# A VOF-Based Conservative Method for the Simulation of Reactive Mass Transfer from Rising Bubbles

# **D.** Bothe<sup>1,2</sup>, **M.** Kröger<sup>1</sup> and H.-J. Warnecke<sup>3</sup>

**Abstract:** In this paper numerical results on reactive mass transfer from single gas bubbles to a surrounding liquid are presented. The underlying numerical method is based on the solution of the incompressible two-phase Navier-Stokes equations. The Volume-of-Fluid method is applied for the description of the liquidgas interface. Within the numerical approach the concentration of the transfer component is represented by two separate variables, one for each phase. Numerical results are in good agreement with experimental data.

### 1 Introduction

Many chemical reactions are heterogeneous, in which case the reaction partners are present in different phases, e.g. in a liquid and in a gas. In order to enable a chemical reaction, at least one of these species has to cross the interface separating the two phases. This process is called reactive mass transfer, if the physical transfer of a chemical component from one phase to the other is followed by a chemical reaction in this phase. A common example is the selective oxidation of cyclohexane where the gaseous oxygen is first dissolved and then reacts with the organic liquid to the desired product. This type of reaction is of high interest, especially for the chemical process industry. The scale-up from laboratory sized model apparatuses to large industrial reactors often turns out to be extremly difficult, in particular because several of the occurring phenomena in such systems are not fully understood. Direct numerical simulations have proven to be a useful addition to small scale experiments and theoretical analysis. Accurate simulations can give insight to local details which are otherwise not achievable. Recently, a lot of work has been devoted to this topic.

VOF-based simulations of purely physical mass transfer across deforming interfaces without chemical reaction have been reported in [Davidson and Rudman

<sup>&</sup>lt;sup>1</sup> Center of Smart Interfaces, Technische Universität Darmstadt, Darmstadt, Germany

<sup>&</sup>lt;sup>2</sup> Corresponding author: bothe@csi.tu-darmstadt.de

<sup>&</sup>lt;sup>3</sup> Department of Chemical Engineering, University Paderborn, Germany

(2002)] and in [Bothe, Koebe, Wielage, Prüss, and Warnecke (2004)]. In the latter paper, transfer of oxygen from air bubbles rising in water or aqueous solutions has been simulated, taking into account the realistic jump discontinuity of the oxygen profiles at the interface. [Onea, Wörner, and Cacuci (2009)] used a simular approach as [Bothe, Koebe, Wielage, Prüss, and Warnecke (2004)] to simulate mass transfer in upward bubble train flow through square and rectangular mini-channels. [Darmana, Deen, and Kuipers (2006)] performed 3D simulations of mass transfer at rising fluid particles using the Front Tracking method. There, the transport resistance inside the fluid particle is neglected, i.e. a constant concentration value inside the bubble is assumed. [Radl, Tryggvason, and Khinast (2007)] performed 2D simulations of deformable bubbles and bubble swarms with mass transfer in non-Newtonian liquids using a semi-Lagrangian advection scheme. To prevent stability problems, a reduced density ratio between gas and liquid is used.

Recently, first papers on numerical simulation of reactive mass transfer appeared. In [Khinast, Koynov, and Tryggvason (2005)], reactive mass transfer at deformable interfaces is examined using a 2D Front Tracking/Front Capturing hybrid method. In [Deshpande and Zimmermann (2006a)], a Level Set based method is used to simulate mass transfer across the interface of a moving deformable droplet. This method is extended to reactive mass transfer in [Deshpande and Zimmermann (2006b)], where an instantaneous chemical reaction occurs inside a moving droplet which leads to a quasi-stationary problem for the mass transfer. In [Radl, Koynov, Tryggvason, and Khinast (2008)], 2D simulations are performed using a Front Tracking method to investigate the effect of different Hatta and Schmidt numbers on the catalytic hydrogenation of nitroarenes for single bubbles and bubble clusters. Based on the numerical approach in [Bothe, Koebe, Wielage, Prüss, and Warnecke (2004)], reactive mass transfer with parallel consecutive reactions at rising bubbles has been analyzed in [Bothe, Kröger, Alke, and Warnecke (2009)] by using local selectivities. The approach from [Bothe, Koebe, Wielage, Prüss, and Warnecke (2004)] employs a single scalar field for any transfer component and this scalar quantity refers to a normalized concentration for which the jump discontinuity can be removed. This has some disadvantages concerning conservativity of the transfer component which can lead to artificial mass transfer. The latter is caused by a relative motion of the interface and the concentration discontinuity due to the use of different advection algorithms. In [Alke, Bothe, Kroeger, and Warnecke (2009)] a new VOF-based approach has been introduced which does not posses these drawbacks. In the present paper, this approach is applied to reactive mass transfer from single rising bubbles.

