EFFECTS OF INTERFACE CONTAMINATION ON MASS TRANSFER INTO A SPHERICAL BUBBLE

Abdellah Saboni¹, Silvia Alexandrova², Maria Karsheva³

¹Laboratoire SIAME-IUT, UPPA, Avenue de l'Université, 64 000 Pau, France
E-mail: abdellah.saboni@univ-pau.fr
²Laboratoire LaTEP-ENSGTI, UPPA, rue Jules Ferry, BP 7511, 64 075 Pau cedex, France
³Department of Chemical Engineering, University of Chemical Technology and Metallurgy, 8 Kl.Ohridski, 1756 Sofia, Bulgaria

ABSTRACT

A numerical study has been conducted to investigate the effects of contamination on mass transfer into a spherical bubble. The influence of the contamination on the mass transfer inside the bubble was studied for different ranges of dispersed phase Reynolds numbers (0.1 < Re_d < 10), Schmidt numbers (0.7 < Sc < 5) and different values of the polar angle \( \theta_{\text{cap}} \), characterizing the extent of a rigid cap at the rear of the bubble (\( \theta_{\text{cap}} = 0^\circ, 30^\circ, 60^\circ, 90^\circ, 120^\circ, 150^\circ \) and \( 180^\circ \)). The results show that the flow and the concentration profiles are strongly dependent on the Reynolds number, the Schmidt number and the stagnant cap angle (\( \theta_{\text{cap}} \)).

Keywords: bubble, mass transfer, interface contamination, stagnant-cap model.

INTRODUCTION

The problem of heat or mass transfer to and from moving bubbles is of interest for a number of engineering applications, including e.g., water treatment, bubbly flows, distillation. It has been observed that the interface contamination modifies the flow around the bubbles or drops in relative motion inside a fluid. The so-called stagnant-cap model has been widely used [1 - 5] to describe the flow around the contaminated bubbles. It assumes that the surfactants (surface active agents) tend to accumulate at the rear of the bubble, forming a cap with an immobile surface, while the rest of the bubble surface remains mobile (Fig. 1). The magnitude of the contamination is described by the polar angle \( \theta_{\text{cap}} \) of the spherical coordinate system with z axis coinciding with the direction of the relative motion of the bubble or drop in the fluid. This model is in agreement with the experimental observations of several authors [6 - 12]. These authors observed the formation of a stagnant cap at the rear of a drop or a bubble contaminated with slightly soluble surfactant (high Peclet number). The case of creeping flow (Stokes flow) past bubbles with stagnant cap has also been investigated [13 - 16]. The effect of the interface contamination was studied for higher Reynolds numbers by Cuenot et al. [1], considering the transient change in flow around a spherical bubble rising in a liquid contaminated by a slightly soluble surfactant. These results confirm the validity of the stagnant-cap model for describing the flow around a bubble, contaminated by slightly soluble surfactants. McLaughlin [17] considered the effect of an insoluble surfactant on flow around a deforming (or deformable) bubble. The stagnant cap model was also used to perform numerical simulations of mass transfer around a bubble in the presence of surfactants in the liquid phase [18 - 20]. Using the cap model, Saboni et al. [21 - 22] investigated the effects of contamination on the flow and mass transfer around a contaminated fluid sphere for Re number values up to 400. Nevertheless, the former studies do not consider the mass or heat transfer into a bubble.

In the present study, the model of mass transfer from a continuous phase to a partially or fully contaminated bubble was developed. First we solve the Navier-Stokes
equations and obtain the flow fields inside and outside a contaminated bubble. Then we use the velocity components for solving the diffusion-convection equation and derive the mass transfer rate into the contaminated bubble.

