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Study of the CO_2 Transfer Rate in a Reacting Flow for the Refined Sodium Bicarbonate Production Process

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  - Comparison with a 1-D common approach
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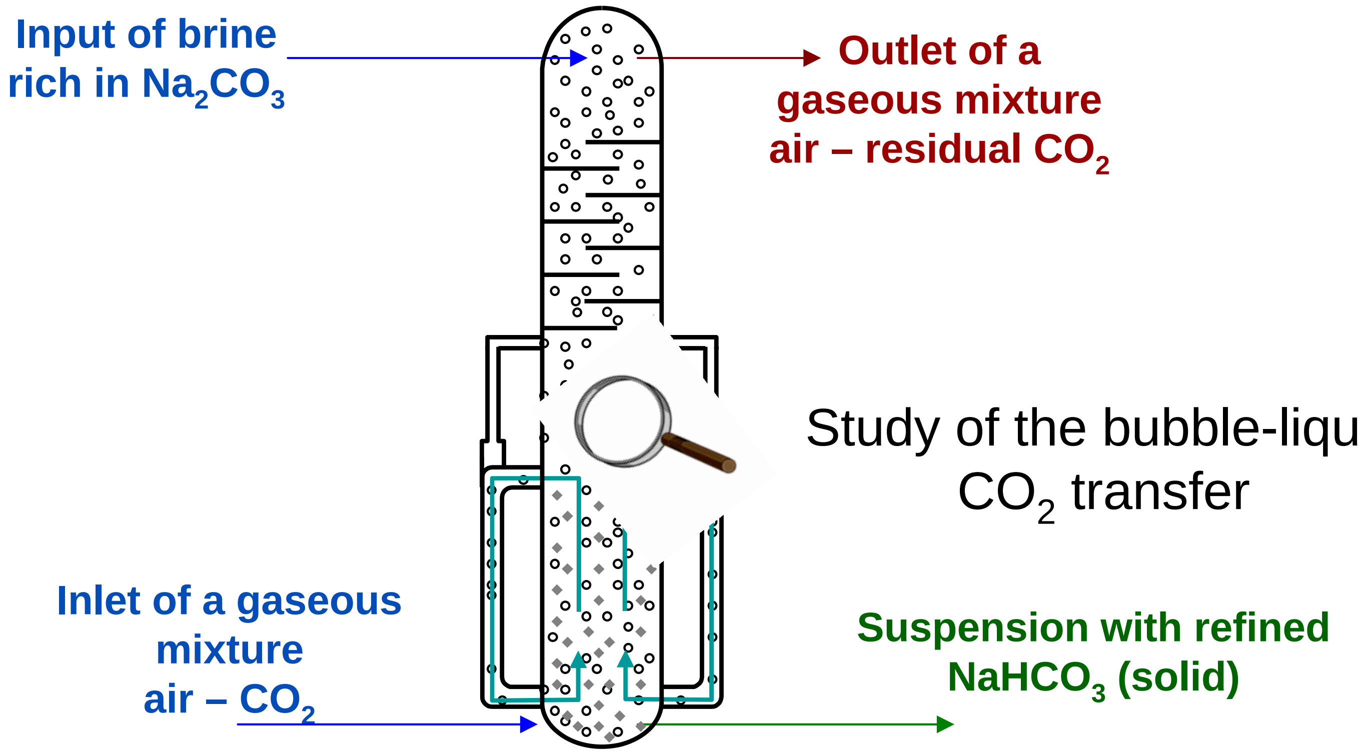
Introduction

