Study of the CO$_2$ Transfer Rate in a Reacting Flow for the Refined Sodium Bicarbonate Production Process

Christophe Wylock (F.R.S.-FNRS research fellow), Aurélie Larcy, Pierre Colinet, Thierry Cartage and Benoît Haut

Chemical Engineering Department
Applied Science Faculty, Free University of Brussels
Table of content

- Introduction
- Modelling
- Simulation results
  - Validation
  - For industrial operating conditions
  - Comparison with a 1-D common approach
- Conclusions and future plan
Refined sodium bicarbonate (NaHCO$_3$) production (Solvay) process in bubble columns (BIR columns)

Limiting step: gas-liquid CO$_2$ absorption
Introduction

- Main resistance: in the liquid phase, where CO₂ takes part to chemical reactions
- This work: modelling of the CO₂ transfer rate from a bubble to the liquid phase

2 reversible chemical reactions:
- \(\text{CO}_2 + \text{NaOH} \rightleftharpoons \text{NaHCO}_3\)
- \(\text{Na}_2\text{CO}_3 + \text{H}_2\text{O} \rightleftharpoons \text{NaHCO}_3 + \text{NaOH}\)
Introduction

- Main resistance: in the liquid phase, where CO$_2$ takes part to chemical reactions
- This work: modelling of the CO$_2$ transfer rate from a bubble to the liquid phase
  - Coupling of
    - Convective transport
    - Diffusive transport
    - Chemical reactions
- Interfacial adsorbed surfactants: change the flow field around the bubble → 2 cases investigated:
  - fully contaminated bubble (no slip)
  - clean bubble (slip)
Table of content

- Introduction
- Modelling
- Simulation results
  - Validation
  - For industrial operating conditions
  - Comparison with a 1-D common approach
- Conclusions and future plan
Modelling

- Incompressible Navier-Stokes mode and Convection and Diffusion mode from the C.E. module
- 2-D axisymmetric geometry
- Computational domain
  - Semi-bubble located at the center of a semi-circular domain
  - Inertial reference frame located at the mass center of the bubble

Dimensionless bubble diameter: \( d_b = 1 \)
Domain diameter: \( 5 \, d_b \)
Modelling

- Governing equations (in vectorial dimensionless form)
  - Navier-Stokes and continuity
    \[
    \begin{aligned}
    (u \cdot \nabla) u &= \nabla \left[ -p + \frac{1}{Re} \left( \nabla u + (\nabla u)^T \right) \right] \\
    \nabla \cdot u &= 0
    \end{aligned}
    \]
  - Mass transport coupled with chemical reactions
    
    \[\begin{aligned}
    \nabla \left( \frac{1}{Pe} \nabla a \right) &= -r_1 - (u \cdot \nabla) a \rightarrow \text{CO}_2 \text{ concentration} \\
    \nabla \left( \frac{1}{Pe} \nabla b \right) &= \nabla \left( -r_1 - r_2 - (u \cdot \nabla) b \right) \rightarrow \text{NaOH concentration} \\
    \nabla \left( \frac{1}{Pe} \nabla c \right) &= \nabla \left( r_1 - r_2 - (u \cdot \nabla) c \right) \rightarrow \text{NaHCO}_3 \text{ concentration} \\
    \nabla \left( \frac{1}{Pe} \nabla d \right) &= \nabla \left( r_2 - (u \cdot \nabla) d \right) \rightarrow \text{Na}_2\text{CO}_3 \text{ concentration}
    \end{aligned}\]

    - 1st reaction rate: \[ r_1 = \frac{Ha_1}{ab - ca} \rightarrow \text{Hatta1 number} \]
    - 2nd reaction rate: \[ r_2 = \frac{Ha_2}{bc - d} \rightarrow \text{Hatta2 number} \]
**Modelling**

- **Meshing**
  - Concentric circular mapped mesh
  - Finer in the vicinity of the interface

  ![Mesh Diagram]

  Thickness: $0.05 d_b$

  The diffusion boundary layer does not lie beyond this zone

- **Solver**: stationnary UMFPACK
Table of content

- Introduction
- Modelling
- Simulation results
  - Validation
  - For industrial operating conditions
  - Comparison with a 1-D common approach
- Conclusions and future plan
1) Validation by comparison of the simulation results WITHOUT reactions with classical correlations from literature

**Drag coefficient**

- COMSOL fully contaminated
- Lapple et al. (eq. 24)
- Clift et al. (eq. 25)
- COMSOL clean
- Hamelie et al. (eq. 22)
- Haas et al. (eq. 23)