**Governing equations**

A contaminated bubble of radius \( a \) is considered. It is moving with uniform velocity \( \mathbf{U}_\infty \) in another immiscible fluid of infinite extent volume (Fig. 1). Since the flow is considered axisymmetric, the Navier-Stokes equations can be written in terms of stream function and vorticity (\( \psi \) and \( \omega \)) in spherical coordinates \( r \) and \( \theta \):

\[
E^2 \psi = \omega_r r \sin \theta
\]  
(1)

and

\[
\frac{\mu_c}{\mu_d} \frac{\rho_c}{\rho_d} \frac{2}{\text{Re}} \left[ \frac{\partial \psi_c}{\partial r} \frac{\partial}{\partial r} \left( \frac{\omega_r}{r \sin \theta} \right) - \frac{\partial \psi_c}{\partial \theta} \frac{\partial}{\partial \theta} \left( \frac{\omega_\theta}{r \sin \theta} \right) \right] \sin \theta = E^2 \left( \omega_r r \sin \theta \right)
\]

where

\[
E^2 = \frac{\partial^2}{\partial r^2} + \frac{\sin \theta}{r^2} \frac{\partial}{\partial \theta} \left( \frac{\sin \theta}{\sin \theta} \frac{\partial}{\partial \theta} \right)
\]

Outside the bubble, the above equations are still valid, but for numerical reasons the radial coordinate \( r \) is transformed via \( r = e^z \), where \( z \) is the logarithmic radial coordinate. The results are, as follows:

\[
E^2 \psi_c = \omega_c e^z \sin \theta
\]  
(3)

and

\[
\frac{\mu_c}{\mu_d} \left( \frac{\partial^2 \psi_c}{\partial z^2} - 3 \frac{\partial \psi_c}{\partial z} \right) = \left( \frac{\partial^2 \psi_d}{\partial r^2} - 2 \frac{\partial \psi_d}{\partial r} \right)
\]

where \( \mu \) is the dynamic viscosity.

iv) Across the stagnant part of the interface (\( \theta > \theta_{cap} \) and \( z = 0 \) or \( r = 1 \)), the tangential velocity is zero:

\[
\psi_c = 0; \quad \omega_c = 0; \quad \psi_d = 0; \quad \omega_d = 0
\]

Equations (1) to (4) subjected to the boundary conditions i) to iv) are solved simultaneously to obtain streamfunction and vorticity values. Once the stream function is known, the velocities can be determined from Eq. (5). The concentrations distribution can then be calculated from the diffusion-convection equation. Since the flow is considered axisymmetric, the unsteady convective mass transfer into a bubble in spherical coordinates \( r \) and \( \theta \) is described by the following dimensionless diffusion-convection equation:

\[
\frac{1}{r^2 \sin \theta} \frac{\partial \psi}{\partial \theta} + \frac{1}{r \sin \theta} \frac{\partial \psi}{\partial r} = 0
\]  
(5)

The boundary conditions to be satisfied are:

i) Far from the bubble (\( z = z_{\infty} \)), undisturbed parallel flow is assumed: \( \omega_c = 0; \quad \psi_c = 0.5 e^z \sin^2 \theta \)

ii) Along the axis of symmetry (\( \theta = 0, \pi \)):

\[
\psi_c = 0; \quad \omega_c = 0; \quad \psi_d = 0; \quad \omega_d = 0
\]

iii) Across the mobile part of the interface (\( \theta < \theta_{cap} \) and \( z = 0 \) or \( r = 1 \)), the following relations account for, respectively: negligible mass transfer, continuity of tangential velocity, continuity of tangential stress:

\[
\psi_c = 0; \quad \psi_d = 0; \quad \frac{\partial \psi_c}{\partial r} = \frac{\partial \psi_d}{\partial r}
\]

\[
\frac{\mu_c}{\mu_d} \left( \frac{\partial^2 \psi_c}{\partial z^2} - 3 \frac{\partial \psi_c}{\partial z} \right) = \left( \frac{\partial^2 \psi_d}{\partial r^2} - 2 \frac{\partial \psi_d}{\partial r} \right)
\]

\[
\frac{\partial \psi_c}{\partial z} = \frac{\partial \psi_d}{\partial z} = 0
\]

iv) Across the stagnant part of the interface (\( \theta > \theta_{cap} \) and \( z = 0 \) or \( r = 1 \)), the tangential velocity is zero:

\[
\psi_c = 0; \quad \psi_d = 0; \quad \frac{\partial \psi_c}{\partial r} = \frac{\partial \psi_d}{\partial r} = 0
\]

\[
\frac{\partial \psi_c}{\partial z} = \frac{\partial \psi_d}{\partial z} = 0
\]
\[
\frac{\partial C}{\partial \tau} + \frac{Pe}{2} \left( \frac{u}{r} \frac{\partial C}{\partial r} + u \frac{\partial C}{\partial \theta} \right) = \frac{\partial^2 C}{\partial r^2} + \frac{2}{r} \frac{\partial C}{\partial r} + \frac{2}{r^2} \frac{\partial C}{\partial \theta} + \frac{1}{r^2} \left( \frac{\partial^2 C}{\partial \theta^2} + \cot(\theta) \frac{\partial C}{\partial \theta} \right)
\]  

