Impact of Variable Gas Mixtures on Bubble Size Distribution and Mass Transfer in Gas Fermentation Reactors

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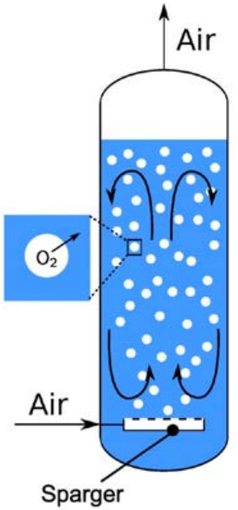
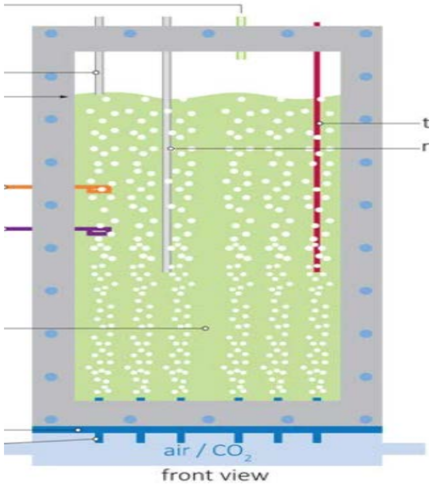
Background

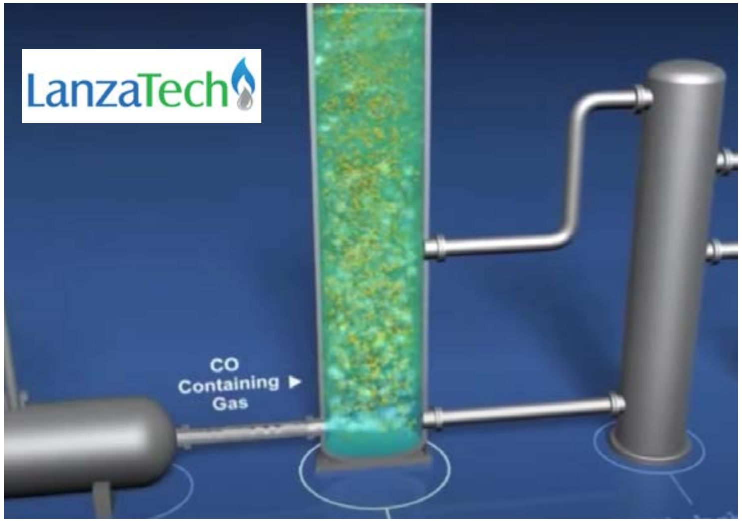
- Bioreactor: microbial action for conversion
  - Food/beverage/Pharmaceutical industry
  - **Biofuels/molecules**
    - **Ethanol/Butane-diol/Methane**
  - **CO_2 capture and conversion**
  - **Syngas fermentation**

  - Fermentation is a large cost contributor





Flat panel bioreactor*



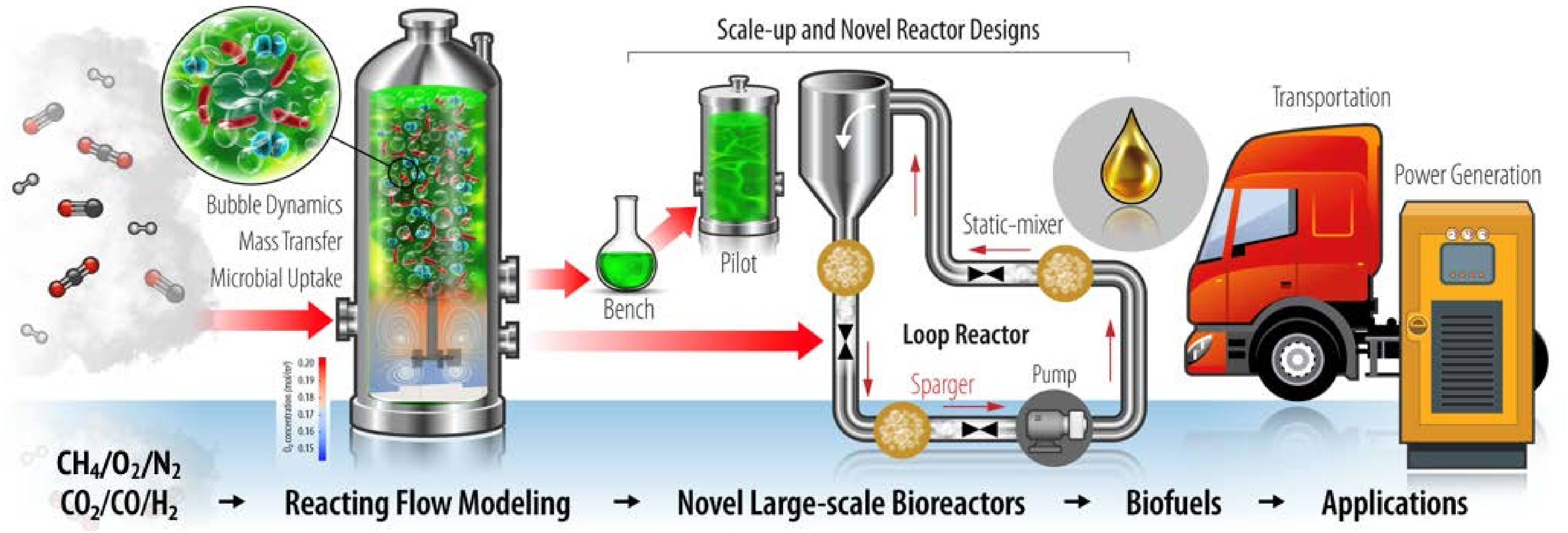
Syngas fermentation** (Lanzatech)

Biomethanation reactor (NREL)

**https://www.youtube.com/watch?v=k3WLwKrEu7c

*Singh and Sharma, Renew. Sust. Reviews, 2012

Project goals



- Enable scaled-up designs and optimization of $CO_2/CO/H_2$ fermenters
  - High fidelity computational models
    - **Focus on bubble dynamics and mass transfer**
    - **Impact of gas mixtures**
    - Coupling with microbial kinetics