### 2 Governing Equations

In the mathematical model it is assumed that the system under consideration consists of two immiscible, incompressible fluids, separated through a moving and deformable interface. The interface between the two phases is presented as a mathematical (sharp) surface of zero thickness and is denoted by  $\Sigma(t)$ . The local balances for mass and momentum can then be written as

$$\nabla \cdot \mathbf{u} = 0 \tag{1}$$

for mass and

$$\rho \,\frac{\partial \mathbf{u}}{\partial t} + \rho \,(\mathbf{u} \cdot \nabla) \,\mathbf{u} = -\nabla p + \rho \,\mathbf{g} + \nabla \cdot \mathbf{S} \tag{2}$$

for the momentum. In this equation, S denotes the viscous stress tensor given by

$$\mathbf{S} = \boldsymbol{\eta} (\nabla \mathbf{u} + (\nabla \mathbf{u})^{\mathsf{T}}). \tag{3}$$

The interface normal unit vector  $\mathbf{n}_{\Sigma}$  points into the continuous phase for the remainder of this paper. This set of equations represents the one-field formulation of the two phase Navier-Stokes equations where the material properties  $\rho$  and  $\eta$  refer to the phase dependent density and viscosity. Additionally, the following jump conditions are satisfied at the interface:

$$[\mathbf{u}] = 0 \tag{4}$$

and

$$[p\mathbf{I} - \mathbf{S}] \cdot \mathbf{n}_{\Sigma} = \sigma \kappa \mathbf{n}_{\Sigma},\tag{5}$$

where  $\kappa = -\nabla \cdot \mathbf{n}_{\Sigma}$  is the curvature (more precisely, the sum of the principal curvatures) and expresses the Laplace pressure jump in stationary cases. Note that the square bracket stands for the interfacial jump according to

$$[\phi](\mathbf{x}_{\Sigma}) = \lim_{h \to 0+} \left( \phi \left( \mathbf{x}_{\Sigma} + h\mathbf{n}_{\Sigma} \right) - \phi \left( \mathbf{x}_{\Sigma} - h\mathbf{n}_{\Sigma} \right) \right)$$
(6)

### where $\phi$ is an arbitrary quantity

It is further assumed that there is at least one transfer component with the volumetric molar concentration  $c_k$ , being soluble in both phases. In case of ideally diluted systems with small pressure gradients the components have no influence on the hydrodynamics and can therefore be considered as passive scalars, transported by the velocity field. The local balance for the species thus reads as

$$\frac{\partial c_k}{\partial t} + \nabla \cdot (c_k \,\mathbf{u} + \mathbf{j}_k) = R_k \quad \text{in} \quad \Omega^c(t) \cup \Omega^d(t), \tag{7}$$

governing the species transport inside the bulk phases. At the interface, the transmission condition

$$[\mathbf{j}_k] \cdot \mathbf{n}_{\Sigma} = 0 \tag{8}$$

holds and the jump condition

$$[\boldsymbol{\mu}_k] = 0 \tag{9}$$

is imposed, where  $\mu_k$  denotes the chemical potential of the species k. The diffusive fluxes are given by Fick's law, i.e.

$$\mathbf{j}_k = -D_k \,\nabla c_k \tag{10}$$

with constant diffusion coefficient  $D_k > 0$ , and the continuity of the chemical potential is expressed by Henry's law, i.e.

$$c_k^d / c_k^c = H_k \tag{11}$$

with Henry coefficient  $H_k$ , where usually  $H_k \neq 1$ . The source term  $R_k$  on the righthand side of equation (7) accounts for chemical reactions.

