1 Introduction

Multiphase flows are of fundamental importance in many reactor concepts in today's chemical and bioprocess industry. Over the last years, increasing volumes of products manufactured in fermentation processes or using heterogeneous catalysis have yielded a need for large-scale reactors, in turn inducing new demand for precise scale-up rules for the transfer of processes from lab-scale to full industrial scale which in the case of bubble columns can mean diameters of several meters.

Design of gas-liquid bubble columns has so far mainly been carried out by means of empirical and semi-empirical correlations which have been gained from experimental data e. g. of mass transfer for bubble columns of different scales [27]. While a strong experimental foundation of such correlations provides security for the applications that they initially were conceived for, transferability to other situations (e. g. different substances or temperature and pressure ranges) is usually very limited. Thus, in many cases trial-and-error schemes or time-consuming scale-up experiments are necessary to achieve satisfactory performance of the large-scale reactor system.

Three-phase flow in bubble columns introduces additional challenges for design and scale-up. Now, not only gas-liquid mass transfer is of concern but additionally, solid fluidization and liquid-solid heat and mass exchange enter the picture yielding a complex interaction of turbulent flow fields, mass transfer and chemical or biochemical reaction inside the reactor. Even modern approaches to reactor scale-up mostly do only consider single aspects out of this multitude, i. e. are restricted to fluiddynamic facets, mass transfer issues or reaction topics [103]. Complete reactor models are still out of reach mostly due to numerical problems in solving the resulting equation systems and – the accelerating development in this field notwithstanding – due to a lack in computational power.

On the measurement side of the issue, recent developments have cast some light into gas distribution and solid fluidization depending on superficial gas velocity and sparger geometry in three-phase bubble columns of pilot-plant scale. Based on work by Sauer and Hempel [101], Lindert et al. [69, 70] and Kochbeck et al. [56], Dziallas et al. [31, 32] have developed a new invasive measurement technique based on a combination of differential pressure measurement and conductivity respectively time domain reflectometry measurement which enables a detailed determination of local gas and solid holdup. Using this new technique, interesting aspects of

sparger geometry and superficial gas velocity influence on local gas distribution as well as solid fluidization in a model system consisting of deionized water, plastic granules and air could be shown.

On the modeling side, Computational Fluid Dynamics (CFD) are gaining importance in general process applications. CFD approaches use numerical techniques for solving the Navier-Stokes equations for given flow geometry and boundary conditions thereby implementing models for flow aspects like turbulence or heat and mass transfer as relevant for the specific modeling task. CFD has been an important tool in air and space industry or vehicle design for a long time [2] where it has to a large extent replaced time-consuming and expensive wind tunnel experiments. Yet, while in these applications single-phase flows are prevailing, modeling applications in chemical and biochemical reactors in most cases include multiphase flows the modeling and numerical treatment of which introduce additional challenges [24, 29]. Therefore, multiphase CFD applications have gained broad attention only during the last decade since increasing computational power available has enabled computations previously considered infeasible. Still, most literature reports are limited to two-phase flows [7], and especially gas-liquid CFD projects often deal only with very low dispersed phase holdups. In effect this means that multiphase CFD still is far away from being a general tool for the practitioner even if recent advances in computational power available in desktop PCs do enable first steps in this direction [45, 116].

Starting point for the project presented in this report was the question how liquid flow structure inside a pilot plant-size bubble column is interacting with local gas and solid holdup, i. e. what influence gassing rate has on liquid flow, how liquid flow is related to solid fluidization and what influence solid loading on liquid circulation and mixing inside the reactor has. For this purpose, the Electrodiffusion Measurement technique (EDM) was to be implemented for reason of its capabilities to deliver two-dimensional liquid velocity values even at high gas and solid holdups inside the 0.63 m diameter, 6 m high bubble column as used by Dziallas et al. [31, 32]. Fig. 1.1 shows the experimental setup.

