Multiscale Computations of Mass Transfer in Bubbly Flows

Abstract

by

Bahman Aboulhasanzadeh

Mass transfer and reaction in the liquid phase of gas-liquid multiphase flows usually takes place at a considerably slower rate than the transfer of momentum, so mass flux boundary layers are much thinner than momentum boundary layers. In Direct Numerical Simulations (DNS) the resolution requirement for flows with mass transfer are therefore significantly higher than for flow without mass transfer and reaction. In this work we develop a multi-scale approach and demonstrate its implementation in 2D to compute the mass transfer from buoyant bubbles, using a boundary-layer approximation next to the bubble and a relatively coarse grid for the rest of the flow. This approach greatly reduces the overall grid resolution required. Then we implement our method in 3D and perform validation of the approach by comparing to experimental data and semi-empirical correlations from the literature. We study the effect of void fraction and bubble interactions on the mass transfer from many bubbles using a 3D implementation of the code. Specifically, we do simulations of single bubbles in periodic boxes and we compare it to the simulation of several bubbles in a larger domain with the same void fraction. Comparisons shows that even though the average Reynolds number of freely moving bubbles drops after a while the mass transfer from the bubbles for most case studies increases slightly when bubbles start wobbling which increases bubble interactions. We also develop a film model to recover the under-resolved viscous forces between colliding non-coalescing droplet.
To

my dearest parents

&

my lovely wife
ACKNOWLEDGMENTS

I would like to thank Professor Grétar Tryggvason for supervising this research and for constantly providing support and ideas. I am always in his debt for patiently teaching me how to do research, write a scientific article, present my work, and being persistent in pursuing my goals. I also want to thank my committee members for their helpful comments and corrections, especially Dr. Dabiri for his helpful discussions and comments and Dr. Ardekani for her valuable advice.

Many thanks to my friends who made living in a different country more pleasant: Hossein Ziayan, Narges Hakim, Mohammad Heydari, Elaheh Rastgoo, Ali Nematbakhsh, Mahdi Agheli, Amin Doostmohammadi, and Billy McGowan. Special thanks to my dear friend, Mohsen Behnam, whom I enjoyed living with and sharing moments for two years. I also want to show my gratitude to my family, specially my father and mother Assadollah and Sakineh who always supported me with their love. Finally, I want to sincerely thank my lovely wife, Vahideh, for her encouragement and support during the course of this study.

This study was funded by NSF Grant CBET-1132410.
CONTENTS

ACKNOWLEDGMENTS ......................................................... iii

FIGURES ........................................................................ vi

TABLES .......................................................................... xi

LIST OF SYMBOLS .......................................................... xii

CHAPTER 1: BACKGROUND .................................................. 1
  1.1 Motivation ................................................................. 1
  1.2 Objective ................................................................. 3

CHAPTER 2: AN EMBEDDED ANALYTICAL DESCRIPTION FOR MASS
DIFFUSION FROM MOVING BUBBLES ............................... 5
  2.1 Numerical Method and Problem Setup ............................. 6
  2.2 A Boundary Layer Description for the Mass Transfer ....... 7
    2.2.1 Derivation ......................................................... 10
    2.2.2 A Simple Example ............................................. 16
  2.3 Results for Buoyant Bubbles ........................................ 17
  2.4 Summary ................................................................. 34

CHAPTER 3: THREE-DIMENSIONAL EXTENSION AND MODEL VALI-
DATION ................................................................. 37
  3.1 Model Extension for Axisymmetric and Three-dimensional Flows . 38
    3.1.1 Axisymmetric Model ........................................... 38
    3.1.2 Three-dimensional Model .................................... 41
  3.2 Results ................................................................. 43
    3.2.1 Taylor Bubbles .................................................. 44
    3.2.2 Ellipsoidal Oscillating Bubbles ......................... 49
    3.2.3 Comparison with the literature ............................. 53
  3.3 Summary ................................................................. 55
<table>
<thead>
<tr>
<th>FIGURES</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.1</td>
<td>The top frame shows that the concentration of mass around a rising bubble is confined to a thin boundary layer around the bubble and thin streams ejected into the wake. The bottom frame shows the boundary layer model introduced in this chapter and the notation explained in the text.</td>
<td>9</td>
</tr>
<tr>
<td>2.2</td>
<td>The solid line shows a fully converged steady-state solution of the one-dimensional advection-diffusion equation. The dashed line shows a one-parameter approximate solution designed to capture the scalar boundary layer.</td>
<td>15</td>
</tr>
<tr>
<td>2.3</td>
<td>The mass transfer in a reversed stagnation point flow. The gradient at the wall, computed from the fully converged finite difference solution and the model are shown in the top frame and the total mass transferred into the liquid is shown versus time in the bottom frame. The flow field is shown in the insert in the bottom figure, where a few streamlines are plotted.</td>
<td>18</td>
</tr>
<tr>
<td>2.4</td>
<td>The mass transferred from a single rising bubble for three different values of the Reynolds number and the Schmidt number after the bubble has reached a steady state rise velocity. The flow field is shown in the left column, the second column shows the mass transfer computed on the same grid as used to find the flow. In the third column the mass transfer is found using the boundary layer approach described in the current chapter and in the last column a very fine grid is used to compute the mass transfer, resulting in essentially fully converged solution.</td>
<td>21</td>
</tr>
<tr>
<td>2.5</td>
<td>The total amount of scalar in the liquid as a function of time for three different cases shown in Fig. 2.4, computed in three different ways: on a coarse grid (dash-dot line), on a fine grid (solid line), and on a coarse grid using the model (solid squares).</td>
<td>24</td>
</tr>
</tbody>
</table>
2.6 Details of the mass transfer for the $Re = 60$, $Sc = 15$ case. In the top frame, the mass in the boundary layer (dash-dot line) and the mass that has left the boundary layer (dashed line) are plotted versus time. Their sum gives the total mass transferred from the bubble (solid line). In the bottom left frame the concentration profile at three points on the bubble: at the top of the bubble, at the side and at the bottom are plotted after the bubbles have reached steady state. The results from the full simulations (solid line) and the results of the model (dashed line) are shown. In the bottom right frame the concentration profile along a horizontal line through the wake of the bubble, one bubble diameter below the bubble, is shown for the finest grid (dashed line), the coarse grid using the model (solid line), and on the coarse grid without the model (dashed-dot line).

2.7 The effect of the Schmidt number on the scalar diffusing from freely moving bubbles, computed using the model described in the text. Top frame, the scalar versus time. Bottom frames, the error, computed as the difference between the mass transfer predicted using the boundary layer approach and the mass transfer predicted by the fine grid finite difference solution, at nondimensional time 10.

2.8 The mass transfer from three freely moving bubbles. The bubbles and the scalar field at a late time are shown at the top, on a grid fine enough to resolve the scalar transfer (right frame) and using the boundary layer description (left frame). The scalar versus time is plotted on the bottom.

2.9 The mass transfer at high Schmidt numbers. The scalar field at a late time is shown at the top, for $Sc = 200, 600$ and 1000, as computed using the boundary layer description combined with a grid based advection of the scalar in the liquid. The total scalar diffused from the bubbles is shown versus time in the graph on the bottom.

3.1 Schematic of the bubble mass boundary layer for the derivation of the boundary layer equation.

3.2 A comparison of the bubble shape for different grid resolutions. Here we use 48, 96, and 192 grid cells across the cylinder half diameter.

3.3 A comparison of the computational and experimental bubble shape for different bubble diameter ratios, $\lambda = 0.99$ and 1.28. The bottom row shows the original images from the experiment while in the top row the corrected binary shape of the bubble is shown. The red vertical lines represent the edge of the cylinder. This figure also shows the mass contour field behind the bubble.
3.4 The Sherwood number, $Sh$ and the Reynolds number, $Re$ versus non-dimensional time for a Taylor bubble with a diameter ratio of $\lambda = 1.199$. The dashed line shows the Sherwood number while the solid line shows the Reynolds number. Horizontal straight lines show the time average of the Reynolds and Sherwood number over the time span of the line.

3.5 A comparison of the computationally predicted Reynolds number (a) and Sherwood number (b) with experimental results for a range of diameter ratio, $\lambda$, from 0.2 to 1.28. The numerical results to the right of the vertical dashed line are from the axisymmetric code for Taylor bubbles while the results to the left of that line are from the 3D code. $d_b$ is the diameter of a spherical bubble with the same volume as the bubble presented here.

3.6 Comparison of computational and experimental bubble shape for bubble diameter ratio $\lambda = 0.68$. On the bottom left the original experimental bubble shape is shown while the top left side shows the corrected binary image of the same bubble. On the right-hand side the result of 3D simulation with the same bubble diameter ratio is shown. The bubble surface is colored with the ratio of mass boundary layer thickness to the boundary layer limit, $\delta/\delta_0$. The mass concentration field, $f$, is also shown. Two solid vertical lines on the sides of the bubble and one in the middle are side edges of the cubical domain while the two dashed line is the edges of an imaginary cylinder with the same hydraulic diameter as the cross section of the domain.

3.7 Comparison of results obtained by the embedded analytical description approach with experimental data and several correlations from the literature, as well as experimental data. Open circles and diamonds represent the calculation for almost spherical bubbles.

4.1 Comparison of mass distribution in the domain for the freely and fixed moving arrays of bubbles for the case with $\varepsilon = 9.0\%$, $\log Mo = -6.5$, $Eo = 5.0$, and at non-dimensional time, $t = 27.9$.

4.2 Reynolds and Sherwood number versus time for the fixed moving array with $\varepsilon = 9.0\%$, $Eo = 5.0$. (a) is for a bubble with $\log Mo = -4.0$ and (b) is for a bubble with $\log Mo = -6.5$. The horizontal dash-dot lines show the time-averaged Reynolds and Sherwood number, over the span of the line on the corresponding graph.
4.3 Reynolds and Sherwood number versus time for the freely moving array with $\varepsilon = 9.0\%$, $Eo = 5.0$. (a) is for array of bubbles with log $Mo = -4.0$ and (b) is for array of bubbles with log $Mo = -6.5$. The horizontal dash-dot lines show the Reynolds and Sherwood number of freely moving array, time-averaged over the span of the line on the corresponding graph. The dashed lines show the statistically steady state Reynolds and Sherwood number for the respective fixed array simulation.

4.4 Average Sherwood and Reynolds number of 8 freely moving bubbles in a periodic domain versus average vertical location of their center of gravity. On the left, 8 snap shots of the bubble positions are shown which corresponds to the locations and values on the right figure. The solid lines represent average Sherwood number while the dashed line shows the average Reynolds number. The vertical solid and dashed lines are the time-average of Sherwood and Reynolds number for the corresponding fixed array, respectively. The non-dimensional parameters are $\varepsilon = 15.5\%$, $Mo = 10^{-8}$, and $Eo = 0.2$.

4.5 Reynolds and Sherwood number for the freely moving array of bubbles and the fixed one for three different void fraction, $\varepsilon$. Left column is for non-deformable bubble, $Eo = 0.2$, while right column is for deformable bubble, $Eo = 5.0$.

4.6 Percentage of the Reynolds and Sherwood number difference between the freely moving array of bubbles and the fixed ones for three different void fraction, $\varepsilon$. Left column is for non-deformable bubble, $Eo = 0.2$, while right column is for deformable bubble, $Eo = 5.0$.

5.1 Schematic of two drops sliding over each other. The dashed line shows the locus of middle points between the two interfaces. The control volume shown on the right is used for the derivation of equation for the evolution of film thickness.

5.2 Change of coordinate system for the calculation of the strain rate tensor.

5.3 Schematic of three-layer profile for the approximation of velocity inside and adjacent to the thin film.

5.4 Schematic of simple problem with opposite moving walls.

5.5 Steady state velocity profile between opposite moving walls with a thin less viscous film in the middle. Red solid line shows the benchmark solution, blue line with circles shows the model with constant viscous stress assumption, and black dashed line shows the coarse calculation without model. Horizontal black dotted lines show the edge of thin film.
5.6 Velocity profile between opposite moving walls with a thin less viscous film in the middle, at three different time, $t = 0.004$, $0.008$, and $0.016$. The left column shows the model solution using a constant viscous stress assumption and the right column shows the model solution using three-layer profile approach. Red solid line shows the benchmark solution, blue line with circles shows the model, black dashed line shows the coarse calculation without model, and the green dotted line shows the three-layer profile. Horizontal black dotted lines show the edge of thin film.

5.7 Schematic of two drop with an initial velocity opposite to each other and with the same magnitude.

5.8 Left droplet horizontal velocity versus time for coarse grid calculation, dashed line, fine grid calculation, solid line, and coarse grid calculation with the model, dotted line.

5.9 Comparison of droplet position after they passed each other. Black droplet shows the fine calculation, while green and red droplets show the coarse grid calculation with and without the model, respectively.

B.1 Shows the mass boundary layer profile at two states: when $\delta < \delta_0$, (a), and when $\delta > \delta_0$, (b). The solid pink shade shows the total mass inside boundary layer, $M_0$, and the dotted area in part b shows the truncated mass inside boundary layer, $M^T_0$.

B.2 The total amount of scalar in the liquid as a function of time for a 2D simulation of mass transfer with $Re = 30$ and $Sc = 20$, computed in four different ways: on a coarse grid (dash-dot line), on a fine grid (solid line), on a coarse grid using the model without correction term (solid squares), and on a coarse grid using the model with the correction term.

C.1 The experimental setup, showing the test section, the piping and controllers.

C.2 The cross section of the test section, the LED lights, and the camera.

C.3 Reconstruction of the bubble shape. The original image (a) is digitized (b) and then split into elliptical disks (c). The disks are reassembled to give a representation of the bubble (d).
### TABLES

<table>
<thead>
<tr>
<th>Table</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.1</td>
<td>Governing Parameters and Initial Conditions Used for the Simulations Discussed in the Text</td>
<td>22</td>
</tr>
<tr>
<td>2.2</td>
<td>A Comparison of the Steady-State Mass Transfer Computed Using the Method Presented In This Work with Correlations From the Literature</td>
<td>32</td>
</tr>
<tr>
<td>3.1</td>
<td>Governing Parameters Used for the Simulation Discussed for the Comparison with the Literature</td>
<td>54</td>
</tr>
</tbody>
</table>
LIST OF SYMBOLS

\( \delta_0 \)  
boundary layer limit

\( \delta \)  
boundary layer thickness

\( \gamma \)  
surface tension

\( \lambda \)  
diameter ratio, \( \lambda = \frac{d_b}{d_c} \)

\( Eo \)  
Eötvös number, \( Eo = \frac{\Delta \rho g d_b^2}{\gamma} \)

\( Mo \)  
Morton number, \( Mo = \frac{g \mu_1^4 \Delta \rho}{\rho_l^2 \gamma^3} \)

\( Sc \)  
Schmidt number, \( Sc = \frac{\mu_l}{\rho_l D} \)

\( Sh \)  
Sherwood number, \( Sh = \frac{k d_b}{D} \)

\( \mu \)  
viscosity

\( \rho \)  
density

\( \sigma \)  
strain rate

\( A \)  
bubble surface area

\( D \)  
mass diffusivity

\( d_b \)  
bubble equivalent diameter

\( d_c \)  
cylinder diameter

\( f \)  
mass fraction/concentration
\( f_\infty \) magnitude of \( f \) outside boundary layer, \( f_\infty = 0 \)

\( f_{\delta_0} \) magnitude of \( f \) at \( \delta_0 \)

\( g \) acceleration of gravity

\( h \) grid spacing

\( k \) mass transfer coefficient

\( M_0 \) total amount of \( f \) in the boundary layer

\( M_0^{ax} \) mass inside boundary layer for axisymmetric flow

\( m_r \) viscosity ratio, \( m_r = \mu_l/\mu_g \)

\( N \) Archimedes number, \( N = gd_b^2 \rho_l \Delta \rho/\mu_l^2 \)

\( n \) coordinate assigned normal to bubble surface (in 2D)

\( n_b \) number of bubbles

\( Pe \) Peclet number, \( Pe = Re \cdot Sc \)

\( r \) radius from axis of symmetry

\( r_r \) density ratio, \( r_r = \rho_l/\rho_g \)

\( Re \) Reynolds number, \( \rho_l u_t d_b/\mu_l \)

\( s \) coordinate assigned with bubble surface

\( s_1 \) first coordinate assigned to bubble surface (in 3D)

\( s_2 \) second coordinate assigned to bubble surface (in 3D)

\( u_n \) component of velocity in the normal direction

\( u_t \) bubble terminal velocity
Subscripts

\[ b \] boundary layer

\[ f \] domain outside the boundary layer

\[ l \] liquid
CHAPTER 1

BACKGROUND

1.1 Motivation

One of the widely used and most interesting processing units in the petro-chemical industry are bubble columns (Furusaki et al., 2001; Deckwer, 1992). They are simple equipments generally consisting of a vertical columns containing different kind of liquids and bubbles are injected from the bottom of these columns. Vertical movement of the bubbles under the influence of buoyancy force leads to interaction of the bubbles and an increase in mass transfer from the bubbles. In spite of the simple structure of bubble columns, there are many aspects of these columns that are not well-understood. Today, we use experimental data and empirical and semi-empirical correlations for their design. While these approaches are in many way satisfactory, with the increase of computational power and the increased availability of direct numerical simulation (DNS) of multiphase flow, it is more insightful to use computational fluid dynamics (CFD) as a tool for designing and optimizing bubble columns and similar equipments.

Computational fluid dynamics has evolved in the last three decades and with the growth of computational power, we are now on our way to be able to simulate many flow situations using direct numerical simulation. However, while the hydrodynamic side of the problem is mostly solved, adding new physics, i.e. mass transfer or chemical reaction, is still a major challenge in performing DNS of new sophisticated problems. The challenging issue in most cases is the wide disparity between
the length and time scales of the flow, on one hand, and the length and time scales of
the new physics on the other hand. For example, when mass diffuses from a bubble
in a bubble column, generally the mass diffusion is at least two or three orders of
magnitude smaller than the momentum diffusion. It means that in order to resolve
the mass transfer which is most important in regions with sharp concentration gra-
dient, i.e. bubble interface, we need to add hundred or more grid points there, which
adds to the computational cost tremendously both by increasing the number of grid
points and decreasing the time step.

