Gas-liquid flow modeling for renewable fuels production

Hari Sitaraman, Malik Hassanaly, Mohammad Rahimi, Yimin Lu, Milo-Parra Alvarez

Research Scientist, National Renewable Energy Laboratory

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Introduction

Renewable electricity generation pathways

Biomass is currently the only resource for carbon-based fuels and chemicals

Image by Dennis Schroeder, NREL

Conversion pathways

High temperature

Thermochemical conversion
- Pyrolysis
- Gasification

Lignocellulosic
Biomass
Eg: Corn stover,
agricultural
waste

products
- Syngas
- methane
- Bio-oil
- ethanol
- diesel

Low temperature

Biochemical conversion
- Enzymatic hydrolysis
- Microbial bioreaction

Unit operations

https://www.energy.gov/eere/bioenergy/sustainable-aviation-fuel-grand-challenge

Feedstock storage and handling

Pretreatment and conditioning

- Make cellulose more accessible

Enzymatic hydrolysis

- Digest cellulose to sugars

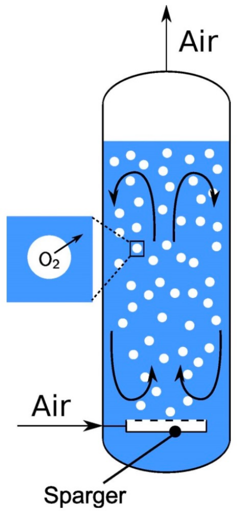
Bioreaction (fermentation)

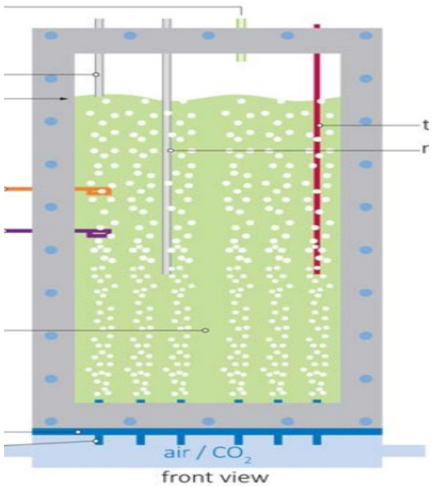
- Convert sugars to products

Catalytic upgrading

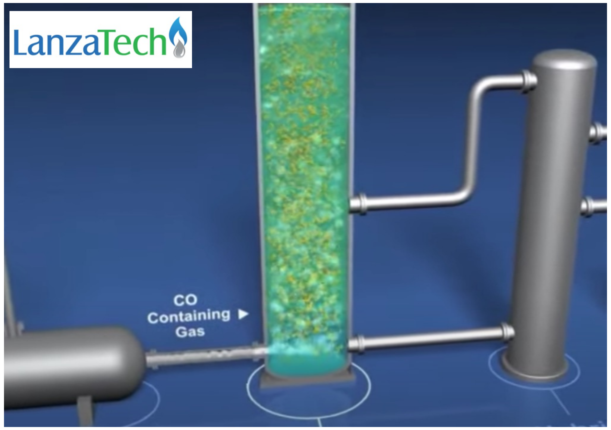
Background

- Bioreactor: microbial action for conversion
  - Food/beverage/Pharma industry
  - **Biofuels/molecules**
    - **Ethanol/Butane-diol/Methane**
    - **Sustainable Aviation Fuels (SAF)**
  - **CO_2 capture and conversion**
  - **Syngas fermentation**
- Fermentation is a large cost contribution
- Improved bioreactor design
  - More engineering than biology
  - Dominant physics: Gas-liquid flow





Flat panel bioreactor*



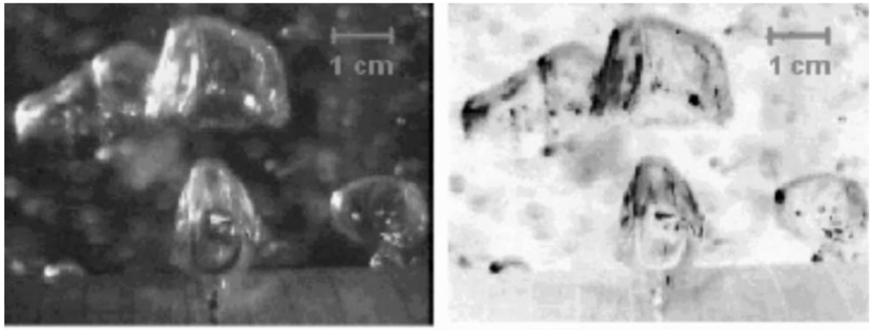
Syngas fermentation** (Lanzatech)

Biomethanation reactor (NREL)

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

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

Challenging multiphase flow questions



Pollia et al., Chemical Engineering Science 57 (2002) 197–205.

Bubble dynamics

How do gas mixtures Impact (e.g. $CO/CO_2/H_2$/air) bubble size distributions and mass transfer?

Microbial kinetics

How does component-wise microbial uptake influence species distribution and hydrodynamics?

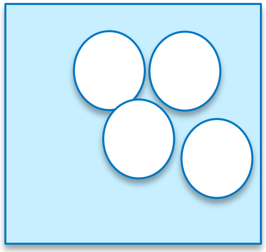
Novel reactor design

How do we design large-scale reactors (~ 500 m^3) with improved mixing and energy requirements?