The reactor could be equipped with a plate sparger (335 holes of 1 mm diameter in triangular layout), a ring sparger (diameter 0.45 m, 12 gassing openings facing to the bottom of the reactor for better solid fluidization) or a central nozzle sparger (diameter 22 mm). Table 1.1 shows properties of the model system as introduced by Dziallas et al. [31, 32] and used for the experiments of the present work.

The model system and especially the solid material have been chosen such as to resemble the flow situation inside a bubble column bioreactor where microorganisms grow immobilized on particles and aeration is used for oxygen intake, mixing and fluidization.

In addition, a Computational Fluid Dynamics (CFD) model based on the experimental results was to be developed that – neglecting mass transfer and chemical reactions for the time being – should be able to reproduce the measured distributions of gas and solid holdup as well as liquid

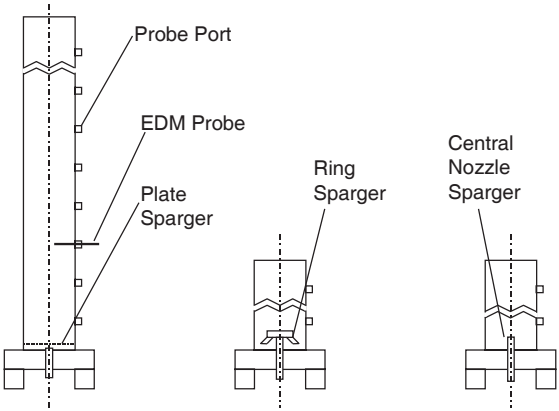

Figure 1.1: Experimental setup: Pilot-plant sized bubble column with different sparger systems; column height 6 m, inner diameter 0.63 m

Table 1.1: Properties of the three-phase model system chosen for the experiments

Liquid Phase	Deionized water with 0.01 mol/L Potassium-Sulfate
Gas Phase	Oil-free pressurized air
Solid Phase	Plexiglass (Polymethyl-Methacrylate, PMMA) particles, density 1200 kg/m^3, cubically shaped, hydraulic diameter 3 mm

velocity thus yielding a flow and holdup prediction tool useful for scale-up calculations as well. Implementation of the model should take place on a common PC workstation such as to show that CFD reactor design studies can be carried out on standard modern desktop hardware at reasonable computation times.

2 Multiphase Flow Fundamentals

2.1 Modeling Fundamentals: The Roots

Modern approaches to modeling of chemical process equipment draw their impetus from two distinct sources. On one side, empirical and semi-empirical reactor modeling approaches based on a long history of dimensional analysis can be found. On the other side, modern Computational Fluid Dynamics (CFD) that have been popular for quite a while in other fields of science (like air and space engineering) are becoming more and more interesting for chemical engineers. Both fields are still coexisting but with computational resources becoming cheaper and more widely available, they soon will develop into one stream of detailed reactor modeling. A first step in this direction has been taken by Bauer and Eigenberger [3] who in their so-called "Multiscale Approach" described the fluid dynamics via CFD computation and included chemical reaction and mass transfer by means of a zone model.

2.1.1 Empirical and Semi-Empirical Models for Bubble Columns

Before the advent of CFD techniques, reactor modeling for chemical and biotechnological purposes was mainly carried out by means of highly simplified, semi-empirical parameter-fitting models. This was due to the fact that with computational resources available until just a short time ago, calculations with more precise models would have taken up a prohibitive amount of time thus being way too expensive for any application of interest. However, if one's interests are limited to certain singular phenomena such as mixing, residence time distribution or averaged mass transfer coefficients for a tightly defined reactor type, a huge body of correlations can be found from the literature [27]. While the limited range of a model's applicability always has to be kept in mind, in the field of two- and three-phase bubble column and airlift loop reactor modeling (and the adjoining area of fluidized bed modeling) a number of approaches have gained large popularity due to their comparatively general validity [103]. Namely these models are:

- Cell models: These models assume circulation cells inside the reactor which are responsible for backmixing processes [26].

