and detachment stages follow the same force balance equation [Eq. (1)], although the expressions for the same force in two stages may be different. The expressions for all the forces under two stages are given in Table 1. The particle-bubble collision force is merely the rate of momentum change of particles colliding with the bubble surface. The suspension inertial force can be derived from the suspension flow field around an accelerating bubble. Luo et al. (1998) quantified the flow field of the suspension around a two-dimensional bubble by using the Particle Image Velocimetry (PIV) technique and obtained the expression for the suspension inertial force, $F_{I,m}$:

$$F_{I,m} = \frac{d\left(\iiint \rho_m u_m \delta V\right)}{dt} = \zeta \frac{d}{dt} \left[\rho_m \left(\frac{4}{3} \pi r_b^3 \right) u_b \right] \quad (2)$$

where the apparent density of the suspension is defined as:

$$\rho_m = \varepsilon_s \rho_s + \varepsilon_l \rho_l. \quad (3)$$

For bubbles formed in liquid-solid suspensions, the coefficient ζ is equal to 3.86 (Luo et al., 1998). When bubbles are formed in liquids, the coefficient ζ is equal to $\frac{11}{16}$, corresponding to the added mass in inviscid liquids (Milne-Thomson, 1955). The detailed descriptions of these forces are given in Luo et al. (1998).

In the expansion stage, the rise velocity of the bubble, u_b , is equal to the bubble expansion velocity, i.e.,

$$u_b = u_e = \frac{dr_b}{dt} \quad (4)$$

and the gas flow rate through the orifice, Q_0 , can be expressed by the following equation:

$$Q_0 = \frac{dV_b}{dt} = 4\pi r_b^2 \frac{dr_b}{dt} \cdot \quad (5)$$

Substituting the expressions of various forces in Table 1 into Eq. (1) and considering Eqs. (4) and (5), the force balance at the end of the expansion stage can be written as:

$$\begin{aligned} \frac{d}{dt} \left[(\rho_g + \zeta \rho_m) \left(\frac{4}{3} \pi r_b^3 \right) \frac{dr_b}{dt} \right] = & \frac{4\pi r_b^3}{3} (\rho_l - \rho_g) g + \frac{\rho_g Q_0^2}{\frac{1}{4} \pi D_0^2} - 6\pi \mu_l r_b \frac{dr_b}{dt} \\ & - \pi D_0 \sigma \cos \gamma - \frac{1}{4} \pi D_0^2 \rho_s \varepsilon_s \left(\frac{dr_b}{dt} \right)^2 \end{aligned} \quad (6)$$

The term on the left-hand side represents the inertial forces of the bubble and suspension. The five terms on the right-hand side represent the buoyancy, gas momentum, viscosity, surface tension and particle-bubble collision forces, respectively.

In the detachment stage, the rise velocity of the bubble is the sum of the expansion velocity (u_e) and the bubble base rising velocity (u):

$$u_b = u + u_e = \frac{dx}{dt} \quad (7)$$

where x is the vertical distance between the bubble center and orifice plate. The gas flow rate through the orifice can be expressed by:

$$Q_0 = \frac{d(V_b + V_{neck})}{dt} = \frac{d \left[\frac{4}{3} \pi r_b^3 + \frac{1}{4} \pi D_0^2 (x - r_b) \right]}{dt} = 4\pi r_b^2 \frac{dr_b}{dt} + \frac{1}{4} \pi D_0^2 \left(\frac{dx}{dt} - \frac{dr_b}{dt} \right). \quad (8)$$

Then, the motion equation of a bubble in the detachment stage can be written as follows:

$$\begin{aligned} \frac{d}{dt} \left[(\rho_g + \zeta \rho_m) \left(\frac{4}{3} \pi r_b^3 \right) \frac{dx}{dt} \right] = & \frac{4\pi r_b^3}{3} (\rho_l - \rho_g) g + \frac{\rho_g Q_0^2}{\frac{1}{4} \pi D_0^2} - 6\pi \mu_l r_b \frac{dx}{dt} - \pi D_0 \sigma \cos \gamma \\ & - \pi r_b^2 \rho_s \varepsilon_s \left(\frac{d(x - r_b)}{dt} \right)^2 - 12 r_b^2 \sqrt{\pi \rho_l \mu_l t} \frac{d^2(x - r_b)}{dt^2} \end{aligned} \quad (9)$$

The terms on the right-hand side represent the buoyancy, gas momentum, viscosity, surface tension, particle-bubble collision and Basset forces, respectively.