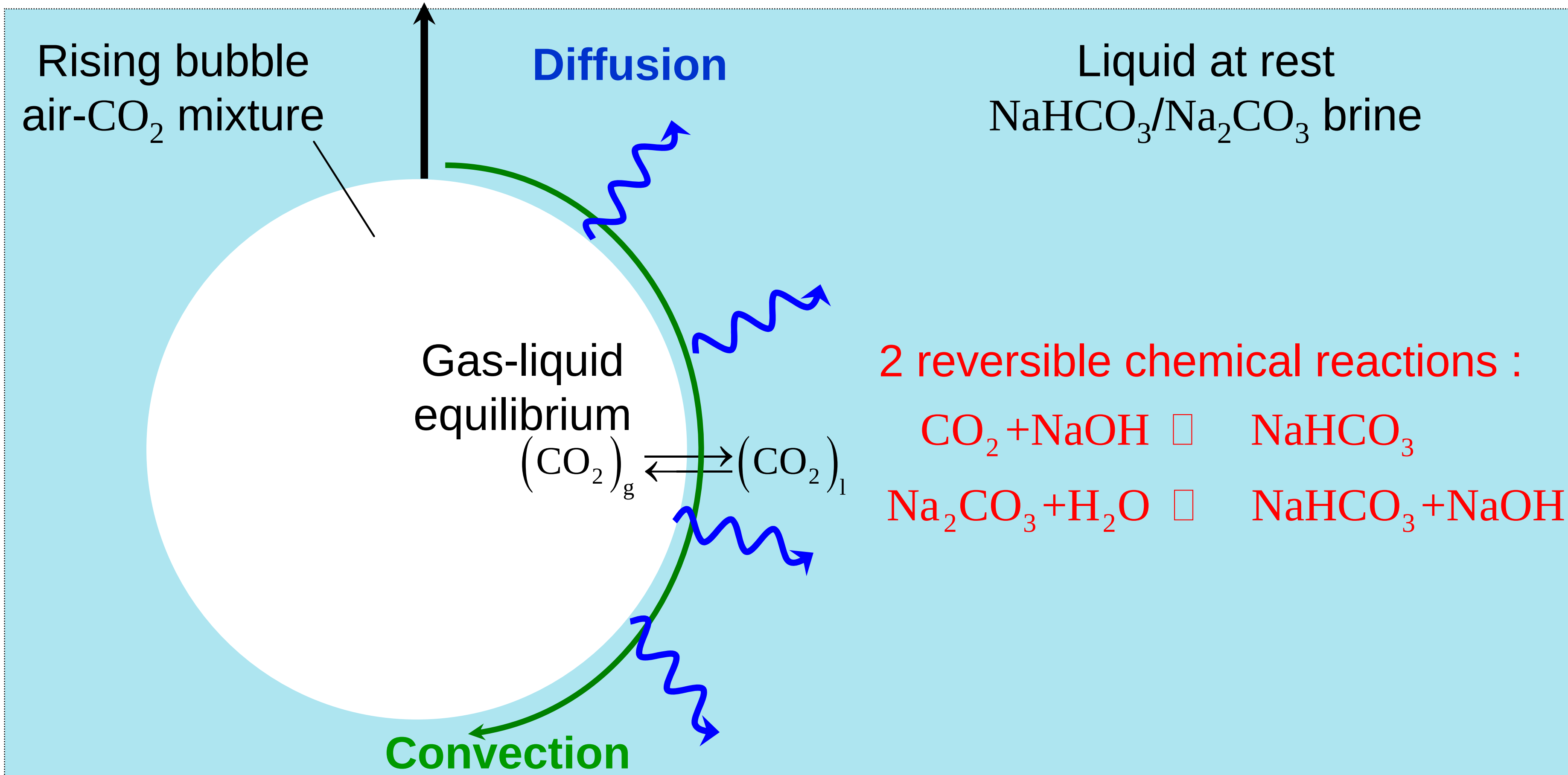
- **Refined sodium bicarbonate ($NaHCO_3$) production (Solvay) process in bubble columns (BIR columns)**
- **Limiting step : gas-liquid CO_2 absorption**



Study of the bubble-liquid CO_2 transfer

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**



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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)

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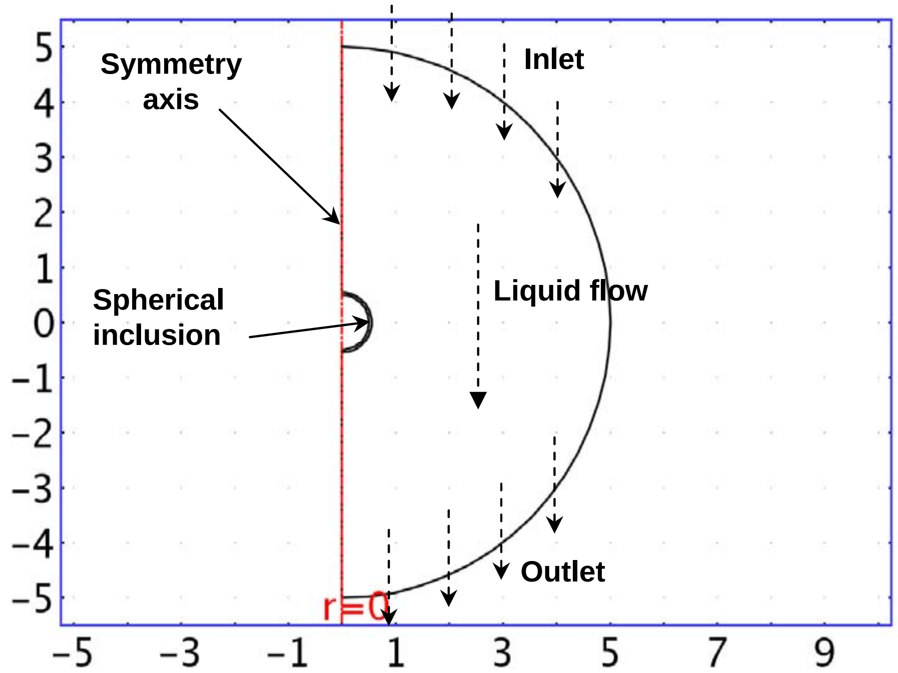
Modelling

- **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: $\tilde{d}_b = 1$

Domain diameter: $5\ \tilde{d}_b$

ULB Modelling

- **Governing equations (in vectorial dimensionless form)**
  - Navier-Stokes and continuity

$$\begin{cases} (\boldsymbol{u}\cdot\nabla)\boldsymbol{u} = \nabla\cdot\left[-p\mathbf{I}+\frac{1}{Re}\left(\nabla\boldsymbol{u}+(\nabla\boldsymbol{u})^{\mathrm{T}}\right)\right] \\ \nabla\cdot\boldsymbol{u}=0 \end{cases}$$

velocity ($\boldsymbol{u}$), pressure (p), Reynolds number (Re)

