FAST REACTIONS IN BUBBLY FLOWS: FILM MODEL AND MICRO-MIXING EFFECTS

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Summary
In our work we focus on the Euler-Lagrange strategy to simulate bubbly flows and fast chemical reactions. We use a film model to account for fast, multi-step chemical reactions near the gas-liquid interface. A combination of such a film model with Euler-Lagrange simulations for bubbly flows has not been used before. Also, we account for micro-mixing effects with respect to chemical reactions in the liquid bulk by solving for the variance of the concentration field. To validate our model predictions for multiphase flow and reactive mixing, we compare our results with literature data and find excellent agreement.

Keywords
Multiphase and particulate reactors, multiscale analysis, dynamics and control of chemical reacting systems

Introduction
Reactive multiphase flows can be found in various applications in the chemical, pharmaceutical and biotechnology industries. Typical examples include catalytic hydrogenations, oxidations, or the aeration of bioreactors. However, a detailed mathematical description of these flows is still challenging, due to the interaction of mass transfer, mixing and (bio-)chemical reactions. All these factors might dominate the yield, selectivity or productivity of the reactor, which complicates the design and optimization of such processes. Clearly, a profound understanding of these influence factors is currently not available. In order to study reactive multiphase flows in detail, experimental and numerical methods have been used in the past. The currently available computational approaches include: (a) a full resolution of all scales with direct numerical simulations (DNS, i.e., first-principles simulations), (b) the use of a filtered set of the momentum equations via so-called Large Eddy Simulations (LES), and (c) the use of the ensemble-averaged Navier-Stokes equations, either steady or unsteady (Reynolds-Averaged-Navier-Stokes approach, RANS). To gain a detailed insight into reactions happening in bubbly flows, we use LES in our work. This strategy allows us to analyze complex reaction networks without performing extensive experimental studies and by keeping the level of modeling relatively low. Thus, this work is one of the first applications of multiscale modeling for reactive mixing in bubbly flows based on the LES approach.

Computational Model
The Euler-Lagrange (EL) approach is used in this work and 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 dispersed phase is tracked explicitly within a Lagrangian frame of reference and Newton’s equation of motion is solved for each individual particle, i.e., bubble. In our work, we have implemented a EL-LES code into the open-source CFD-package “OpenFOAM”.3 The details of our implementation are presented in Radl and Khinast.4 To account for fast chemical reactions near the gas-liquid interface is extremely difficult. This is due to the interaction of mass transfer, multiphase flow and reactions. Furthermore, steep concentration gradients develop near the interface due to the typically high Schmidt numbers of the dissolving species in the liquid phase. Hence, a simplified model for the reaction-convection-diffusion problem near the gas-liquid interface has to be used. For example, the film or penetration theory can be used for this purpose. In our work we use a similar approach as Kenig und Gorak5 together with our detailed LES code. Although the approach of Kenig and Gorak6 is in use for several years, it has not been combined with detailed simulations of the continuous phase.

To analyze inert sub-grid-scale mixing in our simulations, we solve the transport equation of the variance $\nabla Y_i$ of the scalar $Y_i$ (Eqn. 1). This equation is similar to the one presented by Jaberi et al.7 as well as Colucci et al.8 but in

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addition accounts for the liquid-phase hold-up $\varepsilon_L$ as well as for sources due to mass transfer (i.e., the last term on the RHS of Eqn. 1). The SGS scalar dissipation rate $\overline{\gamma}_L$ has been modeled using the IEM (interaction by exchange with the mean) closure as suggested by Colucci et al. $^9$ Also, we consider the source of variance due to the transferred mass with a simple model involving an adjustable parameter $\alpha$.

$$
\frac{\partial (\varepsilon_L \cdot Y_{eff,i})}{\partial t} + \nabla \cdot (\varepsilon_L \cdot \mathbf{u} Y_{eff,i}) = 
$$

$$
\nabla \cdot (\varepsilon_L \cdot \mathbf{D}_{eff,i} \cdot \nabla Y_{eff,i}) 
$$

$$
+ 2 \cdot \varepsilon_L \cdot \mathbf{D}_{eff,i} \cdot \nabla \nabla Y_{eff,i} 
$$

$$
- 2 \cdot \varepsilon_L \cdot \overline{\gamma}_L 
$$

$$
+ 2 \cdot \alpha \cdot Y_{eff,i} \cdot \frac{\Phi_{L}}{Y_{eff,i}} 
$$

In order to account for fast reactions in the liquid phase, we use a presumed PDF method similar to Marchisio $^{10}$ for our LES. Thus, we are able to compute also the outcome of fast chemical reactions by (indirectly) solving the transport equations for the mixture fraction and a reaction progress variable.

**Results**

Our results for the mean liquid-phase flow field in a flat bubble column agree very well with the experimental data of Sokolichin and Eigenberger. $^{11}$ By taking mass transfer into account, we are also able to visualize the concentration field of a dissolving (inert) species (e.g., oxygen) in a bubble column. This has been done by plotting both the filtered concentration as well as the scalar variance field in Figure 1 for two different time instances. Both plots have been made dimensionless with the saturation concentration and the squared saturation concentration, respectively. As can be seen, both, the filtered concentration field as well as the variance field show similar features, i.e., extremely high values near the inlet and at the upper part of the column.

For analyzing fast chemical reactions, we investigated the outcome of a parallel chemical reaction $^{12}$ in a simple micro-mixing device. We compare our findings with the results of Marchisio $^{13}$ for a similar impinging jet reactor and find good agreement.

**References**


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**Figure 1**: Dimensionless concentration (a1, a2) and scalar variance (b1, b2) contour plots after 2 [s] (left) and 10 [s] (right) for an inert scalar.