Ninth International Conference on CFD in the Minerals and Process Industries
CSIRO, Melbourne, Australia
10-12 December 2012

SIMULATION OF COALESCENCE, BREAK UP AND MASS TRANSFER IN GAS-LIQUID SYSTEMS BY USING MONTE CARLO AND QUADRATURE-BASED MOMENT METHODS

Antonio Buffo[1], Daniele L. Marchisio[1*], Marco Vanni[1], Peter Renze[2]

[1] Dept. Applied Science and Technology, Politecnico di Torino, Torino, ITALY
[2] BASF SE, Ludwigshafen, GERMANY

*Corresponding author, E-mail address: daniele.marchisio@polito.it

ABSTRACT

The description of the fluid-dynamics and of mass transfer in gas-liquid systems requires the evaluation of bubble size and composition distributions, dictated by bubble breakage, coalescence and mass exchange with the liquid phase. In our previous work, gas-liquid systems were investigated by coupling CFD with mono-variate population balance equation (PBE) solved with the quadrature method of moments (QMOM). The problem is here extended to the solution of multi-variate PBE. In this work the Conditional Quadrature Method of Moments (CQMOM) is for the first time validated by comparison with Direct Simulation Monte Carlo (DSMC). Results, obtained from the comparison of the two methods, show that the quadrature approximation implicitly used in CQMOM for overcoming the closure problem is very accurate. CQMOM is then implemented in ANSYS/Fluent and OpenFoam and predictions are compared with experiments, resulting in very good agreement.

NOMENCLATURE AND ACRONYMS

G	continuous rate of change of bubble size
h	discontinuous event for coalescence and break up
L	bubble size
$M_{k,l}$	mixed moment of order k and l
n	number density function
$\mathbf{u}_L$	bubble velocity conditioned on size
ϕ	number of oxygen moles in one bubble
$\dot{\phi}$	mass transfer rate

CFD	computational fluid dynamics
CQMOM	conditional quadrature method of moments
DQMOM	direct quadrature method of moments
DSMC	direct simulation Monte Carlo
FV	finite volume
ODE	ordinary differential equations
NDF	number density function
PD	product difference
QBMM	quadrature-based moment method
QMOM	quadrature method of moments

INTRODUCTION

The simulation of gas-liquid systems is very important in the process industry since gas-liquid stirred tanks and bubble columns are encountered in many important chemical processes (such as oxidation, hydrogenation, etc.).

The fluid dynamics of these systems has been widely studied with CFD, by resorting to different multi-phase models, notably the multi-fluid approach. This and other approaches require however an a priori assumption on the mean bubble size to be used and in the simulation, and are strongly limited by the assumption that this value is constant throughout the computational domain.

It is well known however that bubbles are distributed over the different characteristic properties (i.e. internal coordinates) such as size, shape, content of chemical components, etc. These distributions, described by a NDF, change due to the relevant phenomena involved, namely bubble convection, bubble growth or shrink, bubble coalescence and break up and mass transfer between bubbles and the continuous liquid phase.

The main limitation of standard multi-fluid models were overcome when QBMM were introduced (Marchiso et al., 2003a, 2003b; Marchisio and Fox, 2005) and implemented in CFD. These methods, implemented in ANSYS/Fluent were recently applied to the simulation of gas liquid systems, focusing on the effect of accounting for size polydispersity with QMOM on fluid dynamics (Petitti et al. 2007; 2009) and on the quantitative validation of model predictions through comparison with experiments for bubble size distributions (Petitti et al., 2010). More recently, DQMOM was applied to the coupled problem of accounting for size and composition polydispersity by using DQMOM (Buffo et al., 2012a). DQMOM performances were compared with those of CQMOM (Buffo et al., 2012b; 2011) and eventually CQMOM was found more suitable for gas-liquid applications.

