1. Introduction

1.1. Overview

This report documents the work performed for the "Advanced Tomographic Flow Diagnostics for Opaque Multiphase Fluids" LDRD (Laboratory-Directed Research and Development) project and is presented as the fulfillment of the LDRD reporting requirement. Dispersed multiphase flows, particularly gas-liquid flows, are industrially important to the chemical and applied-energy industries, where bubble-column reactors are employed for chemical synthesis and waste treatment. Due to the large range of length scales (10^{-6}-10^{1} m) inherent in real systems, direct numerical simulation is not possible at present, so computational simulations are forced to use models of subgrid-scale processes, the accuracy of which strongly impacts simulation fidelity. The development and validation of such subgrid-scale models requires data sets at representative conditions. The ideal measurement techniques would provide spatially and temporally resolved full-field measurements of the distributions of all phases, their velocity fields, and additional associated quantities such as pressure and temperature. No technique or set of techniques is known that satisfies this requirement. In this study, efforts are focused on characterizing the spatial distribution of the phases in two-phase gas-liquid flow and in three-phase gas-liquid-solid flow. Due to its industrial importance, the bubble-column geometry is selected for diagnostics development and assessment. Two bubble-column testbeds are utilized: one at laboratory scale and one close to industrial scale. Several techniques for measuring the phase distributions at conditions of industrial interest are examined: level-rise measurements, differential-pressure measurements, bulk electrical impedance measurements, electrical bubble probes, x-ray tomography, gamma-densitometry tomography, and electrical impedance tomography. The first four techniques provide either spatially averaged or local information and are discussed in the context of validation. Although already well developed, the fifth technique is not suitable for large-scale flow experiments but is useful for validation efforts. The last two techniques are investigated and discussed in detail, and representative phase-distribution results are presented for gas-liquid and gas-liquid-solid flows in the two testbeds at conditions of interest.

1.2. Motivation

Dispersed nondilute multiphase flow remains one of the most challenging areas in engineering mechanics despite decades of intense research and the economic importance of these flows. Direct numerical simulations of nondilute multiphase flows from first principles are not possible for conditions of industrial relevance due to the wide range of length and time scales inherent in real systems. In the simplest cases imaginable, such as zero-Reynolds-number flow of a liquid suspension of uniform solid spheres (Ingber et al., 1994) or granular flow of uniform spheres without a continuum fluid (Taylor and Preece, 1989), extremely powerful computational platforms such as massively parallel machines are required to simulate physically relevant numbers of particles (say 10^{3}-10^{6}). Complexity is escalated greatly from these cases by progressively considering the following flow classes: turbulent fluid-solid flow (direct numerical simulation of single-phase turbulent flow is now marginally possible under certain conditions for modest Reynolds numbers); nondilute turbulent gas-solid flow, where drag is the primary interphase interaction; nondilute turbulent liquid-solid flow, where the interphase momentum exchange is much more complicated due to the comparable densities of the two phases; nondilute turbulent gas-liquid flow, where the interface between the phases becomes highly distorted (see Figure 1) and experiences continual topological change; and nondilute gas-liquid-solid flow. Geometric complexity and additional physical processes such as heat and mass transfer and chemical reactions further complicate industrial systems.

