

# CFD-based investigation of carbon dioxide absorption in a falling-film micro-absorber

Paris Chasanis, Anna Lautenschleger, Eugeny Y. Kenig  
University of Paderborn, Chair of Fluid Process Engineering,  
Pohlweg 55, 33098 Paderborn, Germany

A rigorous mathematical model is developed to describe chemical absorption of carbon dioxide in a falling-film micro-contactor. Compared to previous models described in literature, hydrodynamics in both phases is accurately captured by the Navier-Stokes equations. By this means, backmixing effects can be accounted for. Furthermore, the interface position is directly determined using the level-set method. The developed model is validated with experimental data obtained from literature and good agreement is established. On the basis of the validated model, detailed sensitivity studies are performed to investigate the impact of process parameters.

## 1. Introduction

In recent years, chemical micro processes have attracted a significant interest of both chemical process industry and research community. These processes occur in equipment with dimensions at micrometer and sub-millimeter scale. As a consequence, high surface areas per unit volume and small diffusion paths are achieved resulting in intensified mass transfer.

In this work, a two-dimensional model is developed to investigate carbon dioxide absorption in a falling-film micro-apparatus. The exact position of the interface is identified by means of the level set method. Afterwards, the computational domain is subdivided into a gas-phase and a liquid-phase part, whereas the interface is fixed at the position determined previously. In this way, the exact velocity and concentration fields are determined by the conservation equations for overall mass, momentum and species. The model is validated by means of experimental data obtained by Zanfir et al. (2004). On the basis of the validated model, studies are performed to investigate the influence of different process parameters.

## 2. Chemical system

In this work, carbon dioxide absorption from a N<sub>2</sub>-CO<sub>2</sub> mixture into a sodium hydroxide solution is investigated. The absorbed CO<sub>2</sub> reacts with OH<sup>-</sup> ions in two steps:





The overall reaction can be written as



The reaction of  $\text{CO}_2$  with water towards bicarbonate and hydrogen ions is only important at low pH values. In this study, we restrict our investigations to alkaline solutions in which the pH value does not fall below 12. Hence, this reaction can be neglected. The almost irreversible reaction (1) is the limiting step, whereas reaction (2) can be considered to be instantaneous (Taghizadeh et al., 2001). The equilibrium of the latter reaction is pH-value dependent. At pH values above 12, carbonic acid exists only in the form of carbonate ions.

### 3. Micro-absorber geometry and mathematical model

The liquid phase is distributed along a vertical reaction plate comprising 64 open-top microchannels with dimensions 0.3 x 0.1 x 66.4 mm. It flows downwards to the withdrawal zone, whereas the gas phase is guided co-currently in a gas-phase chamber. The latter is arranged parallel to the reaction plate and has a depth of 5.5 mm. Figure 1a shows the principle micro-absorber configuration.

It is assumed that all microchannels are characterized by identical conditions, and a two-dimensional model is developed to describe the absorption process in a single microchannel. In a first step, the species transport is neglected, and the exact position of the interface is identified by means of the level set method. This approach is justified, as, due to low  $\text{CO}_2$  inlet concentrations, species transfer does not affect material properties (e.g. density and viscosity) and two-phase flow behaviour. Afterwards, the microchannel is subdivided into a gas-phase and a liquid-phase domain, whereas the interface is fixed at the position determined in the previous step (see Figure 1b). The process under study is isothermal ( $T=20^\circ\text{C}$ ), as reported by Zafir et al. (2005).

The flow of each phase is described by means of the steady-state continuity and Navier-Stokes equations. Velocity and shear stress continuity are assumed at the interface. Finally, the no-slip boundary condition is applied to all walls, whereas velocity values are pre-set at the inlets and pressure is set equal to atmospheric pressure at the outlet. Species transport in both phases is described by conservation equations for all components.