**Sherwood number**

- COMSOL clean
- Loeschl&Calderbank (eq. 27)
- Boussinesq (eq. 28)
- COMSOL fully contaminated
- Clift et al. (eq. 29)
- Clift et al. (eq. 30)

**Separation angle**

- COMSOL fully contaminated
- Clift et al. (eq. 26)

Excellent agreement → validated
Table of content

- Introduction
- Modelling
- Simulation results
  - Validation
  - For industrial operating conditions
  - Comparison with a 1-D common approach
- Conclusions and future plan
1) Validation by comparison of the simulation results without reactions with classical correlations from literature: OK

2) For operating conditions of BIR columns
   - Bubble: 1 mm diameter and rising velocity of 0.2 m/s
     \[ Re = 200 \text{ and } Pe = 100\,000 \]
   - Other parameter values:\(^1\):
     \[
     \begin{align*}
     \alpha &= 0.003 \\
     Ha_1 &= 0.19 \\
     Ha_2 &= 902 \\
     \beta_b &= 4.1 \\
     \beta_c &= 0.9 \\
     \beta_d &= 0.7 \\
     \chi_b &= 64 \\
     \chi_c &= 0.03 \\
     \chi_d &= 0.025
     \end{align*}
     \]
     Study of the CO\(_2\) transfer rate as a function of the Hatta1 number (dimensionless ratio of chemical reaction 1 rate on CO\(_2\) diffusion rate)

\(^1\) correlations from Vas Bhat et al. (2000)
Simulation results

- Simulations of the CO$_2$ concentration field
  - No reactions: $Ha_1=0$ (and $Ha_2=0$)

Fully contaminated bubble

Clean bubble

![Simulation images]
Simulation results

- Simulations of the CO$_2$ concentration field
  - Slow reaction 1: $H \sigma_1 = 0.1$

Fully contaminated bubble
Clean bubble
- **Simulations of the CO$_2$ concentration field**
  - Moderate reaction 1: $Ha_1 = 1$

  Fully contaminated bubble
  
  Clean bubble

Max: 0.9

Min: 0.1

Max: 0.9

Min: 0.1
Simulation results

- Simulations of the CO$_2$ concentration field
  - Fast reaction 1: $Ha_1=10$

Fully contaminated bubble

Clean bubble
Simulation results

- Simulations of the CO₂ concentration field
  → Increasing CO₂ depletion for increasing reaction 1 rate

- Calculation of the CO₂ transfer rate:

  → The CO₂ consumption enhances the CO₂ transfer rate
Table of content

- Introduction
- Modelling
- Simulation results
  - Validation
  - For industrial operating conditions
  - Comparison with a 1-D common approach
- Conclusions and future plan
3) Comparison of the 2-D axysymmetric clean bubble case and a commonly-used 1D-approach of the chemical engineering

- Description of the Higbie approach
  - Liquid flow: mosaic of liquid elements slipping on the bubble
  - Each element stays in contact with the bubble the same time
  - No shear stress in the liquid
  - Diffusion is normal to the interface

\[
\begin{align*}
\frac{\partial a}{\partial t} &= \frac{1}{Pe} \frac{\partial^2 a}{\partial x^2} - r_1 \\
\frac{\partial b}{\partial t} &= \frac{\beta_b}{Pe} \frac{\partial^2 b}{\partial x^2} + \chi_b \left( -r_1 - r_2 \right) \\
\frac{\partial c}{\partial t} &= \frac{\beta_c}{Pe} \frac{\partial^2 c}{\partial x^2} + \chi_c \left( r_1 - r_2 \right) \\
\frac{\partial d}{\partial t} &= \frac{\beta_d}{Pe} \frac{\partial^2 d}{\partial x^2} + \chi_d r_2
\end{align*}
\]

Axis pointed toward the liquid phase in normal direction of the interface
Simulation results

- Comparison results

- The Higbie approach provides an excellent estimation when $Ha_1 > 1$.
# Table of Content

- Introduction
- Modelling
- Simulation results
  - Validation
  - For industrial operating conditions
  - Comparison with a 1-D common approach
- Conclusions and future plan
Conclusion and future plans

- Development of a model of bubble-liquid CO\(_2\) transfer coupled with chemical reactions (for 2 cases):
  - Validation without reaction: excellent agreement
  - Estimation of the chemical enhancement on the transfer rate
  - Excellent comparison for the transfer rate estimation between 2-D clean bubble case and 1-D Higbie approach

- Future plans
  - Extension to larger bubbles (2 - 6 mm)
    - \(400 \leq Re \leq 1200\)
    - Spherical bubble \(\rightarrow\) ellipsoidal-shape bubble
    - Shape coming from experimental observation
  - Comparison with spherical shape \(\rightarrow\) quantification of the shape effect
Thanks for your attention