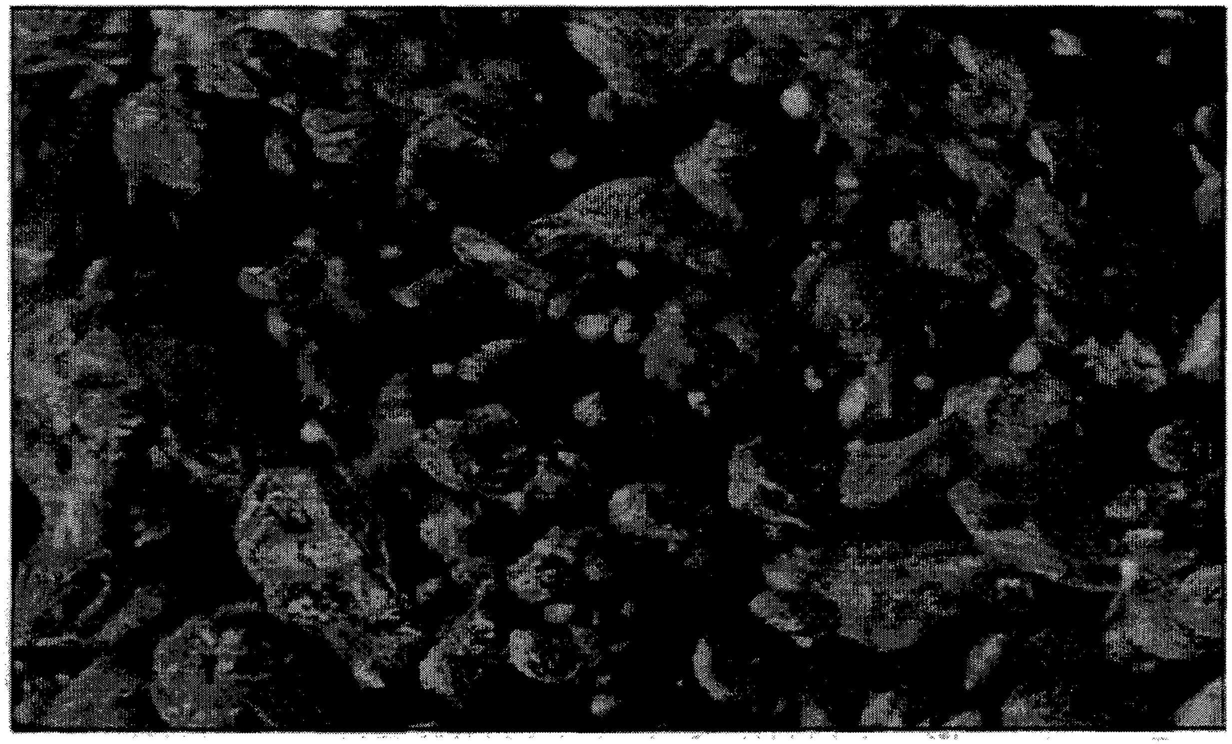


Figure 1. Nondilute turbulent gas-liquid flow.

Curiously, even the terminal velocity of an isolated gas bubble rising in a liquid continues to be a subject of research up to the present time. In a recent paper, Jamialahmadi et al. (1994) present the first properly-dimensioned relation that accurately predicts the bubble terminal velocity U_T for a wide range of parameters and flow conditions:

$$U_T = \frac{U_1 U_2}{\sqrt{U_1^2 + U_2^2}}, \tag{1}$$

$$\text{where } U_1 = \sqrt{\frac{2\zeta}{d_b(\rho_L + \rho_G)} + \frac{g d_b}{2}} \text{ and } U_2 = \frac{(\rho_L - \rho_G) g d_b^2}{18\eta_L} \begin{cases} 1 & \text{polar liquids} \\ \left[\dfrac{3\eta_L + 3\eta_G}{2\eta_L + 3\eta_G}\right] & \text{nonpolar liquids} \end{cases}. \tag{2}$$

Here, ρ_G and ρ_L are the gas and liquid densities, η_G and η_L are the gas and liquid viscosities, ζ is the surface tension, d_b is the diameter of the sphere with the same volume as the bubble, and g is the gravitational acceleration. These relations were developed from heuristic arguments involving Stokes drag on a sphere and interfacial oscillations. The terminal-velocity relation is plotted in Figure 2 for an air bubble in water for various values of surface tension and is surprisingly complex. Nevertheless, Jamialahmadi et al. (1994) show this relation to be in excellent agreement with the experimental data for a wide variety of gas-liquid systems.

