Modeling of Mass transfer and Chemical Reactions in a Bubble Column Reactor Using a Discrete Bubble Model

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Abstract A 3D discrete bubble model is adopted to investigate complex behavior involving hydrodynamics, mass transfer and chemical reactions in a gas-liquid bubble column reactor. In this model a continuum description is adopted for the liquid phase and additionally each individual bubble is tracked in a Lagrangian framework, while accounting for bubble-bubble and bubble-wall interactions via an encounter model. The mass transfer rate is calculated for each individual bubble using a surface renewal model accounting for the instantaneous and local properties of the liquid phase in its vicinity. The distributions in space of chemical species residing in the liquid phase are computed from the coupled species balances considering the mass transfer from bubbles and reactions between the species. The model has been applied to simulate chemisorption of CO₂ bubbles in NaOH solutions. Our results show that apart from hydrodynamics behavior, the model is able to predict the bubble size distribution as well as temporal and spatial variations of each chemical species involved.

1 Introduction

Bubble columns are used in a variety of industrial processes including large-scale production of base chemicals and synthetic fuels. Many processes involve gas-liquid mass transfer with accompanying reactions between the gas and the liquid phase itself or with components dissolved or suspended in it. Despite the widespread application of bubble columns and substantial research efforts devoted to understand their behavior, detailed knowledge on the fluid flow, mass transfer and chemical reactions as well as their interactions are still lacking.

The coupling between these phenomena is illustrated in Fig. 1. The chemical reaction rate depends on the local availability of the species which is determined by the interphase mass transfer process and the mixing induced by the bubbles. The interphase mass transfer depends on the mass transfer coefficient, the specific interfacial area and chemical reaction rate. The mass transfer parameters are a function of the local prevailing hydrodynamics (Cents et al. (2003)), which in its turn are affected by the bubble behavior and variation of physical properties due to in-homogeneous chemical species distributions. It is these complex interactions that makes the overall prediction of performance and scale-up of this type of reactor very difficult.

Figure 1: Inter-dependency diagram of fluid flow, mass transfer and chemical reaction.

In recent years, computational fluid dynamics (CFD) has emerged as a powerful tool for both scientists and engineers. CFD modelling of dispersed gas-liquid two-phase flows has shown remarkable
progress over the last decade. Two models are widely used for describing hydrodynamics of bubble columns, i.e. the Euler-Euler (E-E) model and Euler-Lagrange (E-L) model. The E-E model employs the volume-averaged mass and momentum conservation equations to describe the time dependent motion of both phases (Deen et al. (2001), Sokolichin and Eigenberger (1994), Pan and Dudukovic (2000)). The E-L model on the other hand adopts a continuum description for the liquid phase and additionally tracks each individual bubble using Newtonian equations of motion. This allows for a direct consideration of additional effects related to bubble-bubble and bubble-liquid interaction. Mass transfer with and without chemical reaction, bubble coalescence and re-dispersion can be incorporated directly (Tomiyama et al. (1997), Delnoij et al. (1997), Sokolichin and Eigenberger (1994)).

In this paper we present a detailed 3D model for the hydrodynamics, mass transfer and chemical reactions in an isothermal bubble column operated in the homogeneous regime. The Euler-Lagrange model is adopted to solve the hydrodynamics since incorporation of the bubble size distribution and interphase mass transfer calculation is relatively straightforward. The model is applied to study the coupling of hydrodynamics, gas-liquid mass transfer and chemical reactions in a square bubble column. First we will focus on the hydrodynamics of the column and compare our results with experimental and numerical results obtained by Deen et al. (2001). Next the physical absorption of \( CO_2 \) into water will be addressed and finally the complete model will be used to investigate chemisorption of \( CO_2 \) bubbles into an aqueous solution of sodium hydroxide.

2 Model formulation

The three-dimensional model described in this paper is an extension of the hydrodynamic Euler-Lagrange model of Delnoij et al. (1999), which is applicable to disperse (homogeneous) regimes, which are characterized by low gas velocities and relatively small spherical bubbles that do not coalesce nor break-up. The interphase mass transfer is calculated for each bubble using the surface renewal theory which takes into account both physical and chemically enhanced mass transfer. The spatial distributions of chemical species residing in the liquid phase are computed from the coupled species conservation equations formulated in the Eulerian framework.