- The one-dimensional dispersion model: This model assumes that two- and three-phase flow processes can be modeled as a superposition of convective and dispersive flow where the latter is described in analogy to Fick's first law of diffusion [59, 62, 69, 73, 103, 110, 111]. It is often used in conjunction with gravity terms to model sedimentation effects [25, 101]. More refined versions of this model include sparger and degassing zone effects as well [35].

- Cascade models: These models describe real reactors such as bubble columns or airlift reactors in terms of cascades of ideal continuous stirred tank reactors (CSTRs). Main fitting parameter is the number of tanks in the cascade; backmixing effects can also be included by introducing partial backflow into the model [69].

- The two-dimensional dispersion model in cylindrical coordinates: This model includes radial effects in addition to axial dispersion. In cylindrical coordinates it can be formulated as follows [103]:

$$\frac{\partial \varepsilon_\alpha}{\partial t} = D_{ax,\alpha} \cdot \frac{\partial^2 \varepsilon_\alpha}{\partial x^2} - v_{ax,\alpha,c} \cdot \frac{\partial \varepsilon_\alpha}{\partial x} + \frac{1}{r} \cdot D_{rad,\alpha} \cdot \frac{\partial \varepsilon_\alpha}{\partial r} + D_{rad,\alpha} \cdot \frac{\partial^2 \varepsilon_\alpha}{\partial r^2} \tag{2.1}$$

  A similar equation can be put up for any continuous phase α or dispersed phase β. This model delivers – like its one-dimensional version – only reasonable results for phases which are continuously flowing through the reactor. A simpler model for bubble distribution in three-phase fluidized beds which only considers radial dispersion has been presented by Lee and de Lasa [65].

- Mechanical power balance models: These models calculate liquid circulation velocities from pneumatic power input into the reactor due to gas sparging. This is accomplished by solving force balances including pressure loss and gravitational terms [131].

With the application of the models listed above, great care has to be taken with respect to the areas of their validity. Empirical or semi-empirical models usually are verified only for a limited set of parameter variations. While interpolation between those given values may be viable, extrapolation will always yield results of questionable integrity. This also puts serious limitations to the models' scale-up capabilities.

2.1.2 Computational Fluid Dynamics

Computational Fluid Dynamics (CFD) are an engineering tool which has gained large popularity during the last years. As opposed to the semi-empirical models described above, CFD aims at solving the (complete or simplified) fundamental physical equations that describe a flow phenomenon. The most general form of these equations has been given by Navier and Stokes more than 150 years ago, therefore the set of equations has been aptly named Navier-Stokes equations [2, 120].

These equations encompass mass, momentum and energy balances; they have to be adapted to the specific problem under consideration by additional closure laws [34].

While CFD has been very popular among car manufacturers and in the air and space industry [38], chemical engineers have only recently become aware of the large potential it bears for the development and improvement of process equipment. This is mainly due to the fact that with modeling flow around a car body or an airplane wing, only single-phase flow has to be considered while in most applications in chemical reactors two- and three-phase flows are common. This poses a wealth of new questions and brings about serious difficulties in modeling and numerics.

2.2 Multiphase CFD

In multiphase flow, solving one single mass balance and three momentum balances is no longer sufficient to compute flow fields for all involved phases. While all multiphase CFD approaches do solve these balances for the continuous phase, different ways of treating the dispersed phases have been suggested. However, all models are still way from being reliable tools for the improvement of existing processes or for scale-up considerations. As Sundaresan [116] has pointed out, this is mainly due to the fact that most models have to be numerically solved on grids that are much coarser than the flow structures that one wants to resolve, simply because available computational power is by far not sufficient. This situation is worsened by the fact that unlike with single-phase flow, two-dimensional and steady-state calculations deliver no useful results making three-dimensional transient calculations unavoidable. While a lot of work has already been published on gas-liquid two-phase flow, three-phase flow modeling is still at the very beginning.