In this work CQMOM predictions are for the first time compared and validated with predictions obtained with a more accurate and complete method, namely DSMC, on simplified zero-dimensional systems. Then CMOM is implemented in OpenFoam and used to describe a real three-dimensional bubble column for which experiments are available for validation.

MODEL DESCRIPTION

Population Balance Equation

The gas-liquid system under investigation is described through a number density function, defined so that the following quantity:

$$n(t, \mathbf{x}, \phi, L) d\phi dL \qquad (1)$$

represents the number of bubbles per unit volume and at time t with size bounded in between L and $L+dL$ and composition between ϕ and $\phi+d\phi$. Composition is here described in terms of the absolute number of moles of chemical component (i.e. oxygen) contained within the bubble. The evolution of this NDF is described through a PBE that reads as follows:

$$\frac{\partial n}{\partial t} + \nabla \cdot (\mathbf{u}_L n) + \frac{\partial}{\partial L}(Gn) + \frac{\partial}{\partial \phi}(\dot{\phi} n) = h \qquad (2)$$

where $\mathbf{u}_L$ is the velocity of bubbles with size L, G is the continuous rate of change of bubble size due to mass transfer, $\dot{\phi}$ is the mass transfer rate and h is the rate of change of the NDF due to discontinuous events such as coalescence and break up. Standard sub-models were used for all these terms (for details see Buffo et al., 2011; 2012b). The bubble velocity was calculated by using the classical multi-fluid model. So far only drag, lift and virtual mass forces were considered and for drag an empirical correlation based on the apparent swarm bubble terminal velocity was employed. The mass transfer rate was calculated by using the Higbie's penetration model (and consequently the rate of change of bubble size) by taking the average penetration time equal to the Kolmogorov time-scale. Only coalescence and break up induced by turbulent fluctuations were considered. The coalescence efficiency is calculated as the exponential of the ratio of the contact and drainage time-scales, whereas it is assumed that bubble break up results in two fragments with a parabolic size distribution and equal concentration. It is worth mentioning that most of these rates are calculated based on the local value for the turbulent dissipation rate, obtained in turn from the solution of a standard k-ε model for the gas-liquid mixture.

Conditional quadrature method of moments

CQMOM is based on the simple idea of deriving from Eq.(2) transport equations of the mixed moments of the NDF:

$$M_{k,l}(t, \mathbf{x}) = \int_0^{+\infty} \int_0^{+\infty} n(t, \mathbf{x}, L, \phi) L^k \phi^l dL d\phi \qquad (3)$$

The closure problem generated in deriving the moment equation is overcome by using a quadrature approximation, corresponding to the following assumption for the NDF:

$$n(\phi, L) = \sum_{\alpha=1}^{N_1} \sum_{\beta=1}^{N_2} w_\alpha w_{\alpha,\beta} \delta[L - L_\alpha] \delta[\phi - \phi_{\alpha,\beta}] \qquad (4)$$

where the weights and conditional weights w_α and $w_{\alpha,\beta}$ and the nodes L_α and $\phi_{\alpha,\beta}$ are calculated from the moments with an efficient inversion algorithm. Although the original CQMOM was formulated with an adaptive procedure where no priority was fixed for one internal coordinate on the other, in this case size is more important than composition, therefore N_1 always refer to the number of nodes used for size and N_2 for composition. For example, with $N_1 = 3$ six pure moments with respect to size are necessary to calculate L_1, L_2 and L_3 with the standard PD algorithm (i.e., $M_{0,0}$; $M_{1,0}$; $M_{2,0}$; $M_{3,0}$; $M_{4,0}$; $M_{5,0}$). Then, if $N_2 = 1$, composition moments conditioned on L_1, L_2 and L_3 are calculated by inverting a Vandermonde matrix by using $M_{0,0}$, $M_{0,1}$, $M_{1,0}$, $M_{1,1}$, $M_{2,0}$ and $M_{2,1}$ and eventually $\phi_{1,1}$, $\phi_{2,1}$ and $\phi_{3,1}$ are calculated (by using again the PD algorithm).