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

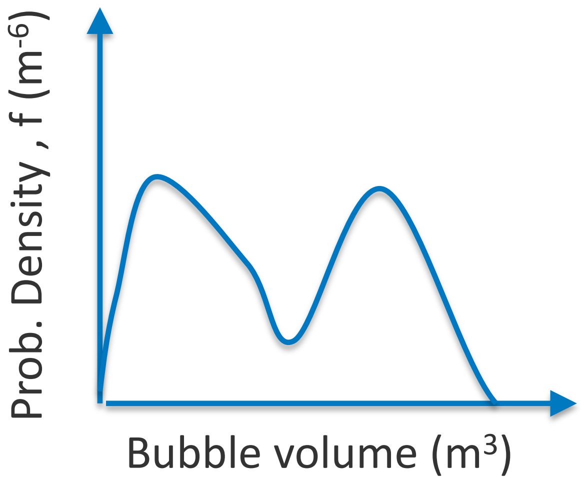
$$\frac{\partial}{\partial t}(\alpha_i \rho_i \mathbf{V}_i) + \vec{\nabla} \cdot (\alpha_i \rho_i \mathbf{V}_i \mathbf{V}_i) = -\alpha_i \vec{\nabla} P + \alpha_i \rho_i \mathbf{g} + \vec{\nabla} \cdot (\alpha_i \bar{\mathbf{R}}_i) + \mathbf{F}_i$$

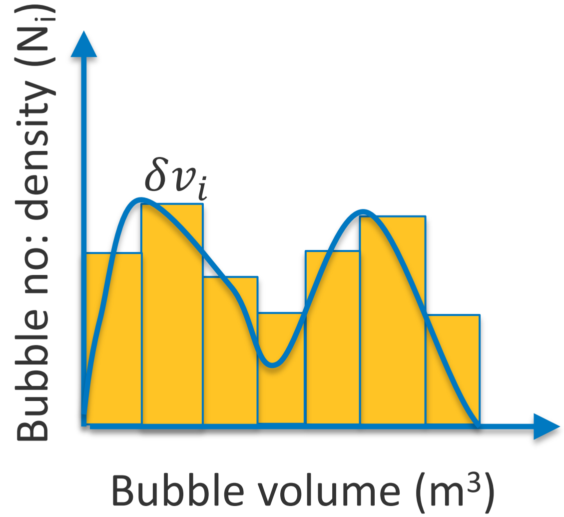
Momentum conservation

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

Species transport within each phase

Bubble size distribution* modeling





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

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

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

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

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

Grace drag model

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

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

species transfer rate

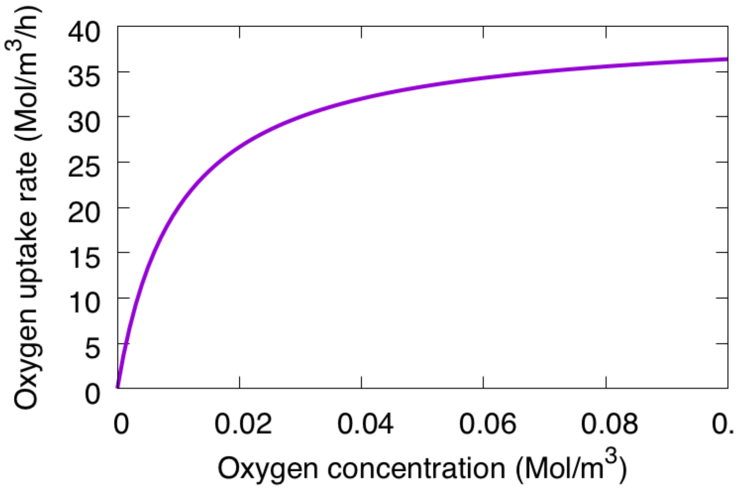
$$C_j^* = \frac{X_{j,G} P}{H_i} \frac{\rho_L}{M_L}$$

Henry's law

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

Microbial uptake (Monod model)

$$\mathrm{OUR} = \mathrm{OUR}_\mathrm{max} \frac{C_{\mathrm{O}_2}}{K_O + C_{\mathrm{O}_2}} \alpha_\mathrm{L}$$



[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

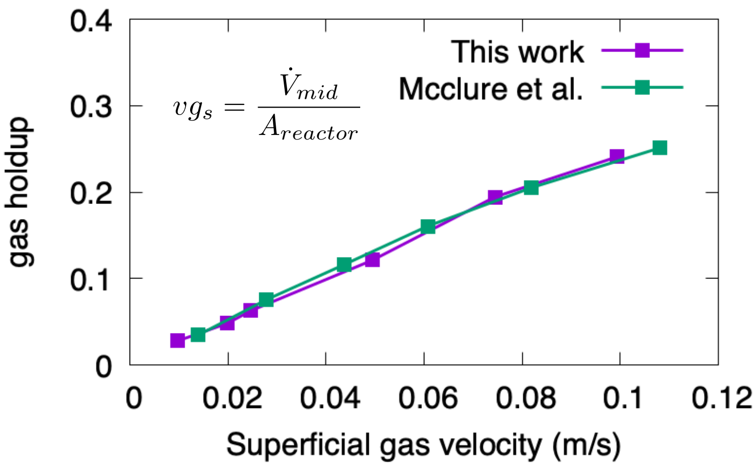
- Transport properties
  - Fermentation broth properties are similar to water
  - Multiphase k-ω SST turbulence model
  - Population balance over 1-5 mm bubbles with 10 classes

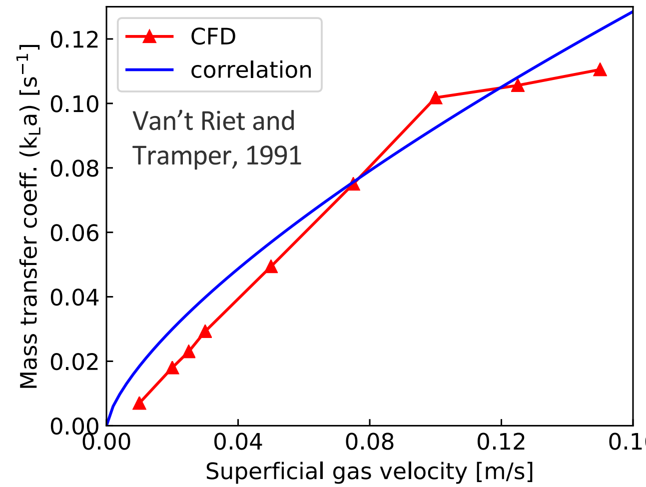
- *multiphaseEulerFoam* in OpenFOAM
  - In-house implementation for
    - Higbie mass transfer model
    - Grace drag model