2.1 Bubble dynamics

The motion of each individual bubble is computed from the bubble mass and momentum equations while accounting for bubble-bubble and bubble-wall interactions via an encounter model of Hoomans et al. (1996). The liquid phase contributions are taken into account by the interphase mass transfer rate \( \dot{m} \) and the net force \( \sum F \) experienced by each individual bubble. For an incompressible bubble, the equations can be written as:

\[
\rho_b \frac{d(V_b)}{dt} = (\dot{m}_{l-b} - \dot{m}_{b-l})
\]

\[
\rho_b V_b \frac{dV_b}{dt} = \sum F - \left( \rho_b \frac{dV_b}{dt} \right) v
\]

where \( \rho_b, V_b \) and \( v \) respectively the density, volume and velocity of the bubble. The interphase mass transfer term \( \dot{m} \) is calculated using a method, which will be described in section 2.4.

The net force acting on each individual bubble is calculated by considering all the relevant forces. It is composed of separate, uncoupled contributions such as gravity, pressure, drag, lift and virtual mass:

\[
\sum F = F_G + F_P + F_D + F_L + F_{VM}
\]
Expressions for each of these forces can be found in Table 1 (see Delnoij et al. (1997) for a discussion on the forces experienced by a bubble).

<table>
<thead>
<tr>
<th>Force</th>
<th>Closure</th>
</tr>
</thead>
<tbody>
<tr>
<td>$F_G = \rho_b V_b g$</td>
<td>—</td>
</tr>
<tr>
<td>$F_p = -V_b \nabla P$</td>
<td>—</td>
</tr>
<tr>
<td>$F_D = -\frac{1}{2} C_D \rho_b \rho_t \pi R_b^2</td>
<td>v - u</td>
</tr>
<tr>
<td>$F_L = -C_L \rho_b V_b (v - u) \times \nabla \times u$</td>
<td>$C_L = 0.5$</td>
</tr>
<tr>
<td>$F_{VM} = -C_{VM} \rho_t V_b \left( \frac{\partial Y_j}{\partial t} - \frac{\partial u_j}{\partial t} \right)$</td>
<td>$C_{VM} = 0.5$</td>
</tr>
</tbody>
</table>

Table 1: Overview of forces acting on a bubble.

### 2.2 Liquid phase hydrodynamics

The liquid phase hydrodynamics is represented by the volume-averaged Navier-Stokes equation, which consists of continuity and momentum equations. The presence of bubbles is reflected by the liquid phase volume fraction $\varepsilon_l$, the source term that accounts for the interphase mass transfer $\dot{M}$, and the interphase momentum transfer $\Phi$:

$$
\frac{\partial}{\partial t} (\varepsilon_l \rho_l) + \nabla \cdot \varepsilon_l \rho_l u = (\dot{M}_b - \dot{M}_l) - \dot{M}_l
$$

$$
\frac{\partial}{\partial t} (\varepsilon_l \rho_l u) + \nabla \cdot \varepsilon_l \rho_l u u = -\varepsilon_l \nabla P - \nabla \cdot \varepsilon_l \tau_l + \varepsilon_l \rho_l g + \Phi
$$

where $g$ is the gravity acceleration, $\rho_l$, $u$ and $P$ respectively the density, velocity and pressure for the liquid phase.

The liquid phase stress tensor $\tau_l$ is assumed to obey the general Newtonian form given by:

$$
\tau_l = -\mu_{eff,l} \left[ (\nabla u) + (\nabla u)^T \right] - \frac{2}{3} \mathbf{I} (\nabla \cdot u)
$$

where $\mu_{eff,l}$ is the effective viscosity. In the present model the effective viscosity is composed of two contributions, the molecular viscosity and the turbulent viscosity:

$$
\mu_{eff,l} = \mu_{L,l} + \mu_{T,l}
$$

where the turbulent viscosity $\mu_{T,l}$ is calculated using the sub-grid scale (SGS) model of Smagorinsky (1963):

$$
\mu_{T,l} = \rho_l (C_S \Delta)^2 |S|
$$

where $C_S$ is a model constant with a typical value of 0.1, $S$ the characteristic filtered strain rate and $\Delta = (V_{cell})^{1/3}$ the SGS length scale.