In recent years there have been considerable amount of research on mass transfer
from moving bubbles. Among those, we can mention the work of Yang and Mao
(2005), Wang et al. (2008), Ganguli and Kenig (2011a,b), and Hayashi and Tomiyama
(2011), who used a level set method to compute the bubble motion, and Davidson
and Rudman (2002), Bothe et al. (2003), Onea et al. (2009), Francois and Carlson
(2010), and Gupta et al. (2010), who used a volume of fluid (VOF) method. Other
authors, such as Mao, Li and Chen (2001) and Figueroa-Espinoza and Legendre
(2010) have used body fitted grids or unstructured meshes (Jung and Sato, 2005)
to allow them to concentrate grid points near the bubble. Most of studies are done
assuming a constant volume for the bubble, except for Hayashi and Tomiyama who
followed the shrinkage of the bubbles as gas diffused to the liquid. In addition to
papers on mass transfer, simulations with reactions have been described by Bauer
and Eigenberger (2001), Haroun, Legendre and Raynal (2010), Wylock et al. (2011),
and Dani et al. (2007). Koynov et al. (2005) performed a detailed simulation of mass
transfer and reaction of a two step reaction using a finer grid for the mass transfer and
reaction equations. Radl et al. (2008) have conducted simulations of the catalytic
hydrogenation of nitroarenes under reasonably realistic conditions using the same
approach as Khinast et al. for a two-dimensional system. Darmana et al. (2006)
implemented the same approach in three-dimensional calculations of mass transfer
from bubbles with relatively modest Schmidt number, $Sc = 1$. While, there have been different approaches for the study of mass transfer and reaction from moving bubbles, they have either been done for a single bubble, two-dimensional systems, or three-dimensional systems with very low Schmidt number.

Processes that take place on a scale much smaller than the dominant scale are common in multiphase flows. For small-scale processes, surface tension and viscosity generally dominate the dynamics, resulting in a relatively simple geometry and flow. These are exactly the situations where analytical or semi-analytical descriptions work well and the obvious strategy is to use such descriptions for these processes and couple them with the numerical description of the rest of the flow. This is, of course, an old idea. Boundary layer theory allows us to compute viscous drag for panel methods; the Hadamarad–Rybczynski solution for small drops in Stokes flows allows us to treat drops as point particles; and thin film models have been used to account for small-scale motion before (see Bossis and Brady, 1984; Davis, Schonberg and Rallison, 1989; Ge and Fan, 2006, for example). It seems likely that coupling analytical description for small-scale process with numerical solutions may have a much broader applicability, but before a general procedure can be established, it is likely that additional examples are needed. Thomas, Esmaeeli and Tryggvason (2010) developed such an approach for the thin film between a drop sliding down an inclined wall. In this work we explain the development, validation and application of similar approach for capturing mass transfer from moving bubbles with thin mass boundary layer around them. We also intend to extend the approach to capture reaction that may occur in the thin film around the bubble in future work.

1.2 Objective

Chapter 2 explains the development of an Embedded Analytical Description (EAD) for capturing mass transfer from moving bubbles. This is done using a two-
dimensional implementation of a front tracking code. We perform several test cases to demonstrate the performance of the EAD. In Chapter 3 the EAD is extended and applied in an axisymmetric and also a three-dimensional code. We perform a direct comparison with experimental result, in addition to comparison with analytical and semi-empirical correlations. Chapter 4 examines the behavior of bubble arrays and the effect of bubble interactions on the mass transfer. Two different flow regimes, almost spherical and deformable bubbles, are studied for three different void fractions. Chapter 5 introduces similar subscale concept for the resolution of thin film between colliding non-coalescing droplets. In the final chapter, chapter 6, we explain possible research directions for future works.
AN EMBEDDED ANALYTICAL DESCRIPTION FOR MASS DIFFUSION FROM MOVING BUBBLES

Here we describe the development of a multiscale approach intended to allow us to incorporate mass diffusion into a direct numerical simulation (DNS) of bubbly flows. The approach is based on the observation that the mass concentration changes very rapidly near the gas-liquid interface, forming a thin mass-boundary layer that controls the diffusion of chemical species from the bubble to the liquid. Since the structure of this boundary layer is relatively simple, its shape and thickness can be predicted accurately with a boundary layer description. Thus, we use the model to predict how much mass diffuses from the bubble into the liquid and then solve an advection-diffusion equation for most of the domain, to determine where the mass goes. For mass transfer, Alke et al. (2010) and Bothe et al. (2011) have used the exact solution of a one-dimensional diffusion problem, fitted to the computational results close to the bubble, to find the mass flux at the surface and Booty and Siegel (2010) have computed the evolution of a soluble surfactant by incorporating a singular perturbation analysis of the fluid next to the interface into a numerical solution of the interface motion. The approach of Booty and Siegel, although limited to Stokes flow, appears to be more closely related to the method presented here than that of Alke et al. In this chapter we focus on the mass transfer only, leaving the reactions for later. Although the approach developed here is intended for use in DNS of many bubbles in turbulent flows, such as those presented by Lu and Tryggvason (2006, 2007, 2008), in this chapter we present results for two-dimensional flows only, focusing mostly on
the initial rise of a single bubble.

2.1 Numerical Method and Problem Setup

For buoyant bubbles the governing nondimensional numbers are the Eötvös number, the Archimedes number, and the ratios of the densities and the viscosities. These numbers are defined in the Nomenclature. For mass transfer we also need to include the Schmidt number, which measures how fast mass diffusion takes place compared to viscous diffusion of momentum. For liquids the Schmidt number is generally very large so mass boundary layers are thin compared to viscous boundary layers.

The simulations discussed in this thesis are done using a front-tracking/finite-volume method where the governing equations are solved on a fixed, regular, mesh, covering both the ambient liquid and the bubbles (Unverdi and Tryggvason, 1992). The interface is marked by connected marker points that are advected with the fluid velocity and a marker function, constructed from the location of the interface, is used to set the density and viscosity of the different fluids. The marker points are also used to compute the surface tension. The method has been used earlier for a large number of simulations of multiphase flows, and both the method and various validation tests have been described in detail in several publications. For applications to bubbly flows, see Tryggvason et al. (2001), Bunner and Tryggvason (2002a,b, 2003), and Esmaeeli and Tryggvason (2005), for example. For other implementation of this method, see van Sint Annaland et al. (2005) and Hua et al. (2008), for example.

The computational domain is fully periodic and we add a force equal to the weight of the fluid to prevent uniform acceleration in the direction of gravity. Here we present computations only for two-dimensional domains. Extension of the model for axisymmetric and three-dimensional flows is presented in Chapter 3.

The mass transfer is governed by an advection-diffusion equation for the mass
concentration $f$,

$$\frac{\partial f}{\partial t} + u \cdot \nabla f = D \nabla^2 f \quad (2.1)$$

and we take the value of $f$ on the bubble boundary, $f_0$, to be given, without loss of generality. We assume that the fluid inside the bubble is well mixed and retains a uniform concentration throughout the simulation. For our case, where the mass diffusion in air is much higher than in water, this should be a reasonable assumption. Thus, it is only the solution outside the bubble that is of interest. As discussed by numerous authors (see Yang and Mao, 2005; Onea et al., 2009, for example) the solution to the original equations for the mass concentration in the gas and the liquid is discontinuous at the bubble surface, with the discontinuity given by Henrys law. However, by rescaling the concentration and the diffusion coefficient in the liquid, the solution can be made continuous. We also assume that the transfer of mass out of the bubble is so small that any change in the bubble volume can be neglected for the time we are following the motion. Including the change of volume for gases with large solubility is important, but remains to be done. Equation (2.1) is solved using a second order ENO method. When we use the boundary layer approach described below to compute the flux of scalar from the bubble to the liquid, we use the same grid resolution as for the fluid dynamics problem. To compute a fully converged solution for the mass transfer without the model, we follow Koynov et al. (2005) and Radl et al. (2008) and use a grid for the advection-diffusion equation that is much finer than the one used for the fluid dynamics part, using a velocity interpolated linearly from the fluid grid.

2.2 A Boundary Layer Description for the Mass Transfer

Mass diffuses from the bubble surface and is then swept by the flow toward the back where it is carried away as the flow separates. The top frame in Fig. 2.1 shows
a rising bubble and the thin mass boundary layer around the bubble. At the back of the bubble the boundary layers converge at flow stagnation points and the mass is ejected into the wake. For high $Sc$, the mass boundary layer is thin and can be difficult to resolve numerically. To derive a description to capture the mass transfer in a boundary layer that is so thin that it is not resolved, we decompose the scalar field into two fields, one resolved on the grid and the other captured by the boundary layer description. Since the advection-diffusion equation is linear, in principle these two fields can evolve independently. Where the boundary layer remains thin, the boundary layer accounts for all the mass that diffuses out from the bubble and the mass field resolved on the grid does not “know” about the mass coming from the bubble. There is, in particular, no flux of mass from the bubble to the grid-mass field. For those parts of the bubble surface where the boundary layer is thick, we transfer the mass from the boundary layer and follow it using advection resolved on the grid. We note that mass is taken away from the boundary layer and given to the grid but not the other way around and that while mass diffuses from the bubble into the boundary layer along its entire length, mass only leaves where the boundary layer is “thick” (see below). In general, we may have several thick segments of the boundary layer along the bubble surface and this would result in several coupling points. Here we assume that the mass concentration outside the bubble is sufficiently low so that it can be neglected when we compute the mass diffusion out from the bubble. While this is true for the examples considered here, it is not always the case. The fluxes from the bubble to the boundary layer depend on the gradient of the total mass field, and would be zero if the bubble was moving in a region with high enough mass concentration. Thus, the transfer of mass into the boundary layer should be found by considering the difference between the value of $f$ at the bubble surface and the value of the grid field, next to the bubble. We believe that this can be accounted for in a relatively straightforward way, but have not done so here.
Figure 2.1. The top frame shows that the concentration of mass around a rising bubble is confined to a thin boundary layer around the bubble and thin streams ejected into the wake. The bottom frame shows the boundary layer model introduced in this chapter and the notation explained in the text.
2.2.1 Derivation

Formally the decomposition of the mass field results in two advection-diffusion equations

\[ \frac{\partial f_f}{\partial t} + \mathbf{u} \cdot \nabla f_f = D \nabla^2 f_f + \dot{S} \]  

\[ \frac{\partial f_b}{\partial t} + \mathbf{u} \cdot \nabla f_b = D \nabla^2 f_b - \dot{S} \]  

(2.2)

where the velocity field is the same and the total scalar field is the sum of the scalar in the boundary layer and everywhere else

\[ f = f_f + f_b. \]

Here, the subscript \( b \) refers to the mass boundary layer and \( f \) denotes to the rest of the domain. Adding the advection-diffusion equations gives Eq. (2.1) for the scalar. The first equation is solved in a standard way using a finite volume method. The second equation is first parabolized. To do so we consider a coordinate system fixed to a material point at the bubble surface, as sketched in Fig. 2.1. One coordinate \((s)\) is aligned with the surface and the other \((n)\) with its normal. The distribution of a scalar diffusing from the bubble is given by the advection diffusion-equation (dropping the subscript \( b \)):

\[ \frac{\partial f}{\partial t} + u \frac{\partial f}{\partial s} + v \frac{\partial f}{\partial n} = D \left( \frac{\partial^2 f}{\partial s^2} + \frac{\partial^2 f}{\partial n^2} \right) \]  

(2.3)

assuming that the boundary layer is sufficiently thin compared to the interface radius of curvature so that all curvature effects can be neglected. The velocity near the origin
can be written as an expansion

\[ u = u_0 + \frac{\partial u}{\partial s} s + \frac{\partial u}{\partial n} n + \cdots \]  
\[ (2.4) \]

\[ v = v_0 + \frac{\partial v}{\partial s} s + \frac{\partial v}{\partial n} n + \cdots. \]

Since the coordinate system is moving with the fluid velocity, the relative velocity at the origin is zero \((u_0 = v_0 = 0)\). We define the strain rate

\[ \sigma = \frac{\partial u}{\partial s} = -\frac{\partial v}{\partial n}, \]

(2.5)

where the second form follows from incompressibility. Substituting Eqs. (2.4) and (2.5) into Eq. (2.3), assuming that all changes in the \(s\) direction can be neglected, and setting \(s = 0\), Eq. (2.3) becomes:

\[ \frac{\partial f}{\partial t} = \sigma n \frac{\partial f}{\partial n} + D \frac{\partial^2 f}{\partial n^2}. \]

(2.6)

Thus, the change of the scalar concentration is, to a leading order, a competition between diffusion in the normal direction, which deposits scalar into the liquid, and straining due to variations in the tangent velocity along the interface, which makes the scalar containing region thinner.

Approximate solutions to Eq. (2.6) can be obtained in many ways. We start by considering the total amount of \(f\) in the boundary layer

\[ M_0 = \int_0^{\delta_0} f \, dn. \]

(2.7)

An equation for the evolution of \(M_0\) is easily derived by integrating Eq. (2.6) across
the boundary layer, giving

\[
\frac{dM_0}{dt} = -\sigma M_0 + \sigma f_0 \delta_0 - D \left( \frac{\partial f}{\partial n} \bigg|_0 - \frac{\partial f}{\partial n} \bigg|_{\delta_0} \right).
\]  

(2.8)

Here the first term on the right is the decrease of \(M_0\) due to the stretching of the boundary layer, the second term is the outflow from the boundary layer when it is thick enough, and the last terms are the increase in \(M_0\) due to diffusion of mass from the bubble and diffusion from the boundary layer. \(\delta_0\) is a numerical parameter giving the maximum thickness of the boundary layer. We can easily derive equations for higher moments of \(f\), or use the integral of \(f\) weighted in other ways to derive additional equations, but here the solution is relatively simple and we would expect that a one-parameter family of profiles can capture the solution reasonably well. Initially we started with a two parameter profile and solved two equations for the zeroth and first moment in order to find the parameters as presented in Appendix A. However, since one of the parameters quickly converged to a constant value, shortly after the boundary layer started evolving, this parameter was set equal to the constant. In the rest of this work we will use

\[
f(n) = \begin{cases} 
  f_0 \left( 1 - 2 \left( \frac{n}{\delta} \right) + \left( \frac{n}{\delta} \right)^2 \right); & n \leq \delta, \\
  0; & n > \delta.
\end{cases}
\]  

(2.9)

Here, \(\delta\) is determined from \(M_0\). For \(\delta \leq \delta_0\), \(\delta\) is the thickness of the boundary layer and can be found by integrating Eq. (2.9) from 0 to \((n/\delta) = 1\), yielding \(\delta = 3M_0/f_0\). For \(M_0 > f_0\delta_0/3\), \(\delta\) is simply a numerical parameter, found by solving a quadratic equation:

\[
M_0 = f_0 \delta_0 \left( 1 - \frac{\delta_0}{\delta} + \frac{1}{3} \left( \frac{\delta_0}{\delta} \right)^2 \right).
\]  

(2.10)

Once the boundary layer becomes thick enough, when \(\delta = \delta_0\), we prevent it from
growing further and instead transfer the mass to the fixed grid. Notice that \( f_{\delta_0} \), the value of \( f \) at the outer edge of the boundary layer, is zero until \( \delta > \delta_0 \), so the sink term in Eq. (2.8) is zero for thinner boundary layers. The derivative at \( n = 0 \), used in Eq. (2.8) is computed directly from the profile, giving

\[
\frac{\partial f}{\partial n} \Big|_0 = -\frac{2f_0}{\delta}.
\]  

(2.11)

Similarly, the diffusion at the outer edge is zero, until \( \delta \geq \delta_0 \). After that it is given by the derivative of the profile (Eq. (2.9)) at \( n = \delta_0 \). At high \( Sc \), the diffusion part of the fluxes out of the boundary layer is generally small and in the simulations presented in Section 2.3 we neglect the last term in Eq. (2.8).

To use the model described above in a simulation of the full bubble motion, we evolve the strained-diffusion mass layer near the bubble interface by solving Eq. (2.8) at every front marker point which is about one-third of grid spacing \((h/3)\) apart from each other. The strain rate is computed by interpolating the velocity onto the front points and differentiating the tangential component along the interface. The scalar at the bubble surface is kept constant throughout the simulations. If \( M_0 \leq f_0\delta_0/3 \), we find \( \delta = 3M_0/f_0 \), which allows us to find the derivative at the bubble surface. For \( M_0 > f_0\delta_0/3 \), we solve Eq. (2.10) for \( \delta \), which also allows us to compute the derivative, as well as the value of \( f_{\delta_0} \). For front points where the boundary layer thickness is equal to \( \delta_0 \) and where \( f_{\delta_0} \) is nonzero, we draw a normal to the front and place a source for the grid mass transfer equation of strength \( \sigma\delta_0f_{\delta_0}\Delta s \), where \( \Delta s \) is the length of the front represented by each front point. In the computations reported below, we have used the so-called area-weighting scheme to distribute the source to the nearest grid points. We have experimented with changing this distance and generally find the result insensitive to its exact value, at least within a reasonable range. For positive strain rates, when the boundary layer is undergoing stretching,
we assume that mass does not flow back into the boundary layer from the grid, even when $M_0 > f_0 \delta_0 / 3$ so the source term in Eq. (2.8), the second term on the right hand side, is set to zero.

To assess how well this model captures the evolution of a strained diffusion layer, we show, in Fig. 2.2, a comparison of the solution of Eq. (2.6) for a one-dimensional situation, found by a finite difference discretization and by the approximate profile (Eq. (2.9)) where the first moment and therefore the thickness have been evolved by Eq. (2.8). The profiles are shown after the solution has reached steady state, for two values of the diffusion coefficient. Obviously the approximate solution is very close to the fully resolved finite difference solution for both values of the diffusion coefficient.

We have experimented with other profiles. A simple linear one gives results that are typically 20%-30% off the exact solution but any other reasonable profile generally gives results that are only a few percentage off. We have also examined a quadratic profile with one additional free parameter, determined by solving for the first moment of Eq. (2.6), and find that it quickly approaches the one given by Eq. (2.9), giving further justification for the use of only the zeroth moment, Eq. (2.8).