Under the conditions described in Section 2, the overall reaction rate can be expressed by a second-order kinetic equation:

$$r = k_{\text{OH}^-} c_{\text{CO}_2, \text{L}} c_{\text{OH}^-, \text{L}} \quad (4)$$

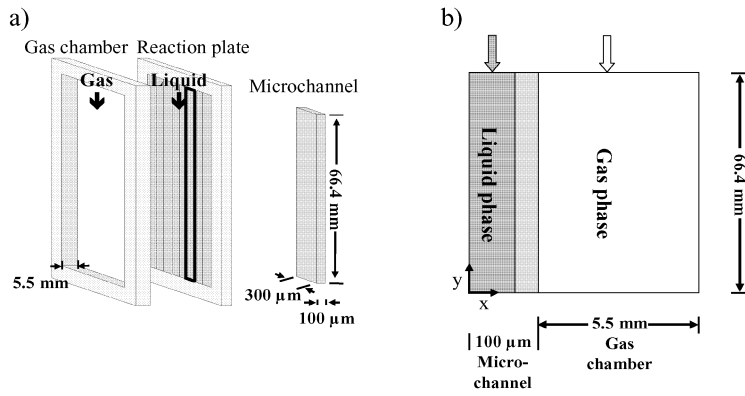


Figure 1. General micro-absorber configuration (a) and scheme of liquid and gas domains of the 2D mathematical model (b).

Thermodynamic equilibrium and the component flux continuity are assumed at the interface.  $\text{CO}_2$  is the only species transferred through the interface. Furthermore, the diffusive flux at the outlets is neglected as it is small compared to the respective convective flux. Finally, all walls are considered to be impermeable and molar concentration values are defined at the inlets.

The mathematical model is implemented in the commercial CFD tool COMSOL Multiphysics by COMSOL AB, which is based on the finite element method.

## 4. Results and discussion

### 4.1 Model validation

The simulated absorption performance is compared with experimental results determined by Zafir et al. (2005). It should be noted that only experiments with low  $\text{CO}_2$  inlet concentrations, in which the pH value of the sodium hydroxide solution does not fall below 12, are considered. However, different NaOH and  $\text{CO}_2$  inlet concentrations as well as different liquid-phase and gas-phase volume flows are investigated. To quantify the separation performance,  $\text{CO}_2$  conversion is defined as follows:

$$U_{\text{CO}_2} = 1 - \frac{N_{\text{CO}_2,\text{G}}|_{\text{outlet}}}{N_{\text{CO}_2,\text{G}}|_{\text{inlet}}} \cdot 100 [\%] \quad (5)$$

where  $N_{\text{CO}_2,\text{G}}|_{\text{inlet/outlet}}$  denote the  $\text{CO}_2$  molar flows entering and leaving the gas-phase chamber.

A parity plot shown in Figure 2a gives a comparison between simulated and experimental results for different process conditions. Good agreement is achieved, which, above all, can be attributed to the accurate description of hydrodynamics. Especially important is that backmixing effects in the gas phase, observed whenever the gas-phase inlet velocity is distinctly smaller than the liquid-phase inlet velocity, can be

properly taken into account (see Figure 2b). These effects have a significant impact on  $\text{CO}_2$  mass transfer and thus, on absorption performance.

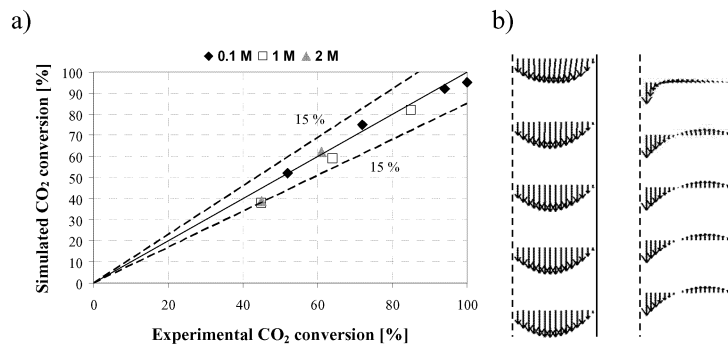


Figure 2. Model validation with experimental data from Zanfir et al. (2005) for different process conditions (a) and gas velocity arrow plots (b): Liquid volume flow rate is 50 ml/h, NaOH inlet concentration in the liquid phase is 0.1 mol/l,  $\text{CO}_2$  inlet molar fraction in the gas phase is 0.001 and the gas volume flow rate is 11.4 l/h (left) and 0.23 l/h (right). The dashed line indicates the gas-liquid interface.