#### **3** Numerical Method

The general approach is to solve the partial differential equations from Section 2 without any additional simplifying assumptions. The governing equations are solved numerically employing the inhouse code Free Surface 3D (FS3D) [Rieber and Frohn (1999)]. A Finite Volume (FV) discretization is used for the spatial discretization and an explicit Eulerian scheme for the time discretization. The computational domain is discretized with a Cartesian, staggered grid, where scalar variables like pressure or concentration are stored as cell centered values and the velocities are stored on the centers of the cell faces. The code employs the Volumeof-fluid (VOF) method. Here, an additional transport equation is solved to keep track of the location of the different phases and, thereby, of the interface. This type of method is a volume tracking scheme, since only the different phases are transported and the interface is geometrically reconstructed from that information. In FS3D, the PLIC algorithm [Rider and Kothe (1998)] is employed for interface reconstruction [Rieber (2004)]. The code uses a one-field formulation of the twophase Navier-Stokes equations in which the volumetric surface tension force is incorporated via the conservative continuum surface stress (CSS)-model of Lafaurie [Lafaurie, Nardone, Scardovelli, Zaleski, and Zanetti (1994)]. The volume fraction transport equation reads as

$$\frac{\partial f}{\partial t} + \nabla \cdot (\mathbf{u} f) = 0, \tag{12}$$

where f is the phase indicator function of the dispersed phase domain  $\Omega^d(t)$ . In the FV discretization scheme employed here, the cell centered value of f corresponds to the dispersed phase fraction inside a computational cell. Within the VOF-method, the phase related material properties are given by

$$\boldsymbol{\rho} = f\boldsymbol{\rho}^d + (1 - f)\boldsymbol{\rho}^c \tag{13}$$

and

$$\eta = f\eta^d + (1-f)\eta^c. \tag{14}$$

### 3.1 Transport of molar species mass

For the computation of the transport of a transfer species k, the concentration is represented by two separate scalar variables according to

$$\phi_k^d(\mathbf{x},t) = \begin{cases} c_k(\mathbf{x},t) & \text{for } \mathbf{x} \in \Omega^d(t) \\ 0 & \text{for } \mathbf{x} \in \Omega^c(t) \end{cases}$$
(15)

and

$$\phi_k^c(\mathbf{x},t) = \begin{cases} 0 & \text{for } \mathbf{x} \in \Omega^d(t) \\ c_k(\mathbf{x},t) & \text{for } \mathbf{x} \in \Omega^c(t). \end{cases}$$
(16)

This allows for a representation of the individual one-sided limits of the concentration at the interface. Within the FV discretization, these scalars are related to the cell volume, i.e. the cell centered values for a grid cell  $V_i$  are given as

$$\phi_k^d(t) = \frac{1}{|V_i|} \int_{V_i \cap \Omega^d(t)} c_k(t) dV \tag{17}$$

and

$$\phi_k^c(t) = \frac{1}{|V_i|} \int_{V_i \cap \Omega^c(t)} c_k(t) dV.$$
(18)

The computation is carried out with a directional splitting, where for each of the three dimensions a one dimensional transport step is calculated consecutively. The order of the directional steps is changed in every time step to reduce systematic errors. Inside the bulk phases the convective transport of the volume fraction f is calculated with a simple first-order upwind scheme. Since f has only one discrete value in the bulk phase, this is accurate.

The concentration of a chemical species, however, can take arbitrary, non negative values. A first-order upwind scheme applied in that case would lead to unacceptable

numerical diffusion. Therefore the convective transfer in the bulk phases is based on the limiter scheme of Van Leer [Van Leer (1979)]. At the interface, the same reconstruction of the interface geometrie (PLIC) is used for a geometric flux computation, analogously as for the f-field. In this way any relative motion between the phase boundary and the surface of the concentration discontinuity is avoided. This ensures that no artificial mass transfer due to convection occurs.

Diffusive fluxes inside the bulk phases are obtained by a standard central differencing scheme. In interfacial cells, different cases have to be distinguished. Diffusion between an interfacial cell and a bulk phase cell is calculated for the appropriate scalar only, using the corresponding diffusion coefficient for that phase. For the diffusion between two interface cells two diffusive fluxes are calculated, one for each phase with the respective diffusion coefficients. The effective area in that case is approximated by the cell face area multiplied by the fraction of the respective phase. To ensure physically reasonable diffusive fluxes into or from cells with very small values of f, the fluxes are restricted. Since diffusion can only take place until the neighboring concentration values coincide, this can be used as a limit for the diffusion in one time step.