### 2.3 Chemical species

The fraction of a chemical species $j$ in the liquid mixture is represented by mass fraction $Y_j^l$. The presence of various chemical species are modeled through a transport equation for each species given by:

$$
\frac{\partial}{\partial t} \left( \varepsilon_l \rho_l Y_j^l \right) + \nabla \cdot \left( \varepsilon_l \left( \rho_l u_j Y_j^l - \Gamma_{el,l}^j \nabla Y_j^l \right) \right) = \left( \dot{M}_b - \dot{M}_l \right) Y_j^l + \varepsilon_l S_j
$$
where \( S^j \) is the source term accounting for production or consumption of species \( j \) due to homogeneous chemical reaction and

\[
\Gamma_{\text{eff}}^j = \frac{\mu_{\text{eff}}^j}{S^j}
\]  

(10)

where \( \text{Sc}^j \) is the Schmidt number of species \( j \) defined as:

\[
\text{Sc}^j = \frac{\mu^j}{\rho_l D^j}
\]  

(11)

where \( D^j \) is the diffusivity of species \( j \).

For a mixture consisting of \( N_s \) species, the liquid density and viscosity are taken as the average of properties of each species as follow:

\[
\frac{1}{\rho_l} = \sum_{j=1}^{N_s} \frac{Y^j_l}{\rho^j_l}; \quad \mu_l = \sum_{j=1}^{N_s} Y^j_l \mu^j_l
\]  

(12)

### 2.4 Mass transfer

The interphase mass transfer in a bubble with radius \( R_b \) is considered to be driven by mass fraction gradients. The mass fraction of a chemical species \( j \) in the liquid phase and bubble are represented by \( Y^j_l \) and \( Y^j_b \) respectively, while the value of both quantities at each side of the bubble-liquid interface is given by \( Y^j_l ^* \) and \( Y^j_b ^* \) (see Fig. 2).

The mass transfer in a bubble due to a concentration gradient of species \( j \) is represented as:

\[
\dot{m}^j_b = E k^j_l A_b \rho_l (Y^j_b ^* - Y^j_l)
\]  

(13)

where \( E \) is the enhancement factor due to chemical reactions, \( A_b \) is the surface area of the bubble and \( k^j_l \) is the mass transfer coefficient for species \( j \), which is determined using a Sherwood relation for a moving bubble (Bird et al. (2002)):

\[
\text{Sh} = \frac{k^j_l d_b}{D^j} = 2 + 0.6415 \left( \text{ReSc}^j \right)^{1/2}
\]  

(14)

where \( d_b \) and \( \text{Re} \) is the diameter and Reynolds number for the bubble.

The mass fraction on the liquid side of the interface can be determined using the Henry constant:

\[
Y^j_l ^* = H^j Y^j_b \rho_b \rho_l
\]  

(15)

where \( H^j \) is the Henry constant for the species \( j \).

The total mass transfer rate is the sum of the mass transfer rates of all species \( j \), thus:

\[
\dot{m}_b = \sum_{j=1}^{N_s} \dot{m}^j_b
\]  

(16)

The mass transfer from the liquid to a bubble can be written as \( \dot{m}_{l\rightarrow b} = \max(\dot{m}_b, 0) \), while the mass transfer from a bubble into the liquid is \( \dot{m}_{b\rightarrow l} = \max(-\dot{m}_b, 0) \).
3 Geometry and boundary conditions

Deen et al. (2001) performed particle image velocimetry (PIV) measurements in a 3D bubble column filled with distilled water. The column has a square cross-section \((W \times D)\) of \(0.15 \times 0.15\ m^2\) and a height \((L)\) of \(0.45\ m\). Air with a superficial gas velocity of \(4.9\ mm/s\) was introduced into the system through a perforated plate. The plate contained 49 holes with a diameter of \(1\ mm\), which were positioned in the middle of the plate at a square pitch of \(6.25\ mm\).