When bubbles are formed under constant flow conditions, only the motion equation of bubble is needed to simulate the bubble formation process. However, when bubbles are formed under variable flow conditions, i.e., a large gas chamber underneath the orifice, the gas flow rate through the orifice varies and depends on the pressure difference between the gas chamber and bubble. In order to simulate bubble formation under such conditions, the additional model equations, such as orifice equation, pressure balance equation at bubble-liquid interface and thermodynamic equation in the gas chamber, are required to account for the pressures inside the bubble and gas chamber. These equations are summarized next.

Orifice Equation

The instantaneous gas flow rate through the orifice depends on the pressure difference between the gas chamber and bubble, and the orifice resistance. The following orifice equation is applicable (Kupferberg and Jameson, 1969):

$$|P_c - P_b| = \left(\frac{Q_0}{k_0} \right)^2. \quad (10)$$

Q_0 is related to the rate of bubble volume change and can be calculated by using Eqs. (5) through (8) for the expansion stage and detachment stage, respectively. The orifice constant, k_0 , is a function of gas flow rate Q_0 , gas density ρ_g , gas viscosity μ_g , orifice diameter D_0 , and orifice plate thickness L . Considering the pressure drop due to the sudden enlargement and contraction at the orifice and the frictional head loss, the following equation can be used to calculate k_0 (McAllister et al., 1958):

$$k_0 = \sqrt{\frac{2}{\rho_g C_g}} \left(\frac{\pi D_0^2}{4} \right) \quad (11)$$

where

$$C_g = 1.5 + 4f \frac{L}{D_0}. \quad (12)$$

Assuming that the flow of gas through the orifice is laminar, the friction factor, f , can be expressed by

$$f = \frac{16}{Re_0} = \frac{4\pi\mu_g D_0}{\rho_g Q_0} \quad (13)$$

and substituting Eq. (13) into Eq. (12), the constant C_g can be estimated by the following equation:

$$C_g = 1.5 + \frac{16\pi\mu_g L}{\rho_g Q_0}. \quad (14)$$

Therefore, the pressure difference between the bubble and gas chamber can be calculated based on Eqs. (10), (11) and (14), if the gas flow rate through the orifice, properties of the gas phase and orifice geometry are known.

Pressure Balance at Bubble-Liquid Interface

The pressure inside the bubble can be obtained by considering the pressure balance at the bubble-liquid interface. The change of pressure along the bubble-liquid interface is mainly due to the inertia, surface tension and viscosity of the liquid, and to the gas momentum, which can be expressed by the modified Rayleigh equation (Pinczewski, 1981):

$$P_b - P_0 = \Delta P_I + \Delta P_\mu + \Delta P_\sigma - \Delta P_m. \quad (15)$$

The average hydrostatic pressure at the bubble surface, P_0 , is approximately expressed by the hydrostatic pressure at the orifice plate as

$$P_0 = P_s + \rho_l gh \quad (16)$$

where P_s is the system pressure and h is the liquid level in the column. The four terms on the right-hand side of Eq. (15) represent the contributions of liquid inertial, viscosity, surface tension, and gas momentum forces, respectively.

For inviscid liquids, the inertial term in Eq. (15) has been expressed in the literature based on the velocity potential around an expanding, rising bubble by Kotake, 1966; Witze et al., 1968; Kupferberg and Jameson, 1969; McCann and Prince, 1969; and Haynes and Gotham, 1982:

$$\Delta P_I = \rho_l \left(r_b \frac{d^2 r_b}{dt^2} + \frac{3}{2} \left(\frac{dr_b}{dt} \right)^2 - gx - \frac{1}{4} \left(\frac{dx}{dt} \right)^2 \right). \quad (17)$$

However, Eq. (17) is valid only for ideal liquids. For highly viscous liquids or liquid-solid suspensions, the flow field around the bubble is different from that for inviscid liquids, and hence the difference in the inertial term. Marmur and Rubin (1976) used the "added mass" concept and the velocity of the interface to define the inertia of the interface. They found that the "added mass" coefficient for the bubble formation process is higher than $\frac{11}{16}$, the theoretical value for a sphere moving in the vicinity of a wall. The reason is that in the bubble formation process, an expansion motion is superimposed on

the translatory motion of the bubble, and the flow field around the expanding and rising bubble is different from that for the theoretical value. In their study, they found that the value of the "added mass" coefficient is 0.85, which agrees well with their experimental results. Luo et al. (1998) used the PIV technique to measure the flow field of liquid-solid suspensions around an accelerating bubble. They found that the flow field in liquid-solid suspensions is quite different from that in liquids, resulting in an increase in inertia. Therefore, in order to use Eq. (15) to simulate the bubble formation in liquid-solid suspensions, the effect of particles on the inertial term must be considered. The pressure change due to the suspension inertia can be expressed by

