PROGRESS IN UNDERSTANDING THE FLUID DYNAMICS OF BUBBLE COLUMN REACTORS

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Abstract

Slurry bubble column reactors (SBCR), due to their superior heat transfer characteristics, are the contactors of choice for conversion of syngas to fuels and chemicals. This study, performed under the DOE sponsored bubble column hydrodynamics initiative, enables us to obtain experimental measurements of liquid velocity, mixing and gas holdup profiles by Computer Aided Radioactive Particle Tracking (CARPT) and gamma ray Computed Tomography (CT) and to determine their dependence on gas superficial velocity, column size and system properties. These measurements can be used to establish a data base which is instrumental in estimating the parameters needed to predict liquid mixing, as described by tracer response curves, in the pilot plant AFDU in LaPorte, Texas. Such a data base is then used to establish the relationship between the developed phenomenological models and the axial dispersion model. Finally, progress in computational fluid dynamics of bubble column flows is briefly reviewed.

Introduction

Bubble columns (BC) and slurry bubble columns (SBC), due to their excellent heat transfer characteristics, have emerged as the reactors of choice for processes in production of chemicals and fuels from syngas obtained from environmentally disadvantaged sources such as coal, waste, etc. (Dudukovic' and Devanathan, 1993). In general, BCs are large diameter cylindrical vessels with length to diameter ratios ranging from 2:1 to 50:1 in which a gaseous reactant is sparged into the liquid reactant or solvent. The presence of suspended fine catalyst particles (typically from $5\,\mu\text{m}$ to $75\,\mu\text{m}$) creates a three phase system - a slurry bubble column (SBC). Characteristic of a BC or SBC is that the superficial liquid velocity is always at least an order of magnitude lower than that of the gas. Hence, it is the gas momentum that dictates the fluid dynamics of the system, and whether net liquid flow is zero (batch), cocurrent or countercurrent to the flow of gas is immaterial as far as the multiphase flow pattern in most of the column is concerned.

Chemical and process engineers, while recognizing the enormous complexity of interacting phenomena that affect the bubble column performance (see Figure 1), until recently relied on a simple approach in their modeling by assuming plug (piston)

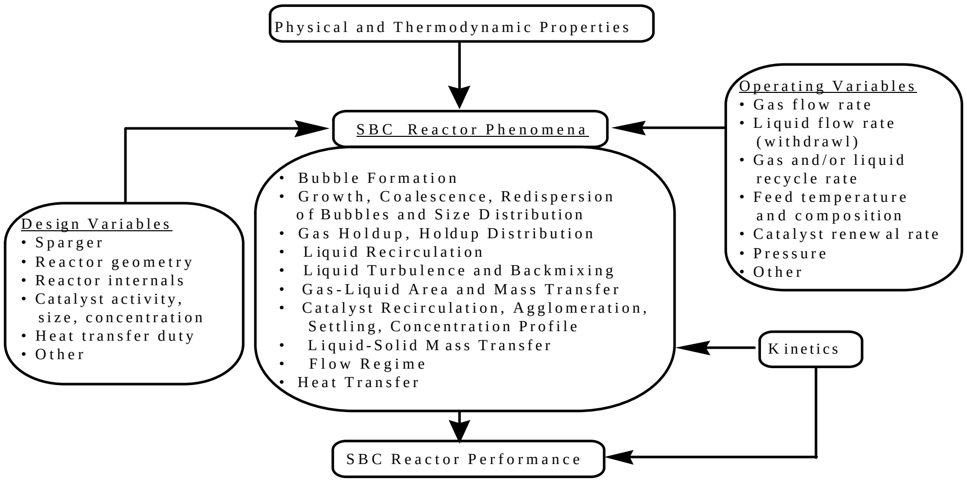


Figure 1. Phenomena affecting bubble column performance

flow of the gas, complete backmixing of the liquid and uniform catalyst distribution. Due to irrevocable trends in the demand for very high reactor volumetric productivity, which requires the use of very high gas superficial velocity of up to 50 cm/s, it is important to provide the measurement techniques and theory (or at least modeling based on observed phenomenological trends) that are capable of elucidating the behavior of large diameter, tall columns at high pressure, high solids loading and at very high gas velocities well into churn turbulent flows. Here we briefly summarize the progress made on characterizing large scale motion and in extrapolating laboratory measurements to operating conditions of interest for churn turbulent bubble columns.