  - Mass transport coupled with chemical reactions

$$\begin{cases} \nabla\cdot\left(-\frac{1}{Pe}\nabla a\right) = -r_1-(\boldsymbol{u}\cdot\nabla)a \\ \nabla\cdot\left(-\frac{\beta_b}{Pe}\nabla b\right) = \chi_b\left(-r_1-r_2\right)-(\boldsymbol{u}\cdot\nabla)b \\ \nabla\cdot\left(-\frac{\beta_c}{Pe}\nabla c\right) = \chi_c\left(r_1-r_2\right)-(\boldsymbol{u}\cdot\nabla)c \\ \nabla\cdot\left(-\frac{\beta_d}{Pe}\nabla d\right) = \chi_d r_2-(\boldsymbol{u}\cdot\nabla)d \end{cases}$$

Peclet number (Pe)

a: CO_2 concentration

b: NaOH concentration

c: $NaHCO_3$ concentration

d: Na_2CO_3 concentration

1st reaction rate: $r_1 = Ha_1^2(ab-\alpha c)$, with Ha_1: Hatta1 number; $\alpha = [CO_2]_{bulk}/[CO_2]_i$

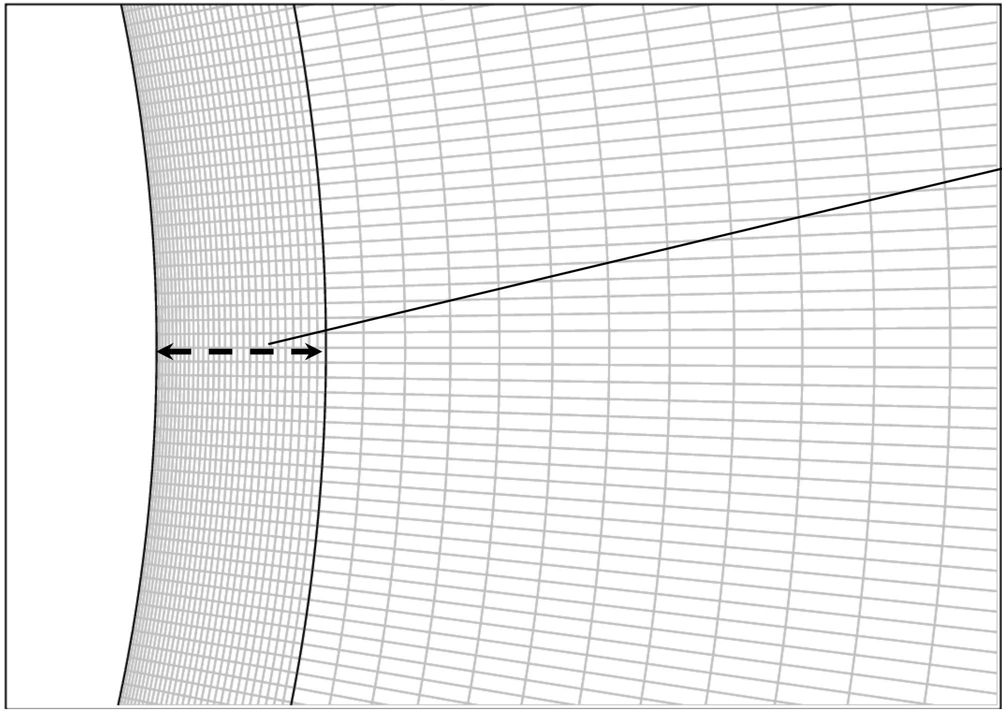
2nd reaction rate: $r_2 = Ha_2^2(bc-d)$, with Ha_2: Hatta2 number

$\beta_b = D_{OH^-}/D_{CO_2}$, $\beta_c = D_{HCO_3^-}/D_{CO_2}$, $\beta_d = D_{CO_3^=}/D_{CO_2}$

$\chi_b = [CO_2]_i/[NaOH]_{bulk}$, $\chi_c = [CO_2]_i/[NaHCO_3]_{bulk}$, $\chi_d = [CO_2]_i/[Na_2CO_3]_{bulk}$

Modelling

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



Thickness : $0.05 \cdot d_b$

The diffusion boundary layer does not lie beyond this zone

- **Solver : stationnary UMFPACK**

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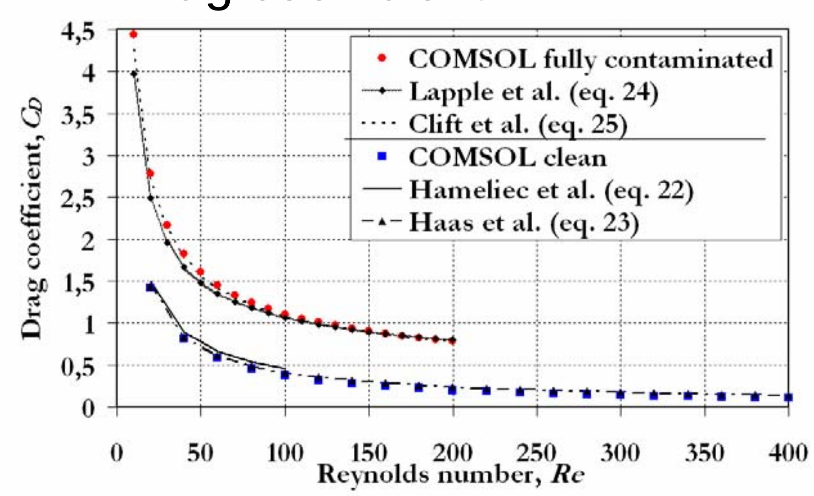
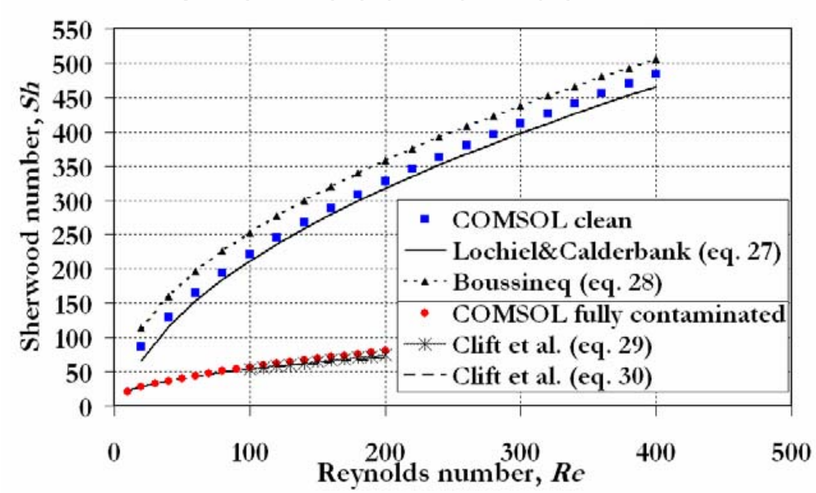
ULB Simulation results