### 3.2 Mass transfer source term

The mass transfer between the phases takes place in those cells carrying a part of the interface and is accounted for by means of source terms for the two scalars representing the full concentration field. In the following, a brief description of two possible ways to calculate the interfacial source term is given. For more details we refer to [Bothe and Kröger (2010)]. The calculation of the source terms is restricted to interfacial cells. Inside those cells, the interface is assumed to be planar. Furthermore, it can be assumed that inside an interfacial cell, the concentration on the dispersed phase side is homogeneous due to the large diffusion coefficient. In a one-dimensional splitted approach, one can then calculate the one-sided interfacial limit value of the concentration for the dispersed phase by simply extrapolating the cell centered value onto the interface. The limit value for the continuous phase can be obtained from Henry's law (11). Together with one neighboring value from the continuous phase, a linear approximation for the gradient of the concentration at the interface can be obtained from this value. Inserting this gradient into Fick's law (10) yields the molar flux of the transfer species across the interface for the continuous phase. Since (8) states that both fluxes have to be equal, this also gives directly the flux for the dispersed phase, leading to a conservative algorithm. In a dimensional splitting scheme, the fluxes are calculated for each direction separately.

The second method is quite similar to the linear gradient method. Here, instead of a

linear approximation a different function is used to estimate the gradient. The type of function used in this subgrid-scale model is obtained from analytical solution describing the mass transfer for an overflown planar and mobile interface [Bird, Stewart, and Lightfood (2002)]. It has the form

$$c_k(x,y) = c_k^c \left(1 - \operatorname{erf}\left(\frac{x}{\delta_y}\right)\right)$$
(19)

with  $\delta_y$  the local thickness of the concentration boundary layer. Here, the latter is determined in such a way that the concentration value in the center of the adjacent continuous phase bulk cell matches the value given by (19). Having adjusted  $\delta_y$ , the local mass transfer rate is then computed using the directional derivative of the nonlinear profile according to (19), thus performing a nonlinear flux correction. It has been shown in simulations that in case of very fine grids both methods yield the same results. In the test case below, a resolution of the boundary layer by at least three grid cells was sufficient. However, with the subgrid-scale model the result can already be obtained with a coarser resolution. Therefore this method is to be preferred. The only drawback here is that in this form it can be only used in situations, where mass transfer resistance in the dispersed phase is small. All numerical results in the next sections are obtained employing the subgrid-scale model.

#### 4 Validation

In this section, comparisons between theoretical and numerical as well as between experimental and numerical result are used to validate the described numerical approach. The first comparison employs the flow field obtained by [Hadamard (1911)] and [Rybczynski (1911)] which is valid for Reynolds numbers below around  $Re_p = 0.3$ , where

$$Re_p = \frac{\rho_L U d_p}{\eta_L} \tag{20}$$

with the particle diameter  $d_p$ , the density  $\rho_L$  and the viscosity  $\eta_L$  of the liquid phase and the terminal rise velocity of the particle U. In Fig. 1 a comparison between the theoretical and simulated concentration profile at the equator of a rising bubble is shown. In the numerical setup the bubble equivalent diameter is set to  $d_p = 4$  mm. The computational domain for this 3D simulation has the dimension of  $4x2x2 d_p$ and is discretized with 64 cells per bubble diameter. To save computation time in 3D simulation runs, only a quater of a bubble is simulated using two symmetry planes. Furthermore, the computational domain moves with the bubble; e.g., when



Figure 1: Comparison of exact (lines) and with the VOF-method obtained concentration profiles at the equator of a rising fluid particle with Re = 0.284, Sc = 1000 (top)