$$\Delta P_I = \frac{F_{I,m}}{A_b} \quad (18)$$

where A_b is the surface area of the bubble and $F_{I,m}$ is the suspension inertial force expressed by Eq. (2). Substituting Eq. (2) into Eq. (18) and considering the different expressions of the bubble velocity in the two stages, the inertial term in Eq. (15) can be modified to

$$\Delta P_I = \zeta \rho_m \left[\frac{r_b}{3} \frac{d^2 r_b}{dt^2} + \left(\frac{dr_b}{dt} \right)^2 \right] \quad (\text{expansion stage}) \quad (19a)$$

$$\Delta P_I = \zeta \rho_m \left[\frac{r_b}{3} \frac{d^2 x}{dt^2} + \frac{dr_b}{dt} \frac{dx}{dt} \right] \quad (\text{detachment stage}). \quad (19b)$$

For a real liquid, it is difficult to obtain theoretically a precise expression of the pressure change across the bubble-liquid interface. The common approach used is to include the terms accounting for the effects of viscosity, surface tension and gas momentum on pressure change in the Rayleigh equation, as shown in Eq. (15). Poritsky (1952) and Miyahara and Takahashi (1984) considered the effect of viscosity from the stress conditions on the bubble surface and derived the expression for the viscous term in the modified Rayleigh equation:

$$\Delta P_\mu = \frac{4\mu_l}{r_b} \frac{dr_b}{dt}. \quad (20)$$

The change in pressure along the bubble-liquid interface due to the surface tension is given by (Levich, 1962):

$$\Delta P_\sigma = \frac{2\sigma}{r_b}. \quad (21)$$

Pinczewski (1981) derived an expression for pressure distribution at the interface due to the gas momentum by assuming that the gas inside the growing bubble follows a circulatory, toroidal motion:

$$\Delta P_{m,\theta} = \frac{l}{2} \rho_g u_\theta^2 \cos\theta \quad (22)$$

where θ is the angle between the radial and vertical directions at any position of the bubble-liquid interface. Integrating Eq. (22) along the bubble surface, the average pressure change due to the gas momentum can be obtained:

$$\Delta P_m = \frac{\int_0^{2\pi} \Delta P_{m,\theta} d\theta}{2\pi} = \frac{l}{4} \rho_g u_\theta^2. \quad (23)$$

Substituting Eqs. (19) through (21) and (23) into Eq. (15), the modified Rayleigh equation can be extended for bubble formation in liquid-solid suspensions.

For the expansion stage:

$$P_b - P_0 = \zeta \rho_m \left[\frac{r_b}{3} \frac{d^2 r_b}{dt^2} + \left(\frac{dr_b}{dt} \right)^2 \right] + \frac{2\sigma}{r_b} + \frac{4\mu_l}{r_b} \frac{dr_b}{dt} - \frac{l}{4} \rho_g \left(\frac{Q_0}{\frac{l}{4} \pi D_0^2} \right)^2. \quad (24a)$$

For the detachment stage:

$$P_b - P_0 = \zeta \rho_m \left[\frac{r_b}{3} \frac{d^2 x}{dt^2} + \frac{dr_b}{dt} \frac{dx}{dt} \right] + \frac{2\sigma}{r_b} + \frac{4\mu_l}{r_b} \frac{dr_b}{dt} - \frac{l}{4} \rho_g \left(\frac{Q_0}{\frac{l}{4} \pi D_0^2} \right)^2. \quad (24b)$$

The instantaneous gas flow rate through the orifice, Q_0 , can be calculated by using Eqs. (5) and (8) for the expansion and detachment stages, respectively. Equation (24) expresses the change of pressure inside the bubble during the bubble formation process in liquid-solid suspensions.

Pressure Change in the Gas Chamber

The change of pressure in the gas chamber, P_c , during the expansion and detachment stages is obtained by assuming an adiabatic or isothermal expansion process of ideal gas and applying the first law of thermodynamics to the gas in the chamber (Wilkinson and van Dierendonck, 1994):

$$\frac{dP_c}{dt} = \frac{\gamma}{V_c} (P_e Q_g - P_c Q_0) \quad (25)$$

where P_e is the pressure at the gas inlet to the chamber. For the adiabatic change, γ is the specific heat ratio of the gas ($\gamma = \frac{C_p}{C_v}$); for the isothermal change, γ is equal to 1.