(6)

where \( Pe \) is the Peclet number \( (Pe = 2aU\infty / D) \), \( a \) is the bubble radius, \( D \) is the diffusivity, \( \tau \) is the dimensionless time \( (\tau = Dt / a^2) \), and \( C \) is the dimensionless concentration \( C = \frac{C}{C_i} \) where \( C_i \) is the concentration at the interface.

The boundary and initial conditions to be satisfied are:

i) Initial condition: \( C(r, \theta, t = 0) = 0 \)

ii) Along the axis of symmetry:

\[
\left. \frac{\partial C}{\partial \theta} \right|_{\theta = 0} = \left. \frac{\partial C}{\partial \theta} \right|_{\theta = \pi} = 0
\]

iii) Across the interface: \( C(r = 1, \theta, t) = 1 \)

The surface and average Sherwood numbers are computed from the mass transfer flux from the surface of the bubble:

\[
Sh_{\text{surface}} = -\frac{2}{\langle C \rangle} \left. \frac{\partial C}{\partial r} \right|_{r = 1}
\]

(7)

\[
Sh = -\frac{1}{\langle C \rangle} \int_0^\pi \left. \frac{\partial C}{\partial r} \right|_{r = 1} \sin \theta \ d\theta
\]

(8)

where the average concentration inside the bubble is computed from the following equation:

\[
\langle C \rangle = \frac{3}{2} \int_0^1 \int_0^\pi C(r, \theta) r^2 \sin \theta \ d\theta \ dr
\]

(9)

Eqs. (1) to (6) together with the above boundary conditions are solved numerically. The elliptic stream function equations are solved iteratively, the parabolic vorticity equations are solved by means of the Alternating Direction Implicit method. Once the stream function is known, the velocities are then determined from Eq. (5). The convection-diffusion equation is solved by means of the Alternating-Direction Explicit (ADE) scheme. Numerical experiments were performed in order to check the grid independence of the solutions and grid size of 181x181 elements in the dispersed phase and 401x181 in the continuous phase. Numerical methods were adopted for grid-free solution throughout the calculations in the present study. In addition, the distance from the bubble to the edge of the computational domain is 100 diameters for small Reynolds numbers and 12 diameters for higher Reynolds numbers, for which the boundary layer thickness is less. The validation of the numerical methods used implied two stages: (1) checking of the numerical solution procedure concerning the hydrodynamics around and inside a bubble, and (2) checking of the numerical method used for the mass transfer. Detailed discussions on the accuracy of the solution procedure employed for the momentum, continuity equations, and diffusion-convection equation were made previously by Saboni and al. [21 - 22].

Fig. 1. Stagnant cap model.
RESULTS AND DISCUSSION

Simulations are made for Peclet numbers encountered in practice consistent with the model assumptions. The Peclet number is defined as $Pe_d = Re_d \cdot Sc$, where $Re_d$ is the dispersed phase Reynolds number. The relation between the dispersed phase Reynolds number and the continuous phase Reynolds number is

$$Re_d = Re_c \frac{\rho_d \mu_c}{\rho_c \mu_d}, \quad \text{where } Re_c = \frac{U_c \delta}{\nu}.$$

For an air bubble in water, with $\mu_d/\mu_c = 1/55$ and $\rho_d/\rho_c = 1/836$, the relation between the two Reynolds numbers is $Re_d = Re/15.2$.

At a temperature of 20°C, the Schmidt number (the ratio) is about 1 for gases, but it is greater for lower temperatures. To account for the effect of the temperature, the numerical computations have been carried out for Schmidt numbers ranging from 0.7 to 5. We consider continuous Reynolds numbers not exceeding ~150. Continuous Reynolds number, $Re_c = 200$, is the highest limit for which the flow rests axisymmetric and for which an air bubble in water remains quasi-spherical. In view of the Schmidt and Reynolds numbers considered, the results are valid for Peclet numbers, $Pe_d \leq 50$.