the bubble's barycenter has been displaced by one grid width h in one basis direction  $\mathbf{e}_i$ , the whole domain is shifted in that direction. Concerning the boundary conditions, the free slip condition is applied at the lateral and the bottom domain boundaries and homogeneous Neumann condition at the upper domain boundary for the velocity, and for the species concentration at all domain boundaries homogeneous Neumann conditions are used. The viscosity of the continuous phase is set to 460 mPa s. The diffusion coefficient of the transfer component is  $0.2 \text{ cm}^2/\text{s}$ inside the dispersed phase and  $3.7 \times 10^{-3}$  cm<sup>2</sup>/s inside the continuous phase. Due to the rather large liquid viscosity, the Schmidt number is quite large. The Reynolds number corresponding to the terminal rise velocity is 0.284 for this setting. The theoretical profile shown in Fig. 1 is obtained from a solution of the species equation outside the spherical bubble in polar coordinates with the given analytical velocity field. Since the problem is stationary in the co-moving frame, the polar angle can be used as the independent (time-like) variable. Hence the profiles on radial rays can be obtained successively for increasing polar angles. The remaining problem is the computation of the concentration profiles in one space dimension, namely on the rays pointing radially outward and starting at the bubble surface. This is also done numerically (with Mathematica), but here the possible resolution is so high that a grid independent solution is easily obtained which approximates the exact solution as close as requested. The profile shows very good agreement in this case. As a measure for the overall mass transfer, the integral Sherwood number

$$Sh = \frac{\beta L}{D} \tag{21}$$

	Re = 0.284
	Sc = 1000
Numerical Result	18.5
Eq. (22)	12.0
Eq. (23)	13.0

Table 1: Comparison of integral Sherwood numbers

can be used. Here  $\beta$  is the mass transfer coefficient, *L* is the particle diameter, and *D* the diffusion coefficient of the liquid phase. There are different theoretical correlations for the Sherwood number for certain special cases:

$$Sh = 1 + \left(1 + 0.546 P e^{2/3}\right)^{3/4},$$
(22)

which is assumed to be valid for  $Re \rightarrow 0$  and arbitrary Peclet numbers Pe = ReSc [Clift, Grace, and Weber (1978)],

$$Sh = 2 + 0.651 \frac{Pe^{1.72}}{1 + Pe^{1.22}},$$
(23)

which is assumed to be valid for  $Re \rightarrow 0$  and  $Sc \rightarrow \infty$  [Oellrich, Schmidt-Traub, and Brauer (1973)]. For the test case described above, the results of the comparison can be seen in Table 1. The agreement for the integral mass transfer is not as good as in for the local profiles. The simulation yields about 50% higher mass transfer than the theoretical correlations. Additional simulations with a finer grid or smaller Schmidt number are needed to clarify this point.



Figure 2: Distribution of oxygen in the wake of a rising bubble. LiF experiment (left)  $d_p = 1.6 \text{ mm}$  with marked bubble and original, numerical 3D Simulation (right)  $d_p = 1.8 \text{ mm}$ .

For physical mass transfer, a high resolution numerical result obtained with the subgrid-scale model is compared with experimental result (Laser-induced fluorescence (LIF) measurement technique) from Schlüter, Hamburg (Germany) and Räbiger, Bremen (Germany). In this method a fluorescence marker is added to the liquid. Fluorescence is induced by a laser pulse and the resulting signal is recorded. In the presence of oxygen the fluorescence is quenched and the intensity of the recorded signal can be related to the concentration of oxygen in the liquid. The dark area on the left is the shadow of the bubble, and the bright area is the reflection of laser light from the surface. Details on this method can be found in [Bork, Schlüter, Scheid, and Räbiger (2001); Kück, Schlüter, and Räbiger (2009)]. An aqueous CMC solution is chosen for the liquid, having a viscosity of  $\eta = 3$  m Pas at 288 K. According to the Wilke-Chang equation [Neue (1997)] the diffusion coefficient of oxygen in this solution at 288 K is  $D_k = 6.9 \times 10^{-6} cm^2/s$  leading to a Schmidt number of about Sc = 4000. The Reynolds number is about Re = 160. Again, only a quater of a bubble is simulated using two symmetry planes. The volume equivalent bubble diameter is 1.5 mm and the cubic computational domain has an edge length of 1.2 cm and is resolved by a cartesian grid with cubic cells with an edge length of about  $23 \,\mu m$ . Fig. 2 shows the numerically and experimentally obtained concentration profiles which compare reasonably well. Since the setup for the experiment differs slightly from that of the simulation, the results are only qualitatively compared.

