Coupled Microbial-Conversion and Computational-Fluid-Dynamics (CFD) Models for Butanediol Production in Micro-Aerated Reactors

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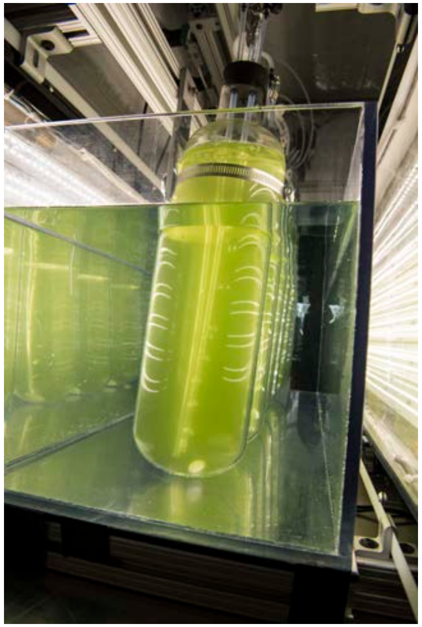
Introduction and Motivation

- Bioreactor: use microbial action for conversion
  - Pharmaceutical industry
  - Waste water treatment
  - **Biofuels and molecules (Research at NREL)**
    - **Ethanol/Butane-diol/Methane**

- Fermentation is a large cost contributor[1]
  - Cost is important: low value products

- Improve economics through bioreactor design
  - More engineering than biology
  - Validated high-fidelity modeling
  - Scale-up/reactor-design optimization
  - Techno-economic analysis

Image by Dennis Schroeder, NREL



Algae bioreactor

Image by Dennis Schroeder, NREL

Biomethanation reactor (NREL)

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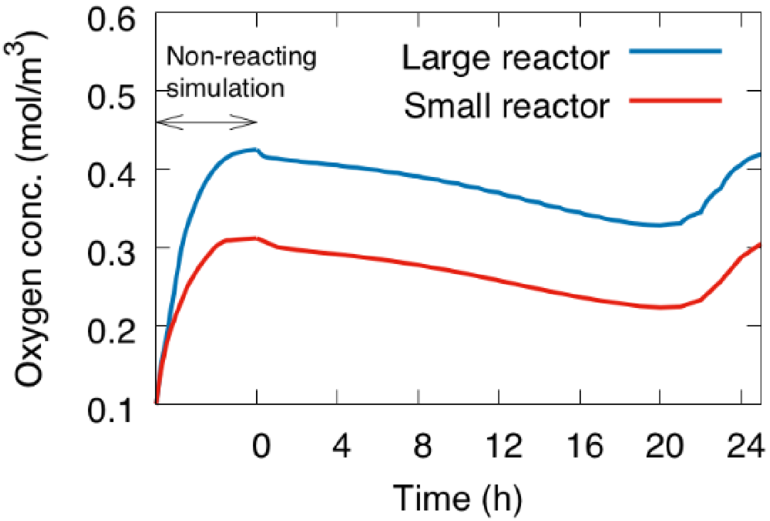
[1] Humbird, D., R. Davis, and J. D. McMillan. "Aeration costs in stirred-tank and bubble column bioreactors." *Biochemical engineering journal* 127 (2017): 161-166.

Micro-aeration for BDO Production

- Central idea:
  - These are not like the traditional bubble columns where lot of air is sparged (superficial velocities of ~1 m/s compared to ~10^{-2} m/s)
  - Only controlled amount of O_2 is required
    - Too much O_2 will trigger creation of wrong products
    - Not enough air will reduce the reaction rate overall, and thus the rate of production of the desired product – butanediol (BDO)
- Method
  - Bubble column CFD + microbial bioreaction
  - Reactions – 5 species (microbe, glucose, xylose, acetoin and BDO)
  - Assume these species are well mixed and O_2 mixing is what limits reactions spatially
- Challenge
  - Long time scales for reactions ~ hours
  - Sub-cycling/operator splitting
    - Solve flow to steady-state
    - Do reactions
    - Redo until final reaction time



Kinetics Model Development and Low-Order Results

Kinetics

Substrate Uptake

$$q_s = q_{s,mx} F_s F_e$$

Sugar Limitation

$$F_s = \frac{C_g + C_x}{C_g + C_x + K_s}$$

Aerobic Limitation

$$F_e = \frac{C_{O_2} + C_A/\beta_e}{C_{O_2} + C_A/\beta_e + K_e}$$

Biomass Growth

$$\frac{dX}{dt} = Y_{X/s} q_s X (1 - \frac{X}{X_{mx}})$$

Electrons Consumed

$$-r_{O_2,e} = \chi_e Y_{O/s} q_s X$$

$$-r_{A,e} = (1 - \chi_e) Y_{A/s} q_s X$$

Substrate Used

$$-r_G = \chi_s q_s X$$

$$-r_{Xy} = (1 - \chi_s) q_s X$$

$$r_{O_2} = r_{O_2,e} + k_L a([O_2]_{sat} - [O_2](x,t))$$

Products Formed

$$r_{A,r} = \chi_p Y_{A/s} q_s X$$

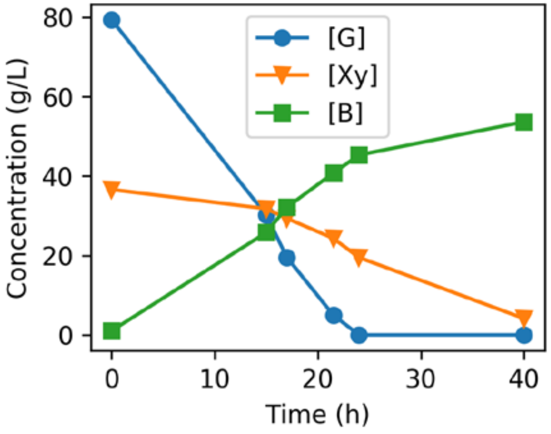
$$r_{B,r} = (1 - \chi_p) Y_{B/s} q_s X$$

Kinetics – Substrate/Product Partitioning

Experimental Phenomena

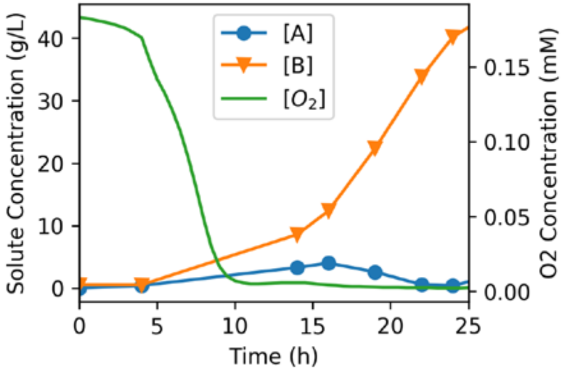
Sugars

Glucose consumed before Xylose



At low O2 concentrations, existing acetoin serves as an electron source

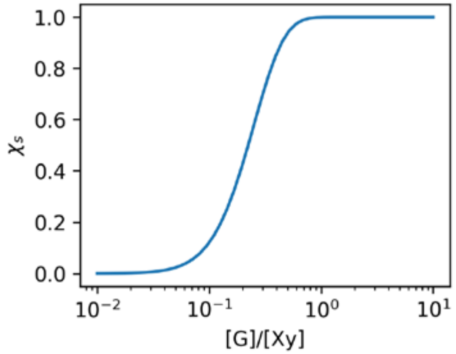
Electrons



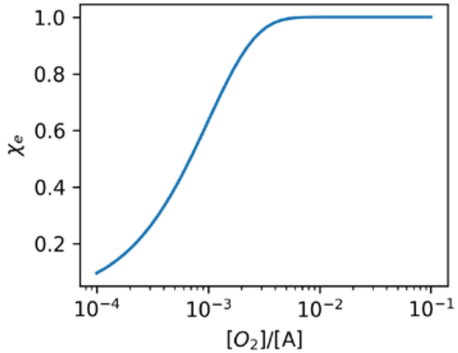
BDO and Acetoin are assumed to be produced in a consistent ratio prior to re-consumption

