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Assessment of Micro- and Mesomixing in Bubble Swarms via Simulations

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Mixing and fast chemical reactions in sparged stirred tanks are of great importance in the chemical industry. If multiple reaction steps occur within one apparatus, a detailed understanding of local mixing is crucial to precisely control the reaction network. However, a single, rigorous framework describing the interaction between reactions, mass transfer and liquid phase mixing is currently not available in the literature. This is mainly because experimental methods as well as numerical simulations are extremely demanding, especially with respect to micro- and mesomixing in bubble swarms. For example, fully resolved 3D simulations of reacting bubble swarms with complex reactions are still challenging. Hence, our previous work (Radl *et al.*, 2008, Radl *et al.*, 2007, Radl and Khinast, 2007) was based on 2D direct numerical simulations of bubbly flows, and three-dimensional effects were ignored completely.
In this work it is our intension to assess micro- and mesomixing effects by using high-fidelity numerical simulations. We use different simulation approaches that allow us to simulate bubbly flows as well as scalar transport in high detail. Our work includes direct numerical simulation of bubble wake phenomena and large eddy simulations combined with Lagrangian bubble tracking to study bubble plume effects. The major achievement presented in this work is the use different concepts for assessing mixing quality in complex gas-liquid flows. Hence, we are one of the first groups that is able to shed some light on the interaction of multiphase flow and local mixing effects.

1. Introduction

Bubbly flows are encountered in a wide range of industrial branches such as the fine chemical, pharmaceutical or petrochemical industry. In all of these branches the correct prediction of mixing in the liquid phase is of high importance. Often the quantification

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of micromixing (i.e., mixing on the finest scales), as well as mesomixing (i.e., mixing on intermediate scales) is of significant industrial relevance.
During the last years, several researchers have analyzed mixing and chemical reactions in multiphase systems. Starting with the early work on non-deformable two-dimensional bubbles (Khinast, 2001), we have refined our methodology within the last years. We are now able to study fully deformable bubble swarms in complex fluids involving arbitrary chemical reactions (Radl *et al.*, 2008, Radl *et al.*, 2007, Radl and Khinast, 2007). In the meantime, other researchers have also made significant progress in the field of multiphase mixing simulation. Some examples include the study of mass transfer from three-dimensional bubbles rising in a stagnant fluid (Darmana, 2006) as well as recent advances for mixing by granular particles (Derksen, 2008). However, a detailed analysis of mixing including mass transfer has not been carried out by these groups. In this paper we highlight a numerical methodology for gaining more insight into this topic.

2. Numerical Method

2.1 Direct Numerical Simulation

In this work we have used a laminar flow solver together with a species transport solver to study mass transfer from a rigid bubble. Hence, we solved the Navier-Stokes equations for an incompressible fluid:

$$\frac{\partial \boldsymbol{u}}{\partial t} + \nabla \cdot (\boldsymbol{uu}) = -\nabla p' + \nu \cdot \nabla^2 \boldsymbol{u} \tag{1}$$

$$\nabla \cdot \boldsymbol{u} = 0 \tag{2}$$

Here **u** denotes the fluid velocity, p' the kinematic pressure (i.e., the pressure divided by the fluid density), and ν is the kinematic viscosity of the fluid. The transport equation for an inert species Y is:

$$\frac{\partial Y}{\partial t} + \nabla \cdot (\boldsymbol{u} Y) = +D \cdot \nabla^2 Y \tag{3}$$

Here D denotes the diffusion coefficient of species Y in the liquid. As we use high-order schemes and a sufficiently fine numerical grid, we fully resolve all details of fluid motion as well as the thin concentration boundary layer near the bubble interface. We therefore refer to these simulations as direct numerical simulations. This methodology follows the ideas of our previous work for two-dimensional elliptical bubbles (Khinast, 2001) and the other literature available in that field (Jung and Sato, 2005). However, our focus here is to analyze mixing in greater detail, which has not been done before.
The geometrical setup for our simulations consists of a spherical bubble that was placed in a sufficiently large cylindrical fluid domain. Appropriate boundary conditions were applied and the set of equations was solved using OpenFOAM (OpenCFD Ltd., 2008).