Measurement of Hydrodynamic Parameters in Churn Turbulent Flow

Phenomenologically churn-turbulent flows can be readily described as follows. At high superficial gas velocities and low liquid superficial velocities in large diameter vessels high gas holdups are reached (typically well in excess of 30%) and large spiraling, transient, vortex like structures move through the column (Hills, 1975, Devanathan, 1991, Chen et al., 1994). These structures contain large voids that favor the central portion of the column. Small bubbles in the vicinity of these large voids are drawn into their wakes. Hence, on the average, gas holdup is larger in the center of the column than at the wall and this holdup profile leads, due to buoyancy forces, to induced liquid recirculation with the liquid, on the average, rising in the center of the column and falling by the walls (Hills, 1974; Devanathan et al., 1990). Small bubbles are also dragged by the upward and downward flowing liquid and those being pulled downwards constantly try to escape creating vigorous mixing in the liquid downflow region. Clearly, to understand this flow regime better and quantitatively, one needs first of all quality experimental information on gas holdup and its distribution, bubble size distribution, liquid velocities and liquid turbulence parameters, including eddy diffusivities. Various techniques for global and local holdup measurements have been recently reviewed by Kumar et al (1997). Both at the Chemical Reaction Engineering Laboratory (CREL) at Washington University and at Sandia National Laboratory gamma ray Computed Tomography (CT) has been implemented to provide time averaged gas holdup distribution in various cross sections of the column (Adkins et al, 1996; Kumar et al., 1995; 1997). It has been established that for vertical columns and carefully designed symmetric distributors gas holdup distribution is axisymmetric and can be represented by the following equation:

$$\varepsilon_g(\xi) = \frac{(m+2)\bar{\varepsilon}_G}{m+2(1-c)}\left(1-c\xi^m\right) \tag{1}$$

where ε_g is point gas holdup, $\bar{\varepsilon}_G$ is the cross-sectional average gas holdup, ξ, is the dimensionless radius, r/R, and m and c are parameters that depend on the operating conditions. It is this gas holdup distribution that gives rise to time averaged liquid circulation.

Techniques available for determination of liquid velocity and mixing parameters were also reviewed by Kumar et al. (1997). The only noninvasive technique that can provide liquid velocity patterns in churn turbulent flow throughout the column is Computed Aided Radioactive Particle Tracking (CARPT). This technique has been implemented at CREL as part of the

DOE Hydrodynamics Initiative Program. The principle of operation and its application to multiphase systems such as bubble columns has been described extensively in the literature (Devanathan et al 1990; Moslemian et al., 1992; Larachi et al 1994; Larachi et al., 1997). The CARPT technique consists of introducing into the flow field a single radioactive particle (gamma ray emitter) of the same size and density as solid particles to be traced, or neutrally buoyant particle if liquid is traced (Devanathan et al, 1990). The bubble column or slurry bubble column is then operated at steady gas and liquid superficial velocity while the position of this single radioactive particle is detected and monitored in time by an array of scintillation detectors strategically located all around the column. The principle of CARPT and processing of information is described in Figure 2. It is important to note that CARPT noninvasively provides the three dimensional velocity field in the whole column. The ensemble averaged velocities for each cell in the column are directly comparable to ensemble averaged velocities of the fluid dynamic simulation codes (Degaleesan, 1997). The difference between instantaneous and average velocity for each cell allows calculation of rms velocities and various turbulence parameters such as turbulent normal and shear (Reynolds) stresses, kinetic energy of turbulence, etc. The comparison of CARPT measured mean velocities with results from the heat pulse anemometer probe (Lubbert and Larson, 1990) at selected points shows good agreement. Comparison of CARPT measured Reynolds stresses and those determined by hot wire anemometry by Menzel et al (1990) also shows similar trends and magnitudes.