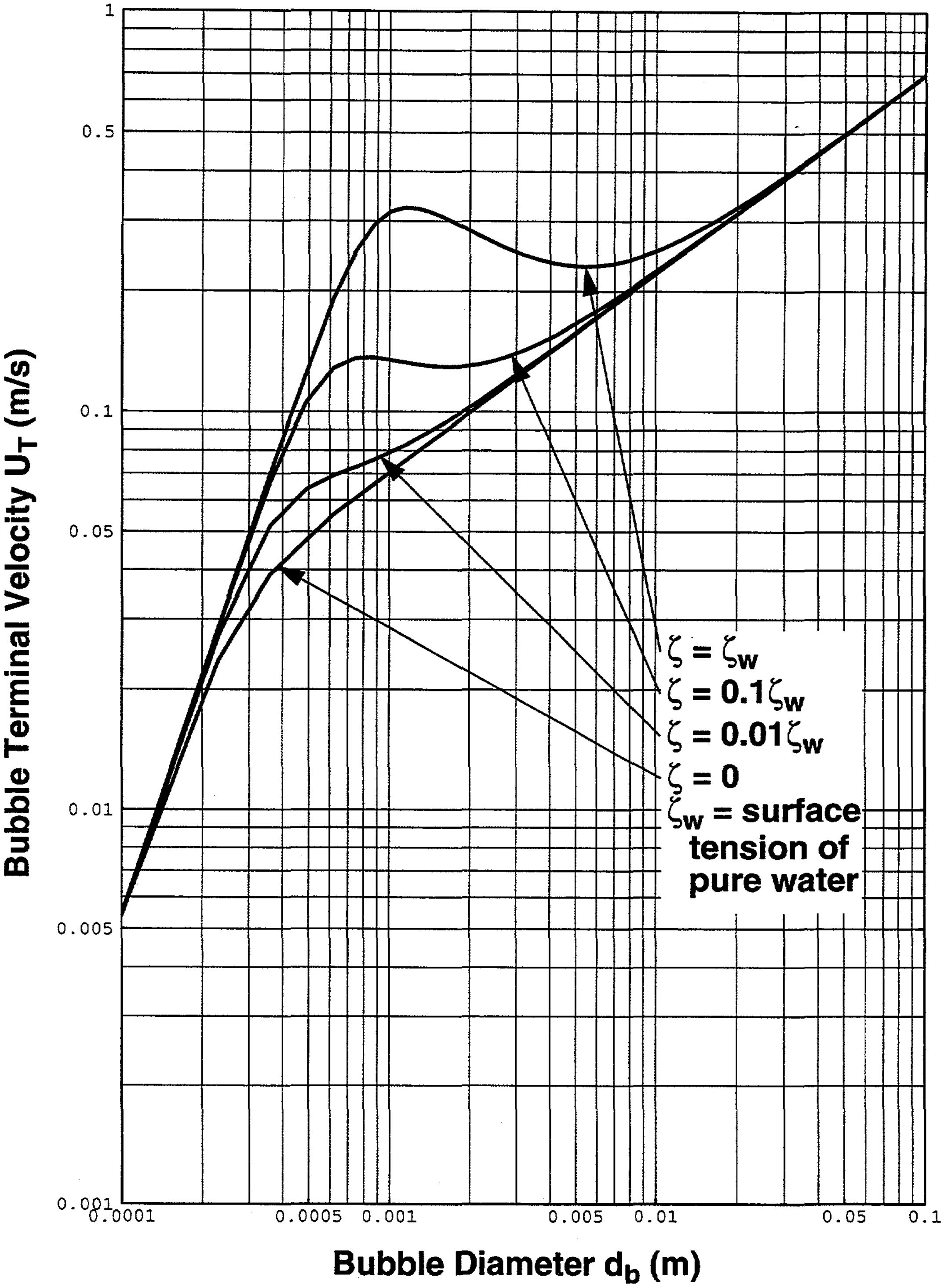


Figure 2. Terminal velocity of an isolated air bubble in water (Jamialahmadi et al., 1994).

