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BUBBLE COLUMN REACTOR DESIGN USING A CFD CODE

Katerina A. Mouza, Nikolaos A. Kazakis and Spiros V. Paras*

Laboratory of Chemical Process and Plant Design
Department of Chemical Engineering
Aristotle University of Thessaloniki
Univ. Box 455, GR 54124 Thessaloniki, GREECE
* *tel*: +302310996174, *e-mail*:paras@cheng.auth.gr, *www*: http://philon.cheng.auth.gr

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Abstract. *This work is motivated by the need to develop reliable predictive tools for bubble column reactor design using a CFD code. Although bubble columns are widely used in industrial gas-liquid operations, their design and scale up is still a difficult task and subject to errors, due to the generally complex structure of the multiphase flow encountered in this type of equipment. Population balance equations combined with a three-dimensional model were used in order to study the operation of a rectangular bubble column. In addition, the mechanisms of bubble coalescence and break-up were considered into the Eulerian-Eulerian simulation, while being applied to a multiple size group model. Computational results have been compared to experimental data and it appears that bubble size distribution, axial liquid velocity and gas holdup can be well predicted at the homogeneous regime for the air-water system. The results acquired for the air-water system were encouraging and the simulations are to be extended to gas-liquid systems where the liquid phase is other than water. However, additional experimental work at the microscopic level combined with theoretical analysis are considered necessary for establishing CFD codes that will successfully predict the behavior of bubble column reactors.*

1 INTRODUCTION

Bubble columns are widely used for conducting gas–liquid reactions in a variety of practical applications in industry such as absorption, fermentations, bio-reactions, coal liquefaction and waste water treatment. Due to their simple construction, low operating cost, high energy efficiency and good mass and heat transfer, bubble columns offer many advantages when used as gas–liquid contactors. In all these processes gas holdup, ε_g, and bubble size distribution are important design parameters, since they define the gas-liquid interfacial area available for mass transfer (α), which is given by:

$$\alpha = \frac{6\varepsilon_g}{d_{32}} \tag{1}$$

where d_{32} is the mean *Sauter* diameter of the bubble size distribution.

Depending on the gas flow rate, two main flow regimes are observed in bubble columns, i.e. the ***homogeneous*** bubbly flow regime and the ***heterogeneous*** (churn-turbulent flow) regime. The homogeneous regime is encountered at relatively low gas velocities and characterized by a narrow bubble size distribution and radially uniform gas holdup and is most desirable for practical applications, because it offers a large contact area[1],[2]. Bubble size distribution and gas holdup in gas-liquid dispersions depend extensively on column geometry, operating conditions, physico-chemical properties of the two phases and type of gas sparger. The design of bubble columns has primarily been carried out by means of empirical or semi-empirical correlations based mainly on experimental data. The scale up of bubble columns is still poorly understood due to the complexity of flow patterns and their unknown behavior under different sets of design parameters such as diameter, height to diameter ratio, gas sparger type, physical properties of the liquid etc[3],[4].

A method to gain more knowledge and detailed physical understanding of the hydrodynamics in bubble columns is Computational Fluid Dynamics (*CFD*). CFD can be regarded as an effective tool to clarify the importance of physical effects (e.g. gravity, surface tension) on flow by adding or removing them at will. An increasing number of papers deal with CFD application to bubble columns[4],[1]. Wild *et al.*[4] cite the most important reasons for this increasing interest:

- Measuring techniques applied to the local hydrodynamics in bubble columns made huge progresses so that more reliable local measurements are now available to check the adequacy of CFD predictions.
- The computer capacity increased.
- The quality of the different CFD program systems has been improved: better numerical methods, more realistic closure laws.

- The occurrence of flow regimes, of deformable gas-liquid interfaces and the lack of knowledge about the closure terms (turbulence, bubble-bubble interactions) has transformed bubble column studies into a benchmark of all gas-liquid dispersed flows.

To analyze the flow pattern of bubble columns both under steady and transient state conditions employing *CFD* simulation, the majority of researchers uses either two- or three-dimensional models, while numerical simulations are usually compared to experimental data.