- Typically simulations performed using
  - 128 Intel Skylake processors
  - 48 hours of run time to simulate 30 seconds for 0.5 million cells

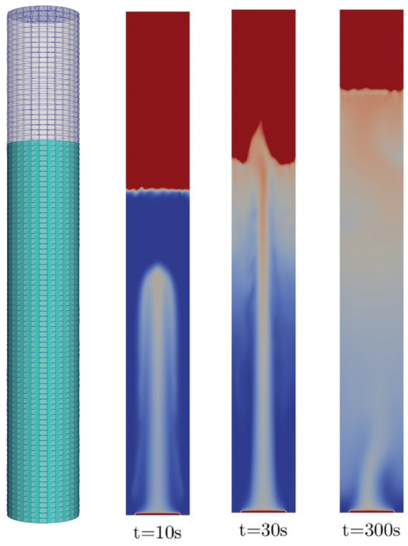
- More details in
  - Rahimi et al., Chem. Engg. Res. Design, 139, 2018
  - Sitaraman et al., Chem. Engg. Res. Design, 197, 2023

Model validation with small-scale bubble column

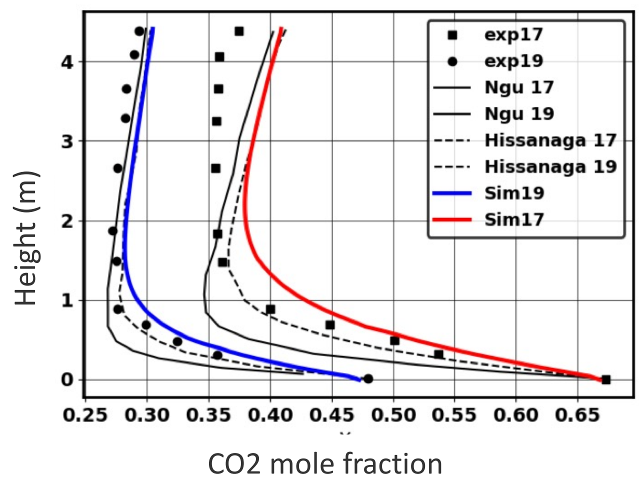




Air validation*



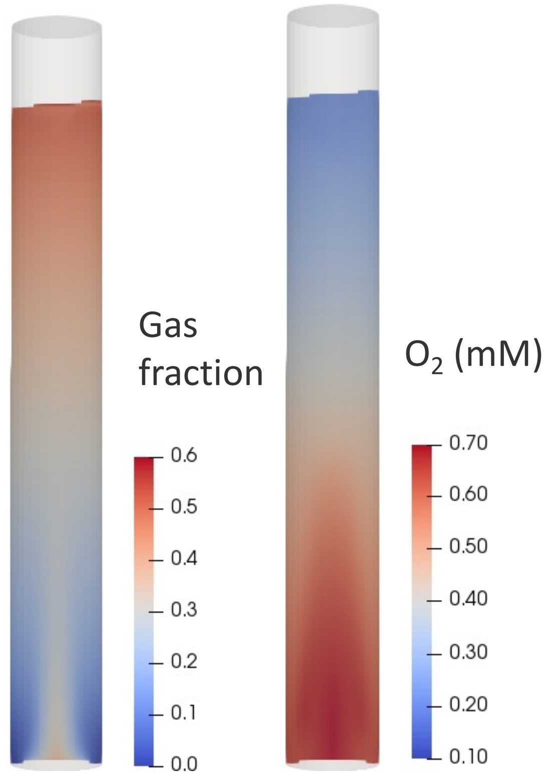
- Air validation: 1 m height, 15 cm diameter bubble column
- CO2 validation: 4 m height, 15 cm dia, co-flow column, with CO_2/N_2 mixture



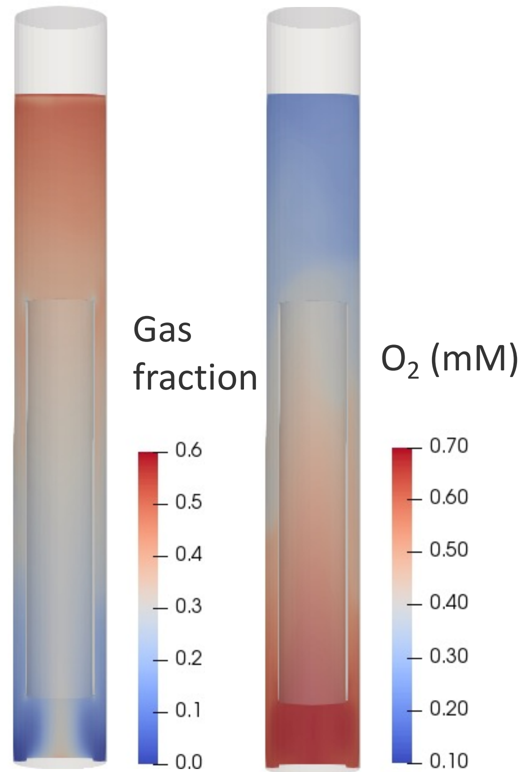
CO_2 validation**

*McClure et al., Chem. Eng. Technol. 36 (12), 2065–2070.
**Comparison with Hissanaga et al., Chem. Engg. Sci, 2020 and Ngu et al., Chem. Engg. Sci., 2022