2.2 Euler-Lagrange Approach

The second approach used in this work refers to a simulation method where the equation of motion for the continuous phase (i.e., the liquid phase) is solved on an Eulerian frame of reference. The disperse phase, i.e., suspended bubbles, is tracked explicitly within a Lagrangian frame of reference. We refer to this approach as the Euler-Lagrange approach (Delnoij *et al.*, 1997).

In our work, we have implemented and tested the Euler-Lagrange approach in the open-source CFD-package "OpenFOAM" (OpenCFD Ltd., 2008) to study liquid-phase mixing. We have included most of the recent developments that can be found in literature including:

- a combined strategy for Lagrangian-to-Euler and Euler-to-Lagrangian mapping inspired by recent literature (Deen *et al.*, 2004).
- We take the volume of the dispersed bubbles into account, i.e., we include the local liquid-phase void fraction in our equations.
- We can use a wide range of sub-grid-scale models for an accurate modeling of the turbulent flow of the liquid phase via Large Eddy Simulation (LES).
- We take all relevant forces between bubbles and liquid, as well as bubbles and the walls into account, as well as
- perform a full two-way coupling between the disperse and the continuous phase for both mass and momentum transfer.

The equations that are solved for the continuous phase are:

$$\frac{\partial \varepsilon_L}{\partial t} + \nabla \cdot \left(\varepsilon_L \cdot \overline{\boldsymbol{u}}_L \right) = 0 \tag{4}$$

$$\frac{\partial \overline{\boldsymbol{u}}_L}{\partial t} + \nabla \cdot \left(\overline{\boldsymbol{u}}_L \overline{\boldsymbol{u}}_L \right) = -\nabla \overline{p}' + \frac{1}{\varepsilon_L} \cdot \nabla \cdot \left[\varepsilon_L \cdot \nu_{eff} \cdot \left(\nabla \overline{\boldsymbol{u}}_L + \nabla \overline{\boldsymbol{u}}_L{}^T - \frac{2}{3} \cdot \boldsymbol{I} \cdot \left(\nabla \cdot \overline{\boldsymbol{u}}_L \right) \right) \right] + \frac{\boldsymbol{\Phi}_L}{\varepsilon_L \cdot \rho_L} \tag{5}$$

This is the filtered Navier-Stokes equation in the variables $\overline{\boldsymbol{u}}_L$ and $\nabla \overline{p}'$ which are the filtered velocity and the filtered kinematic pressure, i.e., the pressure divided by the liquid-phase density, respectively. The liquid-phase void fraction ε_L and the gas-liquid interaction forces $\boldsymbol{\Phi}_L$ are obtained by mapping from the dispersed phase directly. We use the Smagorinsky model with the Smagorinsky constant equal to $C_S = 0.032$ to model sub-grid-scale turbulent motion (Hu and Celik, 2008). A universal wall function (de Villiers, 2006) is used to calculate the turbulent viscosity at the wall.

The species conservation equation for species i in the liquid phase is:

$$\frac{\partial Y_i}{\partial t} + \nabla \cdot \left(\overline{\boldsymbol{u}} Y_i \right) = \nabla \cdot \left(D_{eff,i} \cdot \nabla Y_i \right) + \Phi_{\dot{N}_i} \tag{6}$$

$$D_{eff,i} = D_i + \frac{\nu_t}{Sc_t} \tag{7}$$

Here D_i is the molecular diffusion coefficient, ν_t is the turbulent viscosity and Sc_t is the turbulent Schmidt number. In our simulations we use $Sc_t = 0.7$ (Tominaga and Stathopoulos, 2007). $\Phi_{\dot{N}_i}$ is the species source term from the disperse phase. For the disperse phase, i.e., the bubbles, we solve Newton's equation of motion for each bubble:

$$m_p \cdot \frac{d\boldsymbol{U}}{dt} = \boldsymbol{F}_G + \boldsymbol{F}_p + \boldsymbol{F}_D + \boldsymbol{F}_L + \boldsymbol{F}_A \tag{8}$$