The inclusion of the separation point in the region covered by our boundary layer description deserves a comment. Initially, we did not use a boundary layer description for regions where the layer was thick, but computed the mass transfer directly using the fixed grid. While this worked well for modest Schmidt numbers, at large values the resolution was not sufficiently high and we changed to the current approach. It is well known that the lubrication equations can often be used beyond their “formal” region of applicability. This has been examined recently by Krechetnikov (2010), who argues that the equations are weakly elliptic (instead of parabolic) at the separation point and that this allows them to capture flow around stagnation points, thus providing support for our approach.

Here, we assume a specific profile inside boundary layer, Eq. (2.9), which is not the
Figure 2.2. The solid line shows a fully converged steady-state solution of the one-dimensional advection-diffusion equation. The dashed line shows a one-parameter approximate solution designed to capture the scalar boundary layer.
exact profile. Since we use this profile to derive the equation for the evolution of mass inside boundary layer, in the process of integration from Eq. (2.6) to Eq. (2.8), a small inconsistency is introduced, making the solution dependent of $\delta_0$. In Section 2.3, we study the effect of $\delta_0$ and show that the selection of $\delta_0$ has negligible effect on our results. In Appendix B we explain in more detail how to correct this inconsistency. While, this inconsistency is negligible for the simulation done in this thesis for which concentration outside the mass boundary layer is assumed to be zero, it is important to note that it may introduce larger errors in simulations where the far field has a finite concentration.

2.2.2 A Simple Example

Since the equation controlling the mass transfer is linear, it is relatively easy to generate simple test cases that allow us to assess the main characteristics of the approach. In addition to the 1D problem shown in Fig. 2.2, we have examined the mass transfer from a flat plate in the presence of a reverse stagnation point flow. We consider a rectangular domain, $[-0.5 \leq x \leq 0.5] \times [0 \leq y \leq 1]$, with a velocity given by

$$u(x, y) = \sigma x; \quad v(x, y) = -\sigma y; \quad \sigma < 0$$

and the boundary conditions $f = 1$ at the bottom wall and a zero gradient outflow boundary condition at the top wall. Along the left and the right boundary we specify the $f$ profile, giving smooth but narrow transition from the value at the bottom wall to zero in the interior (specifying $f = 0$ along the sides results in a weak singularity that slows convergence under grid refinement). The advection-diffusion equation is solved by a finite difference method, second order in time and space, and the results compared with those found by the boundary layer approach. The evolution of the
first moment is given by

$$\frac{\partial M_0}{\partial t} + u(x, 0) \frac{\partial M_0}{\partial x} = -\sigma (M_0 - \delta_0 f_\delta) - D \left( \frac{\partial f}{\partial n} \bigg|_0 - \frac{\partial f}{\partial n} \bigg|_{\delta_0} \right),$$  \hspace{1cm} (2.12)

where we have added an advection term to Eq. (2.8) in order to account for the fact that our points are now stationary, rather than moving with the fluid. This equation is integrated using a first order upwind method until the solution has reached a steady state. Fig. 2.3(a) shows the slope at the wall at steady-state as found by the boundary layer approach and the fully converged finite difference solution and Fig. 2.3(b) shows the total mass deposited from the wall, as a function of time. Here, $\sigma = -1.0; D = 0.01$ and $\delta_0 = 0.2$, in computational units. Obviously the results are in good agreement. We have examined this model problem for other values of the various physical and numerical parameters and found similar agreements in all cases. Here we have included the diffusive fluxes when we transfer the mass from the boundary layer to the grid. The error in neglecting the diffusion is generally small (a few percentage for the parameters selected here) and decreases as the diffusion coefficient is reduced.

2.3 Results for Buoyant Bubbles

To examine how the strategy described above works for bubbly flows, we have examined the mass transfer from a rising buoyant bubble, for several different governing parameters. The computational domain is a two-dimensional rectangle, with periodic boundaries. The mass concentration inside the bubbles is kept equal to unity and the surrounding fluid initially has $f = 0$. At time zero the bubbles are circular and stationary and we follow their unsteady motion as they rise due to buoyancy. As the bubbles rise, mass diffuses from the bubbles and is swept to the back of the bubble by the flow. At the back, where the flow separates, the mass is carried away
Figure 2.3. The mass transfer in a reversed stagnation point flow. The gradient at the wall, computed from the fully converged finite difference solution and the model are shown in the top frame and the total mass transferred into the liquid is shown versus time in the bottom frame. The flow field is shown in the insert in the bottom figure, where a few streamlines are plotted.
from the bubble.

In Fig. 2.4 we show the results from several simulations after the bubble has moved several diameters and reached an approximately steady state rise-velocity. Each row is for one set of governing parameters, indicated in the figure. Table 2.1 lists the various physical and numerical parameters for these runs, as well as other runs discussed below. The equations are solved using arbitrary computational units but the results have been nondimensionalized. In this thesis, for all the simulation of buoyant bubbles, time is nondimensionalized by $\sqrt{d_b/g}$, and the total mass is nondimensionalized by the initial mass in the bubble. The first column shows the streamfunction, in a frame of reference moving with the bubble, computed on a grid that is fine enough so that the flow field is fully converged. The next column shows the scalar field as computed on the same grid using a second order ENO scheme for the advection terms and a centered difference approximation for the diffusion terms, as described above. The last column shows the scalar field as computed using a much finer grid, with the velocity field interpolated onto the fine grid. We did a grid resolution study for all the cases. For the hydrodynamic part we compared the bubble shape and position after it had moved several diameters and for the mass transfer we examined the scalar solution using 3, 9, and 27 times finer grid resolution. Since the last refinement, from 9 to 27 times, changed the results by less than one percent, we believe that this is a fully converged scalar field. The third column shows the scalar field as found on the velocity grid (the same grid used in column two) but using the boundary layer description. The thickness of the boundary layer ($\delta$ and $\delta_0$ if $\delta > \delta_0$) is shown by a thin line around the bubble but we have not attempted to plot the scalar distribution inside the boundary layer. Here $\delta_0$ is taken to be $3.5 \times h$, where $h$ is the grid spacing. The scalar is transferred from the boundary layer to the grid at $4 \times h$ and because the ENO scheme used for the advection of the scalar has some numerical diffusion, sometimes scalar “leaks” back into the boundary layer and
sometimes into the bubble, but in all cases this is very small. We have examined the solution for different values of $\delta_0$ as well as distance of the source from the bubble and find only very minor differences. Specifically, for the $Re = 60$, $Sc = 15$ case we found that the total mass transfer changed by less than one percentage when $\delta_0$ was changed from two times the grid spacing to over six times the grid spacing. Similar results were found for other cases.
Figure 2.4. The mass transferred from a single rising bubble for three different values of the Reynolds number and the Schmidt number after the bubble has reached a steady state rise velocity. The flow field is shown in the left column, the second column shows the mass transfer computed on the same grid as used to find the flow. In the third column the mass transfer is found using the boundary layer approach described in the current chapter and in the last column a very fine grid is used to compute the mass transfer, resulting in essentially fully converged solution.
TABLE 2.1

GOVERNING PARAMETERS AND INITIAL CONDITIONS USED FOR THE SIMULATIONS DISCUSSED IN THE TEXT.

<table>
<thead>
<tr>
<th>$Re$</th>
<th>$Sc$</th>
<th>Domain Size Width</th>
<th>Domain Size Height</th>
<th>Bubble Initial Position $(x, y)$</th>
<th>Flow Grid</th>
<th>Mass Transfer Grid</th>
<th>$Eo$</th>
<th>$N$</th>
<th>$Mo$</th>
</tr>
</thead>
<tbody>
<tr>
<td>8</td>
<td>80</td>
<td>$3 \times d_b$</td>
<td>$6 \times d_b$</td>
<td>$1.5 \times d_b, 1.5 \times d_b$</td>
<td>$86 \times 170$</td>
<td>$758 \times 1514$</td>
<td>1.13</td>
<td>174</td>
<td>4.84E-05</td>
</tr>
<tr>
<td>30</td>
<td>20</td>
<td>$3 \times d_b$</td>
<td>$9 \times d_b$</td>
<td>$1.5 \times d_b, 1.5 \times d_b$</td>
<td>$86 \times 254$</td>
<td>$758 \times 2270$</td>
<td>3.24</td>
<td>1984</td>
<td>8.64E-06</td>
</tr>
<tr>
<td>60</td>
<td>15</td>
<td>$3 \times d_b$</td>
<td>$9 \times d_b$</td>
<td>$1.5 \times d_b, 1.5 \times d_b$</td>
<td>$86 \times 254$</td>
<td>$758 \times 2270$</td>
<td>2.81</td>
<td>7031</td>
<td>4.50E-07</td>
</tr>
<tr>
<td>60</td>
<td>15</td>
<td>$9 \times d_b$</td>
<td>$9 \times d_b$</td>
<td>$(2.5 \times d_b, d_b)$, $(4.5 \times d_b, 1.83 \times d_b)$, $(6.11 \times d_b, 2.83 \times d_b)$</td>
<td>$254 \times 254$</td>
<td>$2270 \times 2270$</td>
<td>2.81</td>
<td>7031</td>
<td>4.50E-07</td>
</tr>
</tbody>
</table>

Note: The density and viscosity ratios are taken to be $r_r = m_r = 10$ for all cases.
In the top row, the flow parameters are selected such that the rise Reynolds number is relatively small and the bubble remains essentially circular with a wake that closes near the bubble. Thus, the scalar is deposited into the interior of the flow close to the bubble centerline and we see one scalar stream behind the bubble. In the second row, the Reynolds number is higher and the bubble more deformable, resulting in a recirculating wake. The scalar is therefore injected into the flow at two separation points and we see two scalar streams. In the bottom column, the Reynolds number is even higher and the bubble is more deformable so that the wake is significantly larger. This again results in two separation points and two scalar streams, on either side of the wake.

In all three cases, a comparison of the second and fourth column shows that the scalar stream behind the bubbles is both thicker and longer in column two. This suggests that computing the scalar transfer on the fluid grid results in much more scalar being deposited into the fluid than the converged solution predicts. This is indeed the case as is seen better in Fig. 2.5, where the total scalar transferred from the bubble is plotted versus time for the three cases shown in Fig. 2.4. The top line (dashed-dot) is the total mass transferred as computed on the coarse fluid grid and the thick solid line is the fully converged mass transfer from the fine grid. Obviously these lines are far apart. The mass transfer computed using the boundary layer model on the coarser grid is shown by the line represented by square boxes. For all three cases, the agreement is good, although it is best for the lowest Reynolds number case, where the Schmidt number is highest.

In Fig. 2.6 we examine the mass transfer in more detail, for the $Re = 60$, $Sc = 15$ case. The mass transfer from the bubble is computed by adding the mass in the boundary layer and the mass that has left the boundary layer. In the top frame we show the evolution of those components versus time. The mass in the boundary layer (dot-dot-dash line) quickly reaches a steady state but the mass that has left
Figure 2.5. The total amount of scalar in the liquid as a function of time for three different cases shown in Fig. 2.4, computed in three different ways: on a coarse grid (dash-dot line), on a fine grid (solid line), and on a coarse grid using the model (solid squares).
the boundary layer (dashed line) continues to grow. Adding these quantities gives the solid line, which was shown in Fig. 2.5. In the bottom part of the figure, on the left, we show the concentration profile at three points on the bubble: at the top of the bubble, at the side and at the bottom. Both the results from the full simulations (solid line) and the results of the model (dashed line) are shown. The agreement is good in all cases. The boundary layer description is intended to capture the mass transfer from the bubble, but once the mass has been ejected out of the boundary layer, the mass concentration is evolved on the original coarse grid. Thus, we would expect some difference between the coarse grid and the fine grid results. However, the model ensures that the right amount of mass is injected and it is probably not unreasonable to expect that this improves the accuracy. In the lower right hand frame in Fig. 2.6 we plot the concentration profile along a horizontal line through the wake of the bubble, one bubble diameter below the bubble. Except for the maximum value, the profile computed on the finest grid (dashed line) and on the coarse grid using the model (solid line) are in reasonable agreement, while the profile computed on the coarse grid without the model (dashed-dot line) is not.

Generally we would expect the boundary layer description to become more accurate as the mass diffusion decreases compared with the diffusion of momentum (higher Schmidt number). In Fig. 2.7 we examine how the agreement changes with Schmidt number, when all other parameters are held constant, for two Reynolds numbers. In the top row we show the total mass transferred from the bubble, computed both using a fine grid for the mass transfer as well as on a coarse grid with the boundary layer description. The agreement gets better with increasing Schmidt number, as seen in the bottom row where the percentage error is plotted at nondimensional time 10. We observe that since the boundary layer model is solved using an approximate shape for the concentration profile, the error may not go to exactly zero, even at very high Schmidt numbers. A similar plot for $Re = 13$ (not included) shows a similar trend,
Figure 2.6. Details of the mass transfer for the $Re = 60$, $Sc = 15$ case. In the top frame, the mass in the boundary layer (dash-dot line) and the mass that has left the boundary layer (dashed line) are plotted versus time. Their sum gives the total mass transferred from the bubble (solid line). In the bottom left frame the concentration profile at three points on the bubble: at the top of the bubble, at the side and at the bottom are plotted after the bubbles have reached steady state. The results from the full simulations (solid line) and the results of the model (dashed line) are shown. In the bottom right frame the concentration profile along a horizontal line through the wake of the bubble, one bubble diameter below the bubble, is shown for the finest grid (dashed line), the coarse grid using the model (solid line), and on the coarse grid without the model (dashed-dot line).
but at $Re = 60$, where we only had results for $Sc = 15$, we had difficulties getting
fully converged results for higher $Sc$ numbers using our fully resolved simulations.
We did, however, examine lower $Sc$, for which the error stayed more or less the same.

For the bubbles in Figs. 2.4 to 2.7, we follow the unsteady transient motion until
the bubbles have risen several diameters. The motion is, however, relatively simple
and to examine what happens in a more complex situation we show the results from
a study of the motion of three bubbles at $Re = 60$ and $Sc = 15$ in Fig. 2.8. Figure 2.8
(top) shows the bubbles and the mass at time 12.3, both using the boundary layer
description (left frame) and using a fine grid for the mass transfer (right frame). The
dashed line in both figures shows the path of the bubbles. The total mass transferred
from the bubbles is plotted versus time in Fig. 2.8 (bottom) and again we see that
the use of the model significantly improves the agreement of the coarse grid solution
with the fully converged solution on the fine grid. A careful examination of the
mass away from the bubbles shows a slightly more diffused mass on the coarser grid.
This is due to numerical diffusion in the ENO advection scheme. It is important
to note that the grid used for the evolution of the mass away from the bubbles is
relatively coarse and although Fig. 2.5 shows that injecting the correct amount of
mass improves the results, any structures smaller than the grid will, of course, not
be resolved. A conservative advection scheme should, however, ensure that the right
amount of mass is in the correct place.

For the cases presented so far, we have been able to compare the results using the
boundary layer description with fully resolved computations. As the Schmidt number
increases, this becomes more difficult. We have done a few simulations for much
higher Schmidt numbers, using the boundary layer description to capture the mass
transfer from the bubble, and Fig. 2.9 (top) shows the scalar field at nondimensional
time 12.3 for $Sc = 200, 600$ and 1000. Figure 2.9 (bottom) shows the total scalar
transferred from the bubble as a function of time for all three cases. The flow field
Figure 2.7. The effect of the Schmidt number on the scalar diffusing from freely moving bubbles, computed using the model described in the text. Top frame, the scalar versus time. Bottom frames, the error, computed as the difference between the mass transfer predicted using the boundary layer approach and the mass transfer predicted by the fine grid finite difference solution, at nondimensional time 10.
Figure 2.8. The mass transfer from three freely moving bubbles. The bubbles and the scalar field at a late time are shown at the top, on a grid fine enough to resolve the scalar transfer (right frame) and using the boundary layer description (left frame). The scalar versus time is plotted on the bottom.
is the same as for a single bubble at $Re = 60$ case presented earlier. Obviously, the mass that has diffused from the bubble is very small, the boundary layer is thin, and the scalar away from the bubble is not very visible. To assess the accuracy of these computations we have computed the mass transfer rate at steady state, $k$ (from the slope of the line in Fig. 2.9) and found the nondimensional mass transfer rate, given by the Sherwood number:

$$Sh = \frac{kd_b}{D}$$

where $d_b$ is the diameter of the initially circular bubble. In Table 2.2 we show the results for several simulations, including some of the ones already presented. The first two correlations come from the book by Clift et al. (1978) and hold for low and high Reynolds numbers, respectively, and are developed using analytical approximations for the velocity field. Equation B1 was proposed by Takemura and Yabe (1998) who modified earlier correlations based on experiments and numerical studies of gas bubbles in liquid. The last equation (equation B2), was originally derived by Lochiel and Calderbank (1964), using the velocity field found by Chao (1962), under the assumptions of a spherical bubble, high Reynolds number and freely circulating internal flow. Lochiel and Calderbank compared the predictions of their equation to experimental results and showed that it predicts the measured mass transfer reasonably well. Ponoth and McLaughlin (2000), who used body fitted grids to predict the mass transfer around an axisymmetric bubble, also found that this equation correlated their results for a clean bubble reasonably well. We emphasize that we should not expect an exact agreement between these correlations and our results, both because the correlations are for an axisymmetric or fully three-dimensional bubbles whereas our results are for a two-dimensional one, and also because there are some differences between the different correlations. Nevertheless, Table 2.2 shows that both the trend and the values predicted by our simulations are in reasonably good agreement with the earlier predictions.
Figure 2.9. The mass transfer at high Schmidt numbers. The scalar field at a late time is shown at the top, for $Sc = 200, 600$ and $1000$, as computed using the boundary layer description combined with a grid based advection of the scalar in the liquid. The total scalar diffused from the bubbles is shown versus time in the graph on the bottom.
TABLE 2.2

A COMPARISON OF THE STEADY-STATE MASS TRANSFER
COMPUTED USING THE METHOD PRESENTED IN THIS WORK
WITH CORRELATIONS FROM THE LITERATURE.