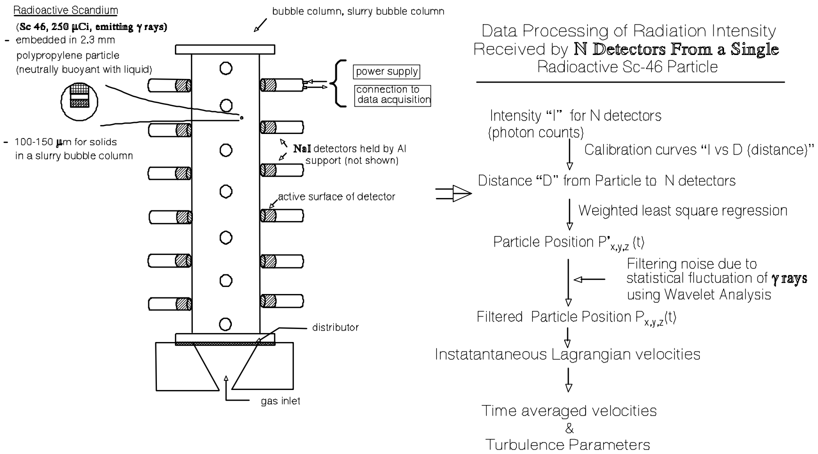


Figure 2. Schematic of CARPT and processing of CARPT information

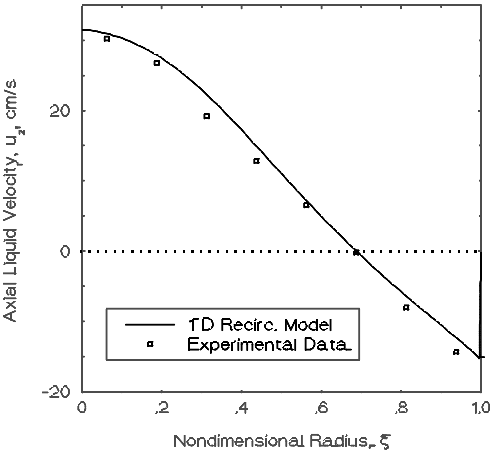
Most significantly since CARPT is a Lagrangian technique, by observing independent sojourns of the particle through a given cell the Lagrangian correlation coefficients can be constructed and eddy diffusivities can be obtained from their basic definition. CARPT directly provides the mean square displacement of the particle tracer and its time derivative for each location in the column. None of the other techniques can accomplish this.

Modeling of Churn Turbulent Bubble Column Flows

Experimental information gathered on churn-turbulent flows in bubble columns can be utilized to develop phenomenological models for liquid and gas flow mixing. Such models should provide a better basis for estimation of reactor performance. The data in columns of large L/D indicate a close to parabolic radial gas holdup profile and fully developed axial time averaged liquid velocity profile with the inversion point in proximity of 0.7 of the column radius (Devanathan, 1991). Axial eddy diffusivities are higher, and approximately constant, in the region of liquid downflow than in the central portion of the column with upward liquid flow. This is not surprising since bubbles trapped in liquid downflow attempt to rise creating increased axial mixing. Radial eddy diffusivities are an order of magnitude or more smaller than the axial ones (Devanathan, 1991; Degaleesan, 1997). This is the result of rapidly spiraling motion of large voids that create most of the turbulence in the axial direction. A recycle with cross-flow and dispersion model (RCDM) emerges as a good approximate representation of liquid flow and mixing (Degaleesan et al, 1996). This model can be extended to gas flow, gas mixing and mass transfer by accounting for large bubbles, which are always in upflow, and for small bubbles in the upflow and downflow region and for their exchange (Wang, 1996; Wang and Dudukovic', 1996; Gupta, 1996). However, while these phenomenological models are useful in utilizing the available fluid dynamic information in assessment of reactor performance, the ultimate goal is to use computational fluid dynamic codes, firmly based on basic principles, in predicting phase velocities and holdup profiles.

At this point it is instructive to ask two questions. First, how well can the fluid dynamics information collected in bubble columns be utilized to predict an independently measured quantity that depends on the flow field, such as the inert tracer impulse response? Second, how well can the CFDLIB codes, with currently available closure forms, match the observed experimental results for the holdup and velocity field in churn turbulent flow?

To address the first question, CARPT data was collected in a 8" diameter column using air-water in churn turbulent flow.

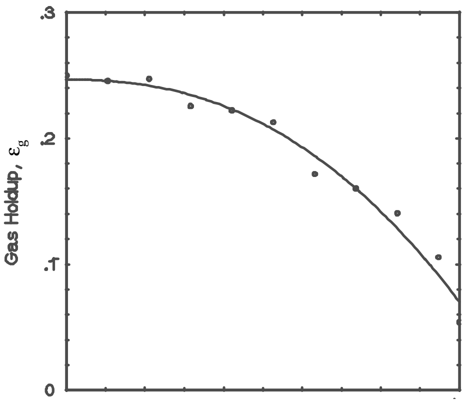


CT scans indicated an axisymmetric gas holdup distribution (Figure 3a) which did not change much with height for at least 3/4 length of the column (in the region about one diameter removed from the distributor and from the liquid and free surface). This holdup distribution drives a large liquid recirculation cell detected by CARPT. The time averaged radial liquid velocity is essentially zero in a large section of the column where a fully developed axial velocity profile of Figure 3b is present. The axial and radial eddy diffusivities are also obtained by CARPT and upon averaging over column height are displayed in Figure 3c. All the information is now available for solving the fundamental ensemble averaged tracer continuity equation which for a nonvolatile tracer takes the form below.