Although direct numerical simulations of realistic multiphase flows are not possible at present (cf. Elghobashi, 1994; Crowe et al., 1996; Jaeger et al., 1996), multiphase flows can be simulated if models are introduced to describe phenomena occurring at unresolved spatial and temporal (subgrid) scales (e.g. Torvik and Svendsen, 1990; Kashiwa et al., 1993; Kashiwa et al., 1994). One such simulation of gas-liquid flow in a bubble column is shown in Figure 3 (Kumar et al., 1995b). Development of subgrid-scale models and correlations typically relies on a combination of theory (to elucidate expected scaling behavior of the modeled phenomena), numerical simulation (to examine microscale mechanisms in detail), and experiment (to search for new phenomena at extreme conditions and to quantify and validate theoretical and numerical predictions). In the continually ongoing attempt to increase the fidelity of numerical simulations employing subgrid-scale models, accuracy requirements for subgrid-scale models become increasingly stringent. As a result, experiments are called upon to deliver extremely accurate and highly detailed information to support the development of sophisticated, high-fidelity subgrid-scale models. This, in turn, necessitates the development of highly sophisticated experimental diagnostics that are capable of providing this type of information.

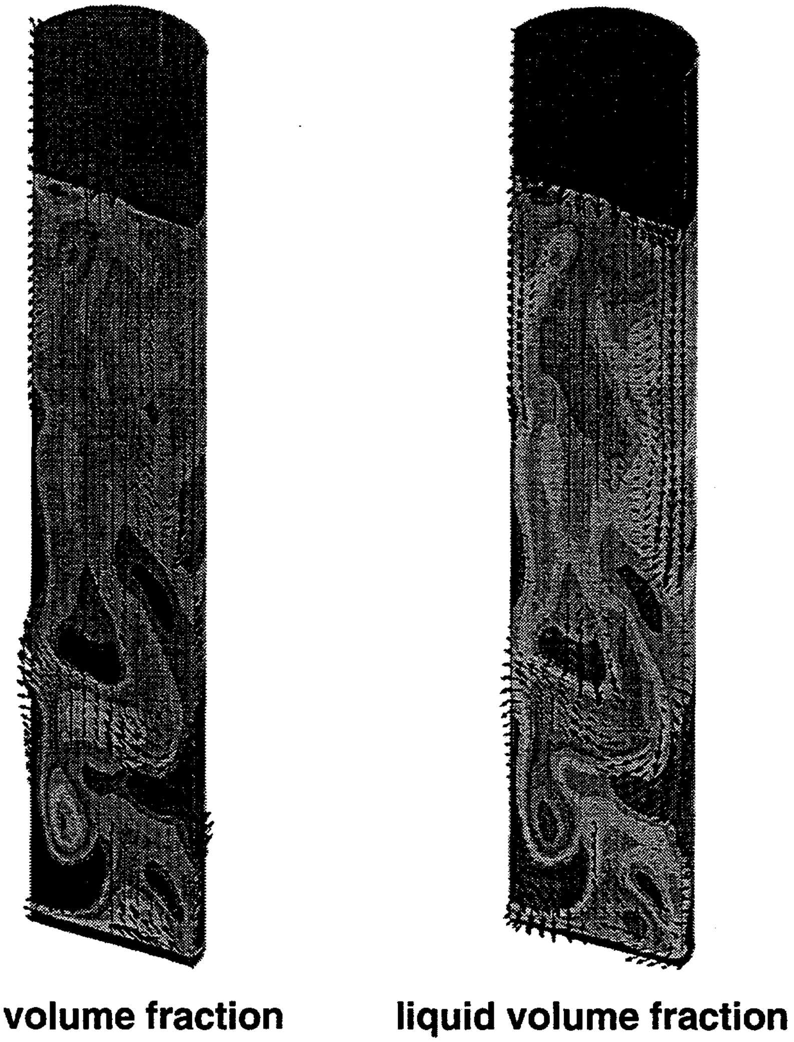


Figure 3. Los Alamos CFDLIB calculation of bubble-column flow (Kumar et al., 1995b).