1) Validation by comparison of the simulation results WITHOUT reactions with classical correlations from literature

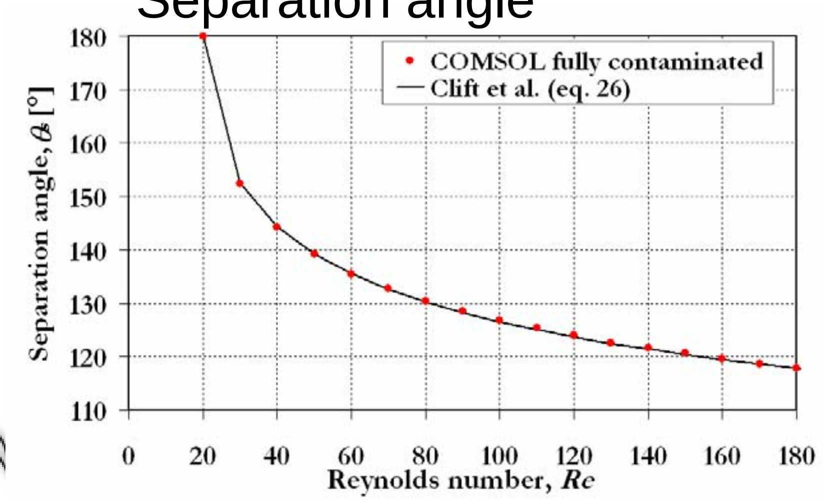
Drag coefficient



Sherwood number



Separation angle



Excellent agreement
→ validated

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Simulation results

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 and Pe = 100 000
   - Other parameter values[1]:

$$\alpha = 0.003 \qquad Ha_1 = 0.19 \qquad Ha_2 = 902$$

$$\beta_b = 4.1 \qquad \beta_c = 0.9 \qquad \beta_d = 0.7$$

$$\chi_b = 64 \qquad \chi_c = 0.03 \qquad \chi_d = 0.025$$

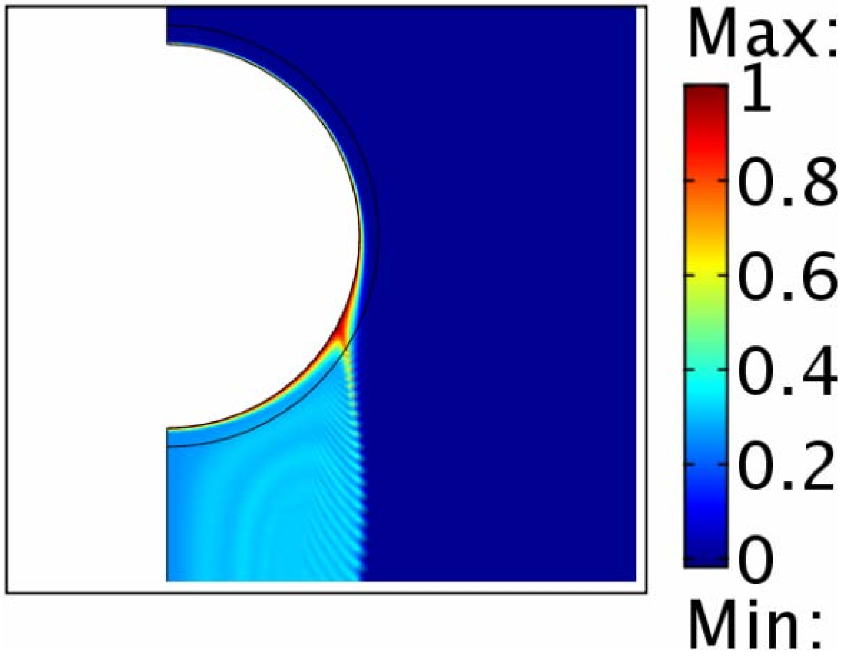
➔ **Study of the CO_2 transfer rate as a function of the Hatta1 number (dimensionless ratio of chemical reaction 1 rate on CO_2 diffusion rate)**

[1] correlations from Vas Bhat et al. (2000)