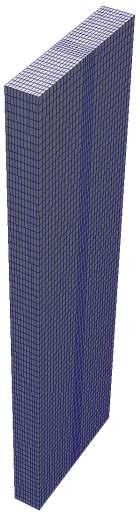

Figure 1: Computational grid of the investigated bubble column. The gas enters the domain (filled with a stagnant liquid) from a rectangular hole in the bottom and exits from the top. The extension of the domain in the three directions is 0.2 × 0.04 × 0.67 m corresponding to a total volume of 0.00536 m^3 and a computational grid of 24640 cells.

TEST CASES AND NUMERICAL DETAILS

As already mentioned the aim of this work is to validate CQMOM performances via comparison with an alternative (more accurate) method and for this purpose DSMC is used.

Two types of simulations were considered. First simple spatially homogeneous systems were considered. These simulations are very useful because they reveal all the interesting local aspects of the investigated phenomena (e.g. coalescence, breakage and mass transfer) and allow for the estimation of the error committed when using CQMOM instead of DSMC (which is considered here as the benchmark solution). Moreover, these simulation can be thought as a representation of a single cell of the computational domain in CFD simulations (since in FV codes each cell of the domain is considered as a perfectly mixed and homogeneous portion of the physical space).

Subsequently a realistic test system was considered. In this work the bubble column experimentally and numerically investigated by Diaz et al. (2008) was simulated. A sketch of the computational domain and of the simulated geometry is reported in Fig. 1.

DSMC simulations were performed with an in-house code, already used for a similar system by Zucca et al. (2007). A detailed description of the algorithm can be found elsewhere (Liffman, 1992) but here we stress only that the number of notional bubbles undergoing randomly selected events was large enough (greater than 10,000) to ensure the statistical reliability of the results.
CQMOM for a homogeneous system results instead in a system of ODE solved here with the standard Matlab integrator ode15s. CQMOM was solved by using a variable number of nodes for the quadrature approximation (from two to four) in order to assess the accuracy of the approximation.
In all cases the studied system was composed by pure water and air, as continuous and disperse phases respectively. Standard physical properties for these phases were considered.
All the simulation were initialized by prescribing the presence of bubbles inside the control volume. The initial bubble population was described as a log-normal distribution with respect to the size with a mean of 3.7 mm and a standard deviation of 15% of the mean value, but with the same oxygen concentration of 8.56 mol m^{-3} in all bubbles, corresponding to the concentration of oxygen in air at 25 ° C and 1 atm. The described initial condition had to be prescribed in the form of initial moments for CQMOM and these values were obtained by the initialization of the DSMC run.
CQMOM was implemented in the CFD code openFoam for the simulation of the bubble column reported in Fig. 1. The implementation in OpenFoam was done in the `compressibleTwoPhaseEulerFoam` solver.

RESULTS AND DISCUSSION

Spatially homogeneous simulations

Figure 2 reports the bi-variate NDF used as initial condition and as boundary (inlet) condition for the homogeneous simulations and the three-dimensional CFD simulations. This NDF represents a population of bubbles characterized by a log-normal bubble size distribution centred at 3.7 mm and by a standard deviation of 15 %. The bubbles are moreover characterized by identical oxygen concentration (and therefore with oxygen content proportional to the volume of the bubble). This condition well represents bubbles exiting a sparger and entering a bubble column.
It is useful to stress here that although the two internal coordinates considered here are bubble size and bubble oxygen content, the representation is done here in terms of bubble size and *bubble oxygen concentration*, since this is easier to understand. Figure 2 reports the bubble size distribution (top), the oxygen concentration distribution (right) and the NDF representation in the bubble size/oxygen concentration plane.