- Supports Department of Energy goals on
  - reducing GHG emissions and sustainable aviation fuel production
  - Accelerate lab research to industrial scale

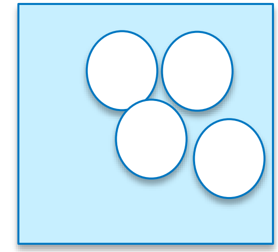
Outline

- Model equations
  - Multiphase-Euler model
  - Bubble size distribution modeling
  - Numerical methods
- Model validation
- Bubble column simulation results
  - Hydrodynamics
  - Mass transfer
  - Sensitivities
- Conclusions

Mathematical model and numerical methods

Multiphase Euler-Euler equations

- Gas and liquid as continuous interpenetrating phases
- Compressible low Mach RANS equations



$$\alpha_{\mathrm{L}} + \alpha_{\mathrm{G}} = 1$$

Volume fraction constraint

$$\frac{\partial}{\partial t}(\alpha_i \rho_i) + \vec{\nabla} \cdot (\alpha_i \rho_i \mathbf{V}_i) = 0$$

Mass conservation

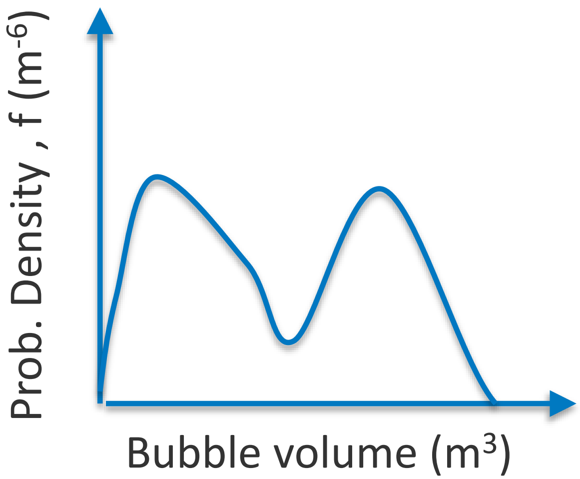
$$\frac{\partial}{\partial t}(\alpha_i \rho_i \mathbf{V}_i) + \vec{\nabla} \cdot (\alpha_i \rho_i \mathbf{V}_i \mathbf{V}_i)$$
$$= -\alpha_i \vec{\nabla} P + \alpha_i \rho_i \mathbf{g} + \vec{\nabla} \cdot (\alpha_i \bar{\mathbf{R}}_i) + \mathbf{F}_i$$

Momentum conservation

$$\frac{\partial}{\partial t}(\alpha_i \rho_i Y_{ij}) + \vec{\nabla} \cdot (\alpha_i \rho_i Y_{ij} \mathbf{V}_i)$$
$$= \vec{\nabla} \cdot (\alpha_i \rho_i \bar{D}_{ij} \vec{\nabla} Y_{ij}) + \dot{R}_{ij}^{\mathrm{MT}}$$

Species transport within each phase

Bubble size distribution* modeling



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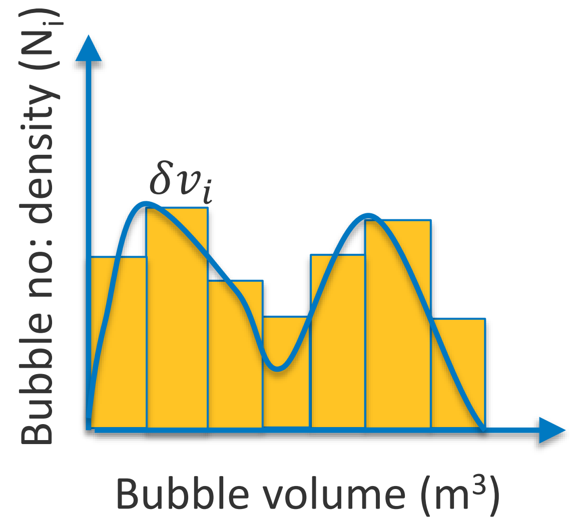


$$number\ of\ bubbles\ per\ unit\ volume\ N_i = f\ \delta v_i$$

$$phase\ fraction\ of\ each\ group\ f_i = \frac{N_i v_i}{\sum_j N_j v_j}$$

$$\frac{\partial n_v}{\partial t} + \nabla \cdot (v_{ph} n_v) = h_v$$

PDF transport equation

$$h_v = \underbrace{\frac{1}{2}\int_0^v n_{v'} n_{v-v'} C_{v',v-v'} dv'}_{\text{coalescence}(+)} - \underbrace{n_v \int_0^\infty n_{v'} C_{v,v'} dv'}_{\text{coalescence}(-)} +$$

$$\underbrace{\int_v^\infty n'_v B_{v'} \beta_{v,v'} dv'}_{\text{breakup}(+)} - \underbrace{n_v B_v}_{\text{breakup}(-)} - \underbrace{\frac{\partial(\dot{v} n_v)}{\partial v}}_{\text{drift}} + \underbrace{\dot{n_v}}_{\text{nucleation}}$$

Bubble dynamics source terms

*Lehnigk et al., AIChE Journal, 68,3, 2022

Drag and mass transfer model

$$F_D = \frac{3}{4}(C_D/d)\alpha\rho_l U_r^2 * sign(U_r)$$

Drag force

$$C_D = f(Re, Eo, \alpha_g)$$

Ishii Zuber drag model

Species mass transfer (Higbie et al. [1])

$$\mathrm{MTR} = k_{\mathrm{L}} a(C_j^* - C_j)$$

Oxygen transfer rate

$$C_j^* = \frac{X_{j,G} P}{H_i} \frac{\rho_L}{M_L}$$

Henry's law

$$k_{\mathrm{L}} = \sqrt{\frac{4D}{\pi} \frac{|\mathbf{u}_{\mathrm{slip}}|}{d_{\mathrm{b}}}} \quad a = \frac{6\alpha_{\mathrm{G}}}{d_{\mathrm{b}}}$$