<table>
<thead>
<tr>
<th>$Re$</th>
<th>$Sc$</th>
<th>Sherwood Number</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Subscale Description</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Before Wobbling</td>
</tr>
<tr>
<td>8</td>
<td>80</td>
<td>17.0</td>
</tr>
<tr>
<td>13</td>
<td>60</td>
<td>18.2</td>
</tr>
<tr>
<td>30</td>
<td>20</td>
<td>17.9</td>
</tr>
<tr>
<td>60</td>
<td>10</td>
<td>20.1</td>
</tr>
<tr>
<td>60</td>
<td>200</td>
<td>89.4</td>
</tr>
<tr>
<td>60</td>
<td>600</td>
<td>153.9</td>
</tr>
<tr>
<td>60</td>
<td>1000</td>
<td>204.3</td>
</tr>
</tbody>
</table>
$Sh = 1 + \left(1 + 0.564Pe^{2/3}\right)^{3/4}$

$Sh = \frac{2}{\sqrt{\pi}} \left[1 - \frac{2.89}{Re^{1/2}}\right]^{1/2} Pe^{1/2}$

$Sh = \frac{2}{\sqrt{\pi}} \left[1 - \frac{1}{2} \left(1 + 0.09 Re^{2/3}\right)^{3/4}\right]^{1/2} (2.5 + Pe^{1/2})$

$Sh = 1.13 \left[1 - \frac{2 + 3\mu_g/\mu_l}{1 + (\rho_g\mu_g/\rho_l\mu_l)^{1/2} Re^{1/2}}\right]^{1/2} Pe^{1/2}$

Clift, Grace & Weber, Low Re (Eq. (3.50)), D1,

Clift, Grace & Weber, High Re (Eq. (5.37)), D2,

Takemura and Yabe (1998), D3,

Lochiel and Calderbank (1964), D4.
The purpose of the model introduced here is to reduce the resolution requirement for DNS of high Schmidt number bubbly flows. This obviously raises the question of how much savings, in computer time, do we expect. Here we have focused on establishing the accuracy of the model and assessing the reduction in spatial accuracy, but not attempted to optimize the overall approach to reach maximum savings. For the three bubble simulations we found that while the fully resolved run took about 11 days on a 3.2 GHz Quad Core Intel Xeon CPU with 12 GB of RAM, the run with the model took about 4 days. However, to keep everything except the grid used for the mass transfer the same, we used identical time steps for both runs. In practice the savings would therefore be larger. For much higher $Sc$, which is where we really need the model, we do, of course, expect the difference to be considerably more significant.

2.4 Summary

The results presented here suggest that the use of a boundary layer description for the mass transfer from the bubble surface, coupled with a finite volume grid based approach for the rest of the flow improves the results on a coarse grid significantly. Conversely, the boundary layer approach allows us to use a much coarser grid for a given accuracy. Indeed, it seems likely that this approach is needed to make simulations of very high Schmidt number systems practical. There are, obviously, several ways in which the approach presented here can be improved. The more obvious ones include:

- Use of a better advection scheme for the finite volume part of the simulation. We used a second order ENO since it is very easy to implement but higher order ENO or WENO schemes are probably needed to eliminate, for example, the slight but unphysical flux of mass into the back of the bubble.

- The parabolized version of the advection diffusion equation used here ignored any curvature effects, as well as fluid shear. Including these terms is likely to improve the results in cases where curvature and/or vorticity are important. As the results presented here show, in most cases these effects are likely to be
The approximate solution of the parabolized equation could be improved. Again, the results presented here suggest that such improvements would only be important in rare cases.

Although we were able to verify our approach for modest values of the Schmidt number by comparing the results using the boundary layer description with converged simulation of the full equations, at higher Schmidt numbers we were only able to do indirect validations by comparing with experimental correlations. Since our simulations are for a two-dimensional system these are not entirely satisfactory and a more thorough validation would obviously be desirable.

As the use of DNS for multiphase flow studies has increased, it has become clear that in many situations the grid resolution is set by isolated small-scale features, such as thin films, threads, drops and—for mass transfer—scalar boundary layers that are much thinner than the viscous boundary layers. Often these features have relatively simple structure and can be described accurately using relatively simple analytical models. For gas bubbles rising in liquids, the resolution needed to capture the mass transfer is usually much higher than what is needed for the fluid dynamics and when the mass transfer is under-resolved the total mass transfer is over-predicted by a large amount. In this chapter we have captured the mass transfer from rising bubbles using a simple boundary layer description for the mass transfer next to the surface of the bubble, coupled with a relatively coarse grid to follow the advection of the scalar far away from the bubble surface. This approach greatly reduces the overall grid resolution required.

Approximating boundary layers by known profiles is a very old idea and although this approach has fallen somewhat out of favor over the last couple of decades (we now generally prefer adaptive grid refinement) it is alive and well in several applications such as wall-functions in turbulence modeling. The fundamental idea of boundary layer modeling is, however, a natural way of accounting for already un-
understood phenomenon in multiscale simulations. When something is already known, why re-compute it? The “philosophy” behind the approach used here is the same as in Thomas, Esmaeeli and Tryggvason (2010) where they used a thin film model to capture the thin film beneath a drop sliding down an inclined plate. While the model and the coupling with the rest of the computed solution was different from the situation here, the use of a multiscale model was found to greatly reduce the required computational effort. As for the thin film in Thomas et al. (2010), there is nothing in the present model that is specific to two-dimensional flow. In the following chapter we present the extension of this description to three-dimension. Most simulations of mass transfer in bubbly flows have been limited to two-dimensional flows due to the fine grid needed for the scalar equation and the use of the present approach should be particularly advantageous for three-dimensional flows.
In Chapter 2, we introduced an embedded analytical description and tested it for two-dimensional flows. The accuracy was established by comparing results using this method with simulations where the mass transfer was computed using grids that were sufficiently fine so the mass transfer was accurately resolved. This approach did, however, restrict the comparison to moderate Schmidt numbers ($O(10)$). In this chapter our multiscale approach for mass transfer from bubbles into liquids is validated by detailed comparisons with experimental results specifically designed to validate the method for high Schmidt number, $Sc = 8260$, and high Reynolds number, $Re \approx 350$. Two classes of bubbles are examined: Taylor bubbles in a pipe and freely rising bubbles. For the first class an axisymmetric code is used, while for the latter a 3D code is used to capture the unsteady manner of a rising bubble for simulations where axisymmetry is not a feasible assumption. We also perform calculations of almost spherical freely rising bubbles for which we compare the computational results with analytical and semi-empirical correlations as well as experimental results from other authors for Reynolds number ranging from 20 to 110 with $Sc = 100$ and 1000. Mass transfer computations using the embedded analytical description approach show good agreement with the experimental results and the correlations. Based on the ratio of the thickness of the mass boundary layer to the grid size used to resolve the fluid flow we estimate that the use of the presented approach reduces the computational cost at least by one or two orders of magnitude, specially when applied in simulations of fully three-dimensional flows. The experimental part of this work is done by
Professor Tomiyama group in Kobe University, Japan, and explained in Appendix C.

3.1 Model Extension for Axisymmetric and Three-dimensional Flows

Here, we compare the results from the model with experimental results for two different types of bubbles, Taylor bubbles and oscillating three-dimensional bubbles. For the Taylor bubbles we use an axisymmetric code to compute the bubble dynamics in a cylinder, while for the oscillating bubbles, where the axisymmetric assumption is not feasible, we use a version of the code written for fully three-dimensional domains. In the following subsection the multiscale model is explained first for the axisymmetric and then for the three-dimensional simulation.

3.1.1 Axisymmetric Model

For the mass boundary layer in the case of axisymmetric flow, for example for the simulations of Taylor bubbles, we define the zeroth-moment of the mass concentration by

\[
M_0^{ax} = \int_0^{\delta_0} r f dn. \tag{3.1}
\]

Here \( n \) is the coordinate normal to the bubble surface, \( r \) is the radial distance from the axis of symmetry, and the upper bound \( \delta_0 \) is the boundary layer limit selected so that it is generally larger than the thickness of the boundary layer \( \delta \).

To extract the boundary layer equation we write the conservative form of Eq. (2.1) in a control volume form for a small segment of the mass boundary layer as shown in Fig. 3.1:

\[
\frac{\partial}{\partial t} (r f dn ds) - u_n r f ds + \left( u_n r f + \frac{\partial (u_n r f)}{\partial n} dn \right) ds
- u_s r f dn + \left( u_s r f + \frac{\partial (u_s r f)}{\partial s} ds \right) dn =
- D \left( \frac{\partial f}{\partial n} \right)_{n=0} ds + D \left( \frac{\partial f}{\partial n} \right)_{n=dn} ds, \tag{3.2}
\]
in which we have neglected diffusion terms in the tangential direction as well as all curvature effects.

Figure 3.1. Schematic of the bubble mass boundary layer for the derivation of the boundary layer equation.

Assuming $\alpha$ to be a Lagrangian marker (and a constant at each front marker point), we have $ds = (\partial s/\partial \alpha) d\alpha$. Expanding and rearranging the first term on the left-hand side of Eq. (3.2), it can be written as:

$$
\frac{\partial}{\partial t} (rfdn\alpha) = r \frac{\partial f}{\partial t} \left( \frac{\partial s}{\partial \alpha} \right) d\alpha + \frac{\partial r}{\partial t} \left( \frac{\partial s}{\partial \alpha} \right) fdn\alpha + \sigma_{ax}r f \left( \frac{\partial s}{\partial \alpha} \right) d\alpha. \quad (3.3)
$$

Here, $\sigma_{ax}$ is the tangential component of tangential derivative of the velocity, or $\sigma_{ax} = \mathbf{s} \cdot \partial(\mathbf{u})/\partial \alpha$. Since the control volume for derivation of the mass boundary layer equation is attached to a Lagrangian marker, the tangential advective terms in
Eq. (3.2) are equal to zero. So, by replacing $ds$, inserting Eq. (3.3) into Eq. (3.2), and simplifying, the boundary layer equation for the mass transfer on the bubble interface can be written as:

$$\frac{\partial rf}{\partial t} + \sigma_{ax}rf + \frac{\partial (u_nrf)}{\partial n} = D \frac{\partial}{\partial n} \left( r \frac{\partial f}{\partial n} \right).$$

(3.4)

An evolution equation for the $M_0^{ax}$ is now derived by integrating Eq. (3.4) in the direction normal to the interface from 0 to $\delta_0$:

$$\frac{dM_0^{ax}}{dt} = -\sigma_{ax}M_0^{ax} - D \left( r \frac{\partial f}{\partial n} \right)_0 - u_{n\delta_0}f_{\delta_0}r_{\delta_0} + D \left( r \frac{\partial f}{\partial n} \right)_{\delta_0}.$$  

(3.5)

The first term on the right-hand side of Eq. (3.5) represents change in the moment due to stretching (or compression) and the second term is the diffusive flux of mass into the boundary layer from the bubble. The last two terms are zero except where the boundary layer thickness exceeds the upper bound of the integral defining $M_0^{ax}$, $\delta > \delta_0$. There they represent sink terms accounting for the advective and diffusive fluxes, respectively, of $f$ out of the boundary layer. To compute the gradient of $f$ we write $r$ as a function of $n$, $r = r_0 + (\partial r/\partial n)n$, where $r_0$ is the $r$ at the interface and $\partial r/\partial n$ is the rate of change of $r$ normal to the interface at that point, which can be simply calculated from the interface orientation. We also assume the same shape of the concentration profile inside the boundary layer as shown in Eq. (2.9). Substituting this profile and $r$ into the definition for $M_0^{ax}$ (Eq. (3.1)) results in the
following expression for $M_0^{ax}$:

$$M_0^{ax} = \begin{cases} 
\frac{f_0r_0\delta}{3} + \frac{1}{12} \frac{\partial r}{\partial n} f_0 \delta^2, & \delta \leq \delta_0, \\
\frac{f_0 \delta_0 \left( 4r_0 \left( 3\delta^2 - 3\delta_0\delta + \delta_0^2 \right) + \frac{\partial r}{\partial n} \delta_0 \left( 6\delta^2 - 8\delta_0\delta + 3\delta_0^2 \right) \right)}{12\delta^2}, & \delta > \delta_0,
\end{cases}$$

(3.6)

which can be solved for $\delta$. The normal derivatives of $f$ are given by differentiating the profile with respect to $n$ and evaluating it at $n = 0$, and $n = \delta_0$, when $\delta > \delta_0$

$$\left. \frac{\partial f}{\partial n} \right|_0 = -\frac{2f_0}{\delta}, \quad \text{and} \quad \left. \frac{\partial f}{\partial n} \right|_{\delta_0} = -\frac{2f_0}{\delta} \left( 1 - \frac{\delta}{\delta_0} \right).$$

(3.7)

Outside the boundary layer, where Eq. (2.9) is solved using a standard conservative finite volume scheme we account for the mass coming from the boundary layer by adding sources to Eq. (2.9), just outside the boundary layer. Generally the boundary layer is thinner than $\delta_0$ for most of the bubble surface so the source terms are confined to a small section of the surface.

3.1.2 Three-dimensional Model

In the three-dimensional case the model equations are the same as those used in our two-dimensional implementation of the model. The only difference is the way we calculate the strain rate, $\sigma$, at the front points. The embedded analytical description that we use here is developed by parabolizing Eq. (2.9) using a boundary layer assumption. To do so we consider an orthogonal coordinate system fixed to a material point at the bubble surface. Two coordinates ($s_1$ and $s_2$) are aligned with the surface and the other ($n$) with its normal. The distribution of a scalar diffusing
from the bubble is given by the advection-diffusion equation:

\[
\frac{\partial f}{\partial t} + u_{s_1} \frac{\partial f}{\partial s_1} + u_{s_2} \frac{\partial f}{\partial s_2} + u_n \frac{\partial f}{\partial n} = D \left( \frac{\partial^2 f}{\partial s_1^2} + \frac{\partial^2 f}{\partial s_2^2} + \frac{\partial^2 f}{\partial n^2} \right)
\] (3.8)

assuming that the boundary layer is sufficiently thin so that all curvature effects can be neglected. The velocity near the origin can be written as an expansion

\[
u_i = \nu_{i0} + \frac{\partial \nu_i}{\partial s_1} s_1 + \frac{\partial \nu_i}{\partial s_2} s_2 + \frac{\partial \nu_i}{\partial n} n + \ldots; \text{ where } i = s_1, s_2, n.
\] (3.9)

Since the coordinate system is moving with the fluid velocity, the relative velocity at the origin is zero \((\nu_{s_10} = \nu_{s_20} = \nu_{n0} = 0)\). We define the strain rate (the stretching of the bubble surface),

\[
\sigma = -\frac{\partial \nu_n}{\partial n} = \frac{\partial \nu_{s_1}}{\partial s_1} + \frac{\partial \nu_{s_2}}{\partial s_2} = \nabla(s) \cdot \mathbf{u}^s,
\] (3.10)

where the second form follows from incompressibility. In order to evaluate the surface divergence of velocity on the interface we use the surface divergence theorem, (Slattery et al., 1990):

\[
\frac{d}{dt} \int_A \psi^s dA = \int_A \left( \frac{d}{dt} \psi^s + \psi^s \nabla(s) \cdot \mathbf{u}^s \right) dA,
\] (3.11)

where \(\psi\) is any scalar or vector quantity, \(A\) is the surface area, the subscript \((s)\) means surface operation, and the superscript \(s\) means the magnitude of quantity at the surface. If we set \(\psi^s = 1\) and simplify, we have:

\[
\nabla(s) \cdot \mathbf{u}^s = \frac{1}{A} \frac{dA}{dt},
\] (3.12)

which is the rate of change of area per unit area. Using Eq. (3.12) and Eq. (3.10), the strain rate, \(\sigma\), can be calculated by evaluating the rate of change of a front element...
Substituting Eqs. (3.9) and (3.10) into (3.8), assuming that all changes in the \( s_1 \) and \( s_2 \) directions can be neglected, and setting \( s_1 = s_2 = 0 \), Eq. (3.8) becomes:

\[
\frac{\partial f}{\partial t} = \sigma_n \frac{\partial f}{\partial n} + D \frac{\partial^2 f}{\partial n^2}.
\]  

(3.13)

Approximate solutions to Eq. (3.13) can be obtained in a similar way as explained in Chapter 2.

3.2 Results

In our calculations, for both Taylor bubbles and ellipsoidal oscillating bubbles, we select the fluid and gas properties in a way to reduce computational cost while replicating the experimental setup. Our test for different viscosity ratios, \( m_r = \mu_l/\mu_g = 10, 50, \) and 300, shows that changing \( m_r \) from 10 to 300 changes either \( Re \) or \( Sh \) by less than 1%. Although for most gas/liquid systems the viscosity ratio is high, we chose \( m_r = 10 \) for all of our computations since this results in faster computations and essentially the same results as the “real” viscosity ratio. On the other hand, while the density ratio, \( r_r = \rho_l/\rho_g \), does not have a significant effect on the Reynolds number, (about 2% decrease for \( r_r = 300 \) compared to \( r_r = 10 \)); the Sherwood number changes by about 4% and 1% when we increase the density ratio from 10 to 50 and 50 to 300, respectively. Since the density ratio of 300 increases the computational cost considerably and only increases the accuracy by 1% we chose \( r_r = 50 \) in our simulations. The other non-dimensional numbers are the Morton number, \( Mo = \Delta \rho g \mu_l^4/\rho_l^2 \gamma^3 = 10^{-7.78} \), and the Eötvös number, \( Eo = \Delta \rho g d_c^2/\gamma = 24.7 \), where \( d_c \) is the cylinder diameter (not the bubble diameter).
3.2.1 Taylor Bubbles

The calculations for the Taylor bubbles that are presented here are done using a front-tracking finite volume code written for an axisymmetric domain. We have examined several bubbles of different sizes. We report the size by the ratio \( \lambda = d_b/d_c \), where \( d_b \) is defined as the diameter of a spherical bubble with the same volume as the bubble that we are simulating. For \( \lambda > 1 \) the bubbles must be elongated to fit into the pipe. We have done grid resolution studies to find the grid independent solution for the fluid flow. For example for the case with a bubble diameter ratio, \( \lambda = d_b/d_c = 1.199 \), we used 48, 96, and 192 grid cells across the half cylinder diameter and the weighted-average terminal Reynolds number was 354.4, 349.8, and 350.2 respectively, which shows a 1.3% and 0.1% change when we increase the grid resolution from 48 to 96 and from 96 to 192, respectively. Figure 3.2 shows a comparison of the computed bubble shape at non-dimensional time \( t = 7.25 \) for the different resolutions. The top part of the bubble matches perfectly for all three resolutions. At the back of the bubble, we have oscillatory behavior so the shapes differ, but this does not influence the general behavior of the bubbles. We also considered the effect of grid resolution on the mass transfer by studying the effect of grid resolution on the Sherwood number after the bubble reaches its terminal velocity. For three grid resolutions, 48, 96, and 192, the resulting Sherwood numbers are 3480, 3552, and 3588 respectively, which shows 2.0% and 1.0% increases when we increase the grid resolution from 48 to 96 and from 96 to 192, respectively. So, altogether we believe that the resolution of 96 grid cells across the half cylinder diameter gives an essentially grid independent solution. The computation is done with a domain size of \( 0.5d_c \times 12d_c \) on a uniform grid in both directions and with no-slip wall boundary condition on top, bottom, and right wall, and symmetric boundary condition on the left wall.