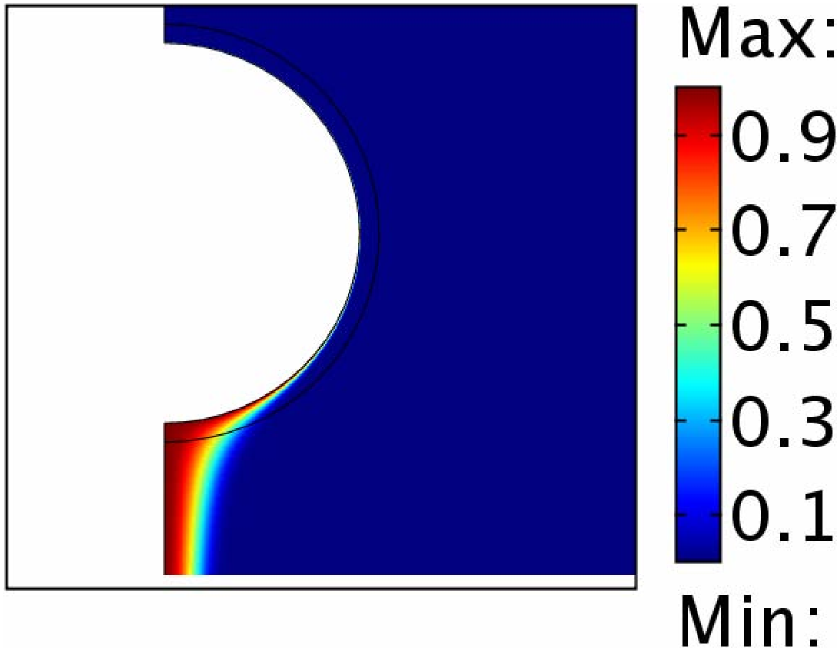
Simulation results

- **Simulations of the CO_2 concentration field**
  - No reactions : Ha_1=0 (and Ha_2=0)