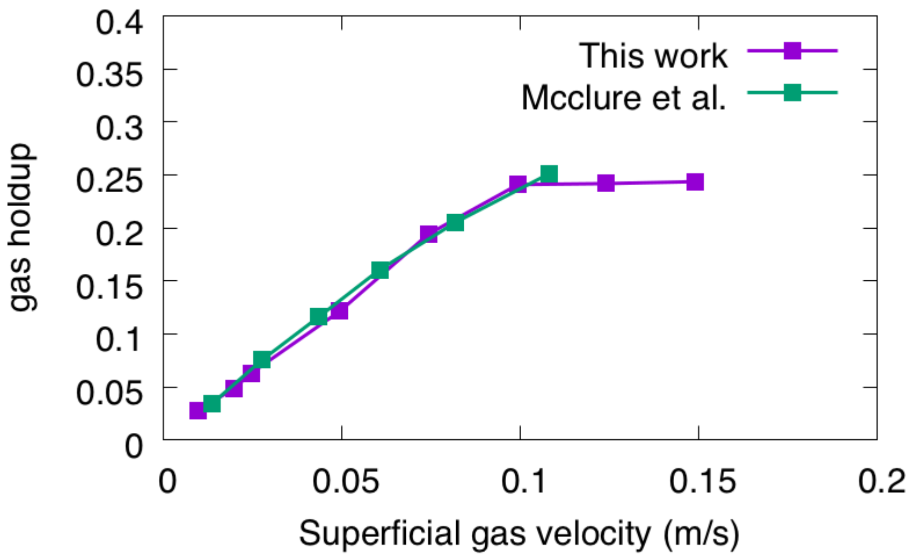
Mass transfer coefficient

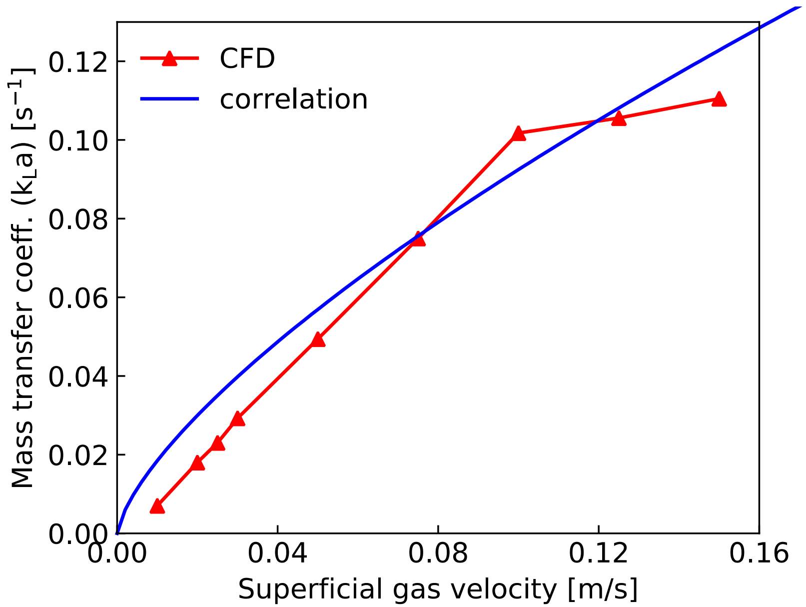
[1] Higbie, R., 1935. The rate of absorption of a pure gas into a still liquid during short periods of exposure. Trans. AIChE 31, 365–389.

Numerical methods and solver

- Transport properties
  - Fermentation broth properties are similar to water
  - Multiphase k-ω SST turbulence model
  - Population balance over 1-5 mm bubbles with 10 classes
- *multiphaseEulerFoam* in OpenFOAM
  - In-house implementation of Higbie mass transfer model
- Simulations performed using
  - 128 Intel Skylake processors
  - 48 hours of run time to simulate 30 seconds for 0.5 million cells
- More details in Rahimi et al., Chem. Engg. Res. Design, 139, 2018

Model validation with small-scale bubble column





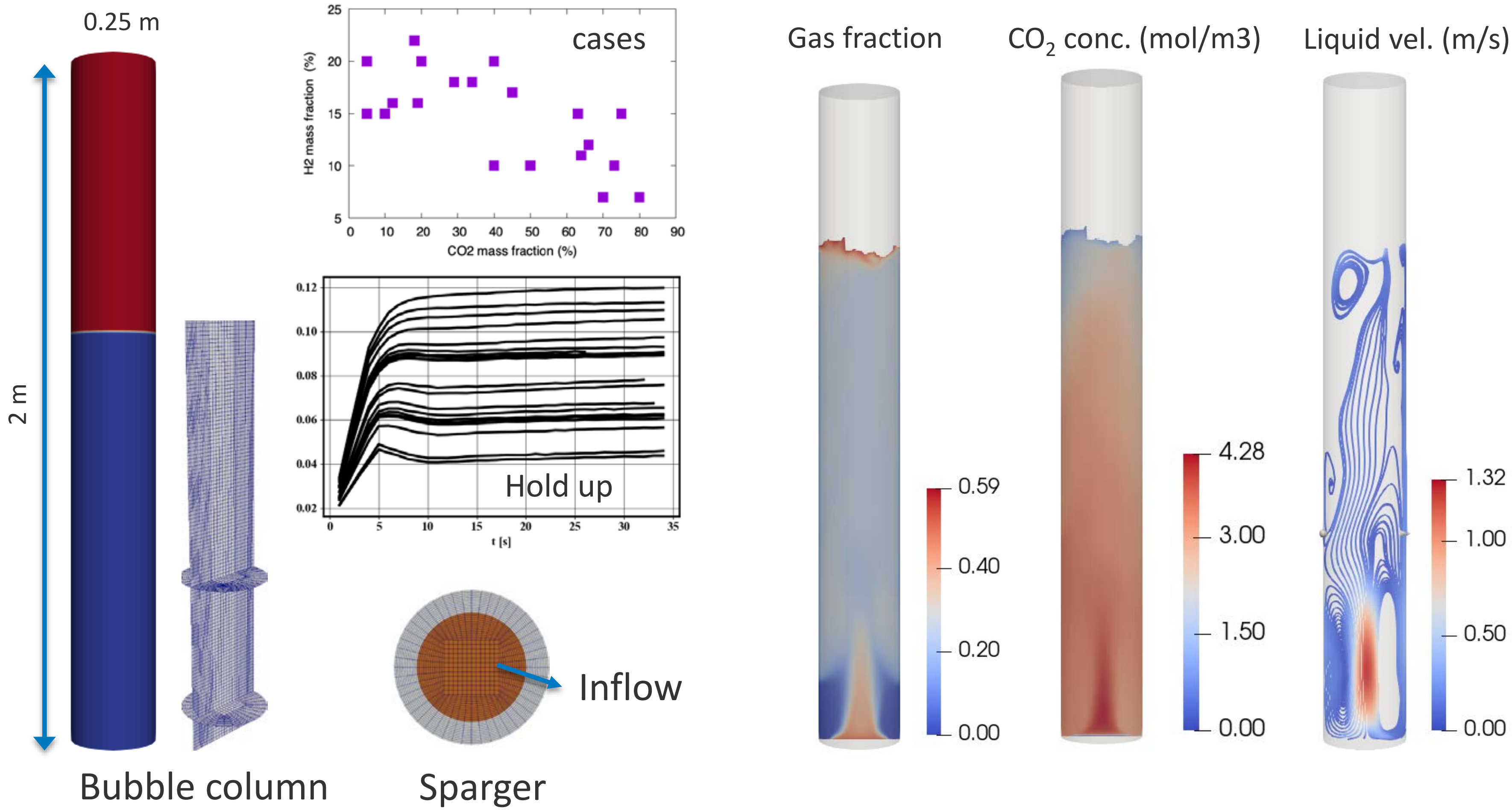
Superficial gas velocity: $vg_s = \frac{\dot{V}_{mid}}{A_{reactor}}$