Fig. 2 shows streamline contours inside and outside a bubble for $Re_d = 1$ ($Re_c = 15.2$) and different stagnant cap angles. For all stagnant cap angles, the contour line plots show no flow separation downstream the bubble. Slight asymmetry is observed between upstream and downstream regions near the bubble. The internal flow departs from symmetry due to the effect of the external flow. Increased contamination causes poor circulation and a dead zone at the rear of the bubble.

Table 1 summarizes the values of the drag coefficient $C_d$ obtained from our calculations, for the range $Re_d = 0.1 - 10$ ($Re_c = 1.52 - 152$) and different stagnant cap angles.

Fig. 2. Stream function contours, inside and outside a bubble for $Re_d = 1$ ($Re_c = 15.2$) and different $\theta_{cap}$. 
cap angles ($\theta_{\text{cap}} = 180^\circ$, $150^\circ$, $120^\circ$, $90^\circ$, $60^\circ$, $30^\circ$ and $0^\circ$). The values for $\theta_{\text{cap}} = 0^\circ$ and $\theta_{\text{cap}} = 180^\circ$ coincide with the drag coefficient for a rigid sphere and a clean bubble, respectively. As expected, the drag coefficient decreases with increasing Reynolds numbers for a fixed stagnant cap angle.

The isoconcentrations’ contours inside a clean bubble ($\theta_{\text{cap}} = 180^\circ$) and inside of a partially contaminated bubble ($\theta_{\text{cap}} = 90, 30^\circ$) for $Re_d = 10$ ($Re_c = 152$) and different Schmidt numbers ($Sc = 0.7, 3, 5$) at time $\tau =$

Table 1. Effect of the interface contamination on the drag coefficient of a bubble.

<table>
<thead>
<tr>
<th>$Re_d$</th>
<th>$\theta_{\text{cap}}$</th>
<th>$0^\circ$</th>
<th>$30^\circ$</th>
<th>$60^\circ$</th>
<th>$90^\circ$</th>
<th>$120^\circ$</th>
<th>$150^\circ$</th>
<th>$180^\circ$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1</td>
<td>18.83</td>
<td>18.73</td>
<td>18.30</td>
<td>16.61</td>
<td>14.00</td>
<td>12.30</td>
<td>12.02</td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>3.26</td>
<td>3.24</td>
<td>3.18</td>
<td>2.78</td>
<td>2.15</td>
<td>1.82</td>
<td>1.78</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>0.87</td>
<td>0.89</td>
<td>0.87</td>
<td>0.67</td>
<td>0.38</td>
<td>0.27</td>
<td>0.26</td>
<td></td>
</tr>
</tbody>
</table>

Table 2. Effect of the interface contamination on the asymptotic Sherwood number of a bubble.

<table>
<thead>
<tr>
<th>$\theta_{\text{cap}}$</th>
<th>$Re_d$ = 1</th>
<th>$0^\circ$</th>
<th>$30^\circ$</th>
<th>$60^\circ$</th>
<th>$90^\circ$</th>
<th>$120^\circ$</th>
<th>$150^\circ$</th>
<th>$180^\circ$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Sc$</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.7</td>
<td>6.57</td>
<td>6.57</td>
<td>6.57</td>
<td>6.57</td>
<td>6.58</td>
<td>6.58</td>
<td>6.58</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>6.57</td>
<td>6.57</td>
<td>6.58</td>
<td>6.60</td>
<td>6.63</td>
<td>6.64</td>
<td>6.64</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>6.57</td>
<td>6.57</td>
<td>6.59</td>
<td>6.64</td>
<td>6.72</td>
<td>6.75</td>
<td>6.76</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>$Sc$</th>
<th>$Re_d$ = 10</th>
<th>$0^\circ$</th>
<th>$30^\circ$</th>
<th>$60^\circ$</th>
<th>$90^\circ$</th>
<th>$120^\circ$</th>
<th>$150^\circ$</th>
<th>$180^\circ$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.7</td>
<td>6.57</td>
<td>6.57</td>
<td>6.62</td>
<td>6.82</td>
<td>7.13</td>
<td>7.30</td>
<td>7.31</td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>6.57</td>
<td>6.58</td>
<td>6.66</td>
<td>7.06</td>
<td>7.66</td>
<td>7.97</td>
<td>8.00</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>6.57</td>
<td>6.59</td>
<td>7.26</td>
<td>9.68</td>
<td>12.31</td>
<td>12.98</td>
<td>13.01</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>6.57</td>
<td>6.62</td>
<td>8.09</td>
<td>12.34</td>
<td>15.28</td>
<td>15.40</td>
<td>15.40</td>
<td></td>
</tr>
</tbody>
</table>