Modeling Formalism

$$\chi_s = P\left(\alpha_s, \beta_s \frac{[G]}{[Xy]}\right)$$

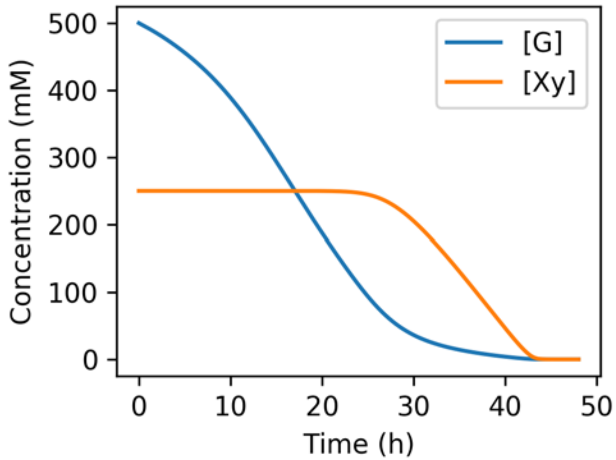


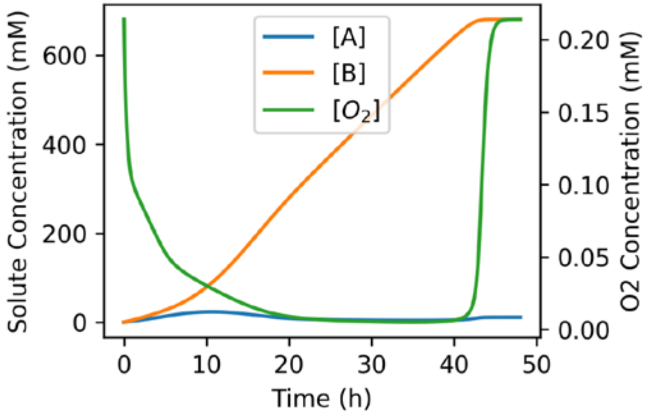
$$\chi_e = P\left(\alpha_e, \beta_e \frac{[O_2]}{[A]}\right)$$



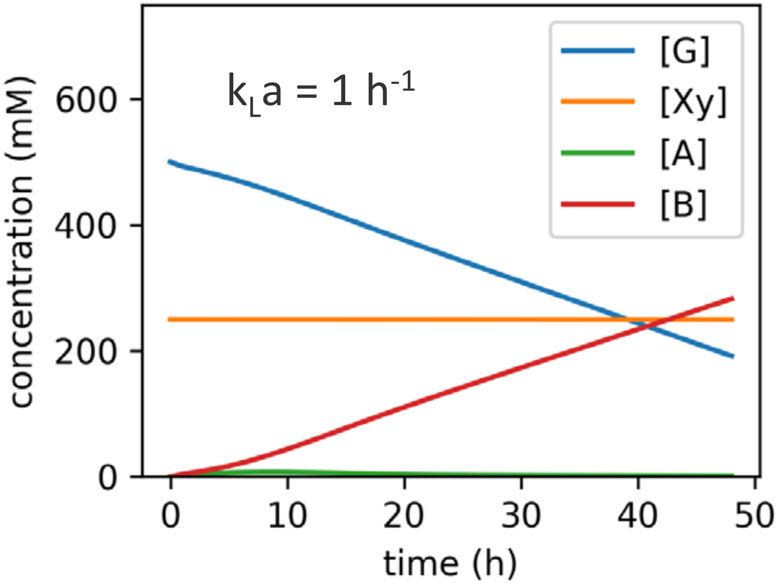
P is the CDF of a gamma distribution

Well-Mixed Model Result



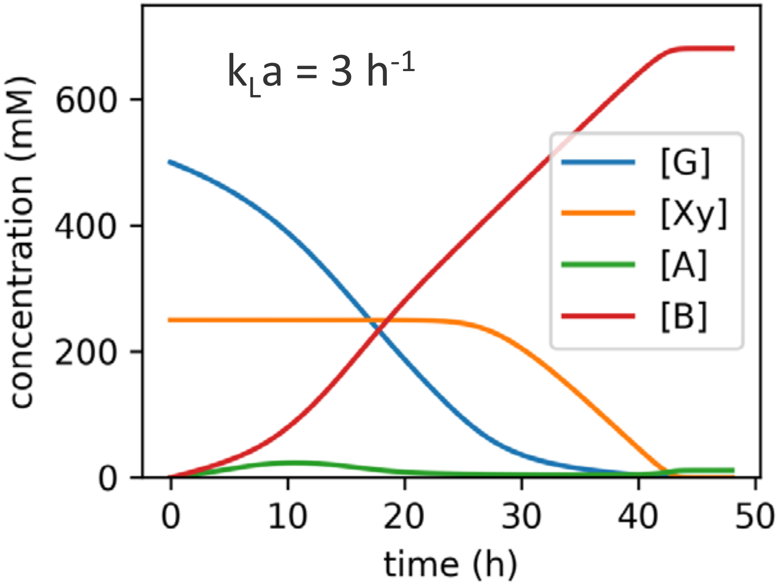


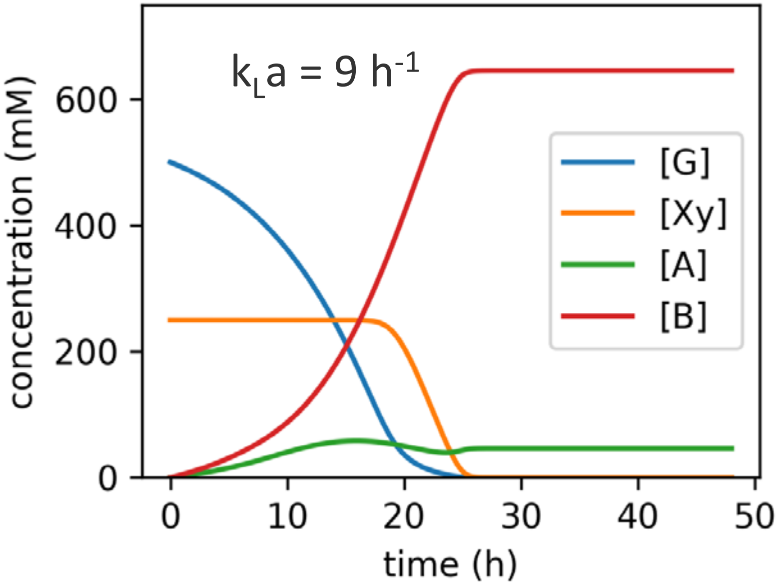
Kinetics Model Exploration – Well-mixed, Constant k_La



Increased oxygen availability:

- Increases overall rate
- Reduces BDO selectivity

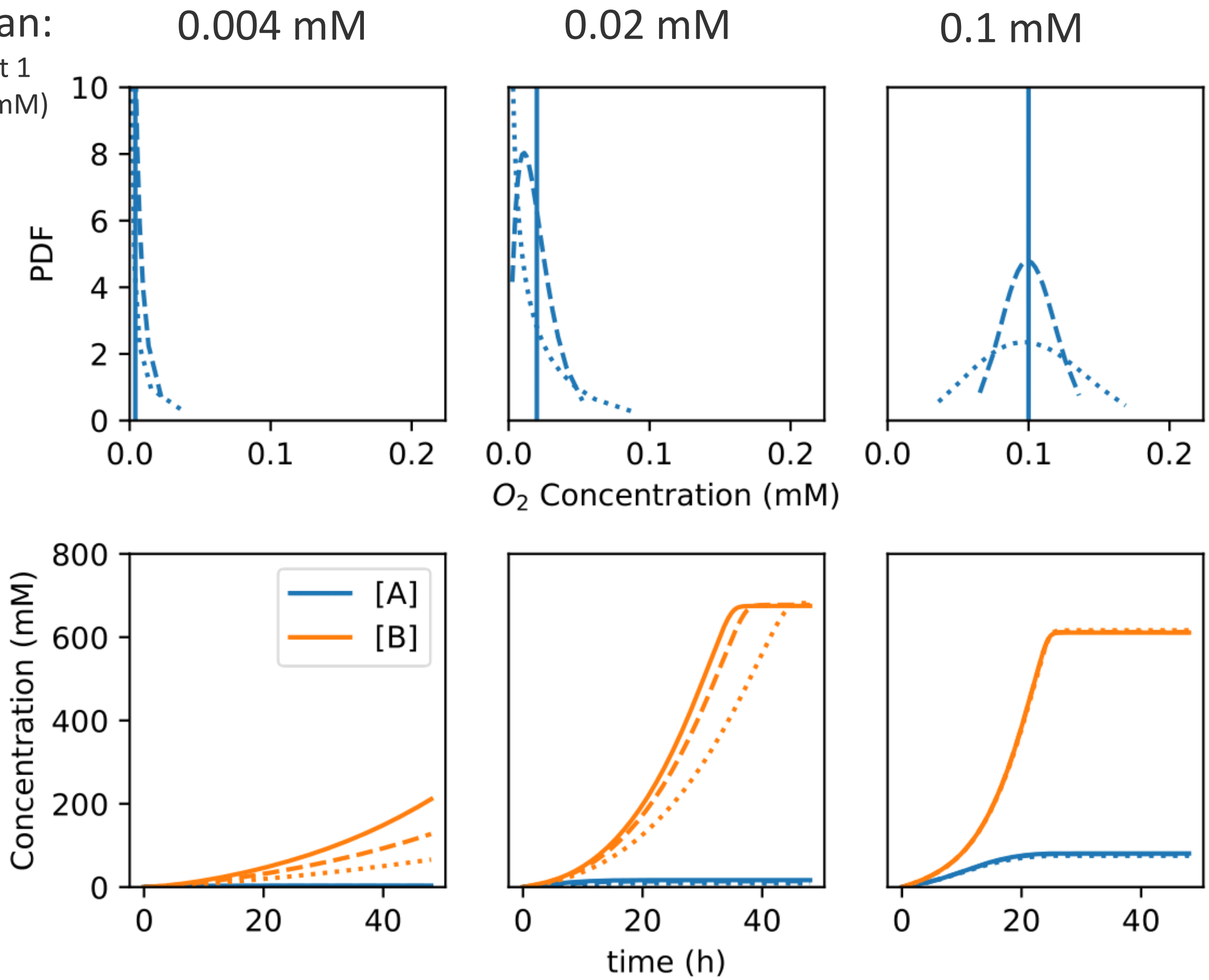




Kinetics Model Exploration – O_2 Distribution

O2 mean:

(Max O2 at 1 bar: 0.21 mM)



O2 assumed to have gamma distribution, where the normalized variance is:

$$\beta^{-1} = \sigma^2/\mu$$

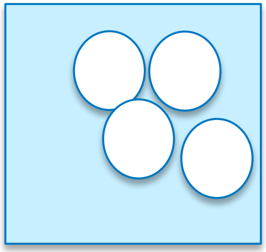
	β^{-1}
Solid	0
Dashed	0.05
Dotted	0.2

Increasing oxygen distribution (holding mean O2 constant) in the reactor reduces overall reaction rate but does not significantly impact product selectivity.

High-Fidelity CFD Sub-cycling Methods

Multiphase Euler-Euler equations

- Gas and liquid as continuous interpenetrating phases
    - Bubble sizes are small compared to reactor dimensions
    - Constant bubble size - 6 mm
- 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

Mass transfer

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

$$\mathrm{OTR} = k_\mathrm{L} a (C^*_{\mathrm{O}_2} - C_{\mathrm{O}_2})$$

Oxygen transfer rate

$$C_i^* = \frac{X_{i,G} P}{H_i} \frac{\rho_L}{M_L}$$

Henry's law

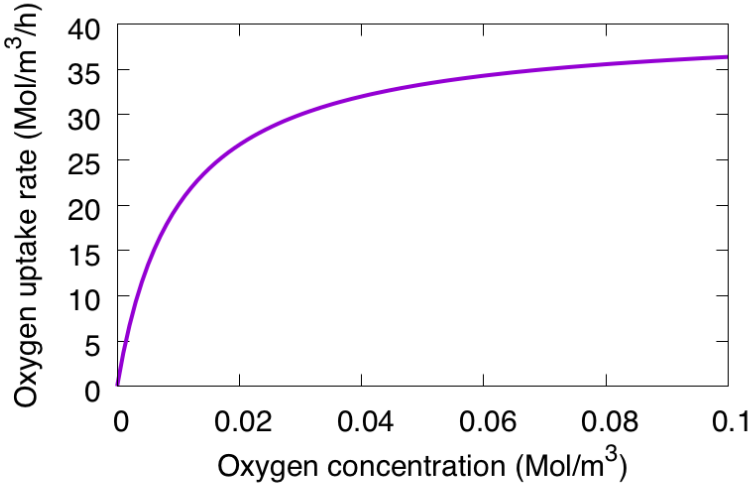
$$k_\mathrm{L} = \sqrt{\frac{4D}{\pi} \frac{|\mathbf{u}_\mathrm{slip}|}{d_\mathrm{b}}} \quad a = \frac{6\alpha_\mathrm{G}}{d_\mathrm{b}}$$

Mass transfer coefficient

Microbial oxygen uptake (Monod model)

$$\mathrm{OUR} = \mathrm{OUR}_\mathrm{max}(X) \frac{C_{O_2}}{K_O + C_{O_2}} \alpha_L$$

Representative oxygen uptake relationship during CFD



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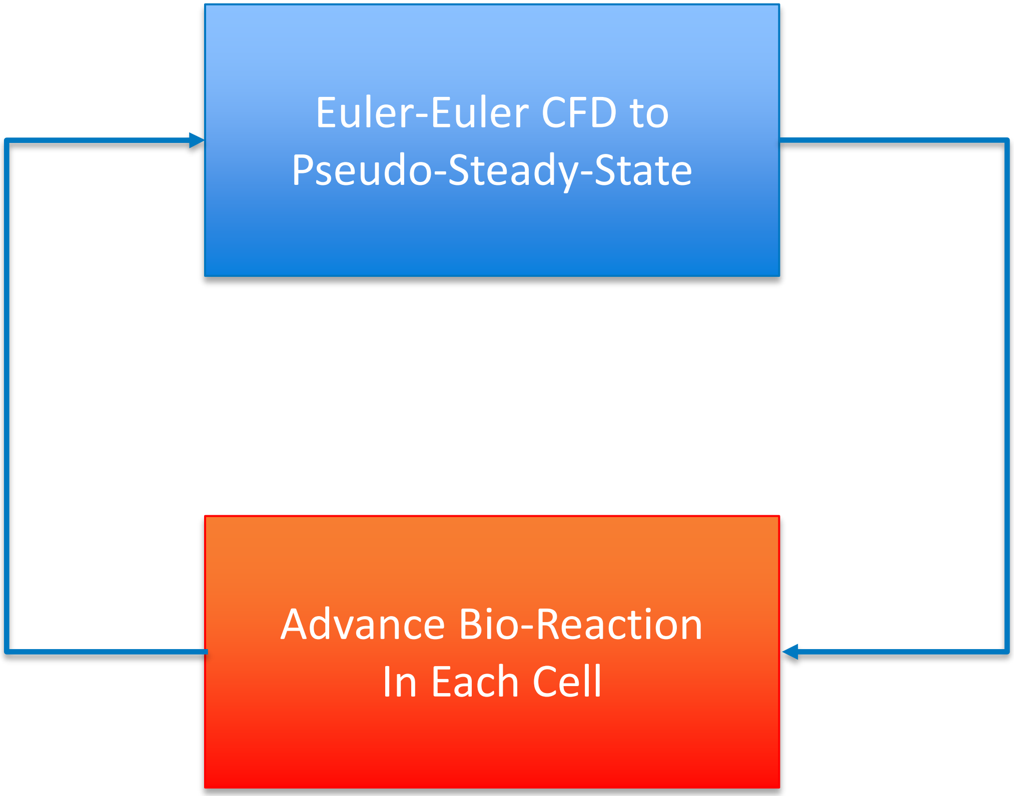
[1] Higbie, R., 1935. The rate of absorption of a pure gas into a still liquid during short periods of exposure. Trans. AIChE 31, 365–389.