- Validation done for a small-scale bubble column (1 m height, 15 cm diameter)
- Average mass transfer coefficient matches *Heijnen and Van't Riet (1984)*[1]
- Gas holdup matches experiments/simulations by Mcclure et al. (2013) [2]

[1] Heijnen, J. J., Van't Riet, K., Apr. 1984. Mass transfer, mixing and heat transfer phenomena in low viscosity bubble column reactors. Chem. Eng. J. 28 (2), B21–B42.
[2] McClure, D. D., Kavanagh, J. M., Fletcher, D. F., Barton, G. W., 2013. Development of a CFD model of bubble column bioreactors: Part one - a detailed experimental study. Chem. Eng. Technol. 36 (12), 2065–2070.

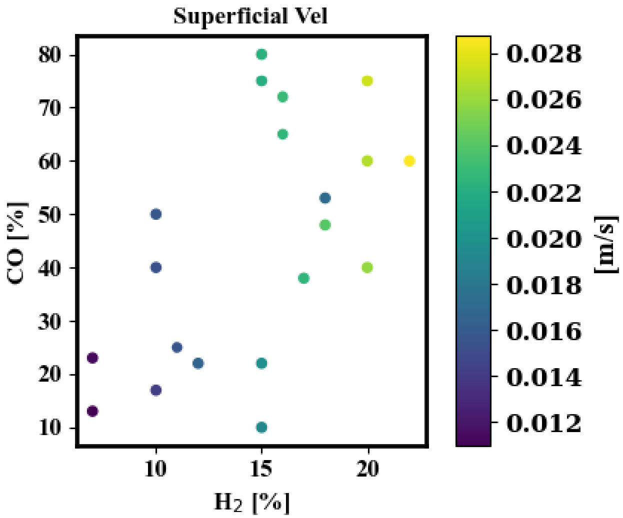
Results

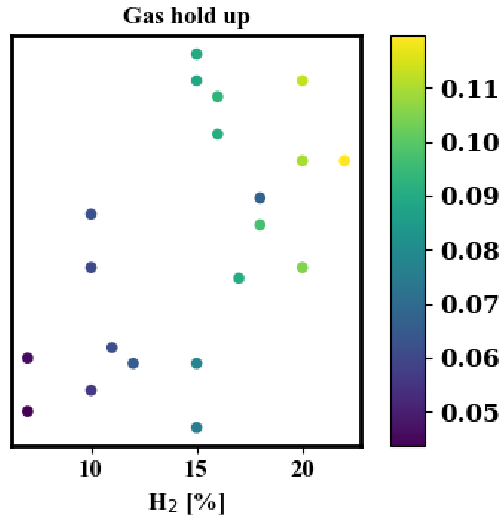