Here m_p is the mass of the bubble and $\boldsymbol{U}$ is the velocity vector of the bubble. F_G, F_p, F_D, F_L and F_A are the gravity force, the pressure gradient force, the drag force, the lift force and the added mass force, respectively. The models and parameters for these interaction forces follow the work of Hu and Celik, 2008, except for the fluid stress term. In our work we consider the stress as a pressure gradient term (in analogy to Delnoij *et al.*, 1997). The lift and added mass coefficient were chosen as C_L=0.53 and C_A=0.5,

respectively. The standard drag curve was used (refer to Hu and Celik, 2008 for details) as well as the standard Sherwood relation (Bird *et al.*, 2002):

$$Sh = 2 + 0.6415 \cdot \sqrt{Re \cdot Sc} \tag{9}$$

Bubble-wall collisions were treated as inelastic collisions with a selectable elasticity. A sophisticated particle tracking routine was used in our work (Kaerrholm, 2008). The "Becker" case (Hu and Celik, 2008) was selected in this work as a prototype for illustrating flow and mixing in bubble swarms.

2.3 Quantification of Mixing

Mixing in the system was analyzed using metrics that characterize the intensity as well as the scale of segregation. Hence, we have computed the intensity of segregation as

$$\sigma^2 = \frac{1}{V} \cdot \int_V \left(Y_i - \overline{Y}_i\right)^2 dV \tag{10}$$

and the scale of segregation (introduced by Bothe *et al.*, 2006) as

$$\Phi = \frac{1}{V} \cdot \int_V \left|\nabla Y_i\right| dV \cdot \tag{11}$$

We have also analyzed the local distribution of these metrics.

3. Results

3.1 Micro-Scale Flow and Micromixing

The results of one of our direct numerical simulation are illustrated in Figure 1 for two different instances in time. As can be seen, the metric Φ has a pronounced maximum at the interface with a complex distribution in the wake of the bubble.

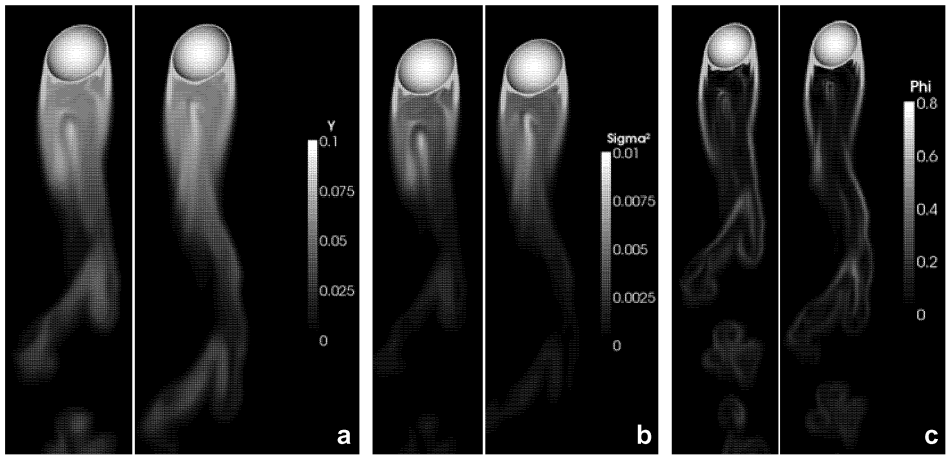

Figure 1: Distribution of (a) the inert scalar, (b) the intensity of segregation and (c) scale of segregation for the flow past a spherical bubble (Re = 1000, Sc =10)

3.2 Results for the Euler-Lagrange Approach

To validate the results of our LES/Euler-Lagrange simulations, we performed a grid sensitivity study as well as compared the results to experimental data (taken from

Sokolichin and Eigenberger, 1999). We quantitatively compare the mean vertical velocity profiles at a relative height of z/h=0.35 for three different mesh configurations (Mesh 0: 122,880 cells; Mesh 1: 269,750 cells; Mesh 2: 820,800 cells). As can be seen from Figure 2, all grids gave excellent agreement with experimental data when averaging was performed over a sufficiently long time interval.