The radius of the bubble at the end of the expansion stage can be obtained by solving Eqs. (10), (24a) and (25) simultaneously under the following initial conditions:

$$t = 0, r_b = \frac{1}{2} D_0, \frac{dr_b}{dt} = 0, P_c = P_s + \rho_m gh + \frac{4\sigma}{D_0}. \quad (26)$$

It is assumed that the bubble is initially a hemisphere having a radius equivalent to the orifice radius. The termination of the expansion stage occurs when various forces are in balance, as given by Eq. (6). The governing equations for the detachment stage, Eqs. (9), (10), (24b) and (25), can be solved by using the final values of the expansion stage as their initial conditions:

$$t = t_e, r_b = r_e, \left. \frac{dr_b}{dt} \right|_{t=t_e} = \left. \frac{dr_b}{dt} \right|_{t=t_e}, x = r_e, \left. \frac{dx}{dt} \right|_{t=t_e} = \left. \frac{dr_b}{dt} \right|_{t=t_e}, P_c = P_{c,e} \quad (27)$$

where subscript e represents the end of the expansion stage. When the neck length reaches r_e , i.e., $x - r_b = r_e$, the bubble detaches from the orifice and the calculation is terminated (Ramakrishnan et al., 1969). The bubble volume at this instant gives rise to the final size of the bubble. The governing equations for both stages are coupled, ordinary differential equations and can be solved simultaneously by using the 4th-order Runge-Kutta method.

Simulation Results

Pressure Effect

The effect of pressure on the initial bubble size predicted by the model is shown in Figure 1; the solid lines represent the model predictions. It was found that the proposed model can reasonably predict the experimental results obtained in this study. The comparison between the model predictions and the experimental data in this study is provided in Figure 2, which shows that the deviation of the predictions is within $\pm 20\%$. The pressure effect on the initial bubble size is strongly affected by the bubble formation conditions. Under variable flow conditions ($N_c > 1$), an increase in pressure significantly reduces the initial bubble size in both the liquid and the liquid-solid suspension. For bubble formation under constant flow conditions ($N_c \leq 1$), the pressure effect is not significant.

Generally, the initial bubble size is determined by the total bubble growth time and bubble growth rate, i.e., the instantaneous gas flow rate entering the bubble Q_0 . In order to analyze the pressure effect on initial bubble size under various formation conditions, it

is necessary to examine pressure effects on the bubble formation time and the gas flow rate entering the bubble. The bubble formation time and instantaneous gas flow rate through the orifice can be estimated by the proposed model. The effect of pressure on bubble formation time in a slurry ($\varepsilon_s=0.18$) is shown in Figure 3. Under variable flow conditions, the bubble formation time is almost independent of the gas velocity and decreases significantly with increasing system pressure; while under constant flow conditions, the bubble formation time only slightly decreases with increasing gas velocity and system pressure. Figure 4 shows the change of gas flow rate through the orifice with time during the bubble formation process. It is seen that at ambient or low pressures, the gas flow rate through the orifice, Q_o , varies significantly with time. At the beginning of bubble growth, Q_o increases rapidly to a maximum, which could be several times higher than the gas flow rate entering the chamber, Q_g . Then, Q_o decreases gradually and approaches a constant value. At high pressures, Q_o increases first to a level slightly higher than Q_g ; it then decreases and quickly reaches a constant flow rate, indicating that the bubble formation is under constant flow conditions. Commonly, when N_c is smaller than 1, the bubble formation is considered to be under constant flow conditions; however, Figure 4 shows that even under such conditions, the fluctuation in gas flow rate still exists at the beginning of bubble growth, due to the existence of a large gas chamber underneath the orifice. Figure 4 also shows that the curve of Q_o versus time shifts to the left when the pressure increases. This indicates that the bubble starts growing earlier under high pressures. Based on the above analysis, for bubble formation under variable flow conditions, reductions in both bubble formation time and gas flow rate through the orifice with increasing pressure result in a significant decrease in initial bubble size.