The column is modelled using the DBM, which was described in the preceding section. The computational grid consists of \(30 \times 30 \times 90\) cells and the flow time step \((\delta t_{\text{flow}})\) is \(1.0 \times 10^{-3}\ s\). Preliminary calculations revealed that this configuration gives a grid and time step independent solution. The boundary conditions are imposed to the column using the ag matrix concept of Kuipers et al. (1993) as can be seen in Fig. 3. The definition of each boundary condition can be seen in Table 2. Each hole in the perforated plate is modeled as a position in the bottom of the column where bubbles with diameter of \(4\ mm\) size enter the column with a fixed velocity.

![Boundary Conditions Diagram](image)

Figure 3: Typical boundary conditions used in simulations with the discrete bubble model. The vertical plane is at \(j = NY/2\) while the horizontal plane is at \(k = NZ\).

<table>
<thead>
<tr>
<th>Flag</th>
<th>Boundary conditions</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Interior cell, no boundary conditions specified</td>
</tr>
<tr>
<td>2</td>
<td>Impermeable wall, free slip boundary</td>
</tr>
<tr>
<td>3</td>
<td>Impermeable wall, no slip boundary</td>
</tr>
<tr>
<td>4</td>
<td>Prescribed pressure cell, free slip boundary</td>
</tr>
<tr>
<td>5</td>
<td>Corner cell, no boundary conditions specified</td>
</tr>
</tbody>
</table>

4 Simulations results and discussion

In order to demonstrate the capabilities of the model, which combines hydrodynamics, mass transfer and chemical reactions, three case studies are presented. The square bubble column introduced in
section 3 is used as a base configuration. The hydrodynamics part of the model is compared with PIV measurements of Deen et al. (2001).

4.1 Hydrodynamics
In this case study, the hydrodynamics of an air-water system are investigated. The gas-liquid flow in a square bubble column is simulated using the hydrodynamic model as described in section 2.1 and 2.2. Air is injected into an initially quiescent liquid as indicated in Fig. 4. Shortly after the bubbles are released into the column, a bubble plume rises through the column and creates a typical mushroom like shape. Due to the gas-liquid momentum coupling, some motion is induced in the liquid phase, and after a while the entire bulk liquid starts to move. The first bubbles escape from the column after 1.5 s. After some time the motion inside the bubble becomes unstable. Strong circulation patterns appear with up flow in the center region and down flow in the corners of the column. Due to the liquid down flow, some bubbles close to the wall are dragged downwards to the lower region of the column before being dragged again to the column surface by the upward flow.

Figure 4: Series of corresponding velocity fields and bubble position obtained from simulation of air-water bubble column at different time after the air was switched on. Gas superficial velocity= 4.9 mm/s.

The bubble plume is meandering in a random fashion. Using animation representation of our simulation results as suggested by Delnoij et al. (1997) we can see that this meandering behavior is due to random formation of vortices close to the column surface. The vortices generate strong down flow, which pushes the bubbles near the inlet region to one side when they reach the bottom of the column. The bubble plume will then move close to one side of the column. This process is repeated, but the vortex formation appears in another location, which will make the bubble plume move in another direction.
Figure 5: (a) Time history of the vertical liquid velocity at the centreline of the column and at a height of $z/H = 0.56$. (b) Comparison of simulated and experimental profiles of the liquid vertical average velocity ($\bar{u}_z$), at a height of $z/H = 0.56$ and a depth $y/W = 0.5$.

A time history plot of the vertical liquid velocity at one point in the column is shown in Fig. 5(a). As can be seen in these figures, the DBM simulation is able to reveal the highly dynamic nature of the bubble column hydrodynamics. As compared to the experimental measurements using laser Doppler anemometry (LDA) technique by Deen et al. (2001), the DBM results are in agreement for both time and velocity scales.