Reaction Subcycling/Operator Splitting



Euler-Euler CFD to Pseudo-Steady-State

Oxygen concentration distribution

Advance Bio-Reaction In Each Cell

Biomass, substrate, and product concentrations give new max O2 uptake

Computational model

- Transport properties

  - Fermentation broth properties are similar to water
  - Grace drag model for bubbles
  - Wilke-Chang diffusion of species
  - Multiphase k-ϵ turbulence model
  - Wall lubrication effects

- Customized solver *bdoFOAM* calls customized solver *TwoPhaseEulerFoam* in OpenFOAM

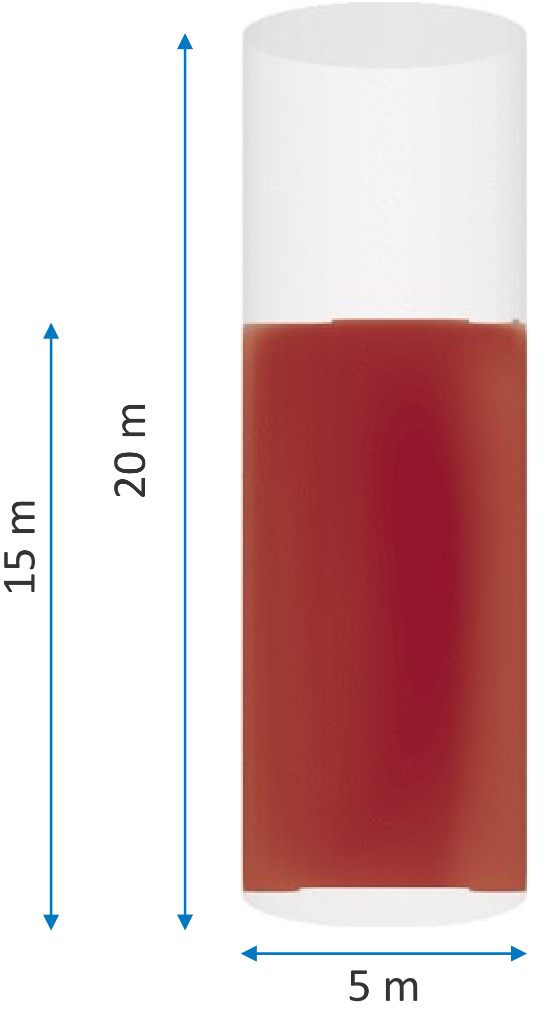
- Simulations performed using

  - 72 Intel Skylake processors
  - 48 hours of run time to simulate 2000-8000 seconds
  - Kinetics step is trivially fast

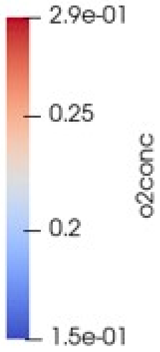
- More details in Rahimi et al., Chem. Eng. Res. Design, 139, 2018

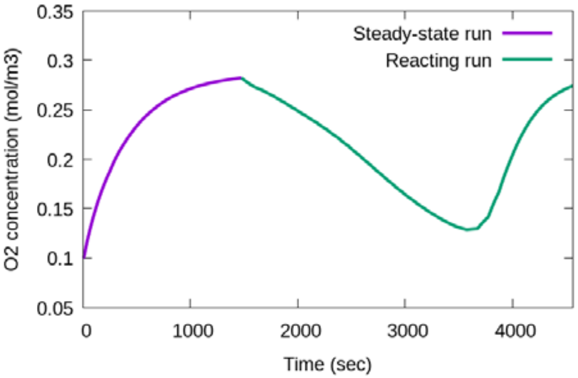
Subcycling Results

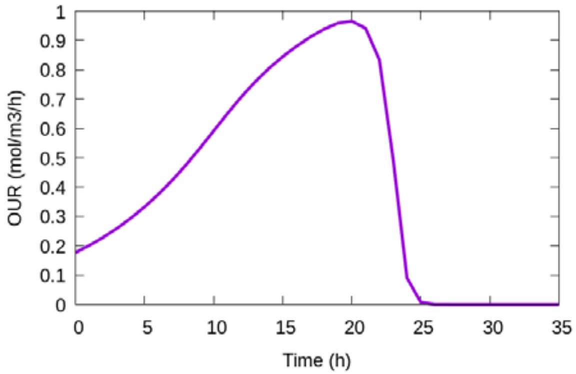
Bubble Column Reactor for BDO Production