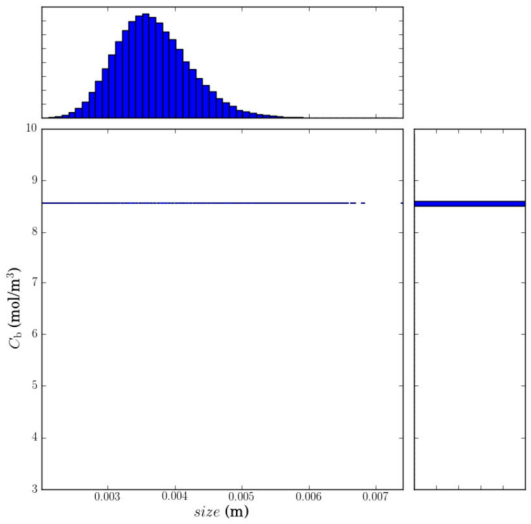

Figure 2: Initial condition for the NDF: log-normal distribution with respect to the size with a mean of 3.7 mm and a standard deviation of 15% of the mean value and with the same concentration of 8.56 mol/m^3 in all bubbles (oxygen in air at 25° C and 1 atm).

Let us now consider the results of a typical DSMC run for the spatially homogeneous case. For the system under investigation bubbles enter with the NDF reported in Fig. 2, break up and coalescence (according to standard kernels) and exchange mass with the liquid, which is initially completely lacking oxygen. For this specific case the mean residence of the bubble was fixed equal to 1 s. As the simulation goes by, bubbles change their size and oxygen content distributions, until the liquid is saturated by oxygen and the bubbles do not change their concentration any more.
Figure 3 shows the NDF during the transient when the oxygen has not yet fully saturated the liquid. As it is seen the action of coalescence and break up is to change the bubble size distribution. In this particular case coalescence prevails over break up and larger bubbles are formed. In addition to that the bubble size distribution becomes wider and wider, when compared to the initial log-normal distribution. As demonstrated by our simulations (not shown here) and as expected, the effect of coalescence and break up is to homogenize the oxygen concentration of the bubbles. In fact, by letting bubbles of different concentrations coalescence and then subsequently break up, forming fragments with the same oxygen concentration, the variance with respect to bubble oxygen concentration is reduced.
The effect of mass transfer is instead to reduce the oxygen concentration in the bubbles and to increase this variance. In fact, as it is easily detectable in the figure, smaller bubbles (having higher surface-to-volume ratios and mass transfer coefficients) exchange mass faster than larger bubbles. As it is well known, this effect can be enhanced by a chemical reaction, that can accelerate mass transfer resulting in even spreader distributions.
Also the effect of convection, namely in this spatially homogeneous system, the effect of introducing bubbles with the inlet NDF and of mixing them with the bubbles within the domain, is to increase this oxygen variance.
This results in the NDF reported in Fig. 3, that as can be easily seen it quite spread for both bubble size and

composition. It is worth mentioning here that the number of notional bubbles used in our DSMC is such that the result reported in Fig. 3 can be considered almost "exact": it is in fact a very accurate solution of the PBE with this combination of kernels and mass transfer rate.

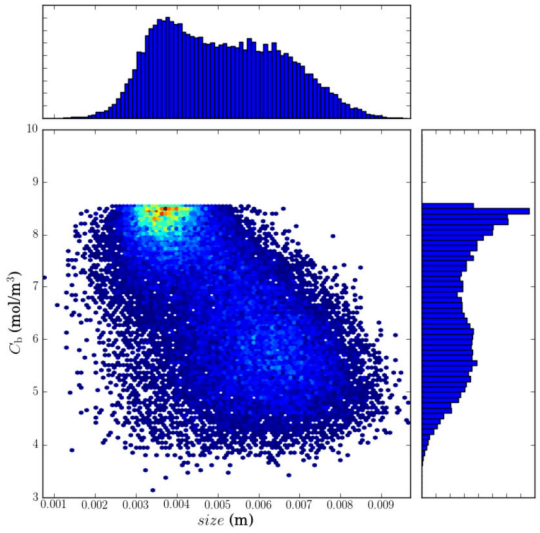

Figure 3: Snapshot of the bubble population after 2.5 s. In the main plot, different colors are used to represent density of bubbles in ascending order from blue to red