The mechanism of pressure effect on bubble formation under variable flow conditions can be explained based on the proposed model. The model shows that pressure can influence the bubble formation process in the following ways: (1) the pressure directly influences the pressure fluctuation in the gas chamber based on the thermodynamic equation; (2) the pressure affects the gas momentum terms in the bubble motion equation and modified Rayleigh's equation through the change of gas density; (3) based on the orifice equation, the pressure can also influence the orifice resistance through the change in gas density, and hence the gas flow rate through the orifice. The model calculation shows that the pressure effect on the orifice constant mainly changes the gas flow rate entering the bubble. With increasing system pressure, the orifice constant becomes smaller due to the increase in gas density, and hence a decrease in the gas flow rate through the orifice. The bubble formation time is determined by the force balance on the bubble and the pressure change in the gas chamber. The increase in pressure results in a higher gas momentum rate and larger pressure fluctuation in the gas chamber, which promote the detachment of the bubble and hence decrease the bubble formation time. The combination of the above effects results in a decrease in bubble formation time and Q_o with increasing pressure, and hence a decrease in initial bubble size.

Under constant flow conditions, the constant gas flow rate through the orifice and the weak dependence of bubble formation time on pressure contribute to an insignificant effect of pressure on initial bubble size (Figure 3). When the bubble is formed under constant flow conditions, the bubble formation process would only be influenced by

pressure through the gas momentum, as illustrated by the bubble motion equation. At low gas velocities, the influence of gas momentum would be negligible, provided the pressure or gas density is low, resulting in an insignificant change in bubble formation time with pressure, as shown in Figure 3. When the gas velocity is high, the influence of gas momentum becomes large. The bubble tends to detach from the orifice earlier due to an increased gas momentum with the increase in gas density, which results in a reduction in bubble formation time with increasing pressure and gas velocity.

Particle Effect

Figure 5 shows the particle effect on the initial bubble size under various bubble formation conditions. Under both constant flow and variable flow conditions, the bubbles formed in the liquid-solid suspension are larger than those formed in the liquid at a given gas velocity for the present system. For bubble formation in the liquid-solid suspension, a further increase in solids concentration only slightly increases bubble size. The proposed model can also predict the trend of the particle effect. Based on the model calculations, the presence of particles in the liquid mainly influences the bubble growth time due to various forces induced by the particles on the bubble surface, such as the suspension inertial force. The particles yield resistant forces for the detachment of bubbles, resulting in longer bubble formation time and hence larger bubble size.

Prediction of Literature Data

To examine the applicability of the proposed model, the model was used to predict the available experimental data in the literature on bubble formation under high-pressure or high gas density conditions. Figure 6 compares the model predictions and the experimental data in the literature, and shows that the proposed model can reasonably predict these data. The average error of the predictions is 18%. Relevant information from various references used in Figure 6 is summarized in Table 2.

Bubble Growth and Pressure Fluctuation in the Gas Chamber

Variations in bubble volume and pressure in the gas chamber with time simulated by the model are plotted in Figure 7. The effect of pressure on the bubble growth curve is shown in Figure 7(a). In the pressure range of 0.1 to 2.5 MPa, bubble formation is under variable flow conditions, and the bubble growth rate varies during the formation process. Under such conditions, with an increase in pressure, the bubble growth rate decreases and the bubble growth time becomes shorter. When the pressure is higher than 2.5 MPa, the bubble growth curves are nearly straight, which indicates that bubble formation is under constant flow conditions. Under such conditions, an increase in pressure does not change the bubble growth rate significantly; however, it reduces the bubble growth time.

A typical variation in pressure in the gas chamber with bubble formation time is shown in Figure 7(b). At the beginning of bubble formation, the pressure in the gas chamber increases first due to the inflow of gas to the chamber and the inertia of liquids. Because of this increase in pressure, the bubble begins to form. When the bubble growth rate is higher than the gas rate supplied to the chamber, the amount of gas and hence the pressure in the chamber begins to decrease. As shown in Figure 7(b), pressure has a significant effect on pressure fluctuation in the gas chamber. At ambient pressure, the

pressure fluctuation in the chamber is relatively small. With an increase in pressure, the pressure fluctuation in the chamber becomes significant. At high pressures, it is also seen that the pressure in the gas chamber becomes constant after a certain time. For example, at a pressure of 2.5 MPa, after 20 ms the pressure in the gas chamber becomes constant, as shown in Figure 7(b). This can be illustrated by considering the change of Q_0 with the growth time. As shown in Figure 4, when the pressure is 2.5 MPa, after 20 ms, the gas flow rate to the bubble is constant and approximately equal to the gas flow rate entering the gas chamber. This results in a constant amount of gas in the chamber, and hence a constant pressure. As shown in Figures 4 and 7, it is clear that even when bubble formation is under constant flow conditions, there still exist fluctuations in pressure in the gas chamber and in the gas flow rate through the orifice at the beginning of bubble formation.