Fully contaminated bubble



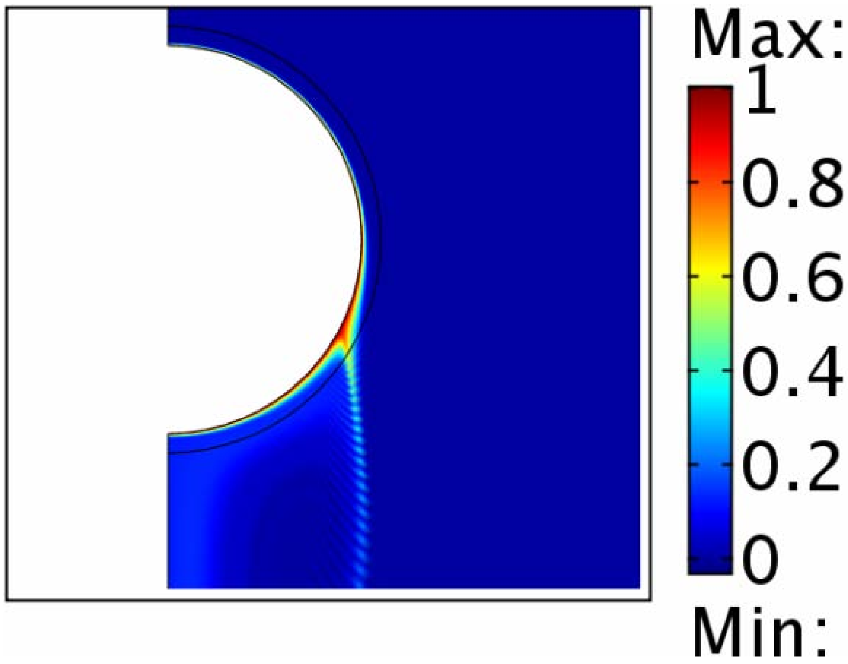
Clean bubble



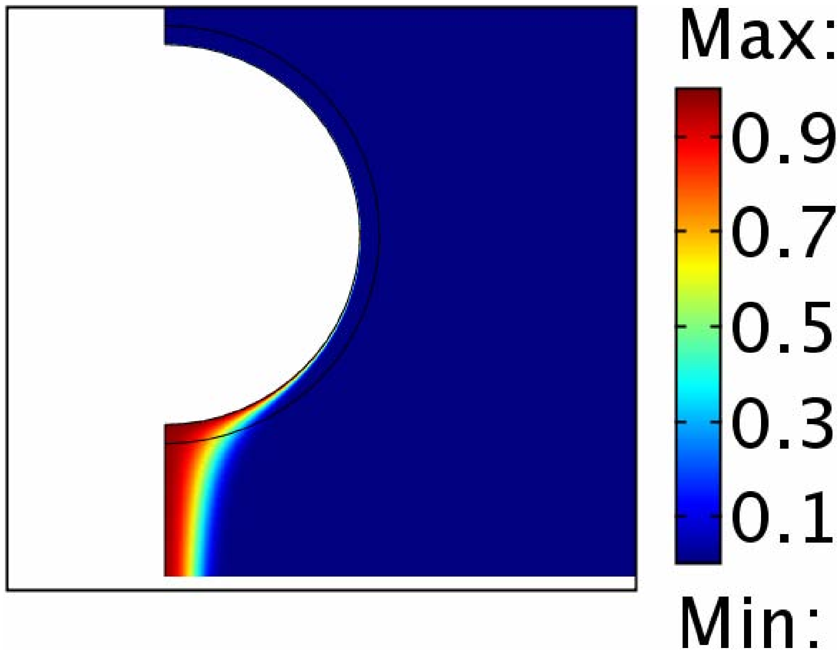
Simulation results

- **Simulations of the CO_2 concentration field**
  - Slow reaction 1 : Ha_1=0.1

Fully contaminated bubble



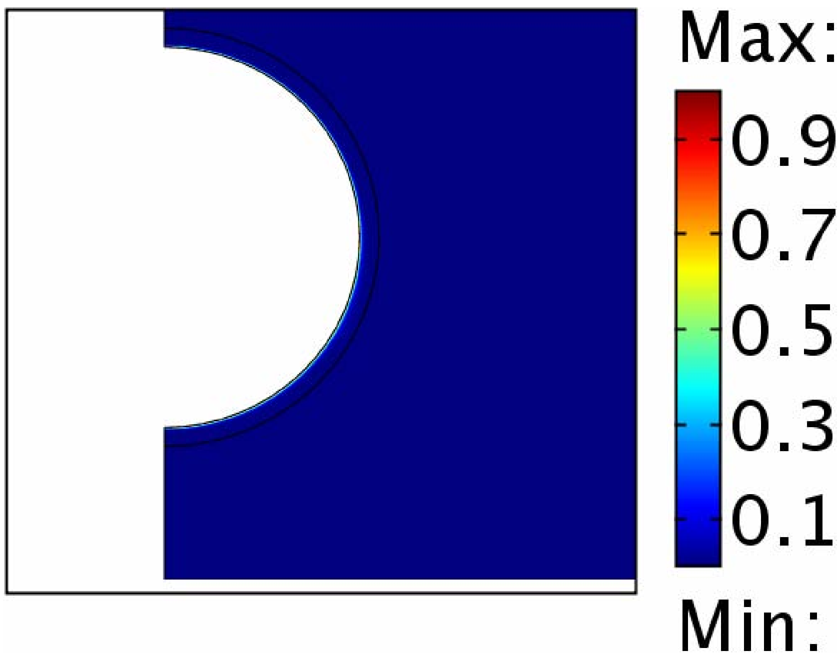
Clean bubble



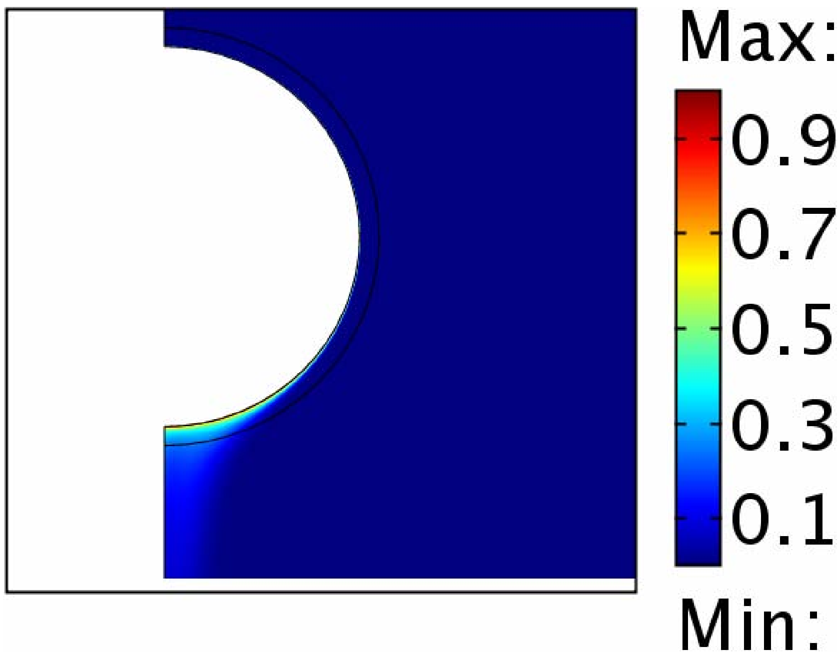
Simulation results

- **Simulations of the CO_2 concentration field**
  - Moderate reaction 1 : Ha_1=1