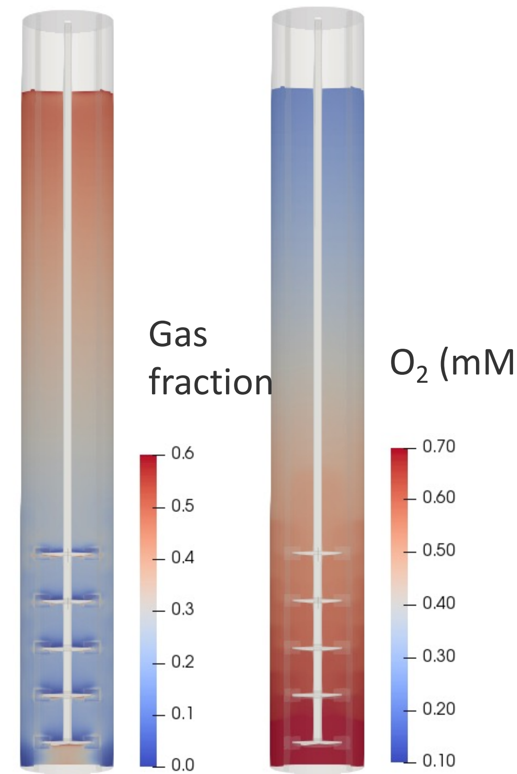
Bioreactors at scale



Bubble column reactor

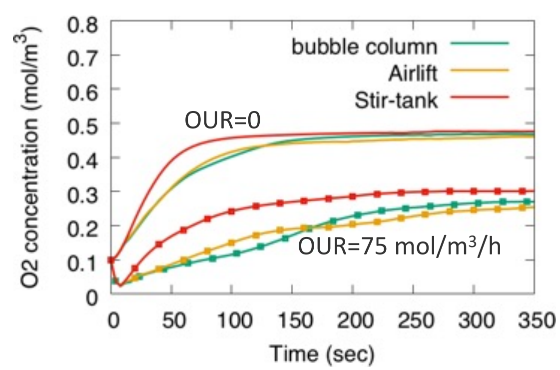


Airlift reactor



Stir-tank

- Explored large reactors – 40 m high bubble column, airlift and stir tanks with
- Bottom boundary – air sparger
- Draft tube and impellers aid better mixing



Two case studies

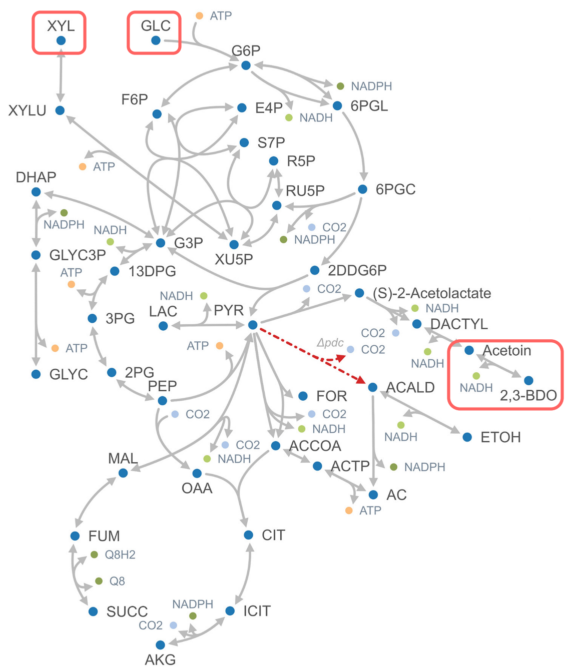
- 2,3 Butanediol production from sugars
- CO_2/syngas conversion to fuels

Butanediol (BDO) synthesis from sugars

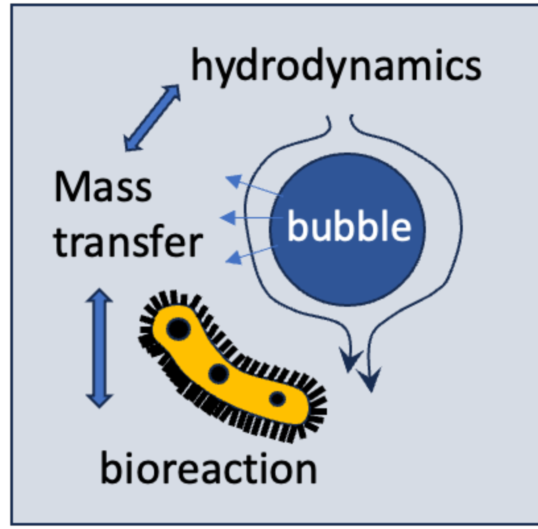
- BDO is a SAF intermediate
- Process scale-up for meeting SAF goals

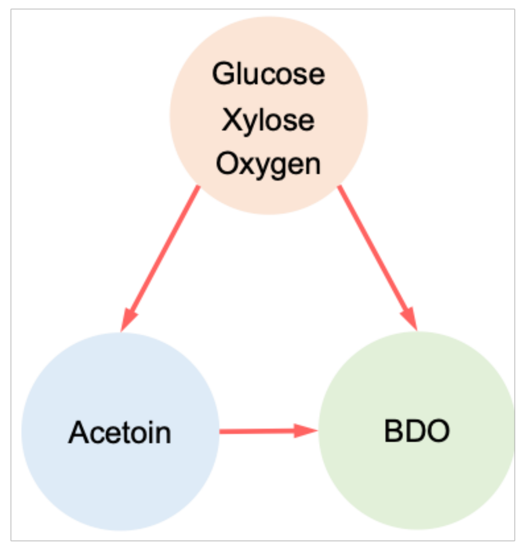
- Needs controlled amount of oxygen
  - High O2 -> side products