It is important to point out here that the NDF presents non-null variances with respect to both internal coordinates and that no algebraic relationship exists between the two variables. In fact, bubbles with the same size can have different concentrations and *viceversa*. This implies that a bivariate PBE is necessary for the accurate description of this gas-liquid system, as accounting only for the size distribution would lead to large errors (especially when non-linear terms due to chemical reactions appears in the governing equations!)
Figure 4 reports instead the very same information when the transient dies out. In fact, after the transient period in which oxygen goes from the bubbles to the liquid, when the liquid is saturated with oxygen, bubbles enter and exit from the domain without exchanging oxygen and the concentration do not vary anymore.
One should notice that coalescence and breakage still have an impact of bubble size distribution (represented on top) due to the fast time scales of these two phenomena. It is important to stress here that this steady-state with uniform bubble oxygen concentration is possible here only because no chemical reactions are present. If for example, oxygen in the liquid were consumed by a chemical reactions the steady-state would resemble the NDF reported in Fig. 3, as demonstrated by our simulations (not shown).
As already mentioned we are interested in describing this system with CQMOM and therefore it is important to discuss how the NDF is represented (or reconstructed) with this methods.

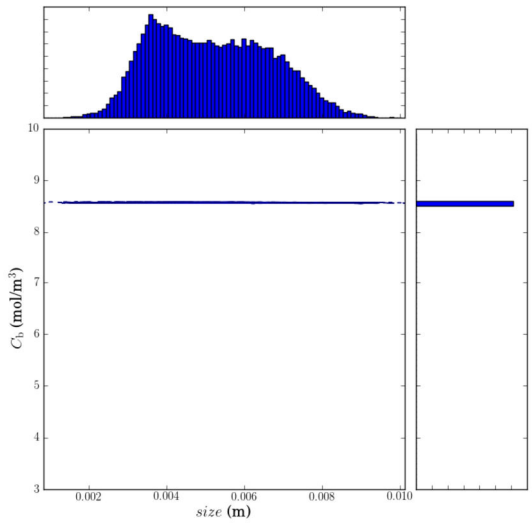

Figure 4: Snapshot of the bubble population after 30s, when the steady state is reached.

A schematic representation is reported in Fig. 5. As it is seen the population of bubbles is presented by a limited (six in this case) number of bubble classes. These bubble classes are characterized by certain nodes, namely some size and oxygen concentration values, and by certain weights, namely the number density of bubbles with these node values. This corresponds to the mathematical representation of the NDF as a summation of (six) Dirac delta functions, as reported in Eq. (4).
Once could think that this representation is very crude and that therefore the evolution of the population of bubbles would be poorly predicted by CQMOM. However, since these bubble classes are placed on nodes of quadrature approximations, the accuracy of the method turns out to be very high.
This is demonstrated in Fig. 6 where the evolution of some mixed moments of the NDF as predicted by DSMC and CQMOM is compared. DSMC predictions are affected by a small statistical noise (black line) whereas CQMOM being a fully deterministic method does not contain any statistical noise (red line).
The moments here reported represent important physical properties of the bubbles. For example $M_{0,0}$ represents the total number bubble density, whereas $M_{2,0}$ is related via the surface shape factor) to the specific surface area of the bubbles. $M_{3,0}$ is related (via the volume shape factor) to the gas bubble volume fraction and $M_{0,1}$ represents to total oxygen (within the bubbles) per unit volume of the multiphase systems. The average oxygen bubble concentration can be calculated from the ratio between $M_{0,1}$ and $M_{3,0}$.