$$\frac{\partial(\varepsilon C)}{\partial t}+\frac{\partial}{\partial z}\left(u_z\varepsilon C\right)=\frac{1}{r}\frac{\partial}{\partial r}\left(r\varepsilon D_{rr}\right)+\frac{\partial}{\partial z}\left(\varepsilon D_{zz}\frac{\partial C}{\partial z}\right)$$

(2)

In arriving at equation (2) we have utilized the Boussinesq’s approximation and approximated the cross-correlation between velocity components and tracer concentration with products of corresponding eddy diffusivity and the mean tracer concentration gradient. Moreover, we assume that eddy diffusivities in the axial and radial direction are those measured by CARPT. Then we can solve the above equation for the distribution of concentration C, since the time averaged liquid holdup profile, $\varepsilon = 1 - \varepsilon_g$, time averaged liquid velocity profile, u_Z, axial eddy diffusivity, D_{ZZ} and radial eddy diffusivity, D_{rr} profiles are all known (Figure 3a,b,c). Then the calculated mixing cup tracer concentration can be properly normalized to provide the exit age density function for the liquid tracer. Comparison of model prediction (with no adjustable parameters) with independently taken tracer data is shown in Figure 3d. The agreement is excellent.



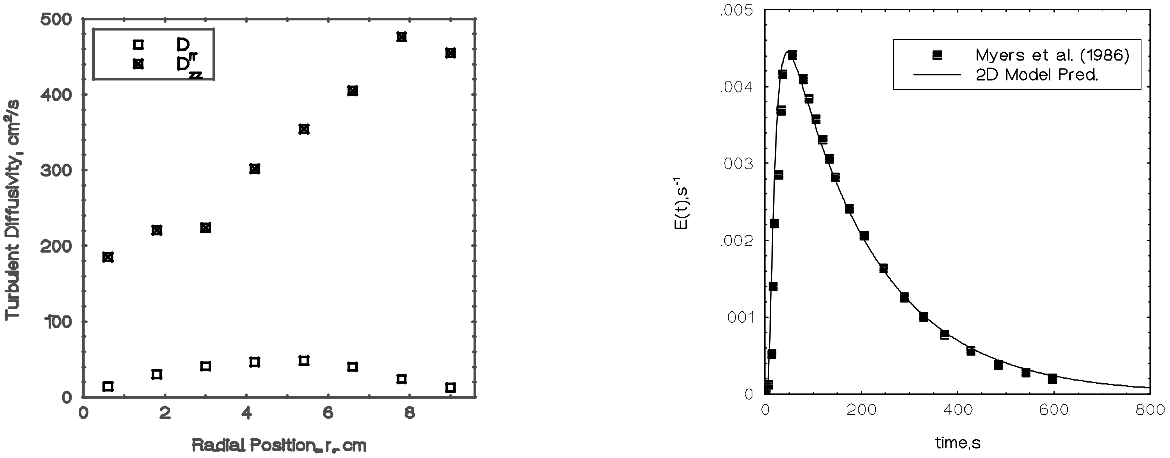
(a) (b)

(c) (d)

Figure 3. Fluid dynamic parameters from CARPT-CT for 8” diameter column, U_g = 10 cm/s
a) Gas holdup profile b) Axial liquid velocity profile c) Axial and radial turbulent eddy diffusivities
d) Comparison of model predicted and measured liquid tracer exit age density function.

This illustrates that a 2D representation of an essentially distinctly 3D flow field is sufficient to predict the overall tracer residence time distribution (RTD). The successful extension of this procedure in interpretation of industrial reactor data has been recently demonstrated by Degaleesan et al (1997). The fundamental 2D model, represented by equation (2), upon radial averaging can be readily reduced to the previously reported recycle with cross flow and dispersion model (RCDM). However, the cross flow and other parameters can now be directly related to the parameters of the fundamental model and quantities measured by CARPT. Nevertheless, in spite of the success of the 2D model (eq (2)) presented here in matching LaPorte tracer data, there is a tendency to avoid the use of models containing a number of parameters that need to be calculated based on extrapolation of an existing data base as is the case here (Degaleesan et al, 1997). For that reason we have also established a relationship between the axial dispersion coefficient and the parameters of the 2D model. It turns out that the axial dispersion coefficient is the sum of properly averaged axial eddy diffusivities and Taylor like diffusivity that includes the liquid recirculation velocity and radial eddy diffusivity (Degaleesan, 1997). This allows a firmer basis for evaluation of the axial dispersion coefficient and the use of a single parameter model familiar to industry.