- Coupling with microbial bioreactions and hydrodynamics

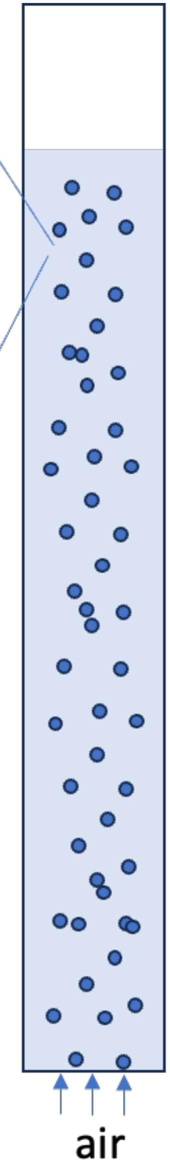


Complex metabolic pathways

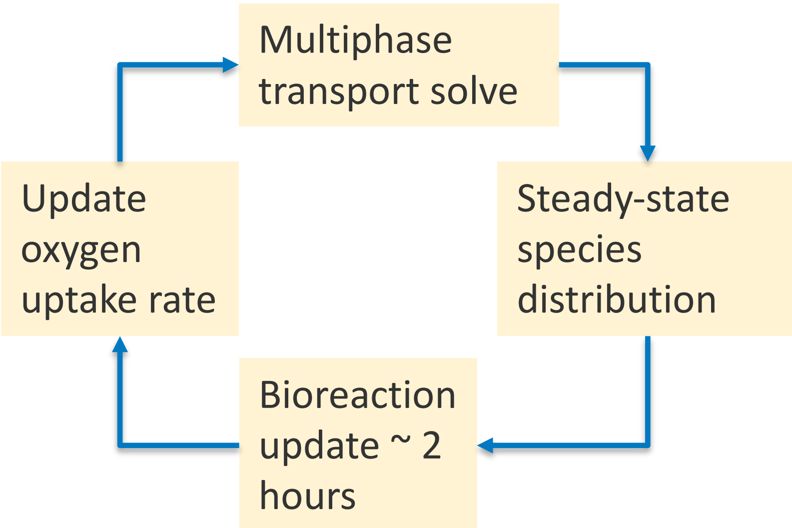




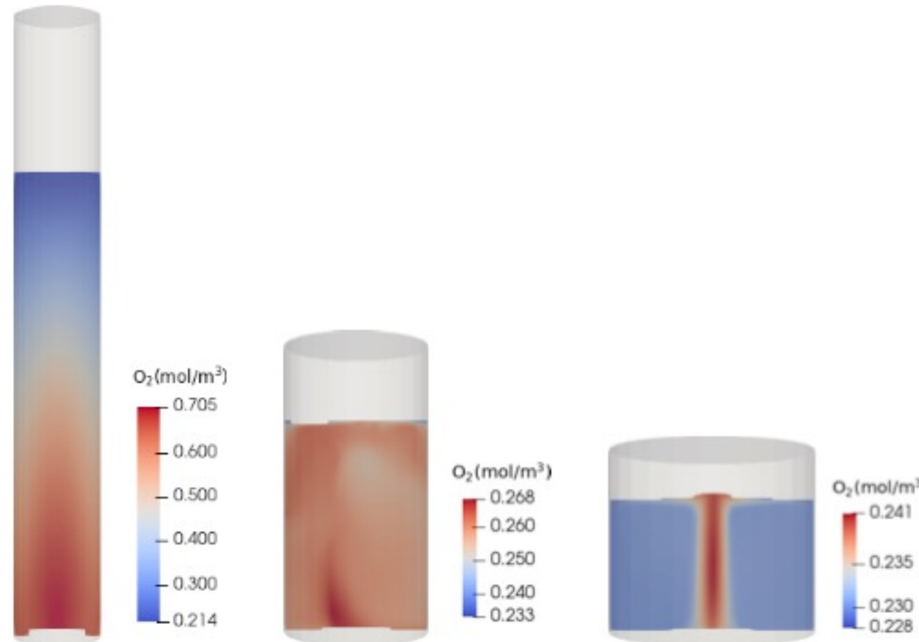
Simplified reaction model



Butanediol (BDO) synthesis from sugars

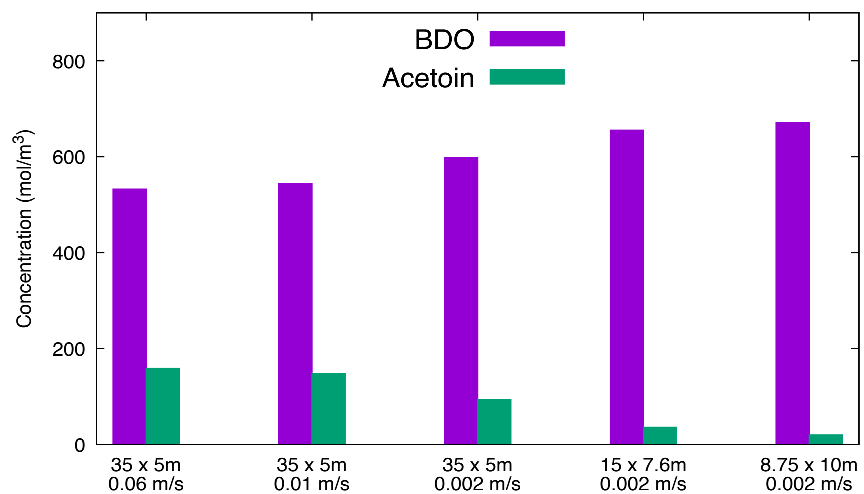


Time step subcycling scheme



- Flow-reaction coupling
  - Disparate time scales: reactions ~ hours, fluid-dynamics ~ 100 sec
  - Time step subcycling

- Lower aeration rates and shorter bubble columns favor higher BDO production
  - Taller columns (case 1) leads to higher O2 concentration at the bottom