Notations

A_b	surface area of bubble, m^2
C_D	drag coefficient, dimensionless
C_g	constant in Eq. (11), dimensionless
C_p	specific heat capacity of gas at constant pressure, $J/(kg \cdot K)$
C_v	specific heat capacity of gas at constant volume, $J/(kg \cdot K)$
D_0	orifice diameter, m
d_b	initial bubble size, m
e	restitution coefficient, dimensionless
f	friction factor, dimensionless
F_σ	surface tension force, N
F_B	buoyancy force, N
F_{Basset}	Basset force, N
F_C	particle-bubble collision force, N
F_D	liquid drag force, N
$F_{I,g}$	bubble inertial force, N
$F_{I,m}$	suspension inertial force, N
F_M	gas momentum force, N
g	gravitational acceleration, m/s^2
h	liquid level in the column, m
k_0	orifice constant, $m^3/(Pa^{0.5} \cdot s)$
L	thickness of orifice plate, m
N_c	capacitance number, dimensionless
P_0	hydrostatic pressure at the bubble surface, Pa
P_b	pressure in the bubble, Pa
P_c	pressure in the gas chamber, Pa
$P_{c,e}$	pressure in the gas chamber at the end of expansion stage, Pa
P_e	pressure at the gas inlet to the chamber, Pa
P_e	pressure at the gas inlet to the chamber, Pa
P_s	system pressure, Pa
Q_0	instantaneous gas flow rate through the orifice, m^3/s

Q_g	volumetric gas flow rate entering the gas chamber, m ³ /s
r_b	bubble radius, m
r_e	bubble radius at the end of expansion stage, m
Re	Reynolds number, $\frac{2\rho_l u_b r_b}{\mu_l}$, dimensionless
Re_o	orifice Reynolds number, $\frac{\rho_g u_0 D_0}{\mu_g}$, dimensionless
T	temperature, K
t	time, s
t_e	time of expansion stage, s
u	rise velocity of bubble base, m/s
u_0	instantaneous gas velocity through the orifice, m/s
u_b	rise velocity of bubble center, m/s
u_e	bubble expansion velocity, m/s
u_g	orifice gas velocity, m/s
u_m	velocity of liquid-solid suspensions, m/s
V_b	volume of bubble, m ³
V_c	volume of gas chamber, m ³
V_{neck}	volume of neck, m ³
x	distance between bubble center and orifice plate, m

Greek Letters

θ	angle between the radial and vertical directions of interface element, rad
ε_l	liquid holdup, dimensionless
ε_s	solids concentration, dimensionless
γ	contact angle between bubble surface and orifice, rad
	specific heat ratio of gas, dimensionless
μ_g	gas viscosity, Pa·s
μ_l	liquid viscosity, Pa·s
ρ_g	gas density, kg/m ³
ρ_l	liquid density, kg/m ³
ρ_m	density of liquid-solid suspensions, kg/m ³
ρ_s	particle density, kg/m ³
σ	surface tension, N/m
ζ	coefficient in Eq. (2), dimensionless
ΔP_l	pressure change along the bubble-liquid interface due to liquid inertia, Pa
ΔP_m	pressure change along the bubble-liquid interface due to gas momentum, Pa
$\Delta P_{m,\theta}$	pressure change at any interface element due to gas momentum, Pa
ΔP_μ	pressure change along the bubble-liquid interface due to liquid viscosity, Pa
ΔP_σ	pressure change along the bubble-liquid interface due to surface tension, Pa