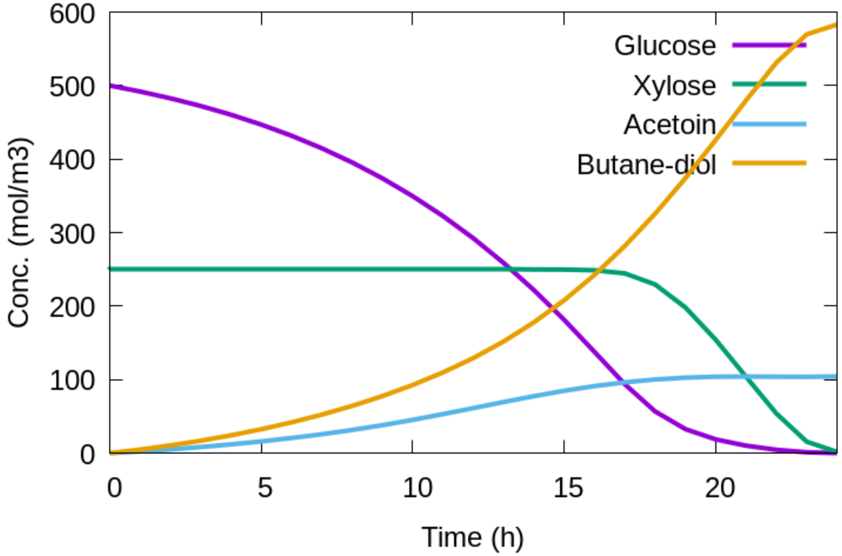


3 m dia sparger

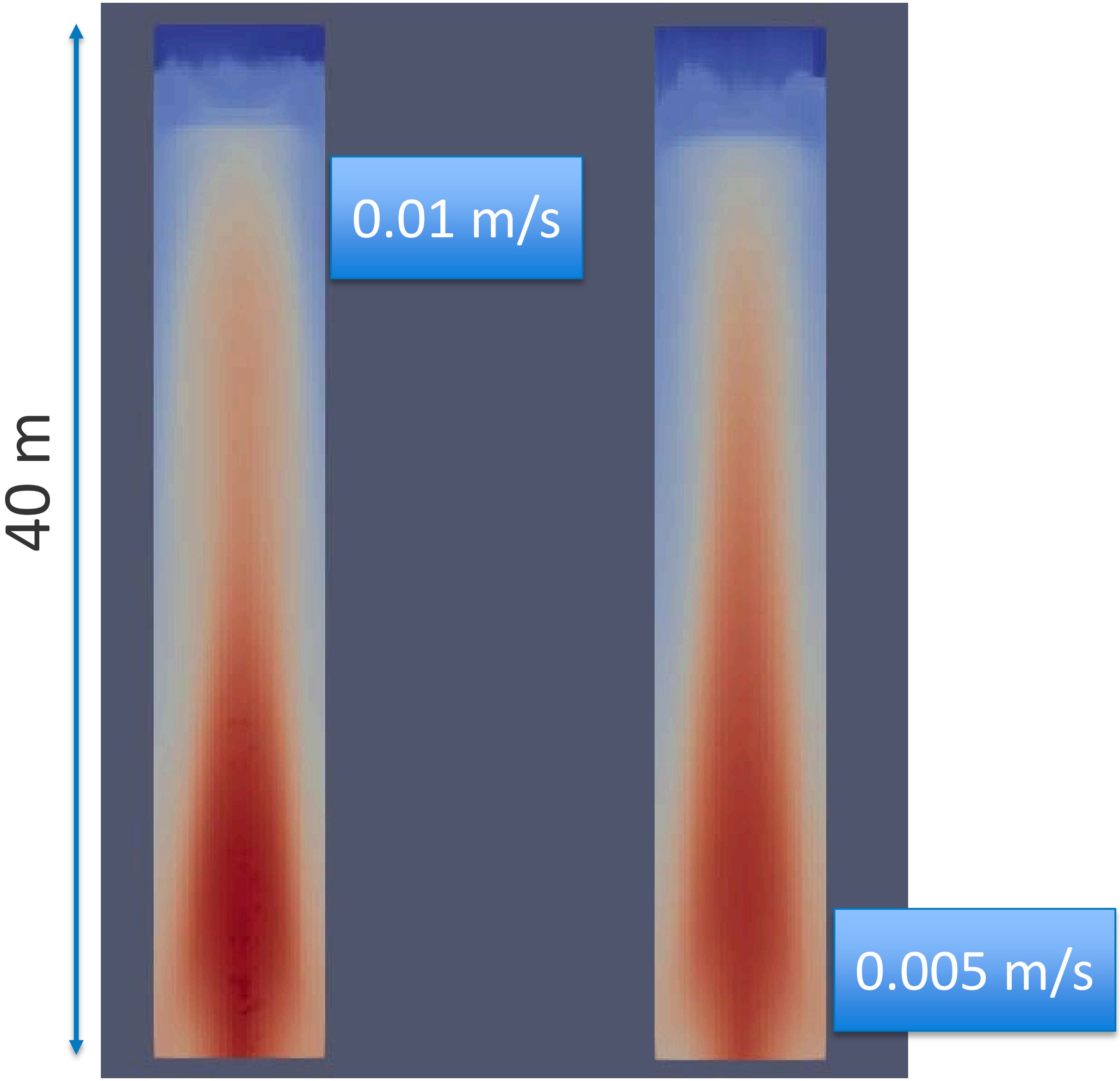


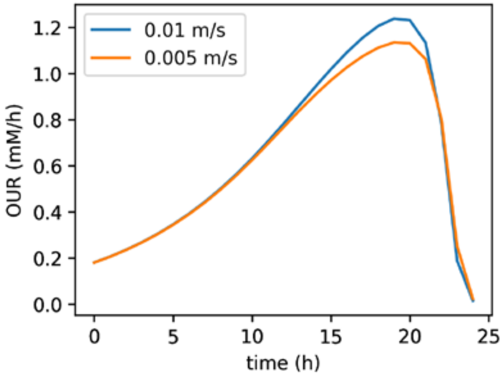


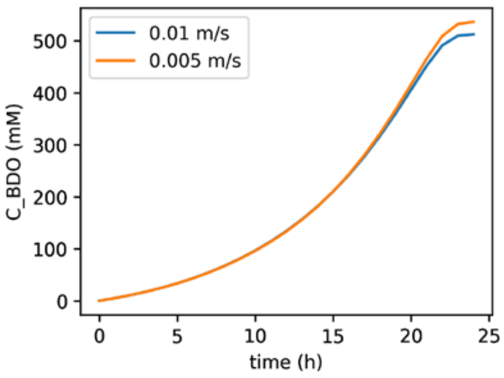


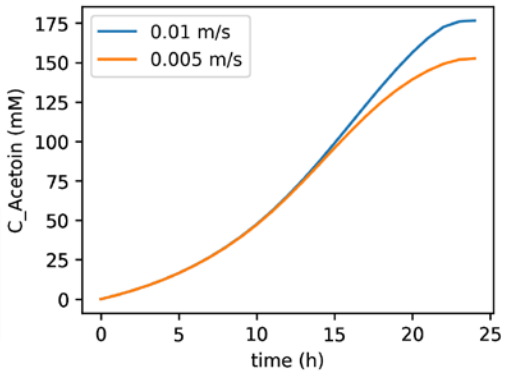


Sparge Rate Comparison

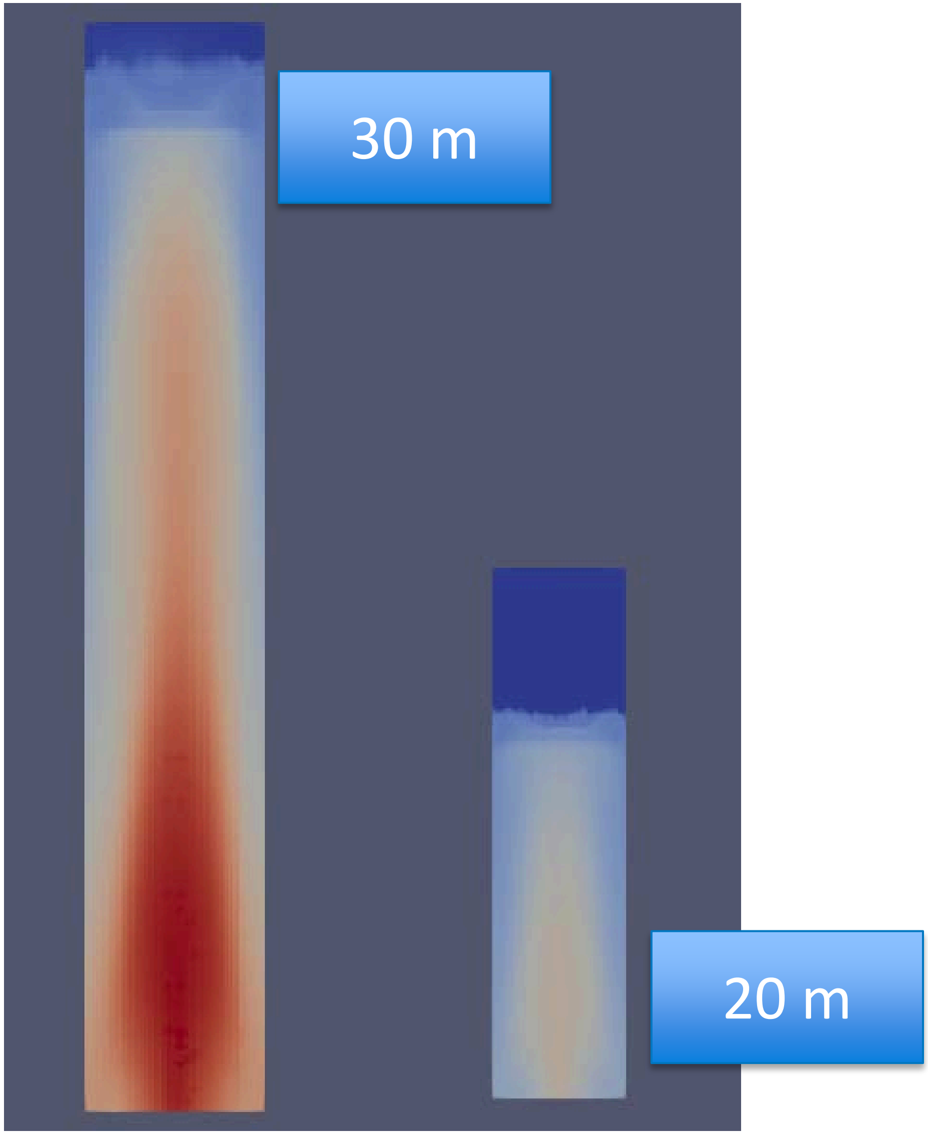


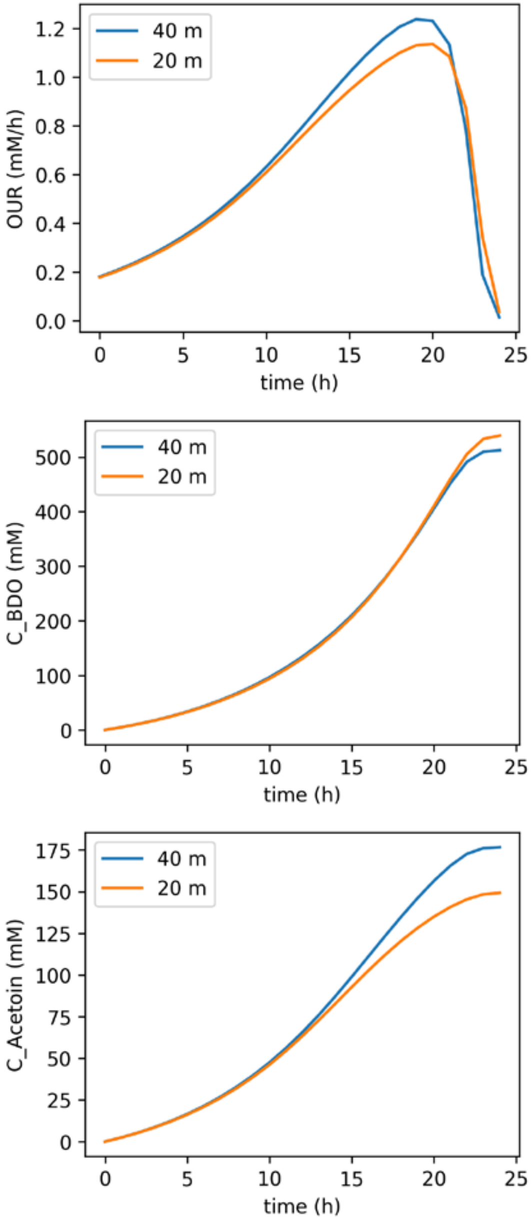






Reactor Height





Conclusions and future work

- **Conclusions**
  - Computational model
    - Kinetics model capturing unique dynamics of *Z mobilis* bench-scale fermentation developed
    - Euler-Euler gas-transfer and OUR model implemented to determine pseudo-steady-state O2 profiles in industrially-relevant bubble columns
    - Subcycling implemented to model batch fermentation in large-scale reactors
  - Results
    - Impact of mean and variance of O2 demonstrated using a simple model