A more quantitative comparison with the experimental measurement is obtained through the long term (i.e. the statistical averaged) quantities. In the present study the time-averaged mean velocity and velocity fluctuations of the simulation are calculated during a $10 - 120$ s interval. The mean velocity is calculated as follow:

$$\bar{u} = \frac{1}{N_t} \sum_{i=1}^{N_t} u_i$$

(17)

where $N_t$ is the number of time steps used in the averaging. The large scale velocity fluctuation is calculated as:

$$u' = \frac{1}{N_t} \sqrt{\sum_{i=1}^{N_t} (u_i - \bar{u})^2}$$

(18)

The time averaged quantities are compared with the PIV measurements of Deen et al. (2001), who also conducted simulations of this column with a two fluid model using the commercial CFD package CFX. Fig. 5(b) shows the profile of the average liquid velocity in the vertical direction while the liquid phase vertical and horizontal velocity fluctuations can be seen in Fig. 6. As can be seen in these figures, the simulation results show good agreement with the experimental data. The average velocity profile gives a maximum value in the center of the column and a negative value close to the wall, which resembles a liquid flow pattern with liquid up-flow in the center region of the column and down flow near the walls. Compared to the PIV measurements, the DBM simulations generally slightly overpredict the average and fluctuation velocities in the center region of the column, while the two-fluid model simulations shows the opposite tendency. The vertical velocity fluctuations, measured with PIV, show a local minimum in the middle, which is also predicted nicely in the DBM
simulation. The velocity fluctuations predicted by the two fluid model, however, do not show this feature.

Figure 6: Comparison of simulated and experimental profiles of the liquid velocity fluctuations, at a height of $z/H = 0.56$ and a depth $y/W = 0.5$. (a): vertical fluctuations ($u'_z$), (b): horizontal fluctuations ($u'_x$).

4.2 Physical absorption

In this case study the physical absorption of $CO_2$ in water is simulated. $CO_2$ gas is fed into the column filled with water. A species transport equation is utilized to track the mass fractions of $CO_2$ in the liquid phase. Initially there is no dissolved $CO_2$ present in the column. During the process of the physical absorption, a relatively low mass transfer rate is experienced by the bubbles.

Fig. 7 shows the transient behavior in the column after the $CO_2$ bubbles are injected into the column. As can be seen, the hydrodynamics are relatively similar to the air-water case as described in the previous test cases. The size of the bubbles is only slightly changed during their presence in the column. A high fraction of dissolved $CO_2$ can be found in the vicinity of the bubbles. The dissolved $CO_2$ is convected by the liquid to other parts of the column.

Fig. 8 shows the concentration of $CO_2$ at the centreline of the column, at a height of $z/H = 0.5$. As can be seen in this figure, the dissolved $CO_2$ is accumulated in time. The interphase mass transfer decreases as the dissolved $CO_2$ concentration increases and eventually diminishes when the equilibrium condition as prescribed by the Henry constant is reached.

4.3 Bubble column under reaction condition

In this case study, we combine all the models described in section 2 to obtain a complete, comprehensive model for a bubble column under reactive conditions. The chemisorption of $CO_2$ into aqueous NaOH solution is chosen as a test case, since it accounts for important phenomena that are encountered in practice.

The reaction mechanism of the chemisorption of carbon dioxide in an aqueous solution of sodium hydroxide is well understood and the reaction kinetics are well documented. The reaction sequence comprises several steps, which are given below:

\[ CO_2(g) \rightarrow CO_2(l) \]  
\[ CO_2 + OH^- \rightleftharpoons HCO_3^- \]
Figure 7: Set of corresponding velocity fields (top), bubble position (middle) and distribution of dissolved CO$_2$ concentration [mol/l] (bottom) obtained from simulation of the physical absorption of CO$_2$ gas in water at various time after the CO$_2$ gas was switched on. Pure CO$_2$ gas with superficial velocity= 4.9 mm/s.
Figure 8: Dissolved CO$_2$ concentration normalized by concentration at equilibrium condition at the centreline of the column and at a height of $z/H = 0.5$. Simulation result of physical absorption of CO$_2$ bubbles in water. Pure CO$_2$ gas with superficial velocity = 4.9 mm/s.