Most or all of the following features characterize the type of diagnostics required for subgrid-scale model development and validation. Although volume-averaged or local measurements can be useful, particularly for diagnostics validation, full-field measurements are needed in order to develop accurate subgrid-scale models. Appreciable degrees of spatial and temporal resolution are desired since multiphase flows are rarely homogeneous in space, even at the macroscale level, and often have large fluctuations. Due to the existence of important microscale phenomena, noninvasive or minimally invasive techniques are particularly desired to avoid distorting the microscale behavior (and perhaps the macroscale behavior in the process) in the vicinity of the measurement. Techniques must be robust and capable of operating in relatively harsh mechanical, thermal, and chemical environments. Direct measurement of the desired physical quantity (e.g. phase volume fraction field) is highly desirable but rarely achievable. Instead, a different physical quantity (e.g. gamma attenuation along paths) is measured, and the quantity of primary interest is subsequently inferred via an assumed physical model (e.g. a linear relationship between phase volume fractions and gamma attenuation coefficient) and a computational algorithm (e.g. tomographic reconstruction of the field from measurements on paths). Thus, it is desirable to have a suite of rather different diagnostic techniques which rely on rather different physical assumptions and processes but which purport to measure the same physical quantity. This becomes essential when more than two phases are present.

1.3. Slurry Bubble-Column Reactors (SBCRs)

At present and in the near future, it is not possible to develop either universal numerical simulation codes or universal diagnostic techniques that cover all known regimes of multiphase flow. Rather, codes and diagnostics are typically developed within a particular context and extended to greater generality when possible. In this study, the context for diagnostics development, validation, and application is taken to be the slurry bubble-column reactor (SBCR) because of its economic and industrial importance (cf. Torczynski et al., 1994; Jackson et al., 1996ab; Dudukovic et al., 1997). Figures 4-5 show a schematic diagram of an SBCR along with a photograph of an actual SBCR (including all its ancillary plant equipment) in LaPorte, TX, operated by Air Products and Chemicals, Inc., for the U. S. Department of Energy. Briefly, an SBCR is a large-diameter, vertically oriented, cylindrical pressure vessel partially filled with a catalyst-laden liquid through which a reactive gas is bubbled (sparged). As the bubbles rise, gas dissolves into the liquid, encounters the catalyst, reacts to form the desired chemical substance, and releases heat to the surrounding liquid if the reaction is exothermic. Important industrial examples include the production of methanol or slurry Fischer-Tropsch wax (a mixture of long-chain hydrocarbons) from syngas derived from coal (principally a mixture of hydrogen and carbon monoxide). To render such processes economically viable, such reactors must be scaled to large sizes (e.g. several meters in diameter and tens of meters in height). Due to the expense of constructing a plant, accurate predictions of hydrodynamic behavior is essential when scaling to large sizes. More specifically, overly conservative scaling can lead to costly overdesign (i.e. an SBCR of far greater volume than required to convert all of its feedstock into product), and overly optimistic scaling can lead to costly, inefficient operation (i.e. an SBCR that is too small to convert all of its feedstock into product) or possibly to catastrophic failure (e.g. from excessive gas channeling through the liquid due to unexpected hydrodynamic effects). The design and scale-up process currently relies on extrapolation from small-scale experiments, where the extrapolation is conservative to minimize the possible impact of encountering a different hydrodynamic regime at larger scale. This process would be greatly facilitated by the availability of validated numerical models of multiphase flow in the parameter ranges of interest, which, as indicated above, requires the capability of acquiring the type of experimental data needed to develop and validate these models at conditions of interest.