    - High-fidelity CFD demonstrated the impact of flow rate and reactor height on product selectivity

- **Future work**
  - Pushing oxygen concentration to lower mean O2 values
    - Model stability
    - Characterize tradeoff between selectivity and productivity
  - Evaluate additional reactor designs to enable high-scale low-mean and low-variance O2 concentration across reactor. Some possibilities:
    - Pump-around loop
    - Shallow channel
  - Evaluate oxygen feed timing strategies to overcome reactor limitations

Acknowledgements

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Energy Efficiency &
Renewable Energy

BIOENERGY TECHNOLOGIES OFFICE

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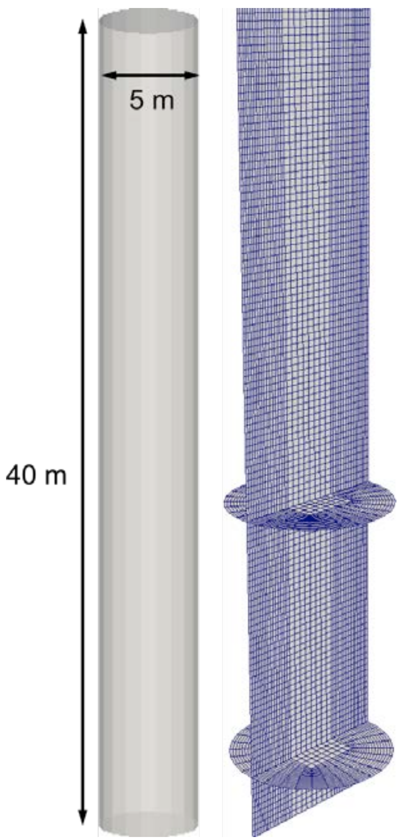
Thank You