2.2.1 Basic Concepts

In multiphase CFD, two main approaches can be discriminated. While all models compute the flow field of the continuous phase using the Navier-Stokes equations, the dispersed phases can either be calculated in a Lagrangian manner as consisting of discrete entities (bubbles, particles or clusters) or as semi-continuous phases where all phases are regarded as interpenetrating continua (Euler-Euler or multi-fluid approach). The **Lagrangian approaches** can be divided according to their treatment of the dispersed phases as follows:

- The Euler-Lagrange Approach: With this concept, bubbles and particles are considered as having a fixed size and shape; computational particles always represent a certain number of real particles. In the numerical solution procedure, the transient Navier-Stokes equations for the continuous phase are solved first; particle velocity and new particle position for the next time step are then calculated using friction laws for every single computational particle

[24, 28, 48]. In an optional third step, two-way coupling effects can be considered where the particles' influence on the continuous-phase flow field are calculated and a new flow field is computed iteratively. Particle-particle or particle-wall interactions can be incorporated into the model as well [112]. As can easily be imagined, the computational effort for this calculation procedure increases drastically with increasing dispersed phase content, leaving its main use so far in the computation of dilute systems or for special applications like residence time distribution calculations which cannot be computed using multi-fluid approaches [64].

- Direct Numerical Simulation (DNS): With the standard Lagrange approach, a particle is considered as occupying only one grid cell at a time giving only one relevant velocity acting on it. In a more refined approach, particles can occupy more than one cell and subsequently experience different velocities simultaneously [30, 67]. In addition, turbulence is resolved directly without any modeling [8, 126]. This yields much more complex particle motions and calls for much finer numerical grids and shorter time steps resulting in an even larger computational demand. Since DNS in general gives very precise results, it is increasingly being used to verify other modeling approaches when experiments are infeasible [9]. A subdivision of DNS called Large Eddy Simulation (LES) tries to reduce computational demand by directly resolving only the larger turbulence eddies and modeling the smaller ones thus enabling calculations on a coarser grid and with larger time steps [126].

- Volume-of-Fluid Methods: In this even more refined approach for the modeling of gas-liquid two-phase flow, bubbles are considered as deformable; even free surfaces can be modeled [43, 68, 117]. One single field of flow vectors is computed; bubbles are distinguished from the liquid by their lower density which means that volume fractions are used to represent surfaces. The sharp density transition at the surface and the fact that rather coarse grids have to be used pose a number of questions related to bubble mass conservation and the calculation of smooth bubble surfaces; for numerical reasons, by now the density ratio between liquid and gaseous phase may not exceed a value of 20 which renders calculations of actual air bubbles in water (density ratio of approximately 800) impossible. While this class of methods has been successfully implemented for the calculation of free surface flows (e. g. bubbles rising to a surface and dissolving there inducing droplet formation [117]), it is still prohibitive for real-life reactor modeling with respect to its immense computational demand.

As can easily be seen from the model descriptions above, all Lagrangian particle-tracking models suffer from high demands on computational power; this renders them rather unsuitable for the computation of multiphase flows in real process applications where dispersed phase holdups are usually high. Therefore, in this project the Euler-Euler or multi-fluid approach will be imple-

mented which allows for the computation of three-phase flow fields even with high solid and gas holdups at reasonable computational expense.

With the **Euler-Euler** or **multi-fluid approach**, a set of Navier-Stokes equations has to be put up for every phase under consideration. This means that if no energy equations are solved (i. e. for isothermal flow conditions which are always assumed here), in three-phase flows 12 partial differential equations (PDEs) have to be solved simultaneously. Depending on which phase interactions are included in the model (e. g. particle-particle or particle-wall interactions or effects due to rotating particles and surface tension), these equations can take on very different appearances [36, 53, 85, 127]. A most complete survey of equation types including terms to even cover flow regime transitions in horizontal evaporator pipes is given by Lahey and Drew [63] and in the book by Drew and Passman [29].

The inclusion of mass transfer and chemical reactions into CFD calculations by means of species balances and reaction kinetic rate equations is fairly straightforward from a conceptional point of view but still suffers from numerical limitations due to the strong coupling of the resulting system of equations. Therefore, CFD approaches which include these effects so far have mainly used simplified semi-empirical models for the description of mass transfer and reactions; e. g. Bauer and Eigenberger [3] have investigated into the possibilities of a so-called "Multiscale Approach" for the description of a nonisothermal parallel-consecutive reaction. In this project, neither mass transfer nor chemical reactions have been included into the calculations.