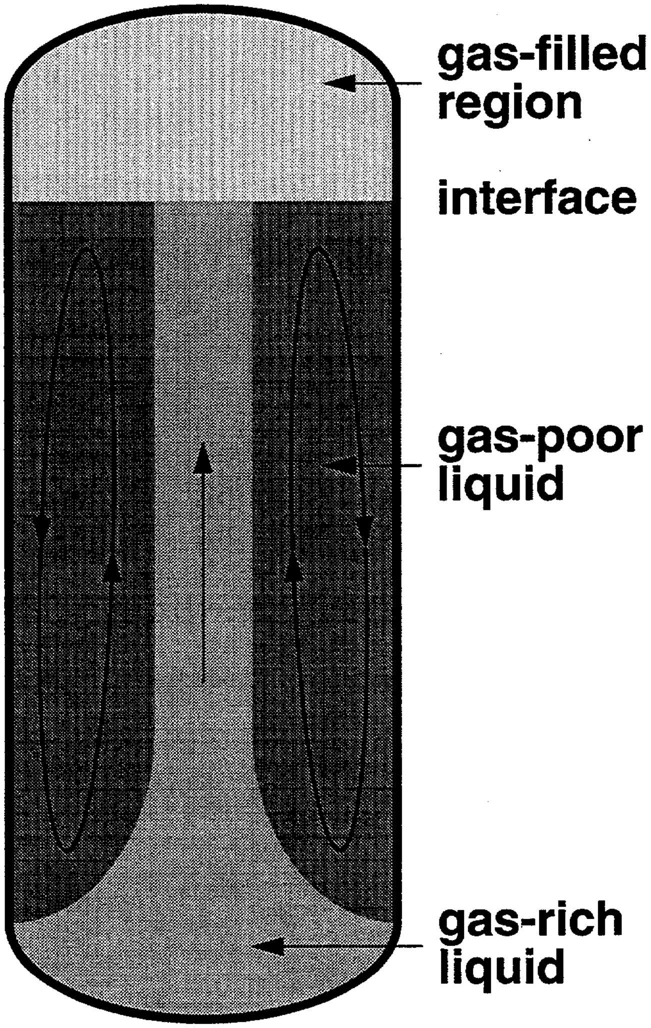
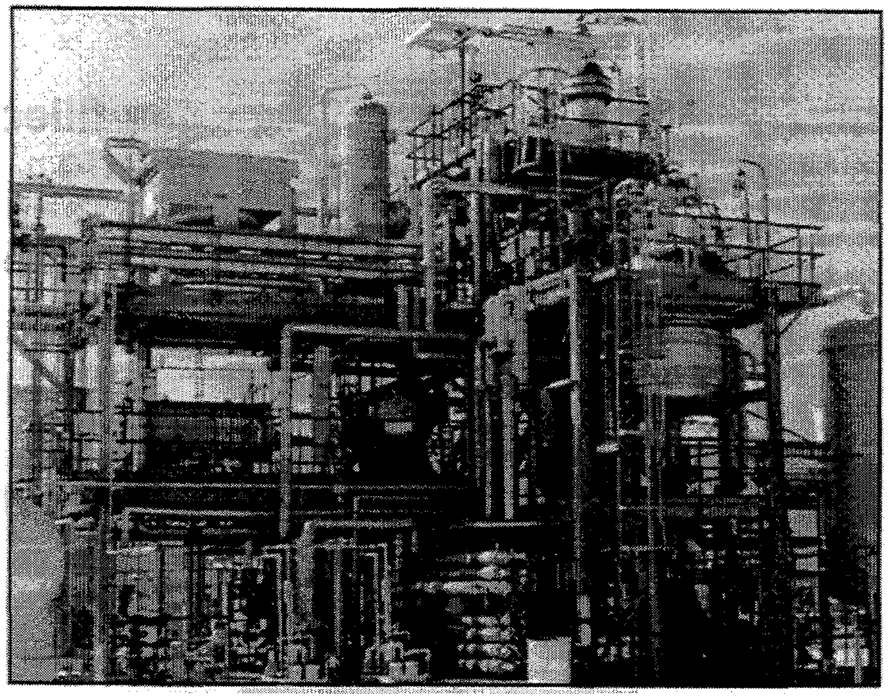


Figure 4. Slurry bubble-column reactor (SBCR) schematic diagram.