Two case studies

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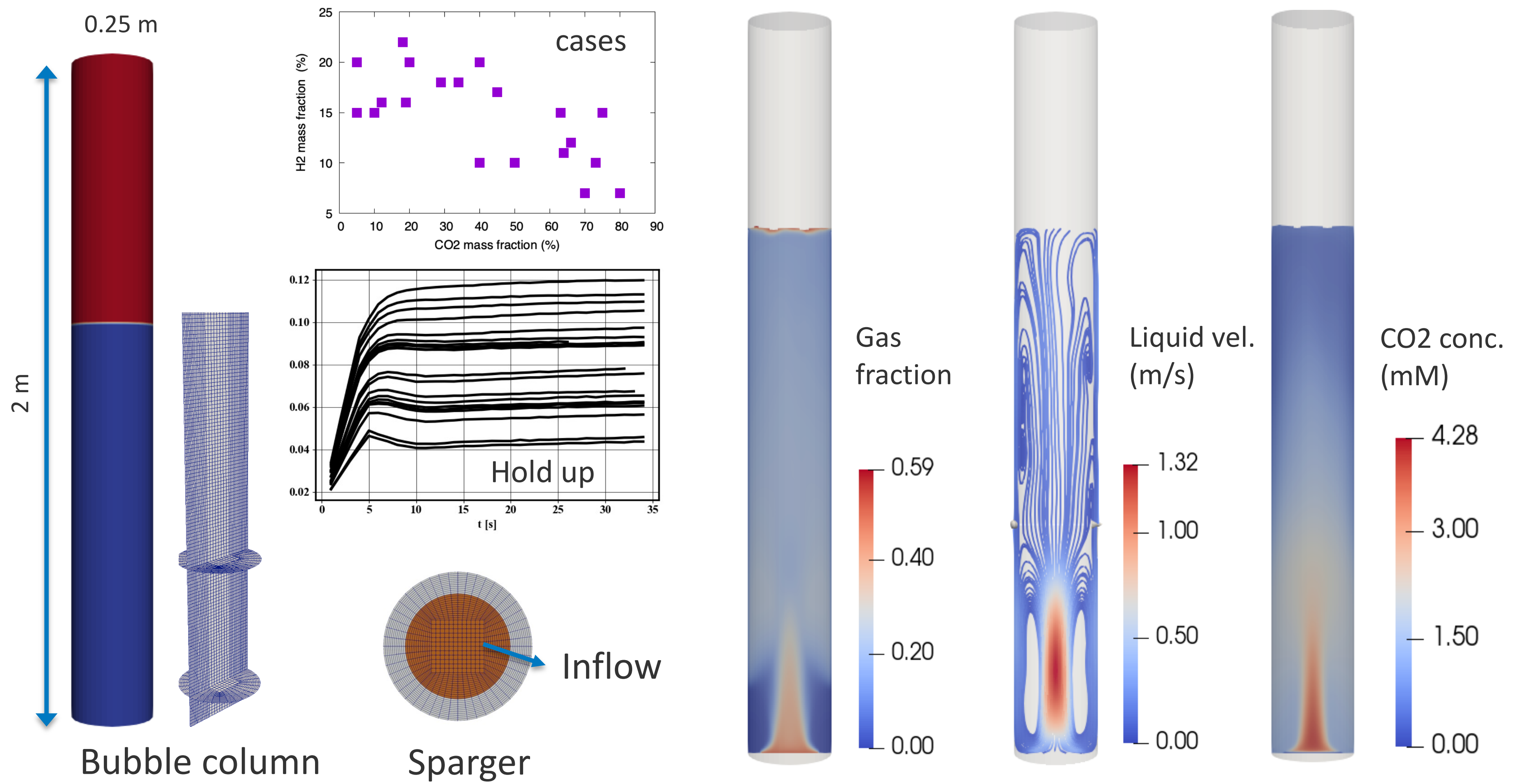
- CO_2/syngas conversion to fuels

Bubble column simulations

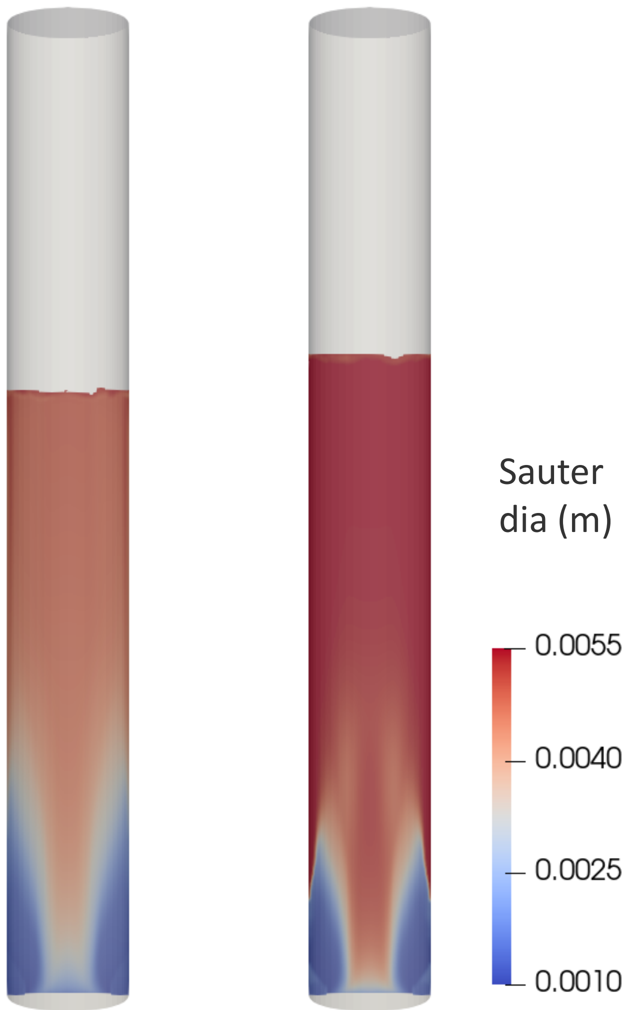


- Bottom inlet with a gas fraction that specifies sparger mass flow rate
- Lateral walls use no-slip condition for liquid and slip for gas
- Vary gas mixture mass fractions (H_2:CO_2:CO) while keeping constant mass flow rate of 0.45 g/s.