Several authors have reported on attempts at solving the two-dimensional multiphase Navier-Stokes equations thereby hoping to be able to reduce computational demand [61, 81, 109]. What they found was, however, that no reasonable, grid-independent results could be obtained leaving three-dimensional computations as the only viable approach. The same outcome is reported on steady-state calculations [28, 79, 93, 109] which only proved that bubble column calculations have to be performed in a transient manner. All of these results are somewhat of a drawback to the initial hope that with multi-fluid calculations computational demand could be reduced drastically.

With these results in mind, the Navier-Stokes equations as used here can be put up in a manner following [83, 93]. The continuity equation without mass transfer and source terms (i. e. without consideration of chemical reactions) in multiphase formulation becomes for both continuous and dispersed phases:

$$\frac{\partial}{\partial t}(\rho_\alpha \varepsilon_\alpha) + \frac{\partial}{\partial x_i}(\rho_\alpha \varepsilon_\alpha u_{\alpha,i}) = 0 \tag{2.2}$$

The momentum balance in multiphase formulation can be written out slightly differently for continuous and dispersed phases. For the continuous phase, it becomes in general formulation:

$$\begin{aligned} \frac{\partial}{\partial t}(\rho_\alpha \varepsilon_\alpha u_{\alpha,i}) + \frac{\partial}{\partial x_j}(\rho_\alpha \varepsilon_\alpha u_{\alpha,i} u_{\alpha,j}) &= -\varepsilon_\alpha \frac{\partial p}{\partial x_i} + \frac{\partial}{\partial x_j} \varepsilon_\alpha \mu_\alpha \left(\frac{\partial u_{\alpha,i}}{\partial x_j} + \frac{\partial u_{\alpha,j}}{\partial x_i} \right) \\ &\quad + \rho_\alpha \varepsilon_\alpha g_i + M_{\alpha,i} \end{aligned} \tag{2.3}$$

Writing down momentum balances for the dispersed phases yields:

$$\frac{\partial}{\partial t}(\rho_\beta \varepsilon_\beta u_{\beta,i}) + \frac{\partial}{\partial x_j}(\rho_\beta \varepsilon_\beta u_{\beta,i} u_{\beta,j}) = -\varepsilon_\beta \frac{\partial p}{\partial x_i} + \frac{\partial}{\partial x_j} \varepsilon_\beta \mu_\beta \left(\frac{\partial u_{\beta,i}}{\partial x_j} + \frac{\partial u_{\beta,j}}{\partial x_i} \right) + \rho_\beta \varepsilon_\beta g_i + M_{\beta,i} \tag{2.4}$$

In the above equations, gas and liquid viscosity are given the actual values valid for the local conditions (temperature, pressure) in the reactor or – in case of the liquid phase – are modified to account for turbulence influence. With solid viscosity, no value or correlation for three-phase flow can be obtained from literature data (solid pressure approaches implementing kinetic theory for fluidized beds [36] can not be transfered in a straightforward manner). It is basically a fitting parameter which has been set constant to the value of water (10^{-3} Pas) for all calculations of this project. This is assumed a legal approximation since test calculations showed that a variation of solid viscosity between 10^{-4} Pas and 10^{-1} Pas did not yield a significant difference in computed time-averaged dispersed phase distribution and liquid flow fields.

Detailed formulation of turbulence and phase interaction terms will be given in section 2.2.4.2 and section 2.2.3, respectively. These terms are of crucial importance for the correct calculation of flow and holdup structure. Even with two-phase flows, laminar calculations cannot deliver grid-independent results [93, 109], thus turbulence models have to be implemented which account for sub-grid size flow structures. In three-phase flows, direct interactions between gas and solid dispersed phases have to be modeled as well in order to achieve correct solid fluidization.