LaPorte Alternative Fuel Development Unit (AFDU), LaPorte, TX



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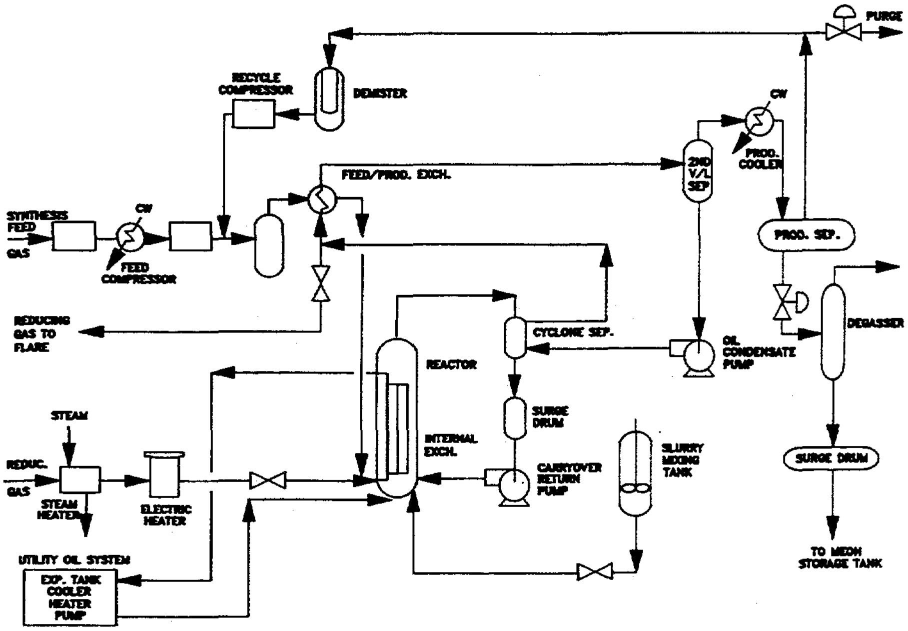

Figure 5. SBCR photograph and process diagram, courtesy of Air Products and Chemicals.

An SBCR is essentially a three-phase bubble column, with gas bubbles and solid particles contained in liquid at nondilute concentrations. Several comprehensive reviews exist in which bubble-column hydrodynamics is discussed in great detail (Shah and Deckwer, 1983; Joshi et al., 1990; Deckwer and Schumpe, 1993; Fan and Tsuchiya, 1993), and many studies exist in which particular features of bubble-column hydrodynamics are examined (e.g. Zuber and Findlay, 1965; Devanathan et al, 1990; Dudukovic et al., 1991; Wilkinson et al., 1992; Kumar et al., 1994; Krishna and Ellenberger, 1996; Dudukovic et al., 1997). The most important phenomenon governing the hydrodynamic behavior of a two-phase gas-liquid bubble column (and of a three-phase gas-liquid-solid bubble column) is buoyancy. A gas distribution that is maximum at the vessel axis and minimum at the vessel side walls is often observed and establishes a liquid circulation as indicated in the SBCR schematic diagram in the absence of forced liquid flow. The strength of this liquid circulation is determined by balancing the rate of momentum production by buoyancy against the rate of momentum transport to the side walls by viscous effects or turbulent mixing. Bubble-column hydrodynamic behavior is often correlated in terms of the gas superficial velocity U_G, which is the total gas mass flow divided by the gas density ρ_G at column conditions (i.e. not at standard conditions) and by the total cross-sectional area of the column, $\pi D^2/4$ for a cylinder of diameter D. Of particular interest is the dependence of the gas volume fraction or "holdup" ε_G on the gas superficial velocity U_G. At low gas flow rates, the homogeneous bubbly flow regime is typically encountered, in which the volume-averaged gas volume fraction increases linearly with gas superficial velocity. At higher gas flow rates, the churn-turbulent flow regime is typically encountered, in which the volume-averaged gas volume fraction increases sublinearly with gas superficial velocity. Unlike the homogeneous bubbly flow regime, the churn-turbulent flow regime is characterized by extreme flow unsteadiness, with velocity fluctuations comparable to average velocity values. The transition between homogeneous bubbly flow and churn-turbulent flow is sometimes explained in terms of the appearance of small numbers of extremely large bubbles in the latter, but this interpretation is not universally accepted. Different types of flow regimes and transitions are possible for different vessel diameters. For example, "slugging" (vertically adjacent regions alternately filled with gas and liquid) can occur for small-diameter vessels. Relatively little information is available for vessels with diameters much in excess of 1 m. The picture is further clouded by the fact that pressure, temperature, viscous effects, surface tension, sparger (gas injector) configuration, and the presence of solid particles all exert significant, and often nonintuitive, influences on bubble-column hydrodynamic behavior.