Literature Cited

- Bier, K., D. Gorenflo, and J. K. Karlsruhe, "Blasenbildung und phasengrenzfläche beim dispergieren von gasen in flüssigkeiten an einzelnen gaszulauffnungen: Teil 2," *Wärme - und Stoffübertragung*, **11**, 217 (1978).
- Haynes, J. B., and D. H. T. Gotham, "Heat transfer during bubble formation," *Int. J. Heat and Fluid Flow*, **3**, 21 (1982).
- Idogawa, K., K. Ikeda, T. Fukuda, and S. Morooka, "Behavior of bubbles in a bubble column under high pressure for air-water system," *Kagaku Kogaku Ronbunshu*, **11**, 253 (1985).
- Kling, G., "Über die dynamik der blasenbildung beim begasen von flüssigkeiten unter druck," *Int. J. Heat and Mass Transfer*, **5**, 211 (1962).
- Kotake, S., "On the mechanism of nucleate boiling," *Int. J. Heat and Mass Transfer*, **9**, 711 (1966).
- Kupferberg, A., and G. J. Jameson, "Bubble formation at a submerged orifice above a gas chamber of finite volume," *Trans. Instn. Chem. Engrs.*, **47**, T241 (1969).
- LaNauze, R. D., and I. J. Harris, "Gas bubble formation at elevated system pressures," *Trans. Instn. Chem. Engrs.*, **52**, 337 (1974).
- Levich, V. G., *Physicochemical Hydrodynamics*, Prentice-Hall, 378 (1962).
- Luo, X., G. Q. Yang, D. J. Lee, and L. S. Fan, "Single bubble formation in high pressure liquid-solid suspensions," *Powder Technology*, **100**, 103 (1998).
- Marmur, A., and E. Rubin, "A theoretical model for bubble formation at an orifice submerged in an inviscid liquid," *Chem. Eng. Sci.*, **31**, 453 (1976).
- McAllister, R. A., P. H. McGinnis, and C. A. Plank, "Perforated-plate performance," *Chem. Eng. Sci.*, **9**, 25 (1958).
- McCann, D. J., and R. G. H. Prince, "Bubble formation and weeping at a submerged orifice," *Chem. Eng. Sci.*, **24**, 801 (1969).
- Milne-Thomson, L. M., *Theoretical Hydrodynamics*, 3rd ed., Macmillan and Co. Ltd., London (1955).
- Miyahara, T., and T. Takahashi, "Bubble volume in single bubbling regime with weeping at a submerged orifice," *J. Chem. Eng. Japan*, **17**, 597 (1984).
- Pinczewski, W. V., "The formation and growth of bubbles at a submerged orifice," *Chem. Eng. Sci.*, **36**, 405 (1981).
- Poritsky, H., "The growth or collapse of a spherical bubble or cavity in a viscous fluid," *A.S.M.E.*, 313 (1952).
- Ramakrishnan, S., R. Kumar, and N. R. Kuloor, "Studies in bubble formation – I: Bubble formation under constant flow conditions," *Chem. Eng. Sci.*, **24**, 731 (1969).
- Tsuge, H., and S. Hibino, "Bubble formation from an orifice submerged in liquids," *Chem. Eng. Comm.*, **22**, 63 (1983).
- Tsuge, H., Y. Nakajima, and K. Terasaka, "Behavior of bubbles formed from a submerged orifice under high system pressure," *Chem. Eng. Sci.*, **47**, 3273 (1992).
- Tsuge, H., Y. Tanaka, and S. Hibino, "Effect of the physical properties of gas on the volume of bubble formed from a submerged single orifice," *Can. J. Chem. Eng.*, **59**, 569 (1981).

- Wilkinson, P. M., and L. L. van Dierendonck, "A theoretical model for the influence of gas properties and pressure on single-bubble formation at an orifice," *Chem. Eng. Sci.*, **49**, 1429 (1994).
- Witze, C. P., V. E. Schrock, and P. L. Chambre, "Flow about a growing sphere in contact with a plane surface," *Int. J. Heat and Mass Transfer*, **11**, 1637 (1968).
- Yoo, D. H., H. Tsuge, K. Terasaka, and K. Mizutani, "Behavior of bubble formation in suspended solution for an elevated pressure system," *Chem. Eng. Sci.*, **52**, 3701 (1997).
- Yoo, D. H., K. Terasaka, and H. Tsuge, "Behavior of bubble formation at elevated pressure," *J. Chem. Eng. Japan*, **31**, 76 (1998).

Figure 1 Effect of Pressure on the Initial Bubble Size ($T=30^\circ\text{C}$, $D_0=1.63\text{ mm}$, $V_c=650\text{ cm}^3$): (a) in the liquid ($\varepsilon_s=0$); and (b) in the liquid-solid suspension ($\varepsilon_s=0.18$) (symbols: experimental data; lines: model predictions)

Figure 2 Comparison Between the Model Predictions and Experimental Data of the Initial Bubble Size under High-Pressure Conditions

Figure 3 The Bubble Formation Time as a Function of Gas Velocity at Various Pressures in the Liquid-Solid Suspension ($\epsilon_s=0.18$)

Figure 4 Effect of Pressure on the Variation of Gas Flow Rate Through the Orifice with Time during the Bubble Formation Process in a Liquid-Solid Suspension ($\epsilon_s=0.18$, $Q_g=2 \text{ cm}^3/\text{s}$)

(a)

(b)

Figure 5 Effect of Solids Concentration on Initial Bubble Size ($T=30^\circ\text{C}$, $D_0=1.63\text{mm}$, $V_c=650\text{cm}^3$): (a) variable flow conditions ($P_s=0.3\text{MPa}$, $N_c=8.8$); and (b) constant flow conditions ($P_s=4.9\text{MPa}$, $N_c=0.5$) (symbols: experimental data; lines: model predictions).