To obtain a solvable system with as many equations as unknowns present, additional closure equations have to be implemented accompanying the 12 partial differential Navier-Stokes equations [29]. These may be algebraic equations or PDEs as well. Since adiabatic gas expansion with increasing vertical position in the bubble column is the main reason for the measured axial gas holdup profiles [31], a CFD model has to consider the gas phase compressibility as well; liquid and solid phases are assumed incompressible. This means that a relationship between gas density and static pressure has to be found; in this project, the ideal gas law has been used where gas constant and temperature are assumed as constant throughout the calculations:

$$\rho_g = \frac{p \cdot \tilde{M}_g}{\tilde{R} \cdot T} \tag{2.5}$$

Here, the molar mass of the gas phase is given the value of air ($\tilde{M}_g$ = 28.8 kg/kmol), the gas constant is set to $\tilde{R}$ = 8.315 kJ/(kmol K). Gravity effects are included in the body force terms of the momentum equations (e. g. $\rho_\beta \varepsilon_\beta g_i$ for dispersed gas or solid phase). While several authors have reported on bubble size models which account for coalescence and break-up effects [22, 66, 77, 78, 108], these effects are neglected here for the sake of simplicity and convergence and bubble size is set to a constant value of 0.008 m. Bubble-bubble interactions and wake effects are neglected as well [54]. Interactions between gas and solid phases, however, have to be modeled in order to obtain correct solids fluidization (see section 2.2.3.2).

Turbulence has an influence on the liquid viscosity μ_α in eqn. 2.3; therefore, a turbulence model has to be included yielding additional algebraic and partial differential closure equations. This will be discussed in detail in section 2.2.4.

2.2.2 Application Examples from the Literature

With the increasing popularity of CFD in the field of process engineering, a lot of interesting applications have been reported on both from university researchers and from industrial CFD users. Birthig et al. [7] give a very broad review of CFD modeling activities in the largest German chemical companies (including Degussa-Hüls, Axiva, Bayer and BASF) proving that these methods have been successfully implemented for apparatus modeling tasks ranging from gas spargers over extruders, impellers, pipe reactors, bubble columns, fixed-bed reactors to spray dryers and separation devices. It is interesting to mention, though, that all their examples consider at most two-phase flows. Further surveys of the current state of CFD modeling activities have been given by LaRoche [64] and Sundaresan [116].

Among the very first CFD applications in the field of process engineering to be reported were Stirred Tank computations [84]. This is due to the fact that even single-phase calculations can deliver interesting results about local velocities and mixing conditions inside the tank – without the numerical problems related to multiphase calculations. Kohnen and Bohnet [58] presented a two-fluid model of solid-liquid flow in a stirred tank and investigated especially into turbulent dissipation; with a very refined model they were able to reduce the error between power input of the stirrer and turbulent power dissipation to less than 20 %. Schütze et al. [105] focused on CFD modeling of Oxygen transfer in a stirred tank bioreactor and succeeded in even including the free surface into their mass transfer calculations.

Results on bubble column CFD calculations have been reported by many groups. Pfleger et al. [93] conducted investigations into the behaviour of a flat laboratory-scale rectangular two-phase bubble column using the Euler-Euler approach under CFX-4.2; main result of their work is that even a flat bubble column cannot be modeled two-dimensionally. This result is supported by the work of Sokolichin and Eigenberger [109] who used their own CFD code but had to implement three-dimensional calculations as well. Both of these projects were carried out at extremely low superficial gas velocities (below 0.01 m/s). Krishna et al. [60, 61] reported on CFD modeling of a pilot-plant size bubble column using CFX-4.2 with the Euler-Euler model as well at higher superficial gas velocities (up to 0.28 m/s). While one of their reports is entitled "*Three-Phase Eulerian Simulation*" [61], the reader would be misled to assume that they included solid particles into their considerations; moreover, two dispersed gas phases were calculated to include the different influences of large and small bubbles. In addition, they calculated integral gas holdups and derived a scale-up correlation for bubble columns of different sizes. Sparger influence on the flow