Spicka *et al.*[5] used a commercial CFD code to simulate a two phase flow in a bubble column both with a stagnant and a flowing liquid phase. Different sets of gas and liquid velocity were applied as inlet boundary conditions and the variation of the local gas volume fraction as well as the liquid velocity profile for an air-water system was studied. To simplify the model they assumed that the bubbles have uniform and constant size in the column, an assumption which is not supported by the experimental data.

As most of the early CFD studies consider monodispersed bubble size distributions ignoring break-up and coalescence mechanisms, their validity is limited. Krishna & Van Baten[6] and Van Baten & Krishna[7] considered a three-phase continuum and assumed that bubbles in the column were either 'small' or 'large', with different velocities, but with no interaction between them. However, when industrial systems are considered, higher superficial gas velocities are encountered, and, thus, both bubble coalescence and break-up must be taken into account in order to better describe the reactor hydrodynamics[4]. In gas–liquid two-phase systems, bubble break-up and coalescence can greatly influence their overall performance by altering the interfacial area available for mass transfer between the phases. So, in order to develop reliable predictive tools for bubble column reactor design, it is essential to obtain some insight into the prevailing phenomena through simulation models based on the bubble formation/distraction mechanisms, i.e. bubble coalescence and break-up[8],[9]. Therefore, Lo[10], Olmos *et al.*[2] and Buwa & Ranade[9] incorporated a coalescence and break-up model into a CFD simulation and calculated hydrodynamic variables such as liquid velocity, gas holdup and bubble size distributions. However, in order to simplify their models, the bubbles injected by the sparger were set to have the ***same*** size, an assumption also made by Shimizu *et al.*[8], who considered that the bubble column reactor consists of a series of discrete 'compartments' in which bubble break-up and coalescence occur and bubbles move from compartment to compartment with different velocities.

Furthermore, since the majority of published data refers to air-water systems, Wild *et al.*[4] suggest that CFD simulations must be validated with gas-liquid systems where the liquid phase is other than water. Dhotre *et al.*[11] developed a two-dimensional CFD model for the prediction of flow pattern in bubble column reactors and studied the effect of the sparger design and column height to diameter ratio on radial gas holdup profiles for three different gas-liquid systems, i.e. air-water, air-aqueous solution of butanol and air-aqueous solution of carboxyl methyl cellulose. Likewise, Van Baten & Krishna[12] carried out fully three-dimensional simulations in the churn-turbulent flow regime, where air and *Tellus* oil, with a viscosity 75 times that of water, were used as gaseous and liquid phase respectively, but without taking into account the break-up or the coalescence mechanism.

Additional experiments and theoretical analyses are needed to establish rigorous criteria for the bubble interaction mechanisms at the microscopic level. Without such criteria, current numerical simulation methods can, at best, be tested by indirect means. So, it is important to develop theoretical frameworks that will enable the coupling of macroscopic and microscopic behaviors into a physically meaningful and accurate model[13].

The intention of this work is to simulate an existing experimental bubble column reactor using a commercial CFD code and to validate the results with the experimental data acquired.

2 MATHEMATICAL MODEL

The type of flow encountered in bubble columns is referred to as polydispersed multiphase flow, i.e. flow in which the dispersed phase covers a broad range of size groups. One of the attributes of polydispersed multiphase flow is that the different sizes of the dispersed phase interact with each other through the mechanisms of break-up and coalescence. To deal with this type of flow, a population balance equation must be formulated. Population balance is a well-established method for calculating the size distribution of a polydispersed phase, including break-up and coalescence effects, a general form of which is:

$$\frac{\partial n_i}{\partial t} + \nabla \cdot \left(\vec{u}_g n_i \right) = B_B - D_B + B_C - D_C \tag{2}$$

where $\vec{u}_g$ is the gas velocity, n_i represents the number density of size group i and the terms on the right-hand side represent the '*birth*' and '*death*' due to break-up and coalescence. The number density n_i is related to the gas holdup ε_g by:

$$\varepsilon_g f_i = n_i V_i \tag{3}$$

where f_i represents the volume fraction of bubbles of group i and V_i is the corresponding volume of a bubble of group i.