Questions? Email me at
James.Lischeske@nrel.gov

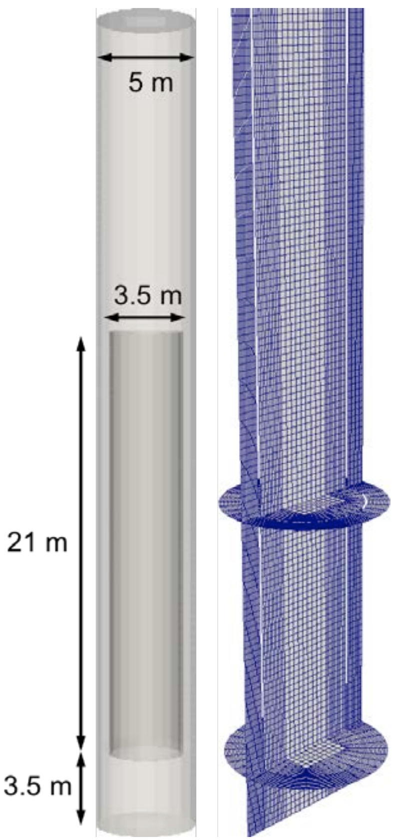
NREL/PR-5100-78334

Appendix

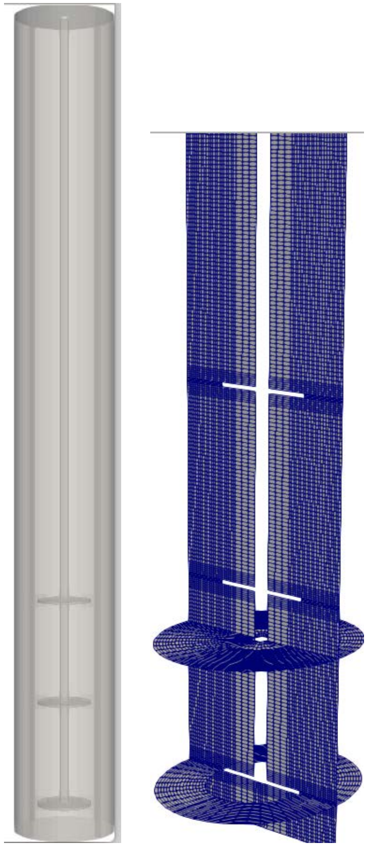
Geometry and meshing



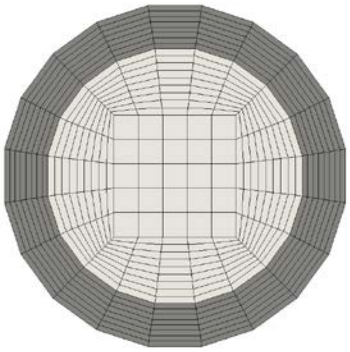
Bubble column



airlift



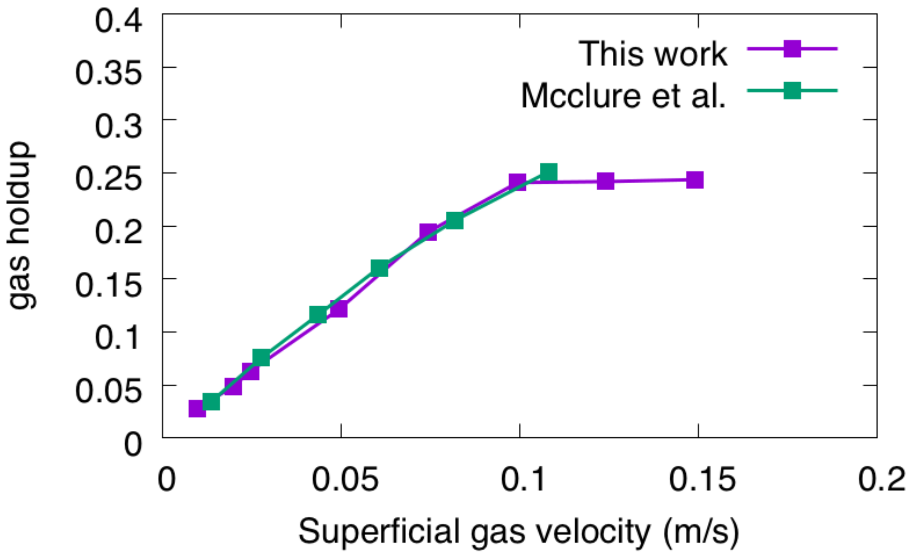
Stir tank

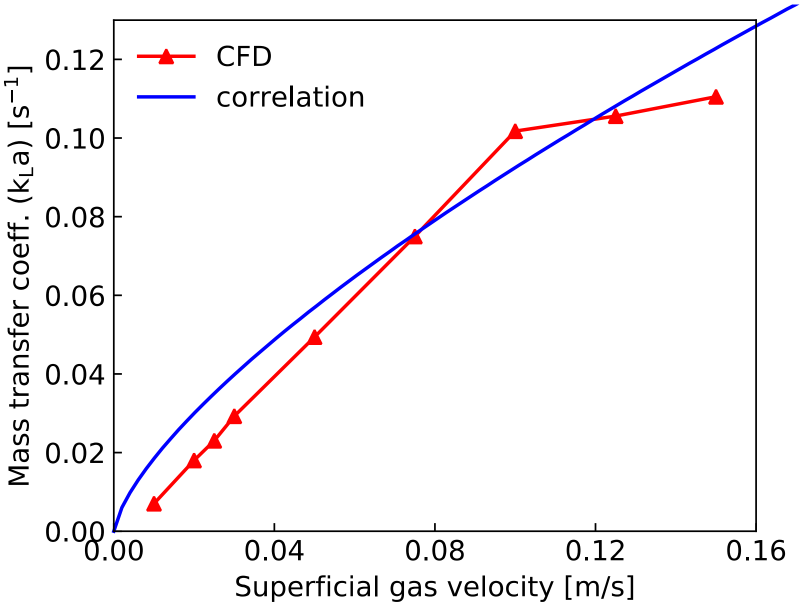


Sparger

- Bottom inlet with a gas fraction that specifies sparger mass flow rate
- Lateral walls use no-slip condition for liquid and slip for gas
- ~ 300,000 cells – sufficient for grid convergent solutions

CFD Model validation with small-scale bubble column





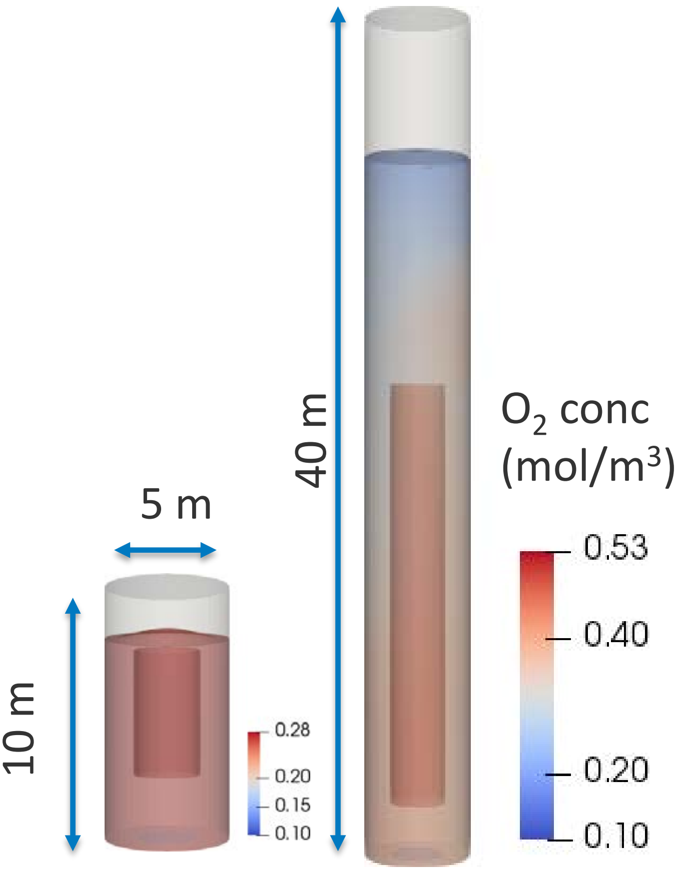
Superficial gas velocity: $vg_s = \dfrac{\dot{V}_{mid}}{A_{reactor}}$

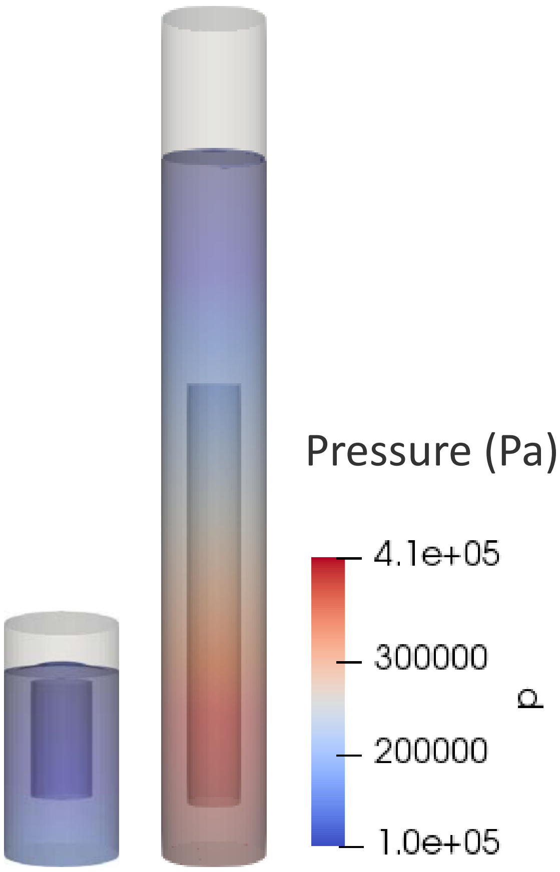
- Validation done for a small-scale bubble column (1 m height, 15 cm diameter)
- Average mass transfer coefficient matches *Heijnen and Van't Riet (1984)*[1]
- Gas holdup matches experiments/simulations by Mcclure et al. (2013) [2]

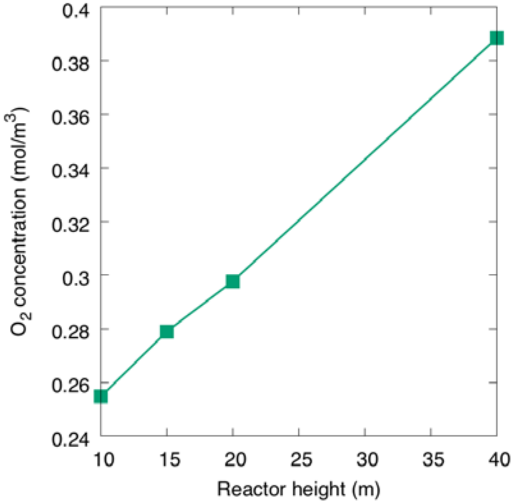
[1] Heijnen, J. J., Van't Riet, K., Apr. 1984. Mass transfer, mixing and heat transfer phenomena in low viscosity bubble column reactors. Chem. Eng. J. 28 (2), B21–B42.

[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.