\[ HCO_3^- + OH^- \rightleftharpoons CO_3^{2-} + H_2O \]  

(21)

At high $pH$ the second reaction is considered to be instantaneous. Thus in the present study the overall reaction can be simplified into:

\[ CO_2 + 2OH^- \rightleftharpoons CO_3^{2-} + H_2O \]  

(22)

Since the first reaction is rate determining, the rate of the overall reaction is taken from the first reaction. The forward and backward reaction rate can respectively be formulated as:

\[ R_{1,1} = k_{1,1} [CO_2] [OH^-] \]  

(23)

\[ R_{1,2} = k_{1,2} [HCO_3^-] \]  

(24)

where $k_{1,1}$ and $k_{1,2}$ are the forward and backward reaction rate constants respectively.

The influence of the chemical reaction on the gas absorption process is usually accounted for by an enhancement factor, $E$. This factor can be calculated numerically by solving the (coupled) diffusion equations in the boundary layer surrounding the bubbles. The equations should be solved for each individual bubble, hence in the case where a large number of bubbles is present this method becomes impractical. In the present study, an approximate solution of the enhancement factor is used. The enhancement factor is approximated in a simple algebraic form as a function of the Hatta number. This approach provides a much cheaper solution for the enhancement factor for each individual bubble.

During the chemisorption process, due to the high interphase mass transfer rate, it is possible that bubbles are completely dissolved in the liquid. In our model a bubble is considered to be completely dissolved in the liquid when the bubble radius is less than 1 $\mu$m, while the bubble growth rate is negative (i.e. the bubble is still dissolving). If a bubble meets these criteria, it will be disposed from the column and no longer being tracked.

The chemisorption problem is simulated using the DBM model. CO$_2$ gas is fed into the column filled with NaOH solution with an initial $pH$ of 14. Three species transport equations are utilized to track mass fractions of CO$_2$(l), OH$^-$ and CO$_3^{2-}$. A summary of the initial conditions and the source terms used in each transport equation is presented in Table 3.

Fig. 9 shows the transient behavior in the column immediately after the CO$_2$ bubbles are injected into the column. Shortly after bubbles are released into the column, a typical mushroom like shape...
Table 3: Initial conditions and source terms used in the species transport equations.

<table>
<thead>
<tr>
<th>$j$</th>
<th>Species</th>
<th>$Y_j(t=0)$</th>
<th>$S_j$</th>
<th>$k_g$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>$CO_2(l)$</td>
<td>$1 \times 10^{-50}$</td>
<td>$-R_{1,1} + R_{1,2}$</td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>$OH^-$</td>
<td>$Y_1(pH)$</td>
<td>$2(-R_{1,1} + R_{1,2})$</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>$CO_3^{2-}$</td>
<td>$1 \times 10^{-50}$</td>
<td>$R_{1,1} - R_{1,2}$</td>
<td></td>
</tr>
</tbody>
</table>

appears. This shape is however, less pronounced compared to the case without absorption as described in section 4.1. Due to the chemically enhanced mass transfer, the bubbles are completely dissolved within $0.15 \text{ m}$ from the bottom and remain in that position for quite a while. Although the bubbles only appear in the lower part of the column, the liquid circulation induced by the bubbles extends to the top portion of the column. Compared to the case without absorption, the liquid phase oscillation is only observed in the higher part of the column. Since no bubbles are present in this region, these oscillations do not interact with bubbles, which makes them less pronounced.

![Figure 9](image)

Figure 9: Set of corresponding velocity fields (top), bubble position (bottom) obtained from simulation of the chemisorption of $CO_2$ gas in liquid $NaOH$ at various time after the $CO_2$ gas was switched on. $pH_0 = 14$, pure $CO_2$ gas with superficial velocity = 4.9 mm/s.