Fully contaminated bubble



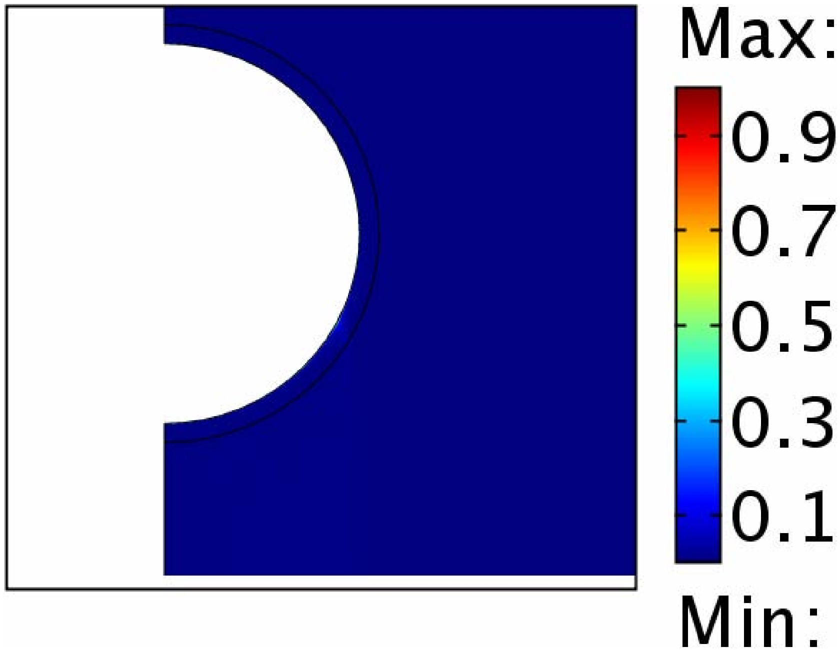
Clean bubble



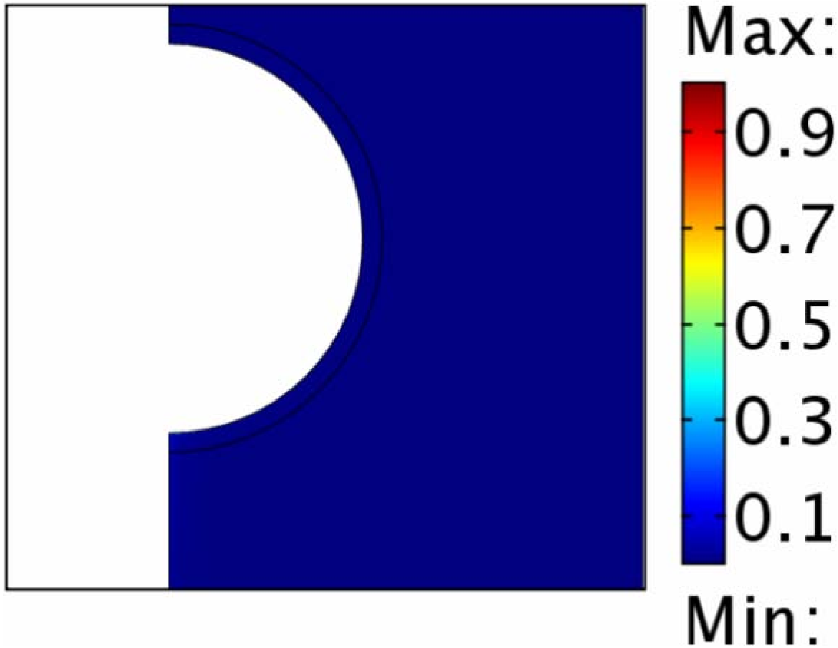
Simulation results

- **Simulations of the CO_2 concentration field**
  - Fast reaction 1 : Ha_1=10

Fully contaminated bubble



Clean bubble



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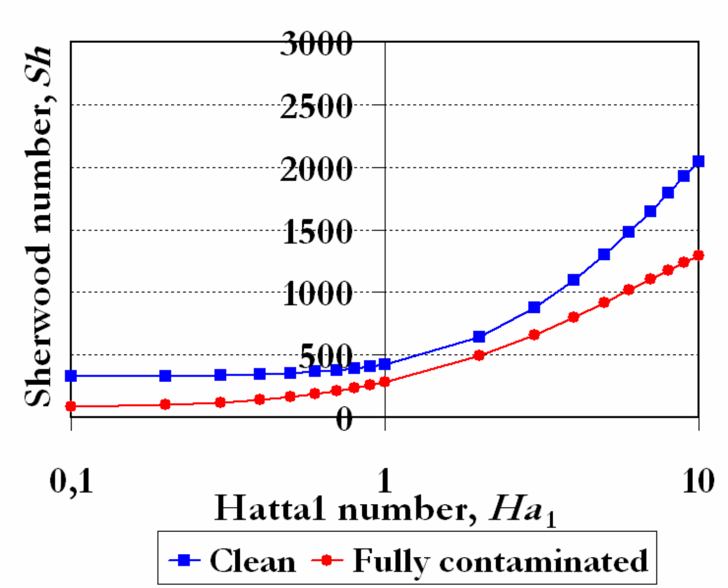
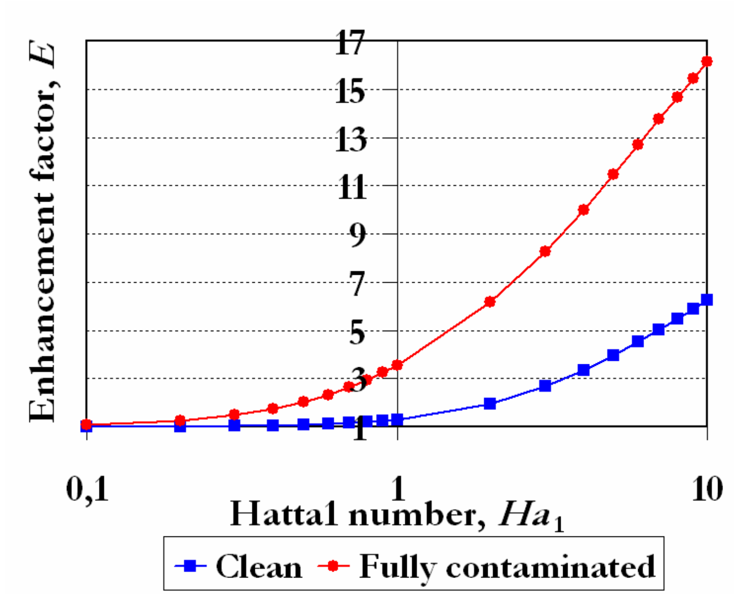
Simulation results

- **Simulations of the CO_2 concentration field**
  → Increasing CO_2 depletion for increasing reaction 1 rate
- **Calculation of the CO_2 transfer rate :**