In Fig. 3.3 the shapes of bubbles with \( \lambda = 0.99 \) and 1.28 from the computation (right) are compared with its counterparts from the experiment (left). The shape
Figure 3.2. A comparison of the bubble shape for different grid resolutions. Here we use 48, 96, and 192 grid cells across the cylinder half diameter.
comparison shows a good agreement between the two. A contour plot of the mass field is also shown. Mass is injected from the boundary layer into the regular domain at regions with large boundary layer thickness (at which points a high concentration of mass is visible), then the deposited mass is advected and diffused in the regular domain by the flow around the bubble. In the case of the Taylor bubbles we generally see one or two coupling points at the back of the bubble. One at the axis of symmetry and the other, which most of the time transfers the largest portion of the mass into the domain, is located somewhere between the axis of symmetry and the rim of the bubble and usually closer to the rim. The location of the latter coupling point changes as the back of the bubble oscillates. The deposited mass is then caught in the vortices at the back of the bubble and advected with the flow.

Figure 3.4 shows the Sherwood number and the Reynolds number of the bubble versus time for the diameter ratio $\lambda = 1.199$. The Reynolds number (top line) reaches a terminal value after a short period of time and while the Reynolds number can be perturbed, like what has happened around time 10-15, as a result of periodic larger oscillations at the back of the bubble, the time averaged Reynolds number remains constant. The Sherwood number (bottom line) is initially very large for a very short period of time that is not visible in this figure due to a sharp concentration gradient at the interface. As the mass concentration in the liquid close to the interface increases, the rate of mass transfer decreases significantly, as seen at time $\approx 1$. Then, as the bubble velocity increases, the flow sweeps the mass close to the interface from the front of the bubble to its back and with the decrease of mass concentration close to the bubble interface, rate of mass transfer increases again and so does the Sherwood number. The Sherwood number increases until the bubble reaches its terminal velocity, after which the mass boundary layer reaches its statistically steady state value. Time averaged values of the Sherwood and the Reynolds numbers are shown as separate horizontal lines over the period that averaging has been done.
Figure 3.3. A comparison of the computational and experimental bubble shape for different bubble diameter ratios, $\lambda = 0.99$ and 1.28. The bottom row shows the original images from the experiment while in the top row the corrected binary shape of the bubble is shown. The red vertical lines represent the edge of the cylinder. This figure also shows the mass contour field behind the bubble.
Figure 3.4. The Sherwood number, \( \text{Sh} \) and the Reynolds number, \( \text{Re} \) versus non-dimensional time for a Taylor bubble with a diameter ratio of \( \lambda = 1.199 \). The dashed line shows the Sherwood number while the solid line shows the Reynolds number. Horizontal straight lines show the time average of the Reynolds and Sherwood number over the time span of the line.

Small wiggles in the instantaneous \( \text{Re} \) number are a result of vigorous oscillations at the back of the bubble.

In Fig. 3.5 (a), the terminal Reynolds numbers for Taylor bubbles with different diameter ratios are shown on the right of the vertical dashed line, both from the computation and the experiment. The results agree reasonably well. The embedded analytical description results on the left of the vertical dashed line are from 3D simulations which are discussed in the next section. In Fig. 3.5 (b), a similar comparison is done for the Sherwood number. In the figure, solid squares show the experimental data while the empty circles represent the result from the computations. The straight line is a linear trend line fitted to the experimental \( \text{Sh} \) and extended for comparison with our results for \( \lambda \) ranging from 0.2 to 1.28. Compared to this trend
line, prediction of the proposed subscale description follows the trend with a relative error of 10% or less.

The results described above suggest that our model can lead to a significant reduction in computational cost for a high Reynolds number, $Re \sim 350$ and very high Schmidt number, $Sc = 8260$. We believe that the error in the description results is mostly from the simple second order profile that we use and it can be reduced by using more sophisticated profiles. Our calculations show that for the parameters studied here, mass boundary layer thickness is generally about 7 times smaller than the flow grid size which means that in order to capture the mass boundary layer accurately we would need to reduce the grid size 15-25 times in order to maintain at least 2-3 grid points in the mass boundary layer. This grid size reduction can lead to enormous computational cost either by using a finer uniform grid or using adaptive mesh refinement (AMR). This can be avoided by using a subscale description like the one we are proposing here.

3.2.2 Ellipsoidal Oscillating Bubbles

When the bubble diameter ratio, $\lambda$, is smaller than about 0.8, it is not feasible any more to assume an axisymmetric bubble. In order to compare the experimental results with the embedded analytical description approach for small diameter ratios, we therefore use the model in a three-dimensional front tracking code. Although the experiment is done in a cylindrical tube, we use a rectangular domain, with no-slip wall boundary condition in all directions. The cross section of the domain is a square selected such that the hydraulic diameter is the same as in the experimental setup. We did a grid resolution study and found that by doubling the grid resolution from 64 to 128 cell across $d_c$ resulted in only 1.5% and 2% changes in $Re$ and $Sh$ numbers, respectively. Therefore we chose a grid resolution of 64 cells across $d_c$ and with a domain size of $d_c \times d_c \times 16d_c$ we use a mesh size of $64 \times 64 \times 1024$. 

49
Figure 3.5. A comparison of the computationally predicted Reynolds number (a) and Sherwood number (b) with experimental results for a range of diameter ratio, $\lambda$, from 0.2 to 1.28. The numerical results to the right of the vertical dashed line are from the axisymmetric code for Taylor bubbles while the results to the left of that line are from the 3D code. $d_b$ is the diameter of a spherical bubble with the same volume as the bubble presented here.
Figure 3.6. Comparison of computational and experimental bubble shape for bubble diameter ratio $\lambda = 0.68$. On the bottom left the original experimental bubble shape is shown while the top left side shows the corrected binary image of the same bubble. On the right-hand side the result of 3D simulation with the same bubble diameter ratio is shown. The bubble surface is colored with the ratio of mass boundary layer thickness to the boundary layer limit, $\delta/\delta_0$. The mass concentration field, $f$, is also shown. Two solid vertical lines on the sides of the bubble and one in the middle are side edges of the cubical domain while the two dashed line is the edges of an imaginary cylinder with the same hydraulic diameter as the cross section of the domain.
A comparison of an experimental bubble shape with computational results is shown in Fig. 3.6. On the bottom left the original shape of the bubble is shown while above it the corrected binary image of it is presented. On the right-hand side, a snapshot from the corresponding simulation is shown. The bubble surface is colored with the ratio of mass boundary layer thickness to the mass boundary layer limit, \( \delta/\delta_0 \). The mass concentration field is plotted for a range of values from 0 to 1, where 1 is the normalized concentration inside the bubble. Two solid vertical lines on the sides of the bubble and one in the middle of the bubble are the edges of the rectangular domain while the two vertical dashed lines are edges of an imaginary cylinder with the same hydraulic diameter as the computational domain.

In Fig. 3.5a, a comparison of the Reynolds numbers from the experiment and the computation, using the 3D code, for different bubble diameters is shown on the left of the vertical dashed line. Here, \( \lambda \) is defined using the hydraulic diameter for \( d_c \). Results from the 3D code show about 11% over-prediction compared to the experimental results. We believe that the difference in the Reynolds number is likely to be caused by the difference in wall effects between the experimental and computational domain. Thus, we see this difference for the ellipsoidal oscillating bubbles where surface tension effects are important but not for the Taylor bubbles where surface tension effects are not as important. We have performed simulation for higher diameter ratio with the 3D code (not showed here) but as the diameter ratio increases above about 0.7, the effect of the wall becomes more important and a comparison with the experimental results become unreasonable. This is clear in Fig. 3.6, where we show the boundaries of the domain and the equivalent hydraulic diameter (dashed line); for bubbles diameter ratios larger than about 0.7 the rim of bubble crosses the equivalent hydraulic diameter which results in different wall effects compared to the experiment. In Fig. 3.5b, on the left-hand side of vertical dashed line, the Sherwood number for different bubble diameters are shown for \( \lambda \) ranging from 0.2 to 0.7 and
the results agree well with the experiment.

### 3.2.3 Comparison with the literature

To further study the performance of the model for fully 3D bubbles, we have also compared its predictions with analytical and semi-empirical correlations, in addition to experimental data from the literature. For simulation in this part, we use less challenging density and viscosity ratio of 10 with a domain size of $3d_b \times 3d_b \times 12d_b$, and grid size of $64 \times 64 \times 256$. We use periodic boundary condition in the direction of gravity and no-slip walls in the other two directions. Calculations are done for Schmidt numbers of 100 and 1000. Other governing parameters are listed in Table 3.1. The comparison of computational results with the ones from correlations and experiments from the literature is shown in Fig. 3.7 and discussed below.

Winnikow (1967) proposed Eq. (3.14) for spherical bubbles with fully developed internal circulation using a boundary layer theory to find the mass transfer,

$$Sh = \frac{2}{\sqrt{\pi}} \left[ 1 - \frac{2.89}{Re^{1/2}} \right]^{1/2} Pe^{1/2}, \quad (3.14)$$

This equation is valid for $Re > 70$, (Clift et al., 1978), and its prediction is shown as dashed line in Fig. 3.7. A similar correlation was suggested by Lochiel and Calderbank (1964):

$$Sh = 1.13 \left[ 1 - \frac{2 + 3\mu_g/\mu_l}{1 + (\rho_g\mu_g/\rho_l\mu_l)^{1/2} Re^{1/2}} \right]^{1/2} Pe^{1/2}, \quad (3.15)$$

which is shown using a dashed-dot line in Fig. 3.7. The model results for spherical bubbles are shown using solid circles, and are in good agreement with Eqs. (3.14) and (3.15) for large Reynolds numbers ($Re > 60$). However, for lower $Re$ numbers the difference between our results and these correlations increases as the $Re$ number decreases since the boundary layer assumption for the velocity profile, used to derive the correlations, is not valid for lower Reynolds numbers.
Takemura and Yabe (1998) studied spherical bubbles and by finding the deviation of $Sh/Pe^{1/2}$ ($Pe = Re \cdot Sc$) calculated using boundary layer theory from their numerical results, they revised the correlation proposed by Lochiel and Calderbank as,

$$Sh = \frac{2}{\sqrt{\pi}} \left[ 1 - \frac{1}{2} \left( \frac{1}{1 + 0.09Re^{2/3}} \right)^{3/4} \right]^{1/2} \left( 2.5 + Pe^{1/2} \right),$$  \hspace{1cm} (3.16)

and comparing with their experimental results, they showed that this correlation is valid for $Re < 100$ and $Pe > 1$. While $Sh/Pe^{1/2}$ is only a function of $Re$ in Eqs. (3.14) and (3.15), in Eq. (3.16) it is also a function of $Pe$ and as a result $Sc$. So here, the correlation is shown for both $Sc = 100$ and $Sc = 1000$ as a dashed-double-dot line and a solid line, respectively. Comparison shows a very good agreement with our results for spherical bubbles both with $Sc = 100$ (open diamonds) and $Sc = 1000$ (open circles) for the range of $Re$ from 20 to 110. The experimental results from Takemura and Yabe (crosses), for spherical oxygen bubble dissolution in silicon oil, are also included and agree well with our predictions.

<table>
<thead>
<tr>
<th>$Re$</th>
<th>19.6</th>
<th>32</th>
<th>51.5</th>
<th>63</th>
<th>85</th>
<th>110</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Eo$</td>
<td>1.0</td>
<td>1.0</td>
<td>1.0</td>
<td>0.8</td>
<td>0.7</td>
<td>0.6</td>
</tr>
<tr>
<td>log $Mo$</td>
<td>-5.5</td>
<td>-6.0</td>
<td>-6.5</td>
<td>-7.0</td>
<td>-7.5</td>
<td>-8.0</td>
</tr>
</tbody>
</table>
Mass transfer from deformable bubbles is different than from non-deformable bubbles. Our calculation for deformable bubbles (not shown here) shows lower Sherwood numbers compared with a spherical bubble with the same Reynolds number. Lochiel and Calderbank (1964) proposed a correlation for the $\frac{Sh_{\text{spheroid}}}{Sh_{\text{sphere}}}$ using a boundary layer approximation for velocity profile around an axisymmetric bubble. Their correlation suggests that for $Re \gg 1$ and $Pe > 1$, the Sherwood number should generally increase with the increase of deformation compared to a spherical bubble, which is opposite to our predictions. However, Figueroa-Espinoza and Legendre (2010) recently performed an extensive numerical study of mass transfer from deformable axisymmetric bubbles for a wide range of Reynolds and Schmidt numbers. Their results show that while deformable bubbles with high $Re$ and $Pe$ numbers show an increase of the Sherwood number compared to non-deformable ones, for low Reynolds numbers ($Re = O(10)$), the Sherwood number decreases with the increase of deformability. This matches with our results for deformable bubbles.

3.3 Summary

Predicting mass transfer from many freely rising bubbles is important for several applications, including bubble columns. While DNS is already providing invaluable insight into the microstructure of bubbly flows and are likely to lead to improved closure models for use in computations of large scale industrial processes, similar simulations of mass transfer are challenging due to the large disparity between the transfer of mass and momentum in liquids. In Aboulhasanzadeh et al. (2012) we introduced a multiscale model, based on an embedded analytical description of the mass boundary layer next to the bubble surface, to allow us to accurately capture mass transfer from bubbles at high Schmidt numbers where the mass boundary layer is much thinner than the momentum boundary layer, without the need to increase the grid resolution. This makes DNS of mass transfer from many freely rising bubbles
Figure 3.7. Comparison of results obtained by the embedded analytical description approach with experimental data and several correlations from the literature, as well as experimental data. Open circles and diamonds represent the calculation for almost spherical bubbles.
feasible. We tested the accuracy of this approach by comparison with fully resolved simulations for two-dimensional flow in Chapter 2. In this chapter we provided further validation by detailed comparison with dedicated experimental studies.

The results from calculation of axisymmetric Taylor bubbles show that using a grid resolution that is enough to resolve the flow, we are able to predict the mass transfer from the bubble for a very large Schmidt number, $Sc = 8260$, using the embedded analytical description proposed here relatively accurately. The agreement can be improved by using more sophisticated profile than the simple second order one that we have used here. Based on the minimum mass boundary layer thickness around the bubble, we estimate that this subscale model leads to a reduction of computational cost of one or two order of magnitude. This is particularly important when dealing with large systems of interacting bubbles. Three-dimensional calculations of ellipsoidal oscillating bubble also show good agreement with the experimental results, further confirming the validity of using our subscale description. We also compared the results from the three-dimensional code with analytical and semi-empirical correlations from the literature, as well as experimental results from other authors, and found good agreement.

In general, our studies show that the embedded analytical description approach proposed here can be an essential element for significantly decreasing the computational cost of simulating mass transfer at the interface of multiphase flow.
CHAPTER 4

EFFECT OF BUBBLE INTERACTIONS ON MASS TRANSFER

The effect of bubble interactions on mass transfer in a multi-bubble system is examined by numerical simulations. Here, we use the multiscale approach, introduced in previous chapters, for the computations of the mass transfer near the bubble surface, in order to reduce the cost, and examine the effect of void fraction and bubble Reynolds number on the mass transfer from bubbles in periodic domains. Specifically, we compare results for a single bubble in a periodic domain with results for several bubbles in a larger domain with the same void fraction. It is shown that even though the average Reynolds number of freely moving bubbles drops after a while, in most cases the mass transfer from the bubbles increases slightly. When bubbles start wobbling, in most cases the increase in bubble-bubble interaction compensate for the decreased Reynolds number.

Considerable research has already been done on the mass transfer from a single bubble, see, Davidson and Rudman (2002); Deshpande and Zimmerman (2006); Figueroa-Espinoza and Legendre (2010); Ganguli and Kenig (2011a); Hayashi and Tomiyama (2011); Juncu (2011); Jung and Sato (2005); Ponoth and McLaughlin (2000); Wang et al. (2008). Most of these studies focused on two-dimensional or axisymmetric flow. Because of the large disparity between length and time scale for the mass transfer compared to the momentum transfer, fully resolved three-dimensional calculation of mass transfer are a challenging undertaking and those available in the literature are done for very low Schmidt number, $Sc = O(1)$, Darmana et al. (2006); Onea et al. (2009).
While much has been done to understand mass transfer from a single bubble, both analytically and numerically, only a limited number of studies have looked at the interaction of bubbles and the effect of void fraction on the mass transfer. Koynov et al. (2005) studied mass transfer and reactions in a multi-bubble system and concluded that for mixing-sensitive reaction networks the interaction of the bubbles impact the reaction selectivity significantly, and Radl et al. (2008) performed simulations of single and multiple bubbles in order to investigate the effect of different Hatta and Schmidt numbers on the catalytic hydrogenation of nitroarenes. Both of these studies examined only two-dimensional systems.

Here, we use the multi-scale method presented in Chapter 2 and further validated with experimental data in Chapter 3 to study the mass transfer in bubble clusters, including the effect of different void fractions and Reynolds numbers.