#### 5 Results

The method described above can be applied to simulate reactive mass transfer with simple and complex reactions inside the phases. As an example, the simulation of the metal catalyzed oxidation of sulfite is considered in this section. The overall reaction scheme for this reaction is given as

$$HSO_3^- + \frac{1}{2}O_2 \xrightarrow{k*} HSO_4^-$$
(24)

with k\* given as the gross reaction rate assumed constant for this reaction. Note that the complete reaction scheme is far more complex, with radical reaction mechanisms involved. For simplification, here the gross reaction is taken into account. In a more abstract notation this results in a reaction scheme of the type

$$A + B \xrightarrow{k*} P. \tag{25}$$

In this scheme A denotes the transfer component (oxygen), B is the dissolved component in the liquid phase (the hydrogen sulfite ion) and P is the desired product (the hydrogen sulfate ion).



Figure 3: Concentration distribution of oxygen



Figure 4: Series of simulations for reactive mass transfer at increasing temperatures. Reaction rate constant from left to right: 0, 100, 1000, 2000, 10000  $[cm^3/mol s]$ 

In the left part of Fig. 3, a snapshot of a LIF-experiment for the sulfite oxidation is shown. It can be seen that the oxygen is depleted in the center of the wake because of the chemical reaction, thus creating a concentration gap inside the wake. The numerical result obtained from a 2D simulation reproduces this gap very well, where so far only a qualitative comparison is possible. The concentration gradient in the surrounding liquid depends on the amount of oxygen that is removed through the reaction. This in turn is influenced by the reaction rate.

In order to simulate the influence of the reaction on mass transfer, a series of simulations with different reaction rate constants is carried out. The equivalent bubble diameter in this case is 1 mm. The viscosity of the liquid phase is 3 mPas, and the Henry coefficient 34.5. This corresponds to a system of oxygen/water at  $25^{\circ}$ C. In this simulations, only k\* is varied, while all other parameters are kept constant. Fig. 4 shows the obtained concentration profiles for reactive mass transfer with increasing reaction rate constant. With increasing k\*, the amount of oxygen in the wake is decreasing, indicating that the reaction is faster than the transport of oxygen inside the bubbles wake. In Fig. 5, the total decrease of species inside the bubble is plotted against time. As expected, these curves get steeper with increasing k\* and



Figure 5: The evolution of species mass for reactive mass transfer at different reaction rate constants

the steepness is proportional to the mass transfer coefficient.

### 6 Conclusions and Outlook

A numerical approach for the simulation of reactive mass transfer is introduced in this contribution. It is validated for the case of physical mass transfer. The method is then applied to the case of a simple reaction and the numerical results are compared with experimental data. Variation of the gross reaction rate constant can be used to simulate the effect of rising temperature on this reaction, which we are currently doing. Since it is quite difficult to measure the velocity at high resolution in experiments, it is also planned to combine the simulated velocity fields with measured concentration fields in order to obtain integral Sherwood numbers from the experiments.

## 7 Acknowledgements

The authors gratefully acknowledge financial support from the Deutsche Forschungsgemeinschaft (DFG) within the DFG-project "Reactive mass transfer from rising gas bubbles" (PAK-119). We also thank Prof. Michael Schüter (TU Hamburg-Harburg, Germany) and Prof. Norbert Räbiger (University Bremen, Germany) for providing the experimental results.

## References

**Alke, A.; Bothe, D.; Kroeger, M.; Warnecke, H.-J.** (2009): VOF-based simulation of conjugate mass transfer from freely moving fluid particles. In Mammoli, AA and Brebbia, CA(Ed): *COMPUTATIONAL METHODS IN MULTIPHASE FLOW V*, WIT Transactions on Engineering Sciences, pp. 157–168.

**Bird, R. B.; Stewart, W. E.; Lightfood, E. N.** (2002): *Transport Phenomena,* 2<sup>nd</sup> ed. John Wiley & Sons, Inc., New York, Chichester, Weinheim, Brisane, Singapore, Toronto.

Bork, O.; Schlüter, M.; Scheid, S.; Räbiger, N. (2001): New phenomena of mass transfer in gas/liquid flows. In *Heat Transfer*, no. November, pp. 1–6.

**Bothe, D.; Koebe, M.; Wielage, K.; Prüss, J.; Warnecke, H.-J.** (2004): Direct numerical simulation of mass transfer between rising gas bubbles and water. In Sommerfeld, M.(Ed): *Bubbly Flows: Analysis, Modelling, and Calculation*, Heat and Mass Tranfer, pp. 159 – 174. Springer, Berlin.