Figure 6 Comparison between the Model Predictions and the Literature Data of Initial Bubble Size under High-Pressure Conditions (For information about literature data, see Table 2.)

Figure 7 Effect of Pressure on the Variation of (a) Bubble Volume and (b) Pressure in Gas Chamber with Time during the Bubble Formation Process ($\epsilon_s=0.18$, $Q_g=2 \text{ cm}^3/\text{s}$)

Table 1 Expressions of the Forces involved in the Bubble Formation Process (Luo et al., 1998)

FORCES	EXPANSION STAGE	DETACHMENT STAGE
F_B	$\frac{4\pi}{3} r_b^3 (\rho_l - \rho_g) g$	Same as expansion stage
F_M	$\frac{\pi}{4} D_0^2 \rho_g u_0^2$	Same as expansion stage
F_D	$C_D (\pi r_b^2) \frac{\rho_l u_b^2}{2} \quad (C_D = 24/\text{Re})$	Same as expansion stage
F_σ	$\pi D_0 \sigma \cos \gamma$	Same as expansion stage
$F_{I,g}$	$\frac{d}{dt} \left[\rho_g \left(\frac{4}{3} \pi r_b^3 \right) u_b \right]$	Same as expansion stage
F_{Basset}	Not applicable	$12 r_b^2 \sqrt{\pi \rho_l \mu_l t} \frac{du}{dt}$
F_C	$\frac{\pi}{4} D_0^2 (1 + e) \epsilon_s \rho_s u_e^2$	$\pi r_b^2 \epsilon_s \rho_s u^2$
$F_{I,m}$	$\frac{d(\iiint \rho_m u_m \delta V)}{dt} = 3.86 \frac{d}{dt} \left[\rho_m \left(\frac{4}{3} \pi r_b^3 \right) u_b \right]$	Same as expansion stage

Table 2 Relevant Information from Various References used in Figure 6 concerning Bubble Formation under High-Pressure Conditions

Reference	System	P _s (MPa)	V _c (cm ³)	D ₀ (mm)	N _c	Q _g (cm ³ /s)	Symbol in Fig. 6
Kling (1962)	N ₂ , Ar/H ₂ O	0.3 ~ 8.1	9.08	1.05	0.01 ~ 0.34	0.1 ~ 5.0	×
LaNauze and Harris (1974)	CO ₂ /H ₂ O	0.1 ~ 2.17	375	4.8	0.09 ~ 2.0	1.0 ~ 30.0	◆
Bier et al. (1978)	N ₂ /H ₂ O	0.1 ~ 2.1	1775	1.6	4.1 ~ 84	0.1 ~ 20.0	◇
Tsuge et al. (1981)	He, N ₂ , Ar, CO ₂ /H ₂ O	0.1	153	1.08	16	0.1 ~ 10.0	—
Tsuge and Hibino (1983)	He, N ₂ , Ar, CO ₂ /H ₂ O	0.1	237	1.08	25	0.1 ~ 10.0	
Idogawa et al. (1985)	air/water	1.0 ~ 5.0	8.1	1.0	0.2 ~ 1.0	0.5 ~ 5.0	○
Tsuge et al. (1992)	N ₂ /H ₂ O	1.0 ~ 8.1	365	1.48	0.3 ~ 2.1	0.1 ~ 5.0	●
Wilkinson and van Dierendonck (1994)	N ₂ /H ₂ O	0.1 ~ 2.1	1775	1.6	0.4 ~ 8.7	0.1 ~ 10.0	▲
Yoo et al. (1997)	air/glycerol/ polystyrene beads	0.1 ~ 8.0	86.2	1.18	0.1 ~ 8.1	0.1 ~ 4.0	■
Yoo et al. (1998)	CO ₂ , N ₂ /H ₂ O	0.1 ~ 8.0	86.2	1.48	0.06 ~ 4.9	0.1 ~ 5.0	+
Luo et al. (1998)	N ₂ /NF fluid /glass beads	0.1 ~ 17.3	Constant flow	1.59	1.0	0.002 ~ 8.0	*
This work (1999)	N ₂ /NF fluid /glass beads	0.1 ~ 8.3	650	1.63	0.3 ~ 26.5	0.1 ~ 15.0	Δ