4.1 Problem Setup

The simulations discussed in this chapter are done using a three-dimensional front-tracking/finite-volume method using our subscale model for the mass transfer, discussed in Chapter 3. Here we assume that the mass concentration outside the bubble is sufficiently low so that it can be neglected when we compute the mass diffusion out from the bubble. In general the mass transfer should depend on the concentration in the surrounding liquid and for bubbles moving in a periodic domains this should increase continuously as the bubbles loose mass. Although accounting for the non-zero concentration in the liquid is, we believe, straightforward in our method, the resulting decrease in mass transfer would prevent us from computing the average mass transfer over a long time. Thus, we ignore the concentration in the ambient liquid when making the boundary layer assumption.
4.2 Results and Discussion

After an extensive testing of the model for two- and three-dimensional flows, here we study the behavior of bubble arrays and the effect of bubble interaction on the mass transfer. We examine eight cases of freely moving bubbles with different governing parameters in a computational domain that is a cubic box with a periodic boundary condition in all directions. The bubbles are initially distributed on a regular grid such that each bubble is an equal distance from their vertical and horizontal neighbors, perturbed in such a way that the initial array breaks up and the bubbles interact freely. To compare the behavior of freely moving bubbles, we have also done similar calculations for one bubble in a small domain with the same void fraction. We refer to these simulations as fixed array calculations, since the horizontal and vertical distance between neighboring bubbles in the periodic domain always remain the same. In Fig. 4.1 we show one frame from a simulation of the regular bubble array on the left and from a simulation with several bubbles on the right. In both cases the bubbles and the concentration of mass in the liquid is shown after the bubbles have risen several bubble diameters. For the regular array it is clear that the concentration is highest in the region between two bubbles rising one behind another, but for the freely evolving array the distribution is more non-uniform. The non-dimensional governing parameters in these simulations are the void fraction, $\epsilon = 9\%$, the Morton number, $Mo = 10^{-6.5}$, and the Eötvös number, $Eo = 5.0$. For all the calculations, here, we use a density and viscosity ratio of 10 and we set the Schmidt number to be 100.

In this study we have done simulations in two regions of parameter space; for deformed bubbles with $Eo = 5.0$ and Morton numbers from $10^{-6.5}$ to $10^{-4}$, and for nearly spherical bubbles with $Eo = 0.2$ and Morton numbers from $10^{-8}$ to $10^{-6}$. In all cases we examine three different void fractions, $\epsilon = 4.0\%, 9.0\%$, and $15.5\%$. Fig. 4.2(a), top frame, shows the Reynolds number versus time for a fixed array
Figure 4.1. Comparison of mass distribution in the domain for the freely and fixed moving arrays of bubbles for the case with $\varepsilon = 9.0\%$, \( \log Mo = -6.5 \), \( Eo = 5.0 \), and at non-dimensional time, \( t = 27.9 \).
simulation with 9% void fraction, $Eo = 5.0$, and $Mo = 10^{-4}$. For this low Reynolds number the bubbles approach terminal velocity monotonically and reach it after time equal to about 20. After that the bubbles rise straight up with a constant $Re$ number and fixed shape. In bottom frame of Fig. 4.2(a) the Sherwood number for the same bubble is plotted versus time. Except for a short initial transient, the Sherwood number also reaches a steady state quickly. Initially, since the bubble is in a clean fluid, the Sherwood number is large but as a mass transfer boundary layer forms next to the bubble, the Sherwood number decreases rapidly. As the bubble rises, the mass that diffuses away from the bubble is swept to the back and into the wake. Eventually the rate of mass diffusion from the bubble is balanced by the advection of mass from the bubble, resulting in a constant Sherwood number.

Figure 4.2(b), top frame, shows a high Reynolds number case in which all the parameters remain the same as in Fig. 4.2(a) except for the Morton number which is decreased to $10^{-6.5}$. Here the bubble velocity increases until its motion becomes unsteady and its velocity decreases by about 25%. Although its average velocity is well defined, the bubble settles down into a persistant wobbly motion. The time averaged Reynolds number is shown as a dash-dot straight line in the plot over the period of averaging. The Sherwood number, as shown in the bottom frame of Fig. 4.2(b), is also oscillatory as a result of the oscillations in the Reynolds number but just as the Reynolds number, the time averaged Sherwood number is a constant, which is shown as a dash-dot line.

In Fig. 4.3(a) the Reynolds and Sherwood number for the freely evolving array, corresponding to the regular array in Fig. 4.2(a), is plotted versus time. In contrast to the fixed array case the initial configuration of the bubbles in the free array breaks up and the bubbles interact with each other in an unsteady way. As a results the time-averaged free array Reynolds number, shown as a dash-dot horizontal line, is about 16% smaller than the time average for fixed array Reynolds number, which is shown as
Figure 4.2. Reynolds and Sherwood number versus time for the fixed moving array with $\varepsilon = 9.0\%$, $Eo = 5.0$. (a) is for a bubble with $\log Mo = -4.0$ and (b) is for a bubble with $\log Mo = -6.5$. The horizontal dash-dot lines show the time-averaged Reynolds and Sherwood number, over the span of the line on the corresponding graph.
a dashed horizontal line. Even though the average Reynolds number for the free array
is significantly lower than for the fixed array case, the average Sherwood number is
approximately the same, or only 1% lower. Thus, even though the Reynolds number
decreases, the increase in interactions between the bubbles compensate in order to
keep the mass transfer similar. Figure 4.3(b) shows the Reynolds and Sherwood
number versus time for a free array with parameters corresponding to those of the
fixed array in Fig. 4.2(b). In this case the average Reynolds number for the free array
is only 9% lower than for the fixed array. As a result, the increase in the interaction
between the bubbles results in a 6% increase of the average Sherwood number. So
even though the Reynolds number decreases, the mass transfer increases as a result
of unsteady bubble interaction.

In domains with freely rising bubbles we often observe clustering. Bunner and
Tryggvason (2003) analysis of micro-structures of bubble distributions shows a ten-
dency of deformable bubbles to collect in vertical “stacks” or “chimneys” while spheri-
cal bubbles have a tendency to form horizontal “rafts.” Figure 4.4 shows an example
of horizontal clustering of bubbles for a freely moving array with 8 bubbles and
$Eo = 0.2$, $Mo = 10^{-8}$, and a void fraction of 15.5%. Here, the average Reynolds
number (dashed line) and the average Sherwood number (solid line) of the bubbles
are shown versus the average location of bubbles on the right side. The location
is non-dimensionalized with the height of the domain. On the left side a snapshot
of the bubbles is shown at 9 different times which correspond to the 9 locations on
the right hand side of the plot. The bubble accelerate until they reach their maxi-
mum average velocity at about location 2, then the average Reynolds number start
decreasing gradually because of bubble interactions and wobbling until the bubbles
reach location 5 where they start to align horizontally in two planes (actually one
plane, since the planes are a domain size apart). When the bubbles start to form
a horizontal cluster we see a large drop in the Reynolds number, which results in a
Figure 4.3. Reynolds and Sherwood number versus time for the freely moving array with $\varepsilon = 9.0\%$, $Eo = 5.0$. (a) is for array of bubbles with $\log Mo = -4.0$ and (b) is for array of bubbles with $\log Mo = -6.5$. The horizontal dash-dot lines show the Reynolds and Sherwood number of freely moving array, time-averaged over the span of the line on the corresponding graph. The dashed lines show the statistically steady state Reynolds and Sherwood number for the respective fixed array simulation.
considerable drop of the Sherwood number (about 16%). As explained by Roghair et al. (2013), the clustering we see here may be a result of the domain size, since $n_b/n_{b,max}$ (where $n_b = 8$, and $n_{b,max}$ is the maximum number of bubbles that can align in a horizontal plane, calculated for this case using Eqn. 6 of Roghair et al. (2013) to be 9.5) is about 0.84, which is close to 1. That means that we may expect to see bubbles clustered in one horizontal plane. However, even if this clustering is a result of the small domain size, it shows that clustering can have a considerable effect on the mass transfer.

The average rise Reynolds and Sherwood numbers for nearly spherical bubbles ($Eo = 0.2$) and deformable ones ($Eo = 5.0$) are plotted in Fig. 4.5 for different Morton numbers and three different void fractions. The top frames show the rise Reynolds numbers and the bottom frames show the Sherwood number. The left column shows results for nearly spherical bubbles and the right column shows results for more deformable ones. The dashed line connects the values for the fixed array and the solid line connects the values for the free values. As expected, the rise velocity of both nearly spherical and deformable bubbles increases with decreasing Morton number and void fraction. The average rise velocities of the freely interacting bubbles are generally smaller than in the fixed array, as examined in more detail in the next figure. The Sherwood number also increases with decreasing Morton number and void fraction.

Figure 4.6 shows the percentage differences between the results for the fixed and the free arrays, plotted in the same way as in Fig. 4.5. For the nearly spherical bubbles the difference increases with the Morton number and while the results for the lowest void fractions are similar, the differences rise more sharply for the highest void fraction. For the lowest Morton numbers the difference is rather significant, with the regular array rising significantly faster than the free one. This is due to the tendency of the nearly spherical bubbles to form horizontal rafts. As the Morton
Figure 4.4. Average Sherwood and Reynolds number of 8 freely moving bubbles in a periodic domain versus average vertical location of their center of gravity. On the left, 8 snap shots of the bubble positions are shown which corresponds to the locations and values on the right figure. The solid lines represent average Sherwood number while the dashed line shows the average Reynolds number. The vertical solid and dashed lines are the time-average of Sherwood and Reynolds number for the corresponding fixed array, respectively. The non-dimensional parameters are \( \epsilon = 15.5\% \), \( Mo = 10^{-8} \), and \( Eo = 0.2 \).
Figure 4.5. Reynolds and Sherwood number for the freely moving array of bubbles and the fixed one for three different void fraction, $\epsilon$. Left column is for non-deformable bubble, $Eo = 0.2$, while right column is for deformable bubble, $Eo = 5.0$. 
number increases and the bubbles become more deformable this trend diminishes and the difference does also. The freely moving deformable bubbles also rise slower than those in a fixed array but there is no clear dependency on the Morton number. The difference does, however, decrease as the void fraction increases. Although the rise velocity of the bubbles depends strongly on whether they are allowed to interact freely or held fixed, in most cases, the effect on the Sherwood number is much smaller. Only for the lowest Morton number and the highest void fraction, for the spherical bubbles, is there a significant difference in the Sherwood number. This is a result of the formation of horizontal rafts as seen in Fig. 4.4.

Intuition suggests that the mass transfer should generally increase with increasing rise velocity and the more unsteady the bubble motion is. This is, indeed, what the data shows. For both the fixed and the free arrays, Fig. 4.5 shows that the Sherwood number increases with the Reynolds number. Comparing the free and the fixed arrays should, in principle, allow us to assess the effect of the fluctuating motion, but the interpretation of the results is complicated by the fact that the rise velocity is reduced. To compare the mass transfer for free and fixed array for spherical bubbles at the same Reynolds number, we need to look at different Morton numbers. Drawing a horizontal line from the first black square in the frame for the Reynolds number for nearly spherical bubbles, for example, intersects the line dashed for the fixed array about a quarter of the way across the plot ($\Delta \log Mo = 0.5$), which give a difference of about 10 in the Sherwood number from the graph below, even though the solid and the dashed line for the Sherwood number in that frame practically coincide. Comparing other points in the graphs in a similar way suggest that for the results presented here, this is generally the case, particularly for the lower Morton numbers. Mass transfer increases with both bubble rise velocity and fluctuations, but when going from a fixed array to a freely moving one the velocity decreases but the fluctuations increase, producing changes in the opposite direction that for the
Figure 4.6. Percentage of the Reynolds and Sherwood number difference between the freely moving array of bubbles and the fixed ones for three different void fraction, $\epsilon$. Left column is for non-deformable bubble, $Eo = 0.2$, while right column is for deformable bubble, $Eo = 5.0$. 
most part cancel each other, as far as the mass transfer is concerned. Although this makes for less interesting results for a scientific publication, the practical implication is actually significant and positive. For bubbles in the parameter range studied here, results for bubbles in a fixed array should give us a reasonable estimate of the mass transfer, alleviating the need for large simulations of many freely interacting bubbles.

4.3 Summary

Here we study the mass transfer of freely moving bubbles and compared them to bubbles in fixed arrays for two different flow regimes: deformable and non-deformable bubbles. In each regime we examined three different void fraction, 4.0%, 9.0%, and 15.5%. The results show that the mass transfer decreases as the Reynolds number decreases and increases as the velocity fluctuations increase, when we go from a fixed array to freely moving bubbles. These two effects tend to balance each other, at least partially. For most cases, except for very low Reynolds numbers, we see a decrease in the Reynolds number but because of increase in bubble interaction we see only a minor increase in the Sherwood number, less than 10 percent. Where we see a decrease in Sherwood number, the difference is less that 5%, except when we see horizontal clustering of the bubbles, for which we see a 17% decrease in the Sherwood number. The effect of void fraction on the mass transfer for deformable bubbles is negligible. However, for very low Reynolds number non-deformable bubbles, because of the increase of the Reynolds number as a function of void fraction, we see considerable increase of the Sherwood number when the void fraction increases and the Reynolds number decreases. For modest Reynolds number the effect of the void fraction is not significant.

We note that here, we have not accounted for the fact that increased mass concentration in the liquid, due to mass transfer from the bubbles, will eventually result in a reduced mass transfer. Here we assume that the mass concentration is zero, outside
the boundary layer. Accounting for this concentration in our approach is relatively
straight forward, but doing so changes the environment in which the bubbles are
moving with time and would make time averages meaningless.
CHAPTER 5

CAPTURING THIN FILM BETWEEN COLLIDING NON-COALESING DROPLETS

In this chapter, a multi-scale approach is used to capture the thin film that appears between two drops sliding over each other. In this approach the points on two adjacent interfaces which are closer than one grid size are detected. Then a coupling force is calculated and added to the Navier-Stokes equations to account for the viscous forces which are not resolved in the thin film region.

We show the effect of low resolution using a 2D problem. With the use of a very high resolution the film is correctly captured and then it is compared to calculations done using a low resolution, with the subscale description that is presented here. The results shows considerable improvements.

5.1 Computational Approach and Multiscale Model

In order to resolve the film the following steps are needed, as explained in the subsequent subsections:

1. detecting the interface points attached to the film
2. finding and updating the film thickness for each front point
3. calculating and adding the sub-scale shear forces

5.1.1 Detecting the Interface Points Attached to the Film

In the front tracking method that is used in this calculations an indicator function, $I(x, y)$, is used to reconstruct fluid properties like the density and the viscosity at...
each time step. The indicator function is a function of position and is equal to 1 inside the drop and 0 in the surrounding fluid. Transition from 1 to 0 occurs smoothly over 2 cells with the interface in the middle. This means that normally the value of the indicator function is about 0.5 at the interface. However, when the two interfaces gets close to each other, as they do when there is a thin film, the magnitude of the indicator function becomes larger than 0.5. We use this to find the front points which are attached to a thin film.

In order to capture the front points which are in the film, the indicator function is first reconstructed on the grid. Then, using a Peskin interpolation, the magnitude of the indicator function is calculated on every front point. If the indicator function at a front point has a value of more than 0.5, it means the two interfaces are closer than three grid cell at that point and the subscale model is needed.

5.1.2 Finding and Updating the Film Thickness for Each Front Point

In order to find and update the film thickness for each front point, if a front point was not attached to a film at the previous time step and now is attached, we search for the closest front point on another interface and then find the closest distance to that interface from that point. In order to reduce the computational effort we only calculate the distance for points which are in a small box around the point of interest.

For the following time step, since we know that this point is attached to a thin film, an equation of conservation of mass is solved to update the film thickness, $h$. To derive this equation we write the conservation of mass for a control volume shown in Fig. 5.1.

$$\rho \bar{u}_s h - (\rho \bar{u}_s h + \Delta (\rho \bar{u}_s h)) = \Delta s \frac{\partial}{\partial t} (\rho h), \quad (5.1)$$

where $\bar{u}_s$ is the average tangential velocity. Since $\rho$ is constant inside the film, rear-
ranging and simplifying yields,

\[ \Delta s \frac{\partial h}{\partial t} = -\Delta(\bar{u}_s h), \]

as \( \Delta s \to 0 \) the equation for film thickness can be written as,

\[ \frac{\partial h}{\partial t} = -\frac{\partial(\bar{u}_s h)}{\partial s}. \] (5.2)

Figure 5.1. Schematic of two drops sliding over each other. The dashed line shows the locus of middle points between the two interfaces. The control volume shown on the right is used for the derivation of equation for the evolution of film thickness.
5.1.3 Calculating and Adding the Subscale Shear Forces

The momentum equation for the tangential component of the velocity, for a control volume along the interface, can be written as:

\[ \rho \frac{Du_s}{Dt} = \rho g_s - \frac{\partial P}{\partial s} + \frac{\partial}{\partial s} \left( \mu \frac{\partial u_s}{\partial s} \right) + \frac{\partial}{\partial n} \left( \mu \frac{\partial u_s}{\partial n} \right). \]  

(5.3)

Since the grid is not resolving the film, instead of \( \mu_f \), \( \mu_f \) is used for the calculation of viscous terms on the grid. Change of \( u_s \) in the \( s \) direction is smooth, so the error from viscous term in the \( s \) direction would be small. However, in the normal direction there is a large velocity gradient because of low viscosity in the film compared to viscosity inside the drops. To correct this under-resolved viscous term, \( \mu_f \) can be written as \( \mu_f = \mu_o + \Delta \mu \), where \( \Delta \mu \) is the viscosity difference between fluid outside and inside the drop. Using this decomposition for the fluid inside the film we have:

\[ \frac{\partial}{\partial n} \left( \mu_f \frac{\partial u_s}{\partial n} \right) = \frac{\partial}{\partial n} \left( \mu_o \frac{\partial u_s}{\partial n} \right) + \frac{\partial}{\partial n} \left( \Delta \mu \frac{\partial u_s}{\partial n} \right) \]

(5.4)

where \( S = \Delta \mu \frac{\partial u_s}{\partial n} \). The first term on the right hand side of Eq. (5.4) is the part of the stress forces that is resolved on the coarse grid inside the thin film. In order to correct the viscous term we need to calculate and add the second term on the right hand side to the Navier Stokes equations.