Finally we need to examine the CFD ability to predict the velocity and holdup field. A good agreement between CFD two-dimensional (2-D) model predictions for holdup distribution and time averaged velocity profiles and PIV collected data in a 2-D column was illustrated for bubbly flow at very low gas velocities (Lin et al, 1996). We compared the axisymmetric 2-D CFDLIB code predictions for time averaged holdup and liquid velocities and data collected in a 3-D column operated in the churn turbulent regime. The experiments were performed by CARPT-CT at CREL in a 3-D, 19 cm diameter column with air-water at atmospheric pressure with batch liquid and air superficial velocity of 12 cm/s. A perforated plate distributor with 0.1% total open area, with holes in a square pitch of 1.25 cm and hole diameter of 0.033 cm, was used. Static liquid height was 95 cm. A 2-D axisymmetric simulation was performed with the CFDLIB codes of Los Alamos (Kashiwa and Gore, 1991). Symmetry at the centerline and free slip at the wall for both phases were used as boundary conditions. Atmospheric pressure was specified at the free surface. Free slip for liquid with the distributor was used while inflow gas condition was prescribed. The following parameters were used for various closure forms: bubble size of 0.5 cm, constant liquid phase mixing length of 1.5 cm, constant gas phase mixing length of 1.5 cm, gas-liquid drag coefficient of 0.44, lift coefficient of 0.01, and added mass coefficient of 0.5. The results were significantly sensitive only with respect to bubble size and clearly the selected 0.5 cm represents some hypothetical mean size since most likely the actual bubble size distribution is bimodal. The CARPT measured time averaged liquid velocity at a particular axial location was in very good. agreement with the computed velocity including the prediction of the inversion point for the velocity profile. However, the simulation vastly underpredicts the holdup magnitude and distorts (flattens) the holdup profile shape



compared to CT measured holdup. The simulation also underpredicts the overall holdup (0.12 versus measured 0.20). If the flat and low in magnitude gas holdup profile, predicted by the 2-D CFDLIB code is used with the liquid mixing length of 1.5 cm which was assumed in the code for the combined one-dimensional momentum balance for the two phases (Kumar et al, 1996), the calculated liquid velocity is much lower than the experimentally observed or 2-D model predicted one. This points to two possible reasons for the discrepancy in measured and predicted holdup. Most likely the interface momentum transfer terms are not modeled correctly, and, hence, expressions for drag, lift and added mass need further attention. None of these appear in the 1-D model for the combined two phases. The other possibility is that in spite of axial symmetry a 3-D flow pattern cannot be adequately captured by a 2-D simulation. Both issues are being explored.

Final Remarks

In churn turbulent two-phase bubble columns of large L/D ratio a long section of the column exhibits a fully developed height invariant time averaged radial gas holdup profile that is almost parabolic and a fully developed liquid velocity profile with the inversion point at about 0.7 of the column radius. Average holdup increases with pressure. CARPT and CT provide unique and relevant information for quantification of this flow rehime.

Utilization of the CARPT-CT measured time averaged velocities and holdup profiles and of the CARPT measured eddy diffusivities leads to a good prediction of tracer concentration distribution in the column (Degaleesan, 1997) and of the tracer residence time distribution. Computational fluid dynamic codes predict the right trend in velocity and holdup profiles but agreement between predicted and measured holdup is poor. Additional work is needed in improving the models for interfacial momentum transfer in churn turbulent flows.