1.4. Scope of Existing and Proposed Diagnostics

The state of a two-phase gas-liquid bubble column is completely determined by specifying the pressure, the temperature, the velocity vector, and the material (gas or liquid) at all points within the vessel for all times, or at least with spatial and temporal resolution sufficient to represent all microscale processes with reasonable accuracy (Dudukovic et al., 1997). Clearly, no diagnostic or set of diagnostics is capable of measuring all of these fields with the required resolution. At present, very few diagnostics exist or have been proposed that make either in situ microscale measurements (e.g. the diameter or velocity of individual bubbles or solid particles) or average measurements of microscale statistical properties (e.g. bubble size and velocity distributions). Some exceptions include electrical and optical probes to measure diameters of individual bubbles and acoustic scattering techniques to measure bubble size distributions (e.g. Duraiswami, 1993). However, significant technical challenges must be overcome to reduce these techniques to routine practice, particularly for nondilute turbulent flow.

Most diagnostics that are currently available or under development focus on making spatially and/or temporally resolved measurements of macroscale properties. For example, pressure and temperature can be measured using various types of transducers having adequate temporal resolution on the macroscale. Adequate macroscale spatial resolution can be achieved by mounting the transducers on supports that can be traversed throughout the region of interest although this is rather invasive. Velocity measurements for nondilute turbulent multiphase flow are much rarer, even at the macroscale. The best example is presented by Dudukovic and coworkers, who have developed computer automated radioactive particle tracking (CARPT) to measure the liquid velocity field (Moslemian et al., 1992; Yang et al., 1994; Dudukovic et al., 1997). In this technique, a small, neutrally-buoyant, radioactive particle is placed within the liquid, and its position is triangulated in real time using multiple radiation detectors. The particle is presumed to move with the liquid, so its trajectory yields both the time-averaged liquid velocity field and liquid-velocity-fluctuation statistics at all locations sampled by the particle, generally the entire liquid-filled region.

Characterization of macroscale material distribution is the target of several existing and proposed diagnostic techniques, which is reasonable considering the importance of material distribution in determining the hydrodynamic behavior of bubble-column and other multiphase flows. Techniques can be divided into two broad classes: (1) diagnostics that provide only spatially-averaged or local information about material distribution; and (2) diagnostics that provide measurements of the macroscale material distribution field throughout a significant portion of the flow. While the first class of techniques provides only limited information, the second class provides the type of information needed for model validation and thus are the focus of this effort. In this second class, spatial resolution is obtained either directly, by measurements at an array of locations covering the region to be examined, or indirectly, by using mathematical techniques such as tomographic reconstruction to infer spatially resolved information from quantities measured on boundaries. Two particular techniques belonging to the second class that obtain spatial resolution indirectly are examined herein: gamma-densitometry tomography and electrical-impedance tomography.

In the remainder of this report, the following subjects are addressed. First, the physical bases and implementations of various diagnostic techniques are discussed, starting with techniques that yield only volume-averaged or local information and concluding with techniques that yield spatially and/or temporally resolved information. The former group includes level-rise measurements, differential-pressure measurements, bulk electrical impedance measurements, and electrical bubble probes, and the latter group includes x-ray tomography, gamma-densitometry tomography, and electrical-impedance tomography. Preliminary validation efforts using these techniques are discussed. Subsequently, advanced testbeds are described, and the results of extensive application of gamma-densitometry tomography and electrical-impedance tomography to these testbeds are presented.