Fig. 3. Stream function contours, inside and outside a bubble for $Re_d = 10$ ($Re_c = 152$) and different $\theta_{\text{cap}}$. 
0.1, are plotted in Figs. 4a, b, c. For Sc = 0.7, the shape of the isoconcentrations’ contours is almost spherical. They are slightly shifted from the center to the rear zone for stagnant cap angles $\theta_{\text{cap}} = 180$ and $90^\circ$. From Fig. 4a it can be seen also that the average concentration in all cases ($\theta_{\text{cap}} = 180, 90, 30^\circ$) is almost 0.77 and corresponds to the value given by the Newnan equation, at time $\tau = 0.1$. This means that transport by convection is very limited and mass transfer occurs mainly by diffusion. For higher Schmidt numbers, convective transport increases, and the isoconcentrations’ contours inside the bubble are deformed under the influence of the internal flow except the case $\theta_{\text{cap}} = 0$, for which the internal flow is very weak. It is evident, that the domain of high concentrations in the downstream region of the bubble is larger in the case of a not contaminated bubble, than that in the case of a partially or totally contaminated bubble.

This can be explained with a higher mass transfer rate for the not contaminated bubble in comparison with a partially or totally contaminated one. On the mobile part of the interface, the mass transfer is facilitated as a result of convection and diffusion. On the other hand, on the immobile part of the interface, the mass transfer is due only to the diffusion.

Table 2 summarizes the asymptotic average Sherwood number (Sh), obtained from our calculations for the range of parameters covered $Re_{\text{d}} = 0.1 - 10$ ($Re_c = 1.52 - 152$), different stagnant cap angles ($q_{\text{cap}} = 180^\circ, 150^\circ, 120^\circ, 90^\circ, 60^\circ, 30^\circ$ and $0^\circ$) and different Schmidt numbers ($Sc = 0.7, 1, 3, 5$). This table demonstrates the influence of the stagnant cap angles on the mass transfer. As expected, the Sherwood number increases with increasing Schmidt number for fixed Reynolds numbers and at given stagnant cap angles. For fixed Reynolds and
Schmidt numbers, the Sherwood number increases with stagnant cap angles increase. From Table 2 it is evident, that the contamination reduces the mass transfer into the bubble for all Reynolds and Schmidt numbers. It can be seen that the average Sherwood number increases with stagnant cap angle increase, and reaches a limit value corresponding to the average Sherwood number, for a clean bubble. The influence of the stagnant cap angle on the mass transfer is more or less important; it depends both on the Reynolds number and the Schmidt number. From Table 2 it appears, however, that the relative difference between the Sherwood values for $\theta_{\text{cap}} = 0^\circ$, $\theta_{\text{cap}} = 180^\circ$ is less than 3% for $Re_\text{a} = 1$. On the contrary, for $Re_\text{a} = 10$, the relative difference can be quite important, approaching 58% for the largest Schmidt number values in this study.

**CONCLUSIONS**

The mass transfer into a contaminated bubble moving in an unbounded fluid is examined numerically. The effect of the interface contamination and the flow, on the concentration profiles inside a bubble, is investigated for different ranges of Reynolds number, Schmidt number and the stagnant cap angle. It was found that the stagnant cap angle influences significantly the flow and mass transfer into a bubble. The value of the total drag coefficient increases as the cap angle decreases and/or as the Reynolds number decreases. Despite the value of the stagnant cap angle, the Sherwood number increases with increasing the Schmidt number for fixed Reynolds numbers. For a fixed Reynolds number, the Sherwood number increases with the Schmidt number increase. For all Reynolds and Schmidt numbers, the contamination reduces the mass transfer. The average Sherwood number increases with a stagnant cap angle increase and reaches a limit value corresponding to the average one, for a clean bubble.

**REFERENCES**