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References

1. Adkins, D.R., Shollenberger, K.A.l, O'Hern, T.J. and Torczynski, J.R., "Pressure Effects on Bubble Column Flow Characteristics", in ANS Proceedings of the National Heat Transfer Conference, THD-Vol. 9, American Nuclear Society, LaGrange Park, IL., pp 318-325 (1996).
2. Chen, R.C., Reese, J., and Fan, F.-S., "Flow Structure in a Three-Dimensional Bubble Column and a Three-Phase Fluidized Bed", AIChE J., 40(7): 1093-1104 (1994)
3. Degaleesan, S., Roy, S., Kumar, S.B. and Dudukovic', M.P., "Liquid Mixing Based on Convection and Turbulent Dispersion in Bubble Columns", Chem. Engng. Sci., 51, 1967-1970 (1996).
4. Degaleesan, S., Dudukovic', M.P., Toseland, B.A. and Bhatt, B.L., "An Improved Mixing Model for Slurry Bubble Column Reactors", paper for presentation at the Banff Eng. Foundation Conf., submitted to I&ECR (1997).
5. Devanathan, N., "Investigation of Liquid Hydrodynamics in Bubble Columns via Computer Automated Radioactive Particle Tracking (CARPT) Facility", D.Sc. Thesis, Washington University, St. Louis, Missouri, USA (1991).
6. Devanathan, N., Moslemian, D. and Dudukovic', M.P., "Flow Mapping in Bubble Columns Using CARPT", Chem. Engng. Sci., 45, 2285-2291 (1990).
7. Dudukovic', M.P. and Devanathan, N., "Bubble Column Reactors: Some Recent Developments", in Chemical Reactor Technology for Environmentally Safe Reactors", (Edited by H.I. DeLasa et al.), NATO ASI Series E: Appl. Sci. 225, 379-408 (1993).
8. Gupta, P., "Fluid Dynamics and Mass Transport in Churn-Turbulent Bubble Columns", Doctoral Proposal, Washington University, St. Louis (1996).
9. Hills, J.H., "Radial Non-uniformity of Velocity and Voidage in a Bubble Column", Trans. Inst. Chem. Eng., 52: 1-9 (1974).
10. Kashiwa, B.A. and Gore, R.A., "A Four Equation Model for Multiphase Turbulent Flow", presented at the First Joint ASME/JSME Fluids Engineering Conference (1991).

11. Kumar, S.B., Devanathan, N., Moslemian, D. and Dudukovic', M.P., "Effect of Scale on Liquid Recirculation in Bubble Columns", Chem. Eng. Sci., 49(24B), 5637-5652 (1994).
12. Kumar, S.B., Moslemian, D and M.P. Dudukovic', "A Gamma-Ray Scanner for Imaging Voidage Distributions in Two Phase Flow Systems", Flow Measurement and Instrumentation, 6, 61-73 (1995).
13. Kumar, S.B., Dudukovic', M.P. and Toseland, B.A., "Measurement Techniques for Local and Global Fluid Dynamic Quantities in Two and Three Phase Systems", in Noninvasive Monitoring of Multiphase Flows (Chaouki, J., Larachi, F. and Dudukovic', M.P., eds.) Ch. 1, p 1-45 Elsevier (1997).
14. Larachi, F., Chaouki, J., Kennedy, G. and Dudukovic', M.P., "Radioactive Particle Tracking in Multiphase Reactors: Principles and Applications", Chapter 11 in Non-Invasive Monitoring of Multiphase Flows, 335-398 (1997).
15. Larachi, F., Kennedy, G. and Chaouki, J., "A Gamma-Ray Detection System for 3-D Particle Tracking in Multiphase Reactors", Nucl. Instr. and Meth., A338-568-576 (1994).
16. Lin, T.-J., Reese, J., Hong, T., Fan, L.-S. "Quantitative Analysis and Computation of Two-Dimensional Bubble Columns", AIChE J., 42(2): 301-318 (1996).
17. Lubbert, A. and Larson, B., "Detailed Investigations of the Multiphase Flow in Airlift Tower Loop Reactors", Chem. Eng. Sci., 45, 3047-3053 (1990).
18. Menzel, T. in der Deide, T., Staudacher, O., Wein, O. and Onken, U., "Reynolds Shear Stress for Modeling of Bubble Column Reactors", Ind. Eng. Chem. Res., 29, 988-994 (1990).
19. Moslemian, D., Devanathan, N. and M.P. Dudukovic', "A Radioactive Particle Tracking Facility for Investigation of Phase Recirculation in Multiphase Systems", Rev. Sci. Instrs., 63, 4361-4372 (1992).
20. Wang, Q., "Modeling of Gas and Liquid Phase Mixing with Reaction in Bubble Column Reactors", M.S. Thesis, Washington University, St. Louis (1996).
21. Wang, Q. and Dudukovic', M.P., "A Two Phase Recycle with Cross-Flow Mixing Model (TRCF) for Churn Turbulent Bubble Column Reactors", paper presented at the AIChE Annual Meeting, Chicago (1996).