Sensitivity to reactor height

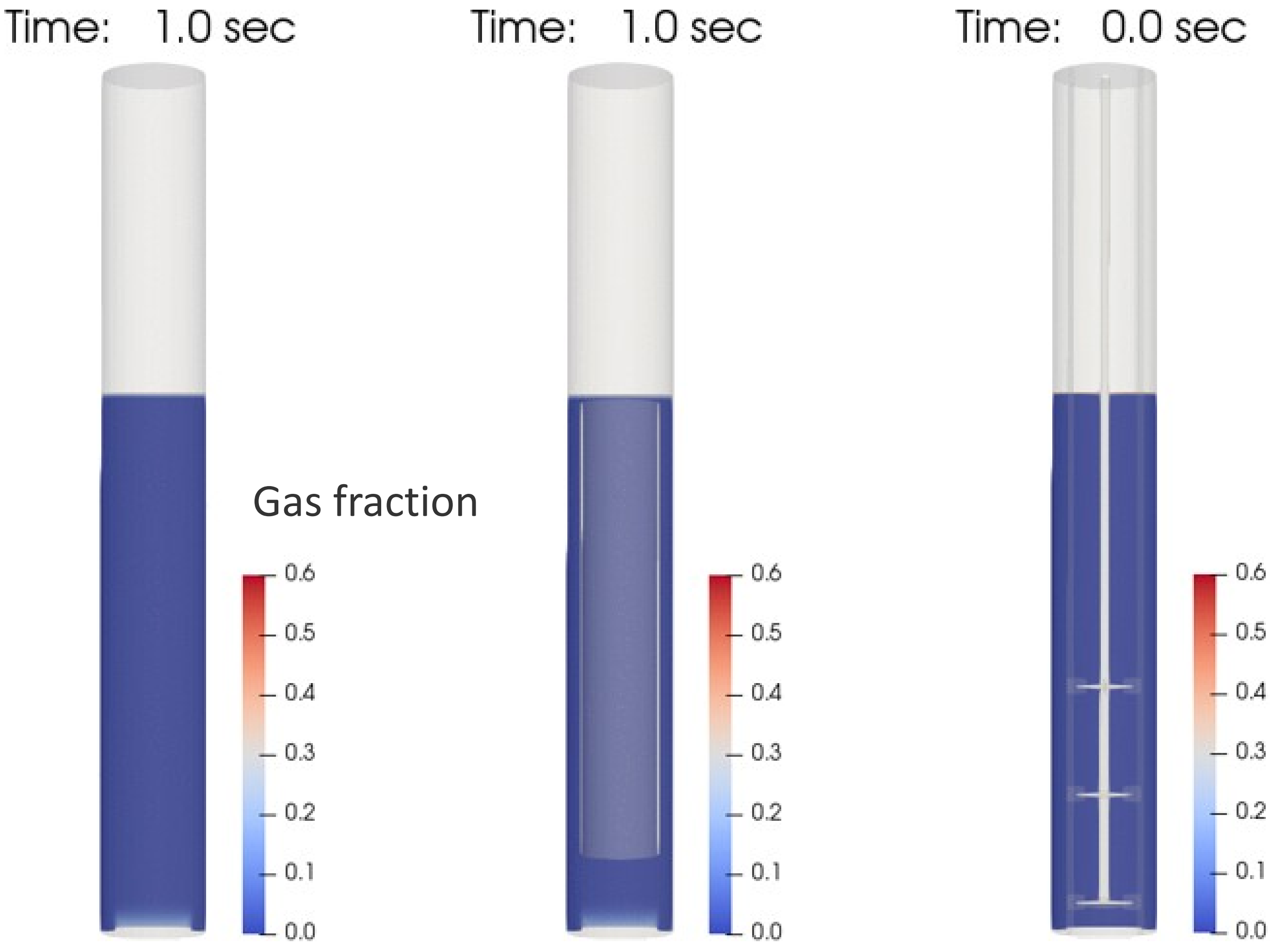






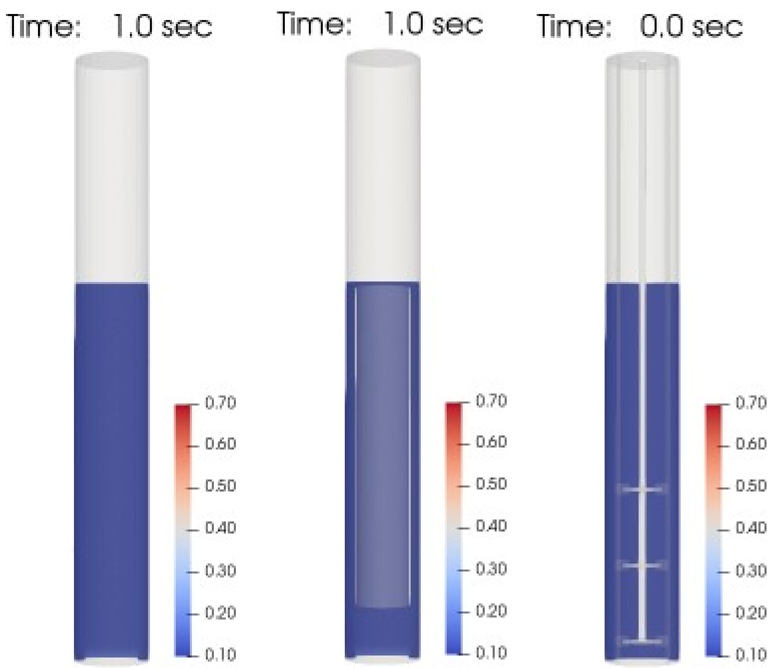
- Cases are at superficial gas velocity of 2 cm/s
- Larger hydrostatic pressure head with greater height
  - Larger oxygen transfer due to higher Henry saturation concentration

Transient fluid dynamics (comparison)

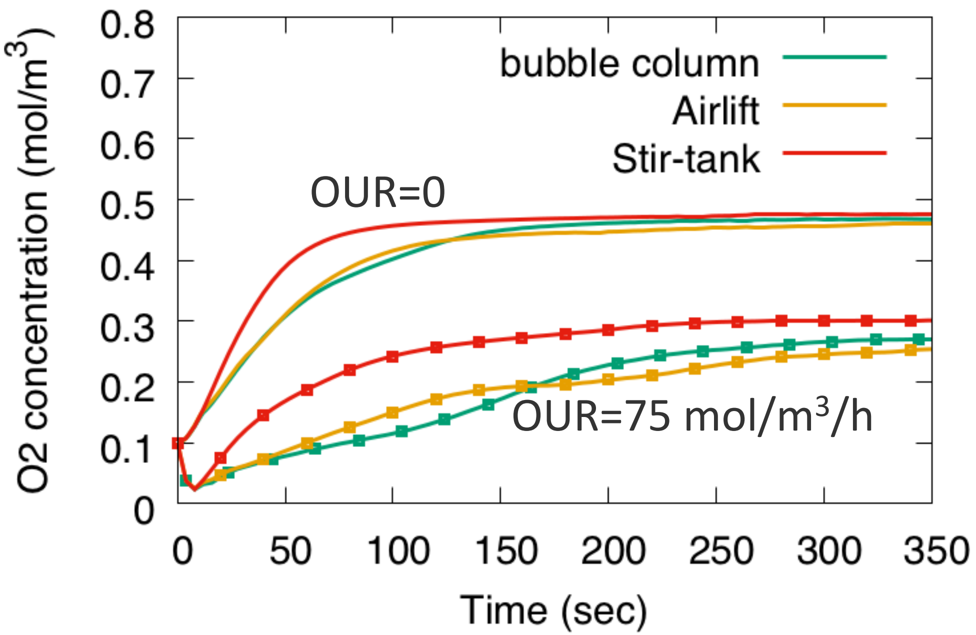


- Superficial gas velocity = 0.1 m/s, impeller speed = 2 rad/s
- Gas hold up is similar for all cases
- Faster time scale to steady state with impellers
- Draft tube and impellers aid better mixing

Oxygen transfer



Oxygen concentration in mol/m^3



- All reactors show almost the same average concentration without microbial uptake
- Higher mass transfer rate in that case of stir tank reactor
- Stir-tank reactor higher average oxygen concentration with microbial uptake

Oxygen limited regions

Bubble column

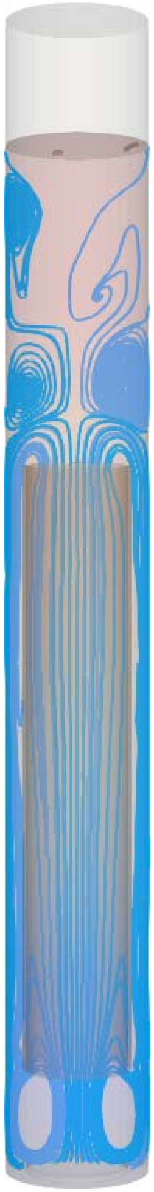
airlift

Stir tank

- Oxygen limited regions are where microbial uptake is sub-optimal < 0.1 mol/m^3
- Radial transport is limited in bubble column, mitigated in airlift and stir tank
- O2 limited regions towards the top and the wall boundaries

Streamlines and mixing

Bubble column

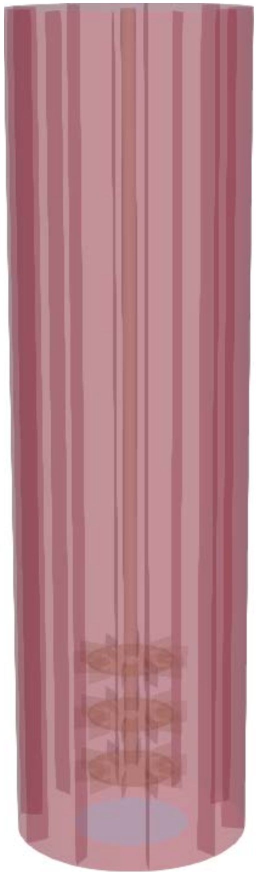


airlift