Bubble column simulations

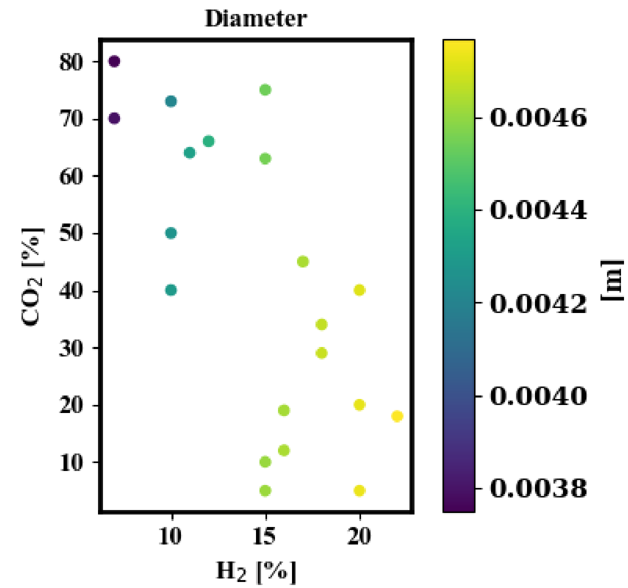


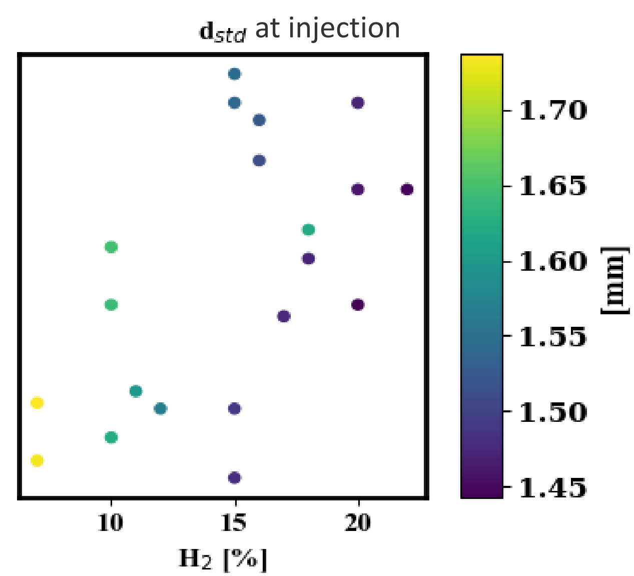
- Bottom inlet with a gas fraction that specifies sparger mass flow rate
- Lateral walls use no-slip condition for liquid and slip for gas
- Vary gas mixture mass fractions (H_2:CO_2:CO) while keeping constant mass flow rate of 0.45 g/s.

Hydrodynamic parameter variations







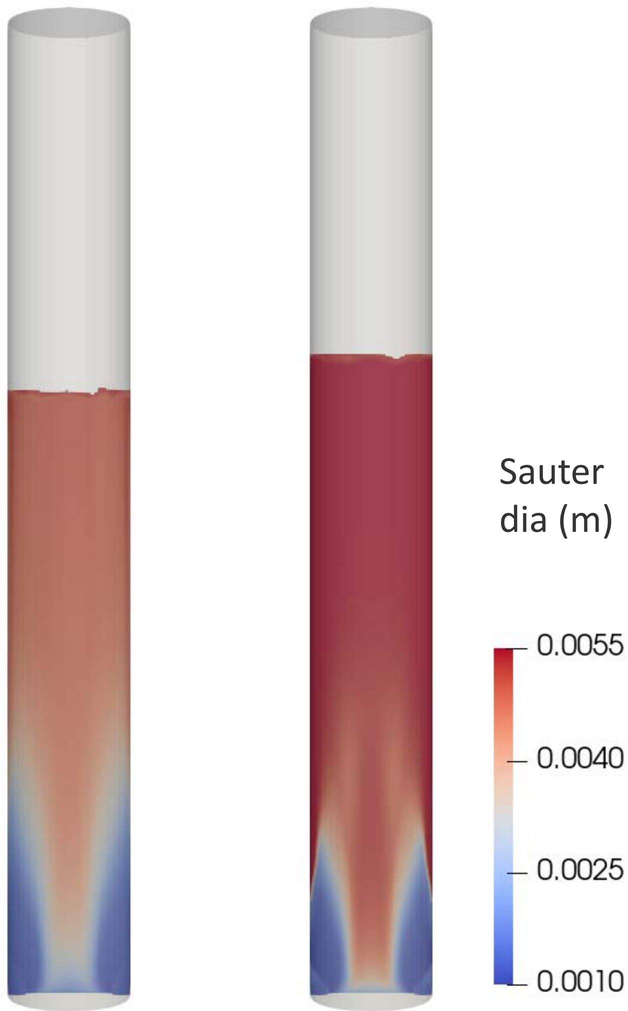


- Higher H_2 fractions result in higher:
  - superficial velocities
  - Gas hold up
  - Mean diameters
  - Greater spread of bubbles at the inlet

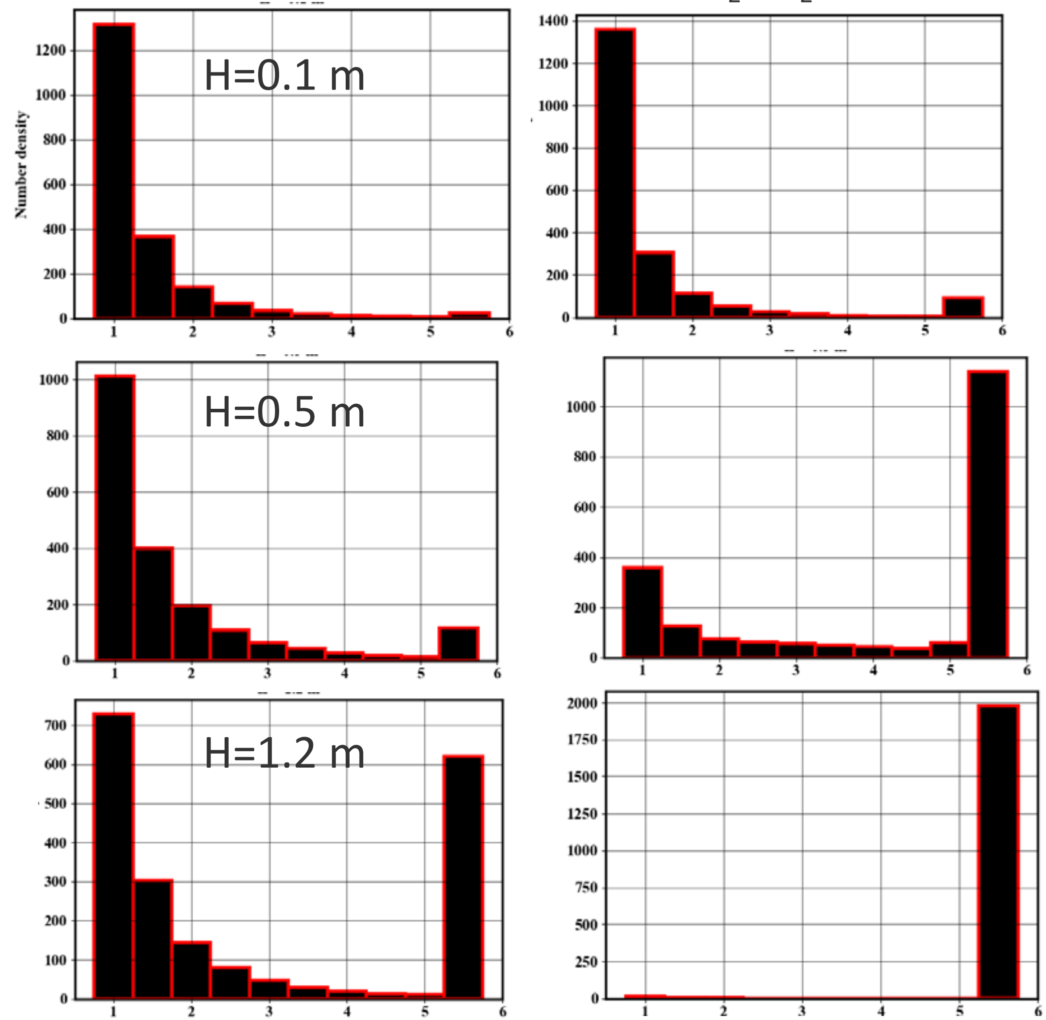
- Same mass flow results in greater volume of H2