## 5. Sensitivity studies

### 5.1 Impact of gas-phase volumetric flow rate

The gas-phase volumetric flow rate is varied between 0.23 and 11.4 l/h, whereas the liquid-phase flow rate is kept constant at 50 ml/h and the NaOH concentration at 0.1 mol/l for all simulations. The  $\text{CO}_2$  inlet molar fraction in the gas phase was 0.001, while the sodium hydroxide solution was  $\text{CO}_2$ -free at the inlet. The achieved conversion degrees are shown in Figure 3. Clearly,  $\text{CO}_2$  conversion increases by reducing gas-phase volumetric flow rate, as just more time is available for the carbon dioxide transport to the liquid phase. High conversion degrees up to almost 100% can be observed for low gas-phase flow rates. On the other hand, for small gas-phase residence times,  $\text{CO}_2$  transfer to the liquid phase is noticeably limited.

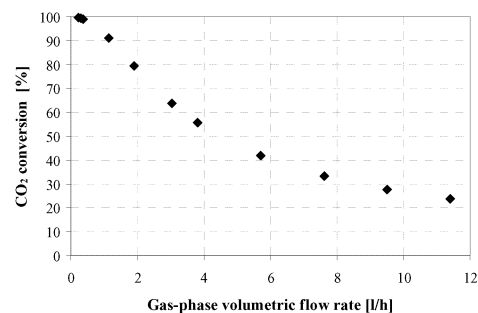


Figure 3. Impact of gas-phase volumetric flow rate on  $\text{CO}_2$  conversion.

### 5.2 Impact of liquid-phase volumetric flow rate

The liquid-phase flow rate is varied between 5 and 200 ml/h, whereas the gas volume flow is set to 3.8 l/h. The NaOH inlet molar concentrations in the liquid phase are set to 0.1 mol/l and the CO<sub>2</sub> molar fraction in the gas phase is 0.001. As can be seen in Figure 4a, the impact of the liquid-phase flow rate on CO<sub>2</sub> conversion degree is relatively low. Only in the region of small liquid-phase flow rates (5-15 ml/h), a noticeable increase of CO<sub>2</sub> conversion degree can be observed. This can be explained by high NaOH conversion degree change in this region (see Figure 4b) resulting in high reaction rate change. A further liquid-phase flow rate increase causes only minimal NaOH conversion changes and, thus, a nearly constant reaction rate is reached. As sufficient amount of OH<sup>-</sup>-ions is now available, the reaction is no longer a limiting factor. Hence, by further increasing the liquid-phase volumetric flow rate, the separation efficiency is hardly influenced.

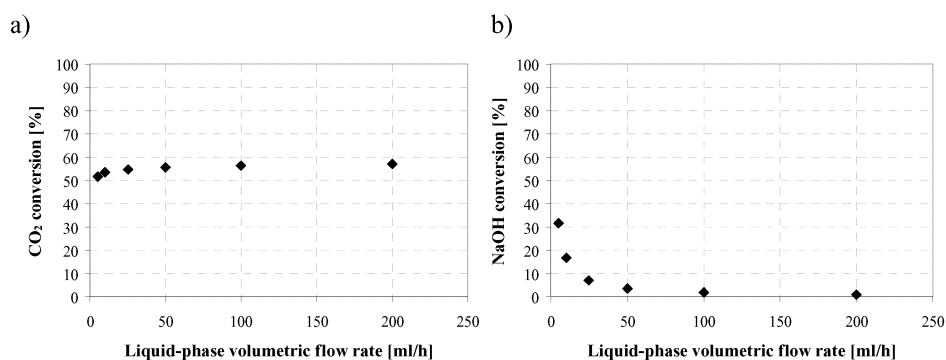


Figure 4. Impact of liquid-phase volumetric flow rate on CO<sub>2</sub> conversion (a) and on NaOH conversion (b).