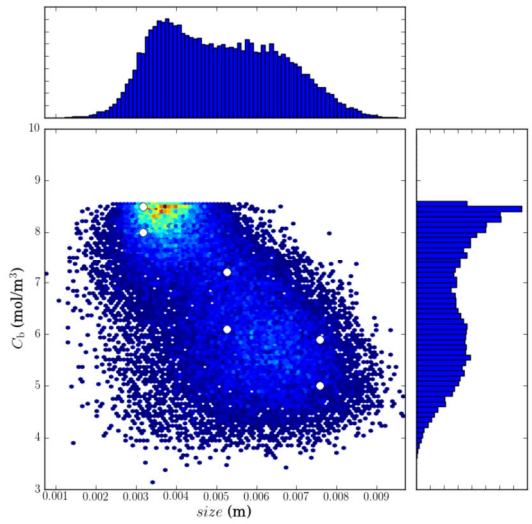

Figure 5: Comparison of the representation of the bubble population with DSMC (colored circles) and CQMOM (white circles).

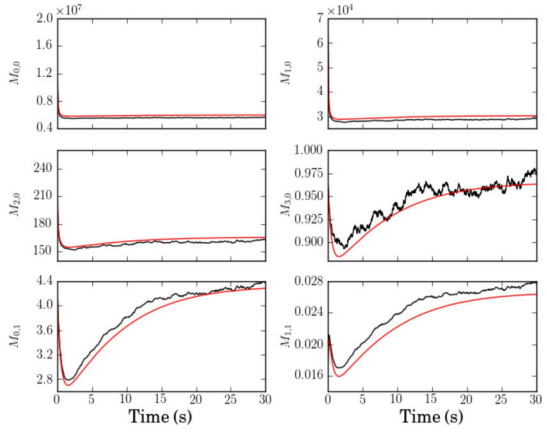

Figure 6: Comparison between Monte Carlo (black line) and CQMOM (red line) simulation in terms of time evolution of certain moments of the bubble population.

The evolution of the moments reveals the prevailing of coalescence over break up (i.e. $M_{0,0}$ decreases), the presence of mass transfer, that causes first an oxygen depletion in the gas phase, resulting in a reduction of both gas volume fraction (i.e. $M_{3,0}$) and total oxygen concentration (i.e. $M_{0,1}$), followed by an increase when the liquid is saturated.

As far as the comparison is concerned it is interesting to notice that CQMOM and DSMC results in very similar predictions. Errors calculated with DSMC (with multiple repetitions to reduce the statistical noise) as a reference resulted in overall integral errors of 5-15 % when using bivariate quadrature approximations with a number of nodes ranging from two to eight (N_1 = 2, 3, 4 and N_2 = 1, 2).

These very small errors come with the great advantage of reducing significantly the computational time. In fact, if in DSMC it is required to track the evolution of 10000 notational bubbles for 10 realizations, resulting in a CPU time (serial simulation) of two days (on a standard Intel Xeon X5650 2.67GHz machine), CQMOM requires the solution of a small moment set (from six to 20 for the abovementioned cases), resulting in CPU time of seconds.

One last important issue concerns the so-called quadrature realizability. In fact, the quadrature approximation is reconstructed from the moments with specific algorithms and for bivariate problems there is no assurance that the nodes are realizable, meaning that they assume allowed and reasonable values (and therefore they sample sensible regions of the internal coordinate space).

One trivial requirement is that both bubble size and concentration nodes are positive. Additional constraints refer to the necessity of respecting physical behaviours. For example, in the case of no mass transfer and uniform inlet bubble oxygen concentration, the nodes for oxygen context (or concentration) have to stay constant, since no oxygen is transferring from one phase to the other.

Our simulations show that this additional condition is respected only when $M_{2,1}$ is included in the moment set, therefore at least three nodes for size (N_1 = 3) must be used.