Knowing \( S = \Delta \mu \frac{\partial u_s}{\partial n} = \Delta \mu \frac{\Delta u_s}{h} \), given the thickness of the film, the velocity jump across the film is needed in order to estimate \( S \); the method for calculating velocity jump is presented in Section 5.1.3.1. Once \( S \) has been found, it’s derivative will be added as a force term to the nodes next to the interface. This approach is suitable for simple horizontal or vertical interfaces, similar to the simple example in Section 5.2.1.
In order to generalize this approach we write the momentum equation in tensor form,

\[
\frac{du}{dt} = g - \frac{1}{\rho} \nabla P + \frac{1}{\rho} \nabla \cdot \mu \tau,
\]

where \( \tau \) is the strain rate tensor,

\[
\tau = \begin{bmatrix} 2 \frac{\partial u}{\partial x} & \frac{\partial u}{\partial y} + \frac{\partial v}{\partial x} \\
\frac{\partial u}{\partial y} + \frac{\partial v}{\partial x} & 2 \frac{\partial v}{\partial y} \end{bmatrix}, \tag{5.5}
\]

Near the interface the viscous term, \( \nabla \cdot \mu \tau \), is under-resolved on a coarse grid. The correction term \( \nabla \cdot (\Delta \mu \tau) \) needs to be added to the node next to the interface to correct the viscous term. In order to estimate \( \tau \) inside the film, a linear velocity profile is assumed as shown in Fig. 5.2. We use a transformation from \( ns \) coordinate to \( xy \) coordinate. Then by inserting the velocities in Eq. (5.5), it gives,

\[
\tau = \begin{bmatrix} -2 \frac{du_s}{dn} \sin \theta \cos \theta & \frac{du_s}{dn} \left( \cos^2 \theta - \sin^2 \theta \right) \\
\frac{du_s}{dn} \left( \cos^2 \theta - \sin^2 \theta \right) & 2 \frac{du_s}{dn} \sin \theta \cos \theta \end{bmatrix},
\]

where \( \theta \) is the angle between a tangent to the interface with the horizontal axis, and \( \frac{du_s}{dn} \) is the rate of change of tangential velocity in the direction normal to the interface, inside the thin film, which is calculated by calculating the velocity jump in the film, explained in the next section. We calculate \( \tau \) on the front points, attached to the film, and then distribute it to the grid using area-weighting interpolation. Having \( \nabla \cdot (\Delta \mu \tau) \) can be easily calculated and added to the momentum equation.
5.1.3.1 Estimating the Velocity Jump Across the Film

In order to estimate the velocity jump across the film, the simplest approach would be to interpolate the viscous stresses outside the film and then use the continuity of the viscous stresses to calculate the viscous stress inside the film and the velocity jump. Our study showed that while this approach gets correct result for a time independent problem, for a transient problem like the one we want to solve here, it can introduce large errors.

In order to include the transient behavior of the film and the fluid next to it, we use a three-layer profile as shown in Fig. 5.3. In this approach we assume a second order velocity profile on each side of the film (in the droplets), and a third order
profile inside the film. The equations for velocity profiles are,

Top profile: \[ u_t(y) = U_t + \frac{\tau_t}{\mu_o} (y - \Delta) + a(y - \Delta)^2, \quad (5.6) \]
Middle (Film) profile: \[ u_f(y) = U_0 + \frac{\tau_0}{\mu_f} y + cy^2 + dy^3, \quad (5.7) \]
Bottom profile: \[ u_b(y) = U_b + \frac{\tau_b}{\mu_o} (y + \Delta) + b(y + \Delta)^2, \quad (5.8) \]

where \( U_t \) and \( U_b \) are the tangential velocities while \( \tau_t \) and \( \tau_b \) are the tangential viscous stresses, all interpolated inside the droplets at distance \( \Delta \) from the interfaces, \( n = \pm \Delta \). To fix the three-layer profile, we need 6 equations to specify 6 unknowns: \( a, b, c, d, U_0, \) and \( \tau_0 \). Continuity of velocity and continuity of viscous stress across interfaces give us 4 equations,

\[
\begin{align*}
\left. u_t \right|_{\frac{\delta}{2}} & = \left. u_f \right|_{\frac{\delta}{2}}, \\
\left. u_b \right|_{-\frac{\delta}{2}} & = \left. u_f \right|_{-\frac{\delta}{2}}, \\
\mu_f \left. \frac{du_f}{dy} \right|_{\frac{\delta}{2}} & = \mu_o \left. \frac{du_t}{dy} \right|_{\frac{\delta}{2}}, \\
\mu_f \left. \frac{du_f}{dy} \right|_{-\frac{\delta}{2}} & = \mu_o \left. \frac{du_b}{dy} \right|_{-\frac{\delta}{2}}.
\end{align*}
\]

(5.9) \quad (5.10) \quad (5.11) \quad (5.12)

We can get two more equations using the tangential momentum equation, simplified by the assumptions that the normal velocity is zero and the tangential rate of change of tangential velocity is negligible,

\[ \frac{\partial u}{\partial t} = \frac{\mu \partial^2 u}{\rho \partial y^2} - \frac{1}{\rho} \frac{dP}{dx}. \quad (5.13) \]

By integrating Eq. (5.13) from \(-\delta/2\) to \(\delta/2\) we get,

\[ \frac{d}{dt} \left\{ U_0 \delta + \frac{c}{12} \delta^3 \right\} = 2 \frac{\mu_f}{\rho_f} c \delta - \frac{\delta}{\rho_f} \frac{dP}{dx}. \quad (5.14) \]
Integrating the first moment of Eq. (5.13) from $-\delta/2$ to $\delta/2$ gets us the last equation to close the system of equation,

$$
\frac{d}{dt} \left\{ \frac{\tau_o}{12 \mu_f} \delta^3 + \frac{d}{80} \delta^5 \right\} = \frac{1}{2} \frac{\mu_f}{\rho_f} \delta^3 d.
$$

(5.15)

Solving Eqs. (5.9) to (5.12) for $a$, $b$, $c$, and $d$ we get,

$$
c = \frac{4 \left( U_b - 2U_0 + U_t \right) \mu_o + (2\Delta - \delta) (\tau_b - \tau_t)}{2\delta \left( \delta \mu_o + (2\Delta - \delta) \mu_f \right)},
$$

(5.16)

$$
d = \frac{2 \left( 4 \left( U_b - U_t \right) \mu_o \mu_f + 4 \mu_o \tau_o + (2\Delta - \delta) \mu_f \left( \tau_b + 2\tau_o + \tau_t \right) \right)}{-\delta^2 \mu_f \left( 2\delta \mu_o + 3 (2\Delta - \delta) \mu_f \right)}.
$$

(5.17)

To simplify the representation, we write $a$ and $b$ as a function of 4 other coefficients, including $c$ and $d$,

$$
a = \frac{\delta (4c + 3d) \mu_f + 4 (\tau_o - \tau_t)}{4(\delta - 2\Delta) \mu_o},
$$

(5.18)

$$
b = \frac{\delta (4c + 3d) \mu_f + 8\tau_o - 4 (\tau_b + \tau_t)}{4\delta \mu_f}.
$$

(5.19)

Assuming that all the variables in Eqs. (5.16) and (5.17), except for the 6 unknown coefficients are independent of time, by taking derivative versus time we get:

$$
\frac{dc}{dt} = -\frac{8\mu_o}{2\delta \left( \delta \mu_o - (\delta - 2\Delta) \mu_f \right)} \frac{dU_0}{dt},
$$

(5.20)

$$
\frac{dd}{dt} = \frac{4 (2\delta \mu_o - \delta \mu_f + 2\Delta \mu_f)}{-\delta^2 \mu_f \left( -2\delta \mu_o + 3 (\delta - 2\Delta) \mu_f \right)} \frac{d\tau_o}{dt}.
$$

(5.21)

Inserting Eqs. (5.16) and (5.17) into Eqs. (5.14) and (5.15), respectively, and simplifying we get,

$$
\frac{dU_0}{dt} = \frac{6 \left( \delta \mu_o - (\delta - 2\Delta) \mu_f \right)}{6 \left( \delta \mu_o - (\delta - 2\Delta) \mu_f \right) - 2\delta \mu_o} \left( \frac{2\mu_f}{\rho_f} c - \frac{1}{\rho_f} \frac{dP}{dx} \right),
$$

(5.22)

$$
\frac{d\tau_o}{dt} = \frac{15 \mu_f^2 \left( -2\delta \mu_o + 3 (\delta - 2\Delta) \mu_f \right)}{2\rho_f \left( -\delta \mu_o + 3 \delta \mu_f - 6\Delta \mu_f \right)} d.
$$

(5.23)
We use these equations to evolve $U_0$ and $\tau_0$, and then use Eqs. (5.16) to (5.19) to calculate the other unknown coefficients, $a-d$.

![Schematic of three-layer profile](image)

Figure 5.3. Schematic of three-layer profile for the approximation of velocity inside and adjacent to the thin film.

5.2 Results

5.2.1 A Simple Example

First, we examine how the subscale model performs on a simple test case. In the 2D domain shown in Fig. 5.4, horizontal walls move in opposite directions with unit velocity. Three layers of two different fluid lays between the two walls with the less viscous one in the middle. The interface between the fluids remains horizontal so the
vertical velocity remains zero. In order to capture the velocity profile between the two wall, a one dimensional momentum equation is solved,

\[ \rho \frac{du}{dt} = \frac{d}{dy} \mu \frac{du}{dy} \tag{5.24} \]

The height of the domain is set to 2 and the thickness of lower viscosity fluid set to 0.08. Initially we set the velocity equal to 1 in the top fluid, -1 in the bottom fluid, and a linear distribution in the middle fluid. The viscosity ratio is set to 50 with \( \mu_f = 0.01 \) and \( \mu_o = 0.5 \). The density of two fluids are set to 1. A very fine grid is used to correctly capture the velocity jump across the thin film, 3024 point in \( y \) direction. The low resolution calculation in which the thickness of the thin film is less than grid spacing, is done with 24 point in the \( y \) direction. To resolve the viscous forces in the thin film, a force term in the direction of velocity is added to the nodes next to the film with the magnitude equal to,

\[ (\mu_f - \mu_o) \frac{\Delta u}{h^2} \]
where $h$ is the horizontal spacing and $\Delta u\big|_{Model}$ is the velocity jump across film, calculated using the model.

Figure 5.5 shows the velocity profile for the steady state solution using a constant viscous stress approximation. Because of the low resolution, calculation without the model reaches a linear profile while the low resolution calculation with the model matches the benchmark solution. Figure 5.6 shows the solution for three different time, $t = 0.004, 0.008, \text{ and } 0.016$, from top row to bottom row. On the left column constant viscous stress approximation is used for the model results while in the right column the three-layer approximation is used. The result for both approaches shows improvement over the coarse calculation without the model. The model with constant viscous stress assumption is not initially capturing the film correctly while the three-layer profile gives very good results in the transient solution. Since in colliding droplets the collision time is usually a short interval, the three-layer profile is suitable to capture the transient solution correctly.

5.2.2 Colliding Droplets

A film between two drops can appear whenever two drops slide over each other but in order to isolate the problem and study the film issue we define the following problem. We study two drops with a diameter $d_d$ for which their center of mass are initially $\frac{12}{7}d_d$ and $\frac{34}{35}d_d$ apart in the $x$ and $y$ directions, respectively, as shown in Fig. 5.7. Initially, the drops are symmetrically placed in a $\frac{40}{7}d_d \times \frac{20}{7}d_d$ periodic domain. At the beginning, the velocity inside the drops are set to $u_x = 1$ and $u_x = -1$ in the left and right drop, respectively. The other parameters used for the calculations are set in order to have density ratio, $r_r = \rho_o/\rho_f = 50$, viscosity ratio, $\mu_r = \mu_o/\mu_f = 50$, and Weber number, $We = \rho_o u^2 d_d/\gamma = 0.25$.

Figure 5.8 shows the horizontal velocity of the left droplet versus time. From time equal 0.5 to 0.8 a thin film appears between droplet which results in sudden decrease
Figure 5.5. Steady state velocity profile between opposite moving walls with a thin less viscous film in the middle. Red solid line shows the benchmark solution, blue line with circles shows the model with constant viscous stress assumption, and black dashed line shows the coarse calculation without model. Horizontal black dotted lines show the edge of thin film.
Figure 5.6. Velocity profile between opposite moving walls with a thin less viscous film in the middle, at three different times, \( t = 0.004, 0.008, \) and \( 0.016 \). The left column shows the model solution using a constant viscous stress assumption and the right column shows the model solution using three-layer profile approach. Red solid line shows the benchmark solution, blue line with circles shows the model, black dashed line shows the coarse calculation without model, and the green dotted line shows the three-layer profile. Horizontal black dotted lines show the edge of thin film.
Figure 5.7. Schematic of two drop with an initial velocity opposite to each other and with the same magnitude.

of horizontal velocity. On the coarse grid since the viscous terms are not resolved correctly, this velocity drop is about twice the actual value calculated from the fine calculation. The model result is shown by dotted line, which improves the results when the film appears. While the model is improving the result during the existence of a thin film, a convergence issue prevents the model to show a better result. The convergence issue prevents getting the same results when the thin film does not exist and makes the drop with coarse grid to see a higher deceleration compared to the fine calculation. We expect to get better results once this issue is resolved. The droplet position at time equal 1 is shown in Fig. 5.9 which shows that the model closely follows the fine solution while the coarse solution without model is left behind because of extra momentum drop. This difference increases for higher viscosity ratio and when the droplets remain in contact for longer period. In larger calculations when lots of droplets interacting with each other this under-resolved solutions can lead to larger errors in predictions.
Figure 5.8. Left droplet horizontal velocity versus time for coarse grid calculation, dashed line, fine grid calculation, solid line, and coarse grid calculation with the model, dotted line.
Figure 5.9. Comparison of droplet position after they passed each other. Black droplet shows the fine calculation, while green and red droplets show the coarse grid calculation with and without the model, respectively.
5.3 Summary

In this chapter we develop a subscale model to recover the under-resolved viscous terms in regions of the domain containing thin films between colliding droplets. An indicator function is used for the detection of front point bounding a thin film. By estimating the velocity jump across the thin film, the correct viscous terms are estimated and the difference with the under-resolved viscous term is added to the momentum equation. In order to estimate the velocity jump in the film, two approaches are tested. In the first approach, the normal viscous stress is assumed to be constant inside and close to the thin film and by interpolating the viscous stress outside the film, the velocity jump is estimated. This approach works well for long term or steady state solution. While the profile in the thin film approaches a linear profile, which makes a constant viscous stress a good assumption inside the film, outside the film the profile is usually non-linear and the estimation of the viscous stress is position dependent. To solve this issue we propose a three-layer profile for the velocity inside and next to the film. Using a cubic profile inside the film and two quadratic profiles next to the film, we use continuity of velocity and viscous stresses across the interface in addition to tangential momentum equation and its first moment inside the film, to solve for the unknown coefficients of the profiles. Then, using the profile we estimate the velocity jump across the film. The model is first tested on a simple case which shows that the three-layer profile can predict the film very well. The model is also tested using a 2D front tracking code in which the result show considerable improvement over the coarse grid results. There are still some remaining issues with the convergence of the code when droplets are far from each other, which affect the performance of the model.
CHAPTER 6

CONCLUSIONS

Bubble columns are one of the most important chemical processing units in the petro-chemical industry and are used to process millions of tons of material every year. Bubble columns generally are simple vertical cylinders containing liquids with bubbles injected at the bottom. The bubbles rise due to buoyancy, interact and mix the liquid and the dissolved gases transferred from bubbles into the surrounding liquid, increasing the reaction rate. In spite of their simple design, the physics in bubble column reactors is complex. Today, we rely mostly on experimental data and semi-analytical correlations for the design of bubble columns, but with increased computational power and the ability to use direct numerical simulation for understanding of multiphase flow, computational studies should be able to help improve the design and optimization of bubble columns.

In this thesis a subscale model for the simulation of mass transfer in bubbly flow was developed where we use a parabolized mass transfer equation for the evolution of mass boundary layer next to the bubble interface. Mass transfer in multiphase flow generally takes place on a much smaller length and time scale than the length and time scale of momentum transfer, resulting in a considerable increase in computational cost if regular approaches, either refining the grid or adaptive mesh refinement, are used for capturing the thin mass boundary layer. In the model developed here, a source term is used in the boundary layer equation and a sink term is used in the mass transfer equation on the grid, to transfer mass from boundary layer into the rest of the domain when and where the boundary layer grows over a predefined length.
First, the equation for the mass boundary layer is derived and tested for simple 1D and 2D case studies. Then it is implemented in a 2D front-tracking code and the accuracy of the results is tested by comparing the results from our model on a relatively coarse grid with a fully converged solution. The fully converged solution is obtained by using a much finer grid for solving the mass transfer equation. Comparison with fully converged solution showed that the model produces less than 10% error for low to moderate Reynolds number, $Re = O(10)$, and Schmidt number, $Sc = O(10)$, showing the model to be effective at predicting the mass transfer from the bubbles. Testing for different Schmidt number with the same Reynolds number, as expected, showed that the accuracy of model improves as the Peclet number increases, $Pe = Re \cdot Sc$. This is promising since in most multiphase flow of interest with mass transfer, the Peclet number is higher than what is used in the 2D comparisons. Initially, a two parameter second order profile was used but one of the parameters always converged to a constant value, shortly after the profile started evolving. We eliminated one of the parameter and a one parameter second order profile is assumed for mass fraction in the boundary layer. The direct comparison between the assumed profile and converged solution shows a good agreement between the profile extracted from the fully resolved calculations and the assumed profile.

While fully converged solution showed the accuracy and validity of the model, it limited the comparison to moderate non-dimensional governing parameters. So, in the next step, the model is extended to axisymmetric and three-dimensional flow in order to compare with experimental results. An axisymmetric implementation of the model is used for comparison with experimental Taylor bubbles in narrow cylinder and a three-dimensional implementation is used for ellipsoidal oscillatory bubbles. Comparing the Sherwood number from our calculation with the experiment shows a relatively good accuracy, less than equal 10% error. The direct comparison with experiment is done for high Schmidt number, $Sc = 8260$, and high Reynolds
number, $Re \approx 350$. Here, we could not do a fully resolved calculation but based on mass boundary layer thickness we estimate that using this method reduces the computational cost at least by one or two order of magnitude, specially when applied in simulations of fully three-dimensional flows. One thing that is not covered here and could be added in future is to include bubble volume reduction as mass is transferred from the bubble. The result for spherical bubbles is compared with correlations from literature which also shows good agreement.