### 5.3 Impact of NaOH inlet molar fraction

The NaOH inlet molar concentration is varied between 0.1 and 3 mol/l. The liquid-phase volumetric flow rate is set to 50 ml/h, whereas the gas-phase flow rate is equal to 3.8 l/h and the CO<sub>2</sub> gas-phase inlet concentration is 0.01. In Figure 5, the CO<sub>2</sub> conversion degree as a function of NaOH inlet molar concentration is shown.

The conversion degree increases until it reaches a maximum at an inlet NaOH molar concentration of about 2 mol/l. Further increasing of NaOH concentration results in a CO<sub>2</sub> conversion decline. This results from the fact that increasing NaOH concentration affects different factors, which impact separation performance oppositely. On the one hand, the reaction rate is increased, which results in higher liquid-phase uptake rate. On the other hand, the density and viscosity of the liquid phase are increased, whereas the diffusion coefficient is decreased. The viscosity and density increase results in larger film thicknesses and, thus, in lower liquid-phase velocities. Consequently, convective supply of OH<sup>-</sup>-ions becomes slower. On the contrary, the diffusion coefficient decrease leads to a decline of diffusive OH<sup>-</sup> supply. The interplay of these effects explains the maximum visible in Figure 5.

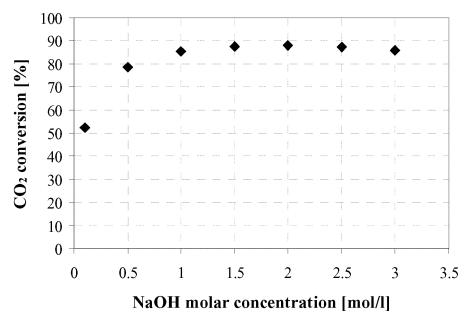


Figure 5. Impact of NaOH liquid phase inlet concentration on CO<sub>2</sub> conversion.

## 6. Conclusions

In this work, carbon dioxide absorption into a sodium hydroxide solution in a microstructured falling-film contactor is investigated numerically. A rigorous 2D-model is developed for this purpose. The model is validated with experimental data from literature and a good agreement between simulated and experimental data is found. On the basis of the validated model, sensitivity studies are performed to investigate the impact of different process parameters on absorption performance. These studies clearly indicate that high CO<sub>2</sub> conversion degrees and simultaneously high efficiency of applied absorbent can be achieved in the investigated apparatus. It is found that, for sufficiently high NaOH amount, the gas-phase CO<sub>2</sub> mass transfer represents the rate-limiting step. In case of low NaOH concentrations, the chemical reaction becomes the limiting factor. The developed model is general enough to be used for a wide range of process conditions in microstructured falling-film contactors. Especially when backmixing effects appear, it should give considerably more accurate prediction compared to simplified methods widely used to describe falling-film processes. The exact localisation of the interface by means of the level set method will enable the development of geometry optimization approaches, which is hardly possible with conventional models employing simplified methods, e.g., the Nusselt solution.

## References

- Taghizadeh M, Jallut C., Tayakout-Fayolle M. and Lieto J., 2001. Non-isothermal gas-liquid absorption with chemical reaction studies Temperature measurements of a spherical laminar film surface and comparison with a model for the CO<sub>2</sub>/NaOH system, *Chemical Engineering Journal*, 82, 143–148.
- Zanfir M., Gavrilidis A., Wille Ch. and Hessel V., 2005. Carbon dioxide absorption in a falling film microstructured reactor: experiments and modeling, *Ind. Eng. Chem. Res.*, 44, 1742-1751.