Spatially (three-dimensional) inhomogeneous simulations

As already mentioned CQMOM was also implemented in OpenFoam (`compressibleTwoPhaseEulerFoam` solver) to simulated the bubble column experimentally investigated by Diaz et al. (2008). Figure 7 reports the contour plot of the gas volume fraction at two different instants. As it is seen the bubble plume oscillates, due to fluid dynamic instabilities. It is interesting to observe that in our simulations this oscillation starts after about 10 seconds from the beginning of the simulation, in accordance with what reported in the work of Diaz et al. (2008). Preliminary results show that the frequency of these oscillations (at different operating conditions) is in accordance with experiments.

As already reported CQMOM allows to simulate the evolution of the NDF describing the polydispersity of the bubbles both in terms of their size and oxygen content.

Figure 8 reports (for a simulation under the absence of mass transfer) a snapshot taken from the evolution of the mean bubble size, calculated as the ratio between $M_{3,0}$ and $M_{2,0}$ (i.e. Sauter bubble diameter).

As it is seen, as soon as bubbles enter the domain, they are broken up by the sudden acceleration due to the buoyancy force. Then coalescence occurs in the upper part of the column on the wake of the plume, where bubbles tend to accumulate.

Moment transport equations are solved within OpenFoam with first-order upwind spatial discretization schemes. In fact, the use of standard higher-order schemes corrupt the moment set hindering the convergence of the simulation.

CONCLUSION

In this work CQMOM and DSMC predictions are compared and results show that CQMOM is an accurate tool for the simulation of bivariate PBE for gas-liquid systems. In particular, extensive validation carried out with realistic kernels and mass transfer rates, have shown that the quadrature formula well approximate the moment source terms. Particular attention should be paid on the moment set to be tracked, since only when some moments are included in the moment set the resulting quadrature is realizable. CQMOM was successfully implemented in opeanFoam, allowing the investigation of a bubble

column for which experiments are available for model validation. The next steps of our work involves the extensive validation of with experiments and the implementation of hybrid higher-order schemes that preserve the validity of the moment set.

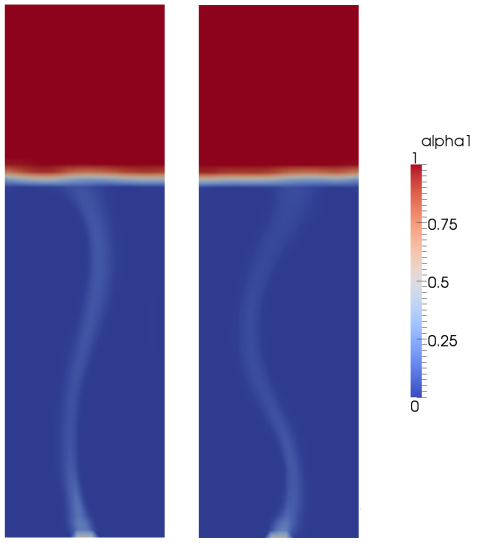

Figure 7: Contour plots of the gas volume fraction in the investigated bubble column at 20 and 25 seconds.

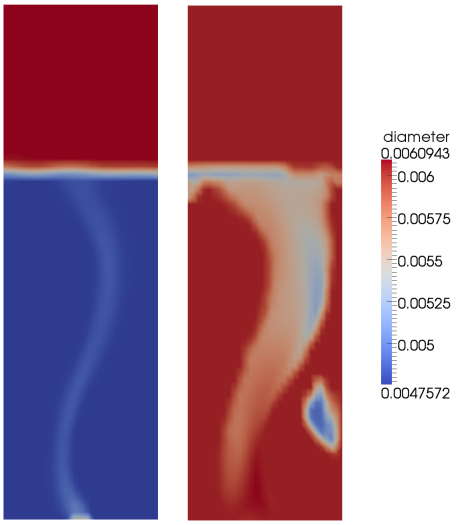

Figure 8: Preliminary result for the mean bubble size (m) on the right, together with the gas volume fraction on the left, at 30 seconds from the beginning of the simulation.

ACKNOWLEDGMENTS

The research project is partially funded by BASF SE. The authors are also grateful to Rodney O. Fox and Miriam Petitti who contributed in different ways to this work.

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