Bubble size distribution variations

Case 1 (less H_2) Case 2 (more H_2)

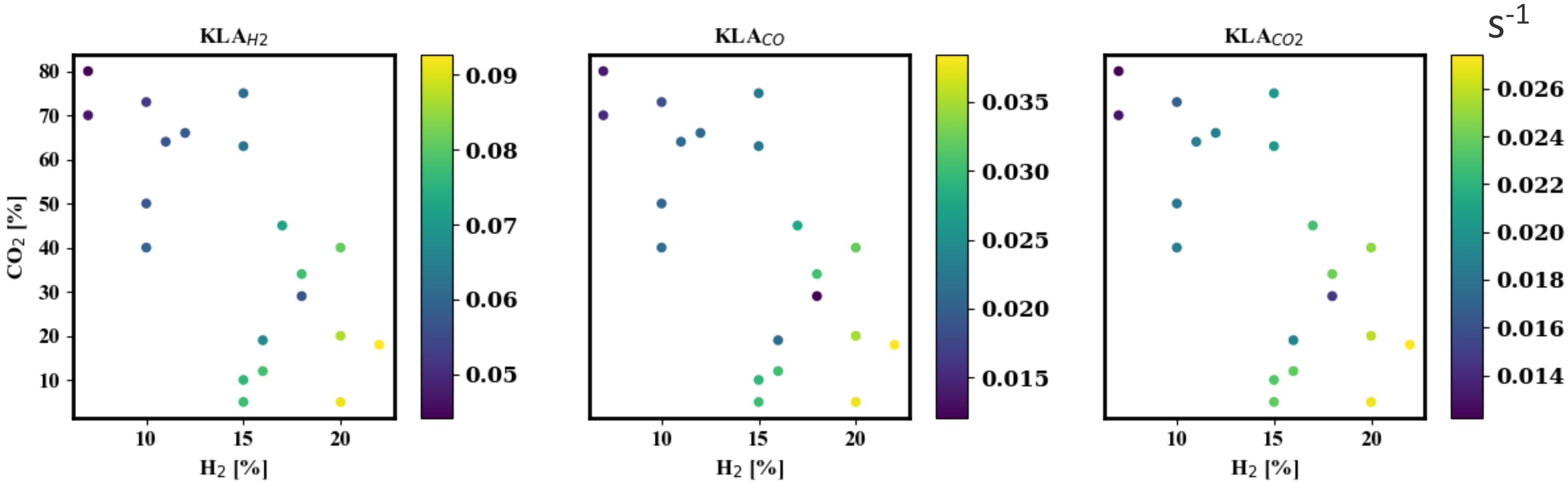


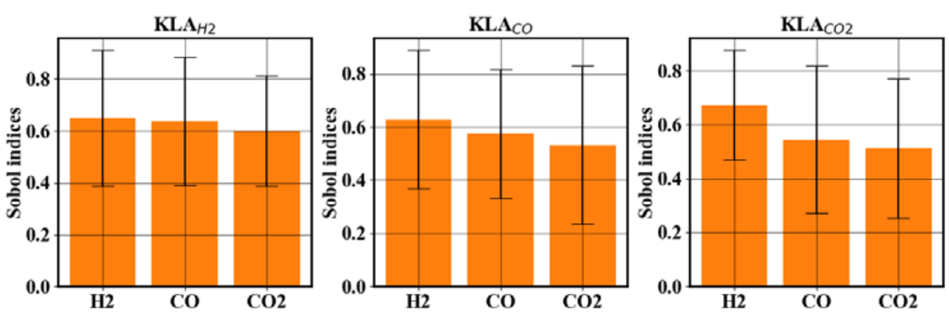
Case 1 - H_2:CO_2:CO=7:80:13 Case 2 - H_2:CO_2:CO=18:34:48



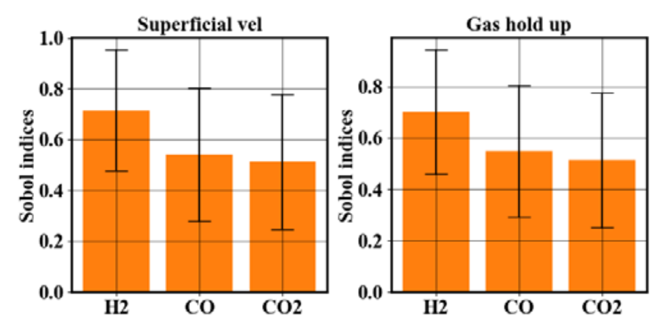
- Higher H_2 fraction at the inlet results in faster bubble coalescence and higher average Sauter diameter