According to Prince & Blanch model[14] the coalescence of two bubbles occurs in three steps. First, the bubbles collide trapping a small amount of liquid between them. Then, this liquid film drains until it reaches a critical thickness. The film then ruptures and the bubbles join together. The coalescence process therefore depends on the collision rate of the two bubbles and the collision efficiency that is a function of the time required for coalescence (t_{ij}) and the contact time (τ_{ij}). Collision is a result of three mechanisms: turbulence (θ_{ij}^{T}), laminar shear (θ_{ij}^{LS}) and buoyancy (θ_{ij}^{B}) and the total coalescence rate is:

$$Q_{ij} = \left(\theta_{ij}^{T} + \theta_{ij}^{B} + \theta_{ij}^{LS}\right) e^{\left(-t_{ij}/\tau_{ij}\right)} \tag{4}$$

The motion of an eddy of the same length scale with the bubble diameter is responsible for the relative motion between bubbles. Collision is also induced by the difference in rise velocities of bubbles with different sizes. Bubble columns usually operate at the homogeneous regime, where due to the narrow bubble size distribution the relative effect of buoyancy can be neglected leading to the assumption that bubbles rise with the same velocity regardless of their size. Collision occurs when a strong circulation pattern develops and, as that happens at gas rates higher than those encountered in the homogeneous regime, laminar shear term can be neglected. Consequently, only the turbulent contributions are included in the model for collision frequency, given by:

$$\theta_{ij}^{T} = n_i n_j S_{ij} (\overline{u}_{ti}^2 + \overline{u}_{tj}^2) \tag{5}$$

where n_i and n_j are the concentrations of bubbles of radius r_{bi} and r_{bj} respectively, $\overline{u}_t$ is the average turbulent fluctuating velocity and S_{ij} is the collision cross-sectional area of the bubbles defined by:

$$S_{ij} = \frac{\pi}{4}(r_{bi} + r_{bj})^2 \tag{6}$$

The time required for coalescence and the contact time are given by ***Eqs.*** (7) and (8) respectively:

$$t_{ij} = \left(\frac{r_{ij}^3 \rho_l}{16\sigma}\right)^{1/2} \ln\frac{h_o}{h_f} \tag{7}$$

$$\tau_{ij} = \frac{r_{ij}^{2/3}}{\varepsilon^{1/3}} \tag{8}$$

where r_{ij}, ρ_l, σ and ε are the equivalent radius, the density of the liquid phase, the surface tension and the turbulent energy dissipation rate respectively. The parameters h_o and h_f represent the film thickness when collision begins and the film thickness when rupture of the film occurs, respectively. The values of the above parameters depend mainly on the physical properties of the liquid phase and have been experimentally computed for the air-water system.

The birth rate of group i due to coalescence of group j and group k bubbles is:

$$B_c = \frac{1}{2}\sum_{j=1}^{i}\sum_{k=1}^{i} Q_{jk}\, n_j\, n_k \tag{9}$$

The death rate of group i due to coalescence with other bubbles is:

$$D_c = n_i \sum_{j=1}^{N} Q_{ij}\, n_j \tag{10}$$

Luo & Svendsen[15] developed a theoretical model for the prediction of drop and bubble break-up in turbulent dispersions. The model is based on the theory of isotropic turbulence and probability and contains no unknown or adjustable parameters. It must be noted that binary break-up is assumed. The break-up rate of bubbles of size i into bubbles of size j is modeled by:

$$g\left(v_j : v_i\right) = 0{,}923\left(1-\varepsilon_g\right)\left(\frac{\varepsilon}{d_j^2}\right)^{\frac{1}{3}} \int_{\xi_{\min}}^{1} \frac{\left(1+\xi\right)^2}{\xi^{\frac{11}{3}}} e^{-\chi_c}\, d\xi \tag{11}$$

where d_j is the bubble diameter, ξ is the dimensionless size of eddies in the inertial sub-range of isotropic turbulence and χ_c is the critical dimensionless energy for break-up, which is proportional to the surface tension and counter-proportional to the density of the liquid phase. The birth rate of group *i* bubbles due to break-up of larger bubbles is:

$$B_B = \sum_{j=i+1}^{N} g\left(v_j : v_i\right) n_j \tag{12}$$

The death rate of group *i* bubbles due to break-up into smaller bubbles is:

$$D_B = g_i \, n_i \tag{13}$$

The mass conservation equation for the liquid and the gas phase, in the Eulerian framework, is given by ***Eqs.*** (14) and (15), respectively:

$$\frac{\partial(\varepsilon_l \rho_l)}{\partial t} + \nabla \cdot (\varepsilon_l \rho_l \vec{u}_l) = 0 \tag{14}$$

$$\frac{\partial(\varepsilon_g f_i \rho_g)}{\partial t} + \nabla \cdot (\varepsilon_g f_i \rho_g \vec{u}_g) = S_i \tag{15}$$

In the above equations, S_i is the net rate at which mass accumulates in group *i* due to coalescence and break-up, ρ_g is the density of the gas phase, ε_l and $\vec{u}_l$ are the liquid volume fraction and velocity respectively. The momentum conservation equation is given by:

$$\frac{\partial(\rho_m \varepsilon_m \vec{u}_m)}{\partial t} + \nabla \cdot (\rho_m \varepsilon_m \vec{u}_m \vec{u}_m - \mu_m \varepsilon_m (\nabla \vec{u}_m + (\nabla \vec{u}_m)^T)) = -\varepsilon_m \nabla p + M_{mn} + \rho_m g \tag{16}$$

where ρ_m, $\vec{u}_m$, ε_m and μ_m represent the density, velocity, volume fraction and viscosity of the *m*th phase respectively, p is the pressure, M_{mn} the interphase momentum exchange between phase *m* and phase *n* and g is the gravitational force. The momentum exchange between gas and liquid phase is given by:

$$M_{\mathrm{lg}} = \frac{3}{4} \rho_l \frac{\varepsilon_g}{d_{32}} C_D (\vec{u}_g - \vec{u}_l) \left| \vec{u}_g - \vec{u}_l \right| \tag{17}$$

where C_D is the interfacial drag coefficient.

3 THE CFD CODE

CFD software packages are based on the use of either 'segregated solvers' or the 'coupled solver' approach. The former solves the underlying equations of Mass, Momentum and Energy in a sequential manner, whereas in the latter the conservation equations are solved simultaneously after being linearized. The 'coupled solver' approach reduces significantly the number of iterations required to obtain convergence, but it does require the solution of a large coupled system of linear equations. To overcome this problem the 'Algebraic Multi-Grid' (*AMG*) solution technique can be applied for the solution of the coupled system of linear equations. Multi-Phase flow models are usually dealt with using a Multi-Fluid approach, where each phase is considered a separate fluid with its own velocity field, while sharing a common pressure field. An extension of the pressure correction schemes (e.g. the IPSA Algorithm) can be used to calculate the volume fractions and the common pressure field. However, most software packages use the Two-Fluid Model, i.e a simplification of the general method that deals with only two fluids, where one velocity field represents the continuous phase and another the dispersed phase. The prediction of bubble sizes is accomplished through the use of a population balance model[16].

In this work a commercial CFD code (namely *CFX*® developed by *AEA*) is employed for bubble column design. The MUSIG (***Mu****ltiple* ***Si****ze* ***G****roup*) model (*CFX*® *5.7* from *ANSYS*) used in this study combines the population balance method with the break-up[15] and coalescence[14] models in order to predict the bubble size distribution of the dispersed phase and it uses the Eulerian-Eulerian two-fluid model. For the type of flow encountered in a bubble column, in which a dilute phase is dispersed in a continuous phase, it is recommended to use the *k-ε* model for the continuous fluid and the *dispersed phase zero equation* model for the dispersed phase[17]. Thus, the turbulent contribution to the stress tensor for the continuous phase (liquid) is evaluated by means of *k-ε* model, with its standard parameters: C_μ=0.09, $C_{1\varepsilon}$=1.44, $C_{2\varepsilon}$=1.92, σ_k=1 and σ_ε=1.3. The Sato's additional term is also used in order to take into account the influence of the dispersed phase on the turbulence of the liquid[18]. The

dispersed phase zero equation model, applied on the dispersed phase, is an algebraic turbulence model and it is represented by:

$$\nu_{td} = \frac{\nu_{tc}}{\sigma_g} \tag{18}$$

where the parameter σ_g is a turbulent Prandtl number relating the dispersed phase (gas) kinematic eddy viscosity v_{td} to the continuous phase kinematic eddy viscosity v_{tc}.