Fig. 10 shows the transient behavior of chemical species distribution in the column. Since bubbles are only present in the lower part of the column, the mass transfer and reactions only take place close to the gas inlet. $CO_2$ gas, which dissolves into the liquid from the bubbles immediately reacts with hydroxide to produce carbonate, hence only very small traces of dissolved $CO_2$ can be found in the vicinity of the bubbles. The hydroxide ions that have been consumed by the reaction as well as the newly formed carbonate ions will be transported by the circulation flow to the top of the column in the center region and return back again through the downflow zone close to the column walls. This
Figure 10: Set of corresponding $CO_2$ concentration [mol/l] (top), $CO_2^{2-}$ concentration [mol/l] (middle) and liquid phase $pH$ [-] (bottom) obtained from simulation of the chemisorption of $CO_2$ gas in liquid NaOH at various time after the $CO_2$ gas was switched on. $pH_0 = 14$; pure $CO_2$ gas with superficial velocity= 4.9 mm/s.
behavior suggests that the reactions taking place in the bottom part of the column are always supplied with relatively fresh reactant, since the product of the reaction is immediately transported by the liquid flow.

\[
\begin{align*}
\text{pH} &= 14 \\
\text{pH} &= 13.8 \\
\text{pH} &= 13.6 \\
\text{pH} &= 13.4 \\
\text{pH} &= 13.2 \\
\text{pH} &= 13.0
\end{align*}
\]

Figure 11: Snapshot of bubble position at time \( t = 10 \) s after the \( CO_2 \) gas was switched on with variation of initial liquid \( NaOH \) concentration. Pure \( CO_2 \) gas with superficial velocity= \( 4.9 \) mm/s.

With time, the \( pH \) is decreasing and the carbonate concentration is continuously increasing. As the \( pH \) decreases, the enhancement factor also decreases resulting in a lower mass transfer rate. In time the position where the bubbles are completely dissolved will slowly move upward. This behavior has been experimentally observed by Fleischer et al. (1996). To investigate whether our model is capable to predict such behavior, a series of simulations with an initial \( pH \) varying from 13 up to 14 has been conducted. This technique is used to save calculation time, since the rate of change of \( pH \) is very low. Fig. 11 shows that the afore mentioned behavior is nicely predicted by the current model. As the \( pH \) decreases the position where bubbles are completely dissolved moves upward and since the model is also able to predict the decrease of \( pH \) in time we can conclude that this behavior can also be captured if we would proceed the simulation for a sufficiently long period.

5 Conclusions

A model that combines hydrodynamics, mass transfer and chemical reaction in a bubble column has been successfully formulated and implemented. The simulation results obtained show that the model can be used to investigate those phenomena in more detail than before.

The hydrodynamics model has been validated using the experimental data of Deen et al. (2001). Both instantaneous and time-averaged liquid velocities predicted by the model are in good agreement with the experimental data. The experimentally observed meandering of the bubble plume is also nicely predicted by the present model.

In the case of the physical absorption of \( CO_2 \) bubbles in water, it was found that the distribution of dissolved \( CO_2 \) is not uniform. Relatively high dissolved \( CO_2 \) was found in the vicinity of bubbles. In time the dissolved \( CO_2 \) is accumulated in the water until the equilibrium is attained.

The model was also used to investigate the chemisorption of \( CO_2 \) gas in \( NaOH \) solution. This process has been studied experimentally by Fleischer et al. (1996). The model is able to reveal liquid phase hydrodynamics, bubble size and position as well as chemical species distribution involved in the reaction. In simulations with initial \( pH = 14 \), bubbles are only present close to the gas inlet. Only very small traces of dissolved \( CO_2 \) can be found in the vicinity of the bubbles while the hydroxide ions that have been consumed and newly formed carbonate ions are transported through the whole
column. In time the pH is decreasing while the carbonate concentration is increasing. The model is also able to predict the position of completely dissolved bubbles due to chemically enhanced mass transfer. Variation of initial pH shows that this position shifts upward in the column as the pH decreases.

Due to the complexity and detailed information that the present model provides, calculation time is still the bottleneck especially for problems which involve a large number of bubbles and chemical species. In the current work, coalescence and break up were not considered. However, appropriate coalescence and break up models can readily be implemented due to the lagrangian treatment of the bubble phase.

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References