Mass transfer effects





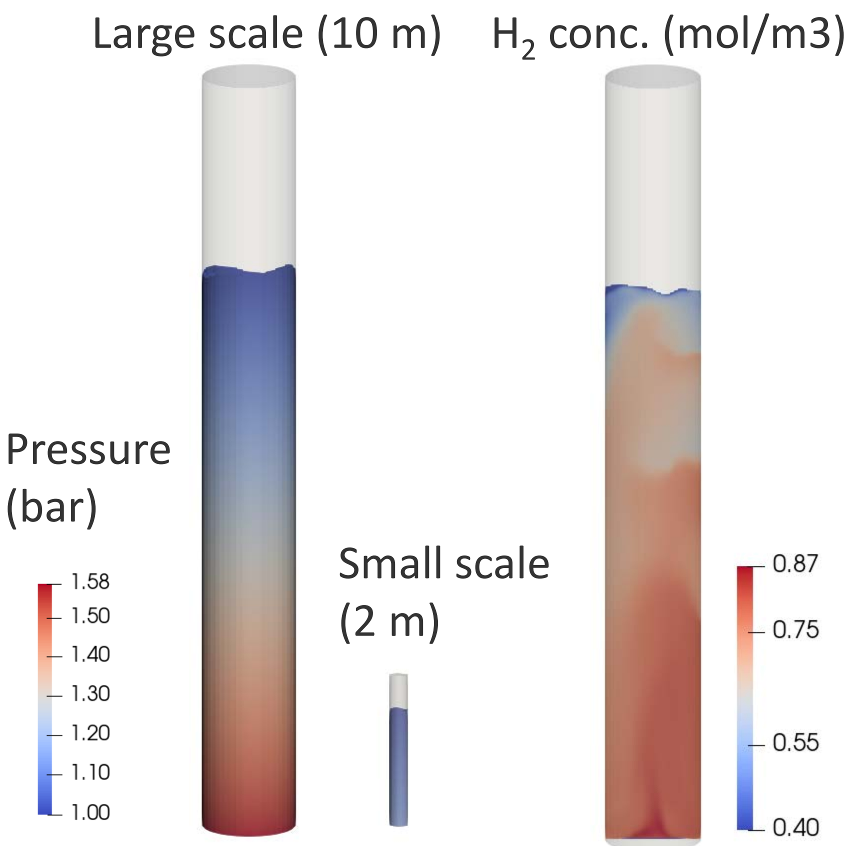
Sobol indices for mass transfer

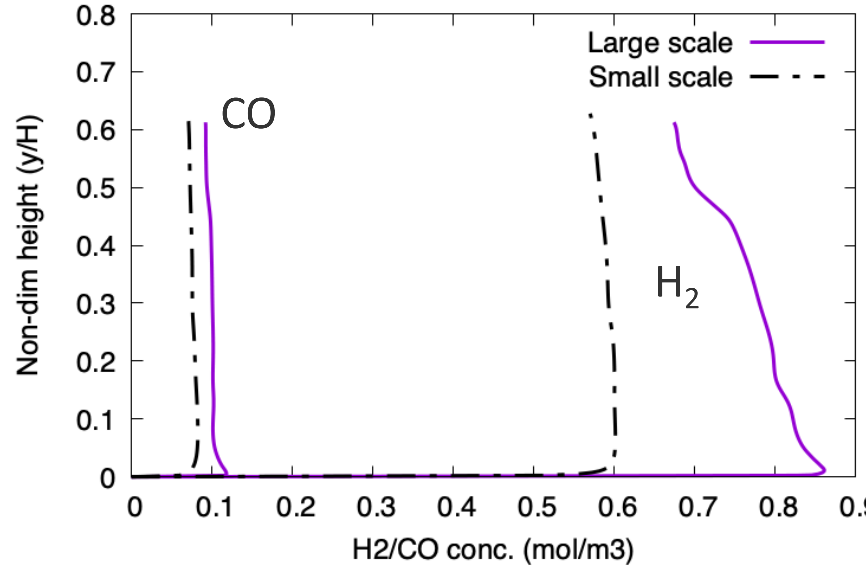


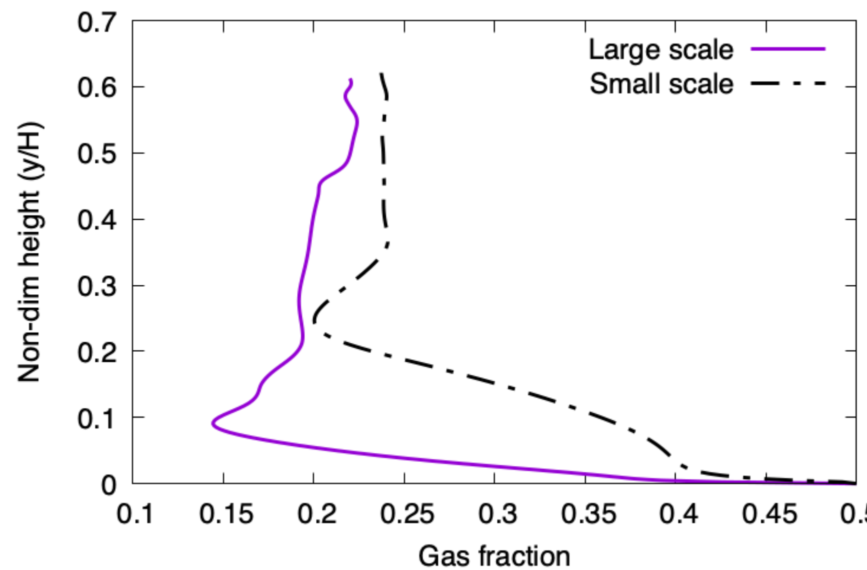
Sobol indices for hydrodynamics

- Higher H_2 results in greater mass transfer for all gases at constant mass flow rate
- Sobol indices – understanding the effect of varying one quantity with respect to total variance, indicates CO/CO_2 have lower impact than H_2

Effect of bubble column height







- Higher hydrostatic pressure head enables greater mass transfer
- Superficial velocity of 5 cm/s is kept the same between cases
- Spatial inhomogenities in species concentration and gas fraction

Conclusions and future work

- **Conclusions**
  - Computational model
    - OpenFOAM based multiphase solver
      - Gas mixtures, bubble size distributions
  - Results
    - Validated small scale bubble column
    - Ensemble simulations with H_2/CO/CO_2 mixtures
    - At constant mass flow rate
      - Greater H_2 fractions increase
        - Superficial velocities, gas holdup and mass transfer
        - Faster coalescence effects
    - Large scale vs small scale bubble column
      - inhomogenous mass transfer/gas fractions at large scale
- **Future work**
  - Microbial kinetics
  - Include product specie, CH_4, which can affect
    - Bubble size distribution and mass transfer
    - Light gas (H_2) gets consumed to heavier gas (CH_4)