**Bothe, D.; Kröger, M.** (2010): A VOF-based method for mass transfer processes at fluidic particles. *in preparation*.

**Bothe, D.; Kröger, M.; Alke, A.; Warnecke, H.-J.** (2009): Vof-based simulation of reactive mass transfer across deformable interfaces. *PCFD*, vol. 9, pp. 325 – 331.

Clift, J.; Grace, M.; Weber, M. (1978): *Bubbles, Drops, and Particles*. Academic Press, New York.

**Darmana, D.; Deen, N.; Kuipers, J.** (2006): Detailed 3d modelling of mass transfer processes in two-phase flows with dynamic interfaces. *Chem. Eng. Technol.*, vol. 29, pp. 1027 – 1033.

**Davidson, M.; Rudman, M.** (2002): Volume-of-fluid calculation of heat or mass transfer across deforming interfaces in two-fluid flow. *Numer. Heat Transfer, Part B*, vol. 41, pp. 291 – 308.

**Deshpande, K.; Zimmermann, W.** (2006): Simulation of interfacial mass transfer by droplet dynamics using the level-set method. *Chem. Eng. Sci.*, vol. 61, pp. 6486 – 6498.

**Deshpande, K.; Zimmermann, W.** (2006): Simulation of mass transfer limited reaction in a moving droplet to study transport limited characteristics. *Chem. Eng. Sci.*, vol. 61, pp. 6424 – 6441.

Hadamard, J. (1911): Mouvement permanent lent d'une sphère liquide et visquese dans un liquide visqueux. *C.R. Acad. Sci. Paris*, vol. 152, pp. 1735 – 1738.

**Khinast, J.; Koynov, A.; Tryggvason, G.** (2005): Mass transfer and chemical reactions in bubble swarms with dynamic interfaces. *AIChE J.*, vol. 51, pp. 2786 – 2800.

**Kück, U.; Schlüter, M.; Räbiger, N.** (2009): Analyse des grenzschichtnahen Stofftransports an frei aufsteigenden Gasblasen. *Ingenieur Technik-CIT*, , no. 1, pp. 1–10.

Lafaurie, B.; Nardone, C.; Scardovelli, R.; Zaleski, S.; Zanetti, G. (1994): Modelling merging and fragmentation in multiphase flows with surfer. *J. Comp. Phys.*, vol. 113, pp. 134 – 147.

Neue, U. (1997): HPLC Columns. Wiley-VHC, New York.

**Oellrich, H.; Schmidt-Traub, H.; Brauer, H.** (1973): Theoretische Berechnung des Stofftransports in der Umgebung einer Einzelblase. *Chem. Eng. Sci.*, vol. 28, pp. 711 – 721.

**Onea, A.; Wörner, M.; Cacuci, D.** (2009): A qualitative computational study of mass transfer in upward bubble train flow through square and rectangular minichannels. *Chem. Eng. Sci.*, vol. 64, pp. 1416 – 1435.

**Radl, S.; Koynov, A.; Tryggvason, G.; Khinast, J.** (2008): DNS-based prediction of the selectivity of fast multiphase reactions: hydrogenations of nitroarenes. *Chem. Eng. Sci.*, vol. 63, pp. 3279 – 3291.

**Radl, S.; Tryggvason, G.; Khinast, J.** (2007): Flow and mass transfer of fully resolved bubbles in non-newtonian fluids. *AIChE. J.*, vol. 53, pp. 1861 – 1878.

**Rider, W.; Kothe, D.** (1998): Reconstructing volume tracking. *J. Comput. Phys.*, vol. 2, pp. 112 – 152.

**Rieber, M.** (2004): Numerische Modellierung der Dynamik freier Grenzflächen in Zweiphasenströmungen, Ph.D. Thesis University of Stuttgart. VDI Verlag, Düsseldorf.

**Rieber, M.; Frohn, A.** (1999): A numerical study on the mechanism of splashing. *Int. Heat and Fluid Flow*, vol. 20, pp. 455–461.

**Rybczynski, W.** (1911): Über die fortschreitende Bewegung einer flüssigen Kugel in einem zähen Medium. *Bull. Inst. Acad. Sci. Cracovie, A*, pp. 40 – 46.

**Van Leer, B.** (1979): Towards the ultimate conservative difference scheme - v. a second order sequel to godunov's method. *Journal of Computational Physics*.