Between the most important parameters employed in the MUSIG model are the initial and final thickness of the liquid film (***Eq.*** (7)) trapped between two bubbles, just before coalescence occurs, since it determines the time required for coalescence during the collision. The values of the initial and final thickness of the liquid film have been determined only for the air-water system and found equal with 10^{-4} and 10^{-8}m respectively[14]. Most of the studies, using the MUSIG model[2],[4], have been conducted without taking into account these parameters. Therefore, it is essential to investigate how the liquid properties such as surface tension, density and viscosity affect the above parameters by studying gas-liquid systems with a liquid phase other than water.

The CFX®5.7 code uses a finite volume method on a non-orthogonal body-fitted multi-block grid. The volumetric gas flow and the gas size distribution, obtained from experimental data, were applied as the inlet boundary condition at the sparger region, while the liquid phase is regarded as stagnant. The 'degassing' boundary condition was applied at the top of the column, which permits only the dispersed bubbles to escape and not the liquid phase[17]. Calculations were performed on an AMD Athlon 2800MHz with 512Mb RAM.

The geometry studied in the present simulations is consistent with an existing bubble column (***Figure 1***), but, to reduce the calculation time, the ¼ of the column is simulated (***Figure 2***) in 3-D due to symmetry. The experimental set-up (***Figure 1***) consists of a vertical rectangular Plexiglas® column 1.5m height, having a square cross section (side length 10cm). For the injection and uniform distribution of the gas phase, a *gas sparger*, i.e. a round metal porous disk, 2.0cm in-diameter, is installed at the center of the bottom plate. The gas sparger used is a *316L SS* porous disk (made by *Mott Corp*.) with nominal pore size of 20μm. The experiments are conducted at ambient pressure and temperature conditions. Each experimental run starts by first filling the column with the appropriate liquid phase up to 80cm above the sparger. All the experiments are performed with no liquid throughput, while the gas phase is injected and distributed into the liquid phase by passing through the porous disk sparger.

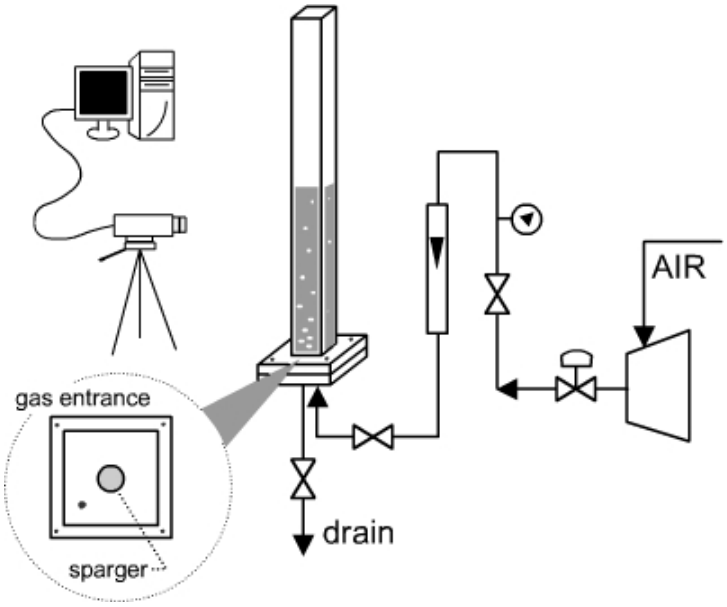

Figure 1. Experimental setup

A high-speed digital video camera (*Redlake MotionScope PCI® 1000S*) is used for direct flow visualisation and for bubble size as well as gas holdup measurements. The recorded images are used to extract *quantitative* information on bubble size distribution and gas holdup values. Approximately 800 bubbles are measured in each experimental run, a number considered to be adequate for statistical calculations. More details about the experimental procedure and methods used are given in a paper by Mouza *et al.*[19].