Bubble size distribution variations

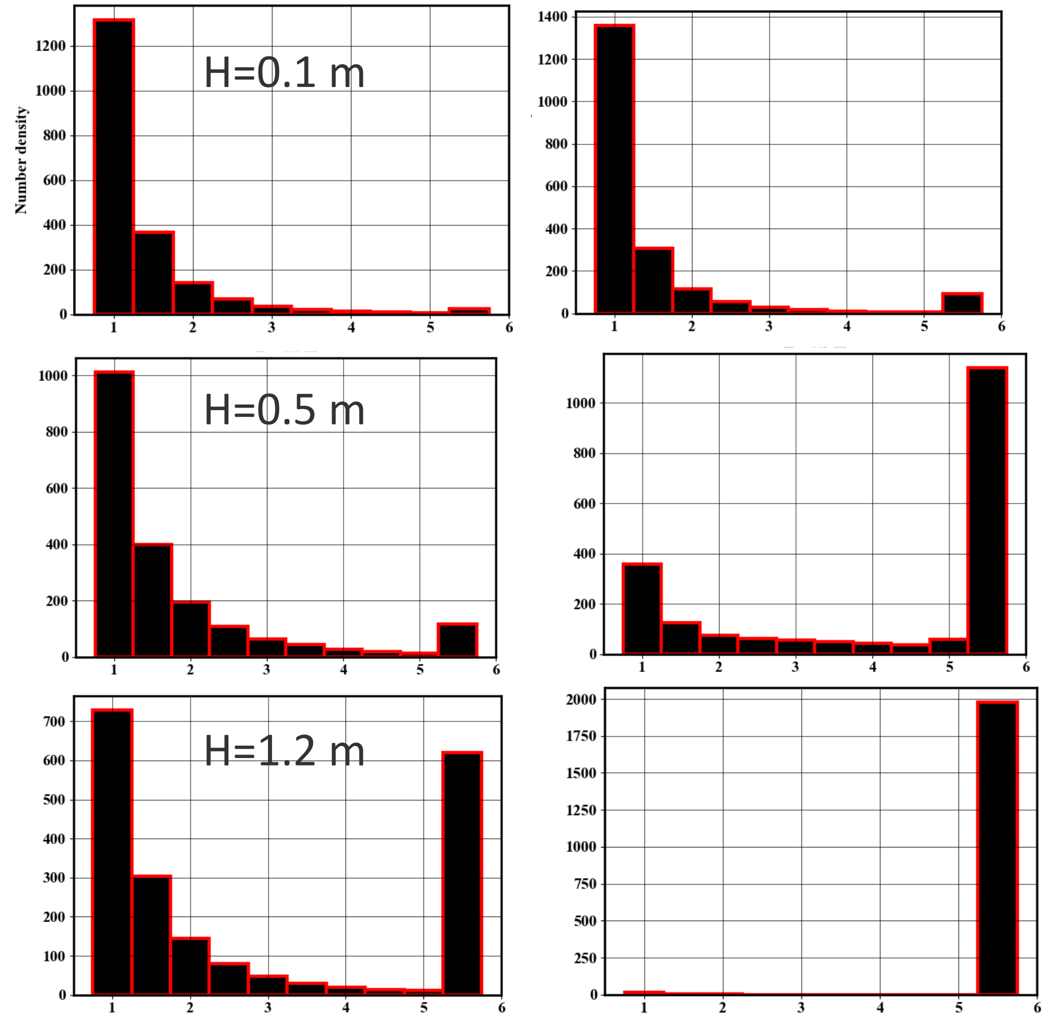
Case 1 (less H_2)

Case 2 (more H_2)