Finally, having a validated method implemented for three-dimensional flows, the effect of bubble interactions on the mass transfer is studied for clusters of bubbles. Bubble volume is assumed to remain constant and the effect of accumulated mass inside the domain is neglected in order to follow the behavior of bubbles for a long period of time and reach a statistically steady state. To examine the effect of bubble interactions, a simulation of one bubble in a periodic domain, called fixed arrays, is compared with a simulation of eight bubbles in a larger domain, free arrays, with the same void fraction. The results show that while the rise Reynolds number decreases after bubbles start wobbling and interacting, the increase in bubble interactions compensate for the decrease in Reynolds number and for most cases the mass transfer increases slightly. The effect of void fraction on mass transfer from deformable bubbles is negligible. However, for very low Reynolds number, as Reynolds number increases with the increase of void fraction we see a considerable increase in the Sherwood number when the void fraction increases and Reynolds number decreases. The most significant change in mass transfer is observed when there is a horizontal clustering of bubbles. Here we see a 17% decrease of the Sherwood number.

On a separate topic, colliding non-coalescing droplet is studied for which a thin film appears between the droplets. Under-resolution of these films will lead to incorrect momentum drop. Here, a model is developed to recover the under-resolved viscous forces in thin films. The model is tested for simple 2D cases for which it gives
promising results. The implementation for 2D colliding droplets shows improvements over the under-resolved simulation but still needs more work to be completed.

The work done in this thesis shows the possibility of developing new subscale models by using analytical or semi-analytical descriptions, which use our understanding of some of the physics in the flow to bridge the gap between our computational power and the computational power needed to do large scale engineering problems. While computational power is increasing everyday, the engineering challenges also grow larger. The basic “philosophy” here is that we should not spend our time computing what can be understood physically in a simpler way.

Future work will be in several directions. The first direction would be the improvement of current method by adding the effect of mass concentration next to the interface, which makes the study of several different topics in bubble column possible. One of the important topics that needs more attention is the effect of bubble clustering, horizontally and vertically, on the mass transfer in large domains.

The second direction would be to develop similar model for the mass transfer and reaction next to the bubble interface. The reaction in bubbly flow is one of the challenging problems that can put a burden on computational simulation of bubble columns.

The third direction would be to improve the profile used for mass fraction inside the mass boundary layer. While second order profile produce good results for our simulations, the accuracy of the profile degrades as the boundary layer grow at stagnation points. One possibility for improving the method would be using a hybrid profile that changes its characteristic as boundary layer grows over the boundary layer limit. Other possible directions could be in subscale modeling for film boiling or in simulation of variable surface tension interface, considering surfactant advection diffusion in the liquid phase and it’s transfer from liquid to interface and vice versa.
APPENDIX A

TWO PARAMETER MASS TRANSFER MODEL

As explained in the text, we began with a two parameter second order profile,

\[
f(n) = \begin{cases} 
  f_0 \left(1 - a \left(\frac{n}{\delta}\right) - (1 - a) \left(\frac{n}{\delta}\right)^2\right); & n \leq \delta, \\
  0; & n > \delta,
\end{cases}
\]

(A.1)

where we need two parameters, \(a\) and \(\delta\) to fix the profile, in contrast to the one parameter profile used in this thesis where we only need \(\delta\). We want to solve Eq. (2.6) that is,

\[
\frac{\partial f}{\partial t} = \sigma n \frac{\partial f}{\partial n} + D \frac{\partial^2 f}{\partial n^2}.
\]

(A.2)

Approximate solution of equation Eq. (A.2) can be obtained by considering the zeroth moment,

\[
M_0 = \int_0^{\delta} f dn,
\]

(A.3)

and the first moment,

\[
M_1 = \int_0^{\delta} nf dn,
\]

(A.4)

of \(f\). We should note that in this initial version of the model, there was no boundary layer limit, \(\delta_0\). Equation for the evolution of \(M_0\) is easily derived by integrating Eq. (A.2) inside the boundary layer,

\[
\frac{dM_0}{dt} = -\sigma M_0 - D \left. \frac{\partial f}{\partial n} \right|_0.
\]

(A.5)
Similarly, by multiplying Eq. (A.2) by $n$ and integrating in the boundary layer we get,
\[
\frac{dM_1}{dt} = -2\sigma M_1 + D f_0. \tag{A.6}
\]

Inserting Eq. (A.1) into Eq. (A.3) and Eq. (A.4), yields,
\[
M_0 = f_0 \delta^4 - \frac{a}{6}, \tag{A.7}
\]
\[
M_1 = f_0 \delta^2 \frac{3 - a}{12}. \tag{A.8}
\]

Solving Eq. (A.7) and Eq. (A.8) together for $\delta$ and $a$, gives:
\[
\delta = \frac{3M_0 + \sqrt{9M_0^2 - 12f_0M_1}}{f_0}, \tag{A.9}
\]
\[
a = \frac{-3M_0^2 + 8f_0M_1 + M_0\sqrt{9M_0^2 - 12f_0M_1}}{2f_0M_1}. \tag{A.10}
\]

The procedure for evolving the boundary layer is similar to what is described in Section 2.2.1. When we solved the boundary layer using the two parameter profile presented here, $a$ always approaches 2 shortly after the boundary layer started evolving. Thus, we eliminate $a$ by putting $a = 2$, into the two parameter profile.
APPENDIX B

CORRECTION TO THE MASS TRANSFER MODEL

Since we use an approximate profile inside the mass boundary layer, we introduce a minor inconsistency in the process of integrating Eq. (A.2), which means the solution will be dependent on the boundary layer limit, \( \delta_0 \). Although the effect of \( \delta_0 \) on our results is shown to be negligible in Section 2.3, here we derive the mass boundary layer equation in a different way that the solution would be independent of specified \( \delta_0 \).

B.1 Derivation

Writing \( M_0 \) and \( M_0^T \) as functions of \( \delta \) and \( \delta_0 \), gives,

\[
M_0 = \int_0^{\delta} f dn = \frac{f_0 \delta}{3}, \tag{B.1}
\]
\[
M_0^T = \int_0^{\delta_0} f dn = f_0 \left( \delta_0 - \frac{\delta_0^2}{\delta} + \frac{\delta_0^3}{3\delta^2} \right). \tag{B.2}
\]

Then, by writing \( M_0 \) as a function of \( M_0^T \)

\[
M_0 = M_0^T + \frac{f_0 \delta}{3} - f_0 \left( \delta_0 - \frac{\delta_0^2}{\delta} + \frac{\delta_0^3}{3\delta^2} \right). \tag{B.3}
\]

Inserting Eq. (B.3) into Eq. (A.5), yields,

\[
\frac{dM_0^T}{dt} + \frac{df}{dn}\bigg|_0 = -\sigma M_0^T - \sigma \left[ \frac{f_0 \delta}{3} - f_0 \left( \delta_0 - \frac{\delta_0^2}{\delta} + \frac{\delta_0^3}{3\delta^2} \right) \right] - D \frac{df}{dn}\bigg|_0,
\]
Figure B.1. Shows the mass boundary layer profile at two states: when \( \delta < \delta_0 \), (a), and when \( \delta > \delta_0 \), (b). The solid pink shade shows the total mass inside boundary layer, \( M_0 \), and the dotted area in part b shows the truncated mass inside boundary layer, \( M_0^T \).
or

\[
\frac{dM_0^T}{dt} = -\sigma M_0^T - D \frac{df}{dn}\bigg|_0
\]

\[
\begin{aligned}
&= - \left\{ \frac{d}{dt} \left[ \frac{f_0\delta}{3} - f_0 \left( \frac{\delta_0 - \frac{\delta_0^2}{3\delta^2} + \frac{\delta_0^3}{3\delta^2}}{\delta} \right) \right] + \sigma \left[ \frac{f_0\delta}{3} - f_0 \left( \frac{\delta_0 - \frac{\delta_0^2}{3\delta^2} + \frac{\delta_0^3}{3\delta^2}}{\delta} \right) \right] \right\}.
\end{aligned}
\]

(B.4)

Putting Eq. (B.1) into Eq. (A.5), we know that \( \frac{d\delta}{dt} = -\sigma\delta - \frac{3Df}{f_0} \frac{df}{dn}\bigg|_0 \). Expanding and factoring \( \text{1} \), gives,

\[
\begin{aligned}
\text{1} &= f_0 \frac{1}{3} \frac{d\delta}{dt} - f_0 \frac{\delta_0^2}{\delta^2} \frac{d\delta}{dt} + f_0 \frac{2\delta_0^3}{3\delta^2} \frac{d\delta}{dt} = f_0 \left( \frac{1}{3} - \left( \frac{\delta_0}{\delta} \right)^2 + 2 \left( \frac{\delta_0}{\delta} \right)^3 \right) \frac{d\delta}{dt}
\end{aligned}
\]

\[
= f_0 \left( \frac{1}{3} - \left( \frac{\delta_0}{\delta} \right)^2 + 2 \left( \frac{\delta_0}{\delta} \right)^3 \right) \left( -\sigma\delta - \frac{3Df}{f_0} \frac{df}{dn}\bigg|_0 \right).
\]

(B.5)

Inserting Eq. (B.5) into \( \text{2} \) and simplifying, \( \text{2} \) becomes,

\[
\begin{aligned}
\text{2} &= -\sigma\delta_0 f_0 \left( 1 - 2 \frac{\delta_0}{\delta} + \left( \frac{\delta_0}{\delta} \right)^2 \right) - 3D \frac{df}{dn}\bigg|_0 \left( \frac{1}{3} - \left( \frac{\delta_0}{\delta} \right)^2 + 2 \left( \frac{\delta_0}{\delta} \right)^3 \right)
\end{aligned}
\]

\[
= -\sigma\delta_0 f_0 \left( \frac{1}{3} - \left( \frac{\delta_0}{\delta} \right)^2 + 2 \left( \frac{\delta_0}{\delta} \right)^3 \right).
\]

(B.6)
By differentiating the profile versus $n$, we have $\frac{df}{dn}\bigg|_0 = \frac{-2f_0}{\delta}$, so,

$$3 = -D \frac{-2f_0}{\delta} \left(1 - 3 \left(\frac{\delta_0}{\delta}\right)^2 + 2 \left(\frac{\delta_0}{\delta}\right)^3\right) = -D \frac{-2f_0}{\delta} \left(1 - \frac{\delta_0}{\delta}\right) \left(1 + \frac{\delta_0}{\delta} - 2 \left(\frac{\delta_0}{\delta}\right)^2\right)$$

$$= -D \frac{df}{dn}\bigg|_{\delta_0} \left(1 + \frac{\delta_0}{\delta} - 2 \left(\frac{\delta_0}{\delta}\right)^2\right).$$

(B.7)

Inserting Eq. (B.7) into Eq. (B.6) and then Eq. (B.6) into Eq. (B.4), the equation for the evolution of mass inside boundary layer can be written as

$$\frac{dM_0}{dt} = -\sigma M_0 - D \frac{df}{dn}\bigg|_0 + \sigma \delta_0 f_{\delta_0} + D \frac{df}{dn}\bigg|_{\delta_0} \left(1 + \frac{\delta_0}{\delta} - 2 \left(\frac{\delta_0}{\delta}\right)^2\right).$$

(B.8)

B.2 Discussion

Comparing Eq. (B.8) with Eq. (2.8), shows that they are the same except for a correction term, $C_{corr} = \left(1 + \frac{\delta_0}{\delta} - 2 \left(\frac{\delta_0}{\delta}\right)^2\right)$, for the diffusive flux at the edge of boundary layer limit. If we substitute $x$ for $\frac{\delta_0}{\delta}$, $C_{corr} = 1 + x - 2x^2$ and by differentiating,

$$\frac{dC_{corr}}{dx} = 1 - 4x,$$

which means the extremum of $C_{corr}$ occurs at $x = 1/4$ where $C_{corr} = \frac{9}{8} = 1.125$. It means, by integrating the Eq. (2.6) from 0 to $\delta_0$, using an approximate profile, we introduce a small $\delta_0$ dependency. In this thesis we have not included the correction term in our calculation. However, we tested a couple of calculations with the correction term and the change of the mass transfer was negligible. Fig. B.2 shows one of the cases from Fig. 2.5 with $Re = 30$ and $Sc = 20$. Comparing the result with correction term, dotted line, and the result without the correction term, solid squares, shows negligible effect from the correction.

We should note that this correction term could make significant contribution if
the same method is used for a reverse profile where at the interface \( f \) has a small value and outside the boundary layer \( f \) has a finite value.

Figure B.2. The total amount of scalar in the liquid as a function of time for a 2D simulation of mass transfer with \( Re = 30 \) and \( Sc = 20 \), computed in four different ways: on a coarse grid (dash-dot line), on a fine grid (solid line), on a coarse grid using the model without correction term (solid squares), and on a coarse grid using the model with the correction term.
APPENDIX C

EXPERIMENTAL STUDY

In Chapter 3, we compare our axisymmetric and 3D results directly with the experimental results. In this section we bring the explanation of the experimental setup and evaluation of mass transfer coefficient, which is done by Professor Tomiyama’s group in Kobe University, Japan. This chapter is published as part of a collaborative journal article with Tomiyama’s group in Chemical Engineering Science, Aboulhasanzadeh et al. (2013), and we include it here for completeness.

C.1 Setup

Figure C.1 shows a schematic of the experimental apparatus, which consists of a test section, lower and upper tanks, two high-speed video cameras, two LED light sources, two optical filters, four z-axis stage actuators and a digital fiber sensor. The test section is a vertical pipe with an inner diameter, \( d_c \), of 12.5 mm and a length of 2000 mm, made of fluorinated-ethylene-propylene (FEP) resin whose refractive index, 1.338, is close to that of water, 1.333. The FEP pipe is installed in an acrylic duct and the gap between the duct and the pipe is filled with water. This allows the bubbles, to be observed with minor optical distortion. The very small distortions that do take place are corrected with image post-processing.

A predetermined amount of CO\(_2\) gas (99.9 vol.% purity), whose volume is measured by using a gastight syringe, is stored in a hemispherical cup. A single bubble is released by rotating the cup. As shown in Fig. C.2, front and side images of a bubble
are recorded by using two synchronized video cameras (frame rate: 250 frame/s, exposure time: 1000 µs, spatial resolution: 0.04 mm/pixel), which are mounted on two actuators. Two color LED lights and optical filters reduce the overexposure caused by light reflection at the bubble surface. All the actuators are synchronized with the video cameras by a relay control circuit.

An image processing method, Tomiyama et al. (2002), is used to measure instantaneous bubble diameters and positions. An example of the original images is shown in Fig. C.3 (a). The front and side images are transformed into binary images as shown in Fig. C.3 (b). The method assumes that all the horizontal cross-sections of the bubble are elliptical. The height of each elliptical disk is one pixel and its physical length is $\Delta l_p$. The lengths of the major and minor axes of the disk are obtained from the lengths of the front and side images in the horizontal plane ($R_i$ and $G_i$ in Fig. C.3 (c)). The resultant elliptical disks are piled up in the vertical direction to reconstruct a three-dimensional bubble shape as shown in Fig. C.3 (d). The sphere-volume equivalent bubble diameter $d_b$ is therefore

$$d_b = \left[ \sum_{i=1}^{N} \frac{3}{2} R_i G_i \Delta l_p \right]^{1/3},$$  \hspace{0.5cm} (C.1)

where $N$ is the total number of disks in the vertical direction of the bubble image. Uncertainty estimated at 95% confidence in measured $d_b$ is ±2.1%.

C.2 Evaluation of the Mass Transfer Coefficient

The mass transfer coefficient $k_L$ and the Sherwood number $Sh$ are evaluated from the rate of decrease in $d_b$. Assuming that the flow is isothermal, the rate of mass transfer from a bubble to water is given by

$$\frac{dm}{dt} = -k_L A (C_s - C),$$  \hspace{0.5cm} (C.2)
where $m$ is the total moles of CO$_2$ in a bubble, $A (= \pi d_b^2)$ is the bubble surface area, $C_s$ is the CO$_2$ concentration at the bubble surface, and $C$ is the CO$_2$ concentration in the glycerol-water solution. By assuming that $C_s$ follows Henry’s law and $C$ is negligible, they obtain
\[ k_L = -\frac{1}{\pi d^2} \frac{H - P(z)X}{C_L P(z)} \frac{dm}{dt}, \]  
(C.3)

where $H$ is the Henry constant (205 MPa for CO$_2$ at $T = 298 K$), $C_L$ is the H$_2$O concentration (37.3 kmol/m$^3$), $X$ is the mole fraction of CO$_2$ which can be postulated to be unity because the dissolution rate in the CO$_2$-50% glycerol-water solution system is extremely low, and $P$ is the pressure inside a bubble given by
\[ P(z) = P_{atm} + \rho_t g h(z) + 4\gamma / d_b. \]  
(C.4)

Here, $P_{atm}$ is the atmospheric pressure, $\rho_t$ is the liquid density, $g$ is the magnitude of the gravitational acceleration, $h(z)(= 1.85 - z(t))$ is the distance from the free surface to the bubble center (see Fig. C.1), and $\gamma$ is the surface tension. By assuming that CO$_2$ is an ideal gas, $dm/dt$ can be expressed in terms of $P$ and $d_b$ as follows:
\[ \frac{dm}{dt} = \frac{\pi}{6RT} \frac{d(P d_b^3)}{dt}, \]  
(C.5)

where $R$ is the universal gas constant. Substituting Eq. (C.5) into Eq. (C.3) and evaluating $d(P d_b^3)/dt$ by using a centered difference approximation between $t_1$ and $t_2$ yields
\[ k_L = \frac{(H - P_{12})(P_2 d_{b2}^3 - P_{1d_{b2}}^3)}{6RT(t_2 - t_1)d_{b12}^2 C_L P_{12}}, \]  
(C.6)

where the subscripts, 1 and 2 represent times, $t_1$ and $t_2$, respectively, and subscript 12 denotes a value at time $(t_1 + t_2)/2$. The above equation is used to evaluate $k_L$ and the Sherwood number,
\[ Sh = \frac{k_L d_b}{D_c}, \]  
(C.7)
where $D_c$ is the diffusion coefficient of CO$_2$ in the liquid ($0.546 \times 10^{-9} \text{ m}^2/\text{s}$).

Figure C.1. The experimental setup, showing the test section, the piping and controllers.
Figure C.2. The cross section of the test section, the LED lights, and the camera.
Figure C.3. Reconstruction of the bubble shape. The original image (a) is digitized (b) and then split into elliptical disks (c). The disks are reassembled to give a representation of the bubble (d).