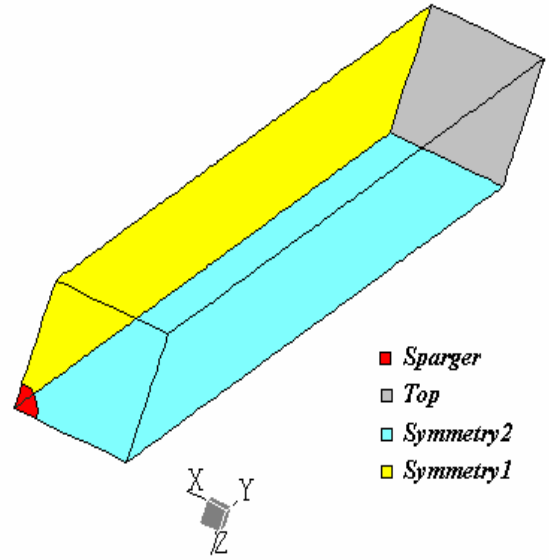

Figure 2. Geometry of simulated bubble column.

4 RESULTS AND DISCUSSION

The air-water system at the homogeneous regime has been studied computationally so far, since this regime is of practical interest. Moreover, experimental data are not available for the code validation at the heterogeneous regime, due to experimental difficulties.

For the sake of simulation, the bubble sizes were distributed into twelve groups (bins) with diameters between 0 and 7mm. The simulation results are compared with the experimental data[19] acquired in our Lab. The experimental distribution, which follows a *log-normal* distribution, was imposed as initial condition in the CFD simulation and the corresponding distribution at 40cm above the sparger was predicted. ***Figure 3*** shows the experimental and the calculated bubble size distribution over the sparger for a gas flow rate Q_G=0.7x10^{-5}m^3/s. The two distributions seem to be practically the same, a fact that implies that bubble interaction mechanisms have no significant effect for this system of fluids and for the gas flow rates tested.

Figure 4 presents the liquid phase axial velocity variation along the column radius. The calculated values are in fairly good agreement with the experimental data of Vial *et al.*[20], who conducted axial velocity measurements of the liquid phase using LDA in a cylindrical bubble column with fine porous sparger. It is obvious that the CFD simulation also predicts the liquid recirculation into the bubble column.

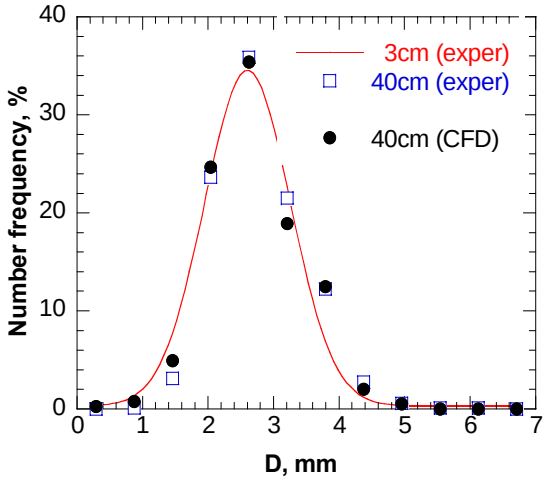

Figure 3. Bubble size distribution at various distances above gas sparger for air-water system [Q_G=0.7x10^{-5} m^3/s]

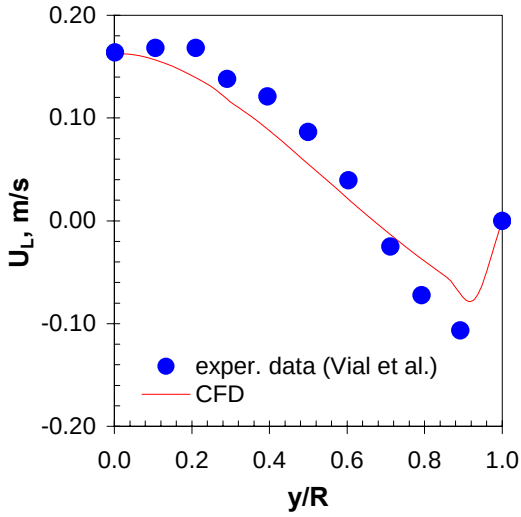

Figure 4. Comparison of experimental mean axial liquid velocity[20], along the column radius (R), to CFD predictions

Figure 5 presents the effect of the superficial gas velocity (U_{GS}) with respect to the column cross section on the overall gas holdup in the bubble reactor. The comparison of the experimental data with the simulation predictions reveals that the CFD code calculates satisfactorily the gas holdup at the homogeneous regime but underpredicts it at the heterogeneous regime, where the model assumptions of equal bubble velocity and low gas flow rates do not hold true.