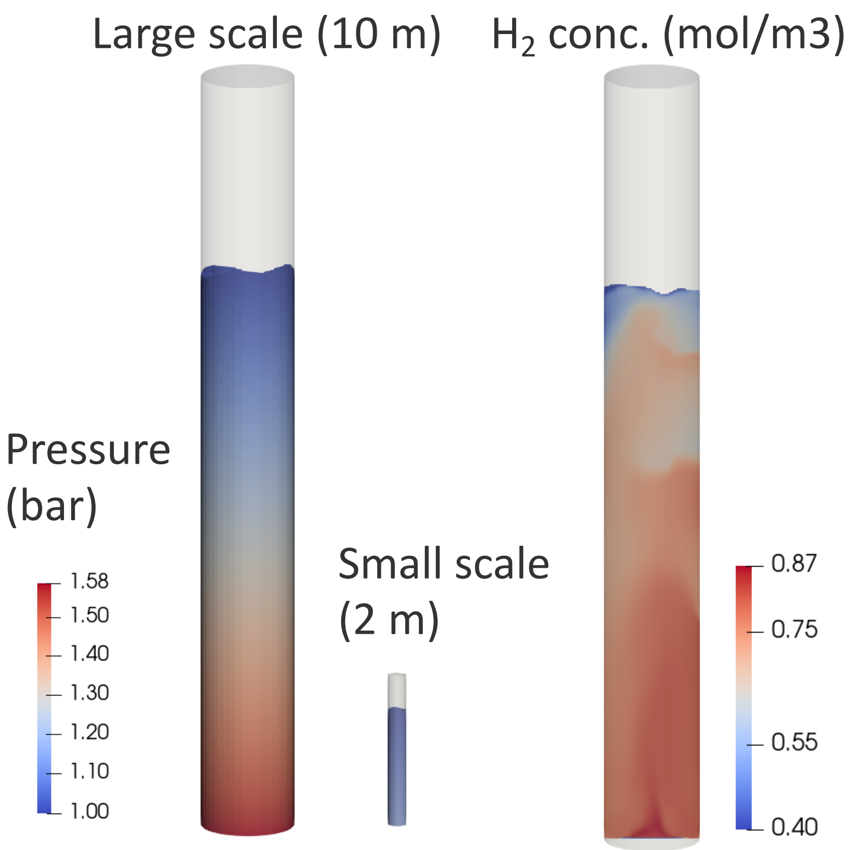
Case 1 - H_2:CO_2:CO=7:80:13

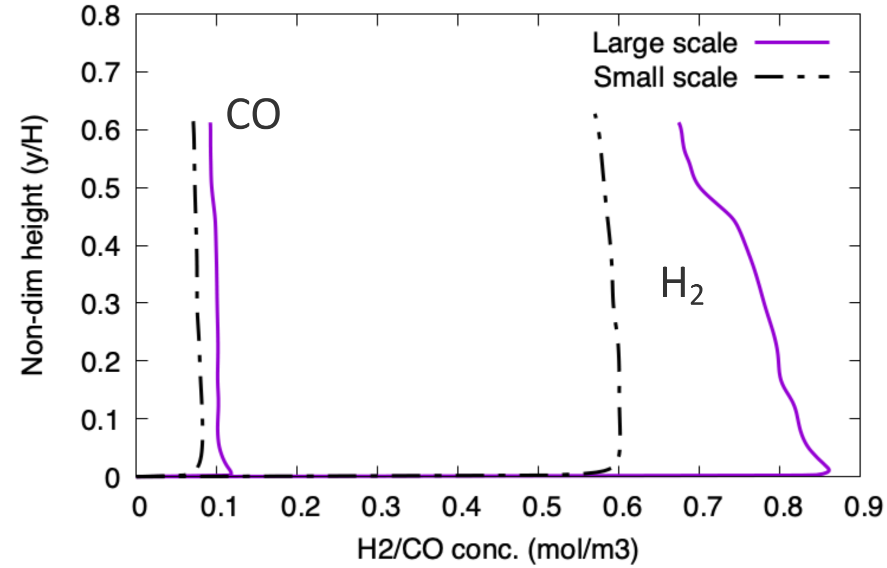
Case 2 - H_2:CO_2:CO=18:34:48

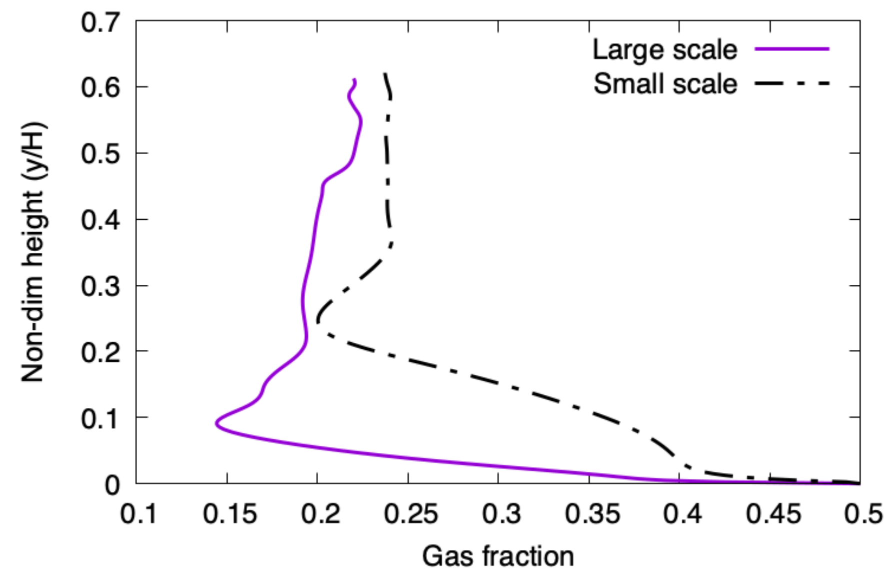


- Higher H_2 fraction at the inlet results in faster bubble coalescence and higher average Sauter diameter

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 with microbial uptake
  - Results
    - Validated small scale bubble column
    - Comparison between bubble column, airlift and stir-tank reactors
      - Better mixing in stir tanks
      - Greater pressure heads leads to greater O_2 transfer
    - Butanediol reactors
      - Multiphase coupling to microbial bioreactions
      - Lower aspect ratio reactors are favorable at scale
    - CO2/syngas reactors
      - H2 has a strong impact on multiphase dynamics

- **Future work**
  - Higher fidelity LES
  - Bubble size distribution models for mixtures
  - Novel reactor designs: e.g. looping reactors

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BIOENERGY TECHNOLOGIES OFFICE

NREL
Transforming ENERGY

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- Computational resources at NREL

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Thank you

Literature

- **Methods and validation papers**

  - Rahimi, M. J., Sitaraman, H., Humbird, D., & Stickel, J. J. (2018). Computational fluid dynamics study of full-scale aerobic bioreactors: Evaluation of gas–liquid mass transfer, oxygen uptake, and dynamic oxygen distribution. *Chemical Engineering Research and Design*, *139*, 283-295.

  - Sitaraman H, Lischeske J, Stickel J & Lu Y (2023), A reacting multiphase computational flow model for 2,3-butanediol synthesis in industrial-scale bioreactors, *Chemical Engineering Research and Design (in press)*

- **Reactor comparison, conversion pathways and optimization**

  - H Sitaraman, M Hassanaly, M Parra-Alvarez, M Rahimi and J Stickel, Impact of Variable Gas Mixtures on Bubble Size Distribution and Mass Transfer in Gas Fermentation Reactors, presented at AIChE Annual Meeting 2022, Phoenix, AZ, USA https://www.nrel.gov/docs/fy23osti/84546.pdf

  - H Sitaraman, M Rahimi, J Lischeske, J Stickel (2019), Multiphase Reacting Flow Simulations and Optimization of Commercial-Scale Aerobic Bioreactors In *2019 AIChE Annual Meeting*. AIChE.

  - H Sitaraman, J Lischeske, M Rahimi, J Stickel (2020, November) Coupled Microbial-Conversion and Computational-Fluid-Dynamics (CFD) Models for Butanediol Production in Micro-Aerated Bioreactors in *2020 AIChE Annual Meeting*

  - M Rahimi, H Sitaraman, J Stickel, K Harrison, N Dowe (2021), Computational fluid dynamics simulation of biomethanation in a large-scale gas bioreactor in *2021 AIChE Annual Meeting*