Acknowledgements

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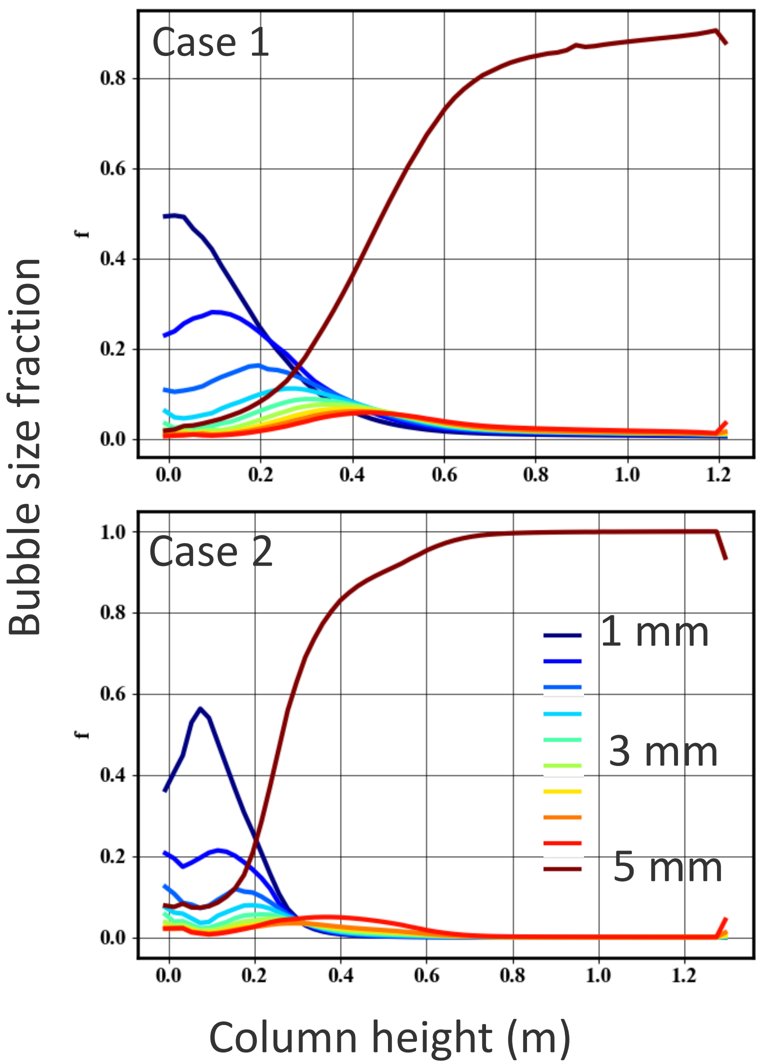
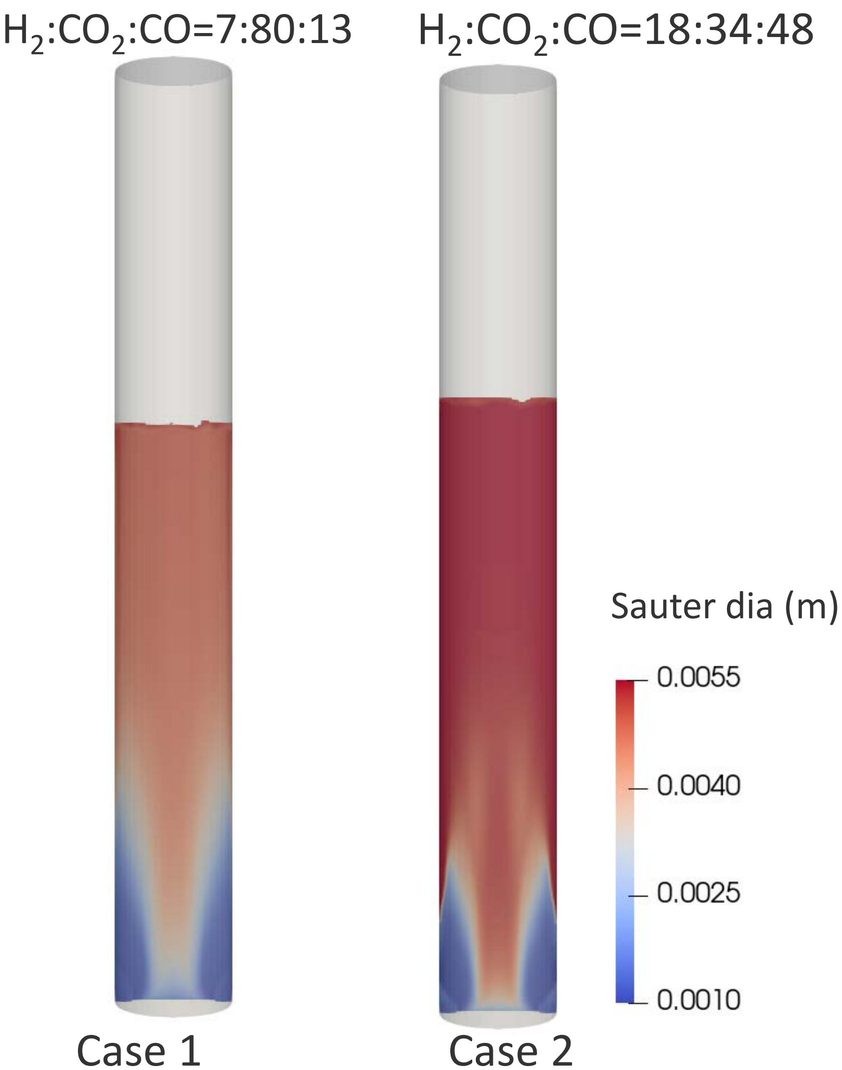
Thank You

Bubble size distribution variations





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