In contrast to the air-water system, the bubble interaction mechanisms are important for systems with liquids having surface tension and/or viscosity values different from these of water (e.g. low surface tension butanol-solutions or high viscosity glycerin-solutions)[19]. In ***Figure 6*** the bubble size distribution changes with the distance from the gas sparger when the liquid phase is a glycerin in water solution (66.7%), indicating that coalescence and break-up do occur inside the column. Preliminary results proved, as it was expected, that the CFD model can not give reasonable result by just using the physical properties of the liquid phase without adjusting the parameters h_o and h_f of ***Eq.*** (7), which control the coalescence mechanism.

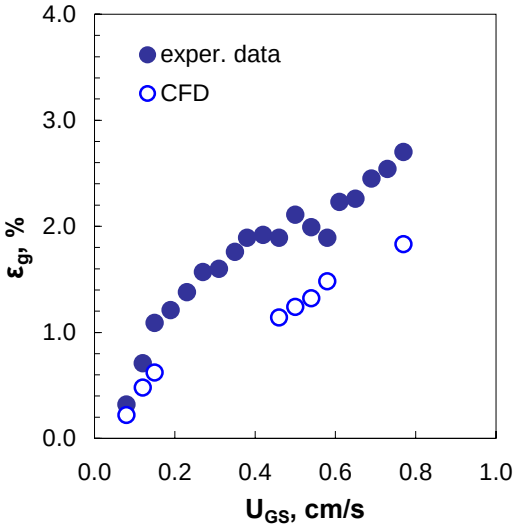

Figure 5. Comparison of experimental gas holdup values[19] to CFD predictions

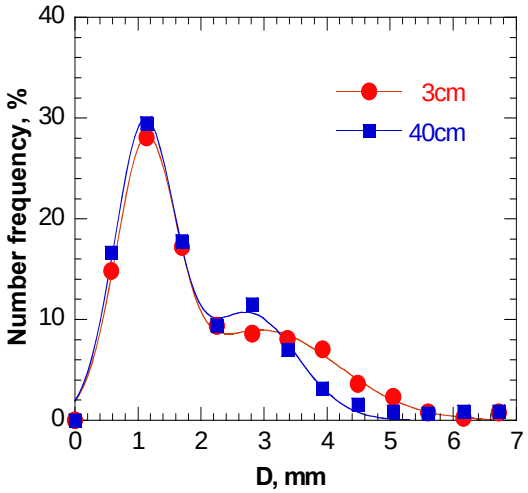

Figure 6. Bubble size distribution at various distances above gas sparger for air-glycerin (66.7%) system [$Q_G=0.7x10^{-5}m^3/s$]

5 CONCLUSION

Population balance combined with coalescence and break-up models were taken into consideration, simulating the operation of a bubble column, using a CFD code. Computational results have been compared to experimental data and it appears that bubble size distribution, axial liquid velocity and gas holdup can be well predicted at the homogeneous regime for the air-water system. In order to extend the validity of the CFD codes to other flow regimes and include systems with various physical properties, there is need for better models concerning interaction between bubbles. Experimental work combined with CFD simulations would lead to correlations of universal validity between the adjustable parameters and the physical properties of the liquid phase.

New experimental data must be collected concerning the bubble interaction mechanisms in gas-liquid systems with the liquid phase being other than water. These data at the microscopic level combined with theoretical analysis are considered necessary in order to establish rigorous criteria for the coalescence and breakage of bubbles. Thus, the coupling of macroscopic and microscopic behaviors into physically meaningful and accurate models will permit the successful prediction of bubble column behavior, instead of trying to define the adjustable model parameters simply by comparison with experimental data.

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