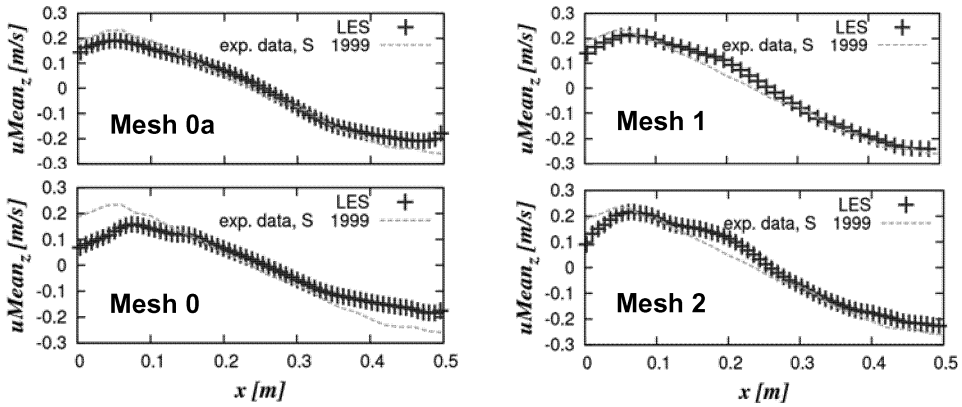

Figure 2: Comparison of mean vertical velocity profiles at z/H=0.35 for three different grids averaged over 35 [s]. The data averaged over a substantial longer time interval of 100 [s] for Mesh 0 is indicated as "Mesh 0a".

3.3 Mesomixing

Selected results of our Euler-Lagrange simulations are shown in Figure 3. The interesting result is that the intensity of segregation σ^2 (Figure 3, right) has a pronounced peak after approx. 25[s]. Also, the distribution of Φ is complex and shifts position during time. These results will be important for simplified modeling of bubble columns and stirred reactors.

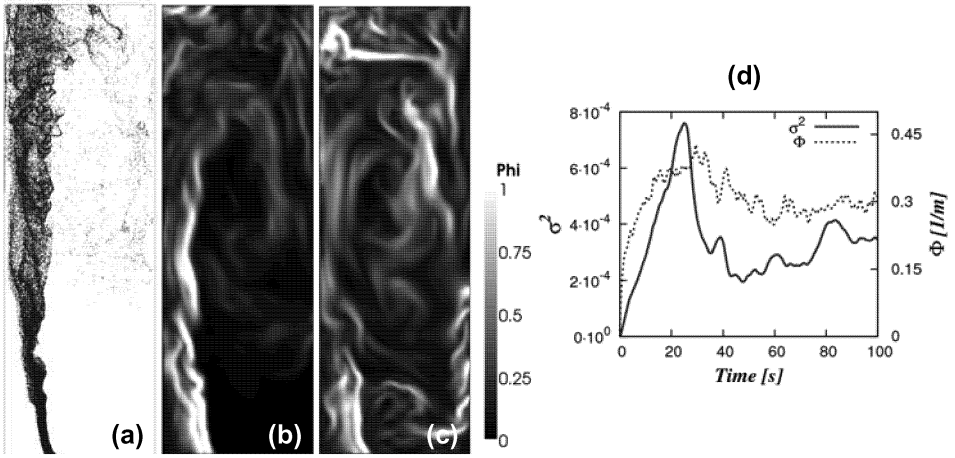

Figure 3: (a) distribution of bubbles after 5[s]; distribution of the scale of segregation Φ after (b) 5[s] and (c) 40[s]; (d) intensity and scale of segregation vs. time.

4. Acknowledgement

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