Sherwood number



Enhancement factor



→ The CO_2 consumption enhances the CO_2 transfer rate

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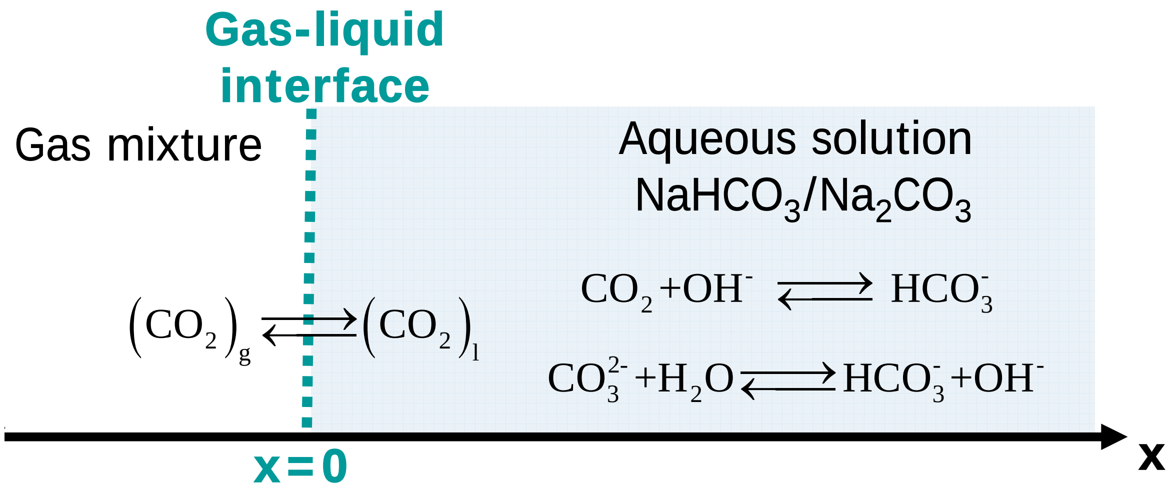
Simulation results

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 : mosaïc 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

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

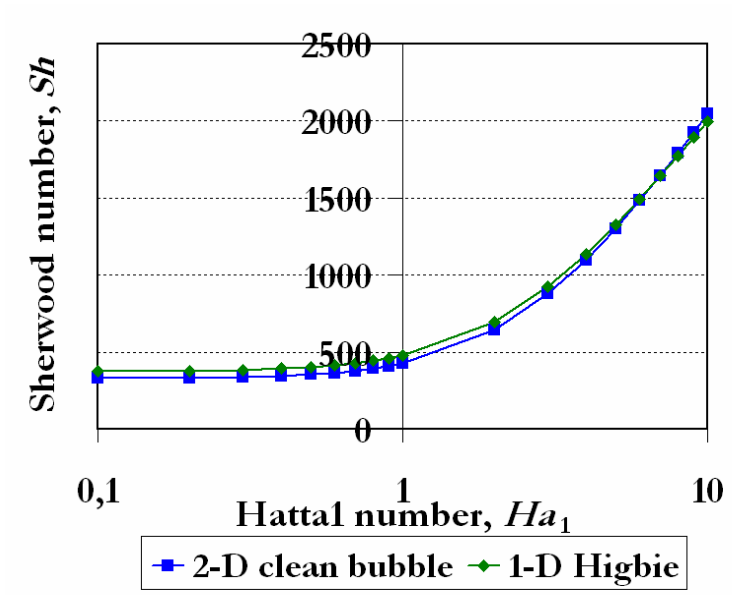


Axis pointed toward the liquid phase in normal direction of the interface

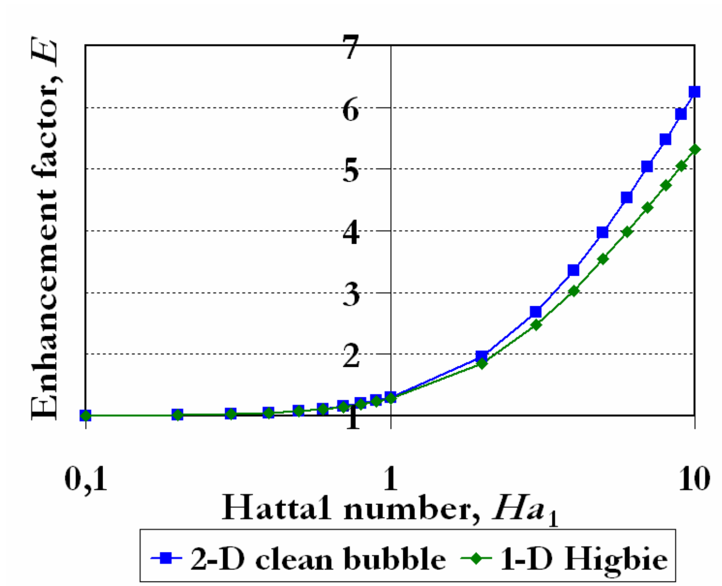
Simulation results

- **Comparison results**

Sherwood number



Enhancement factor



- The Higbie approach provides an excellent estimation
- Tend to slightly underestimate the chemical reactions effect when $Ha_1 > 1$

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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 → ellipsoidal-shape bubble
    - Shape coming from experimental observation
  - Comparison with spherical shape → quantification of the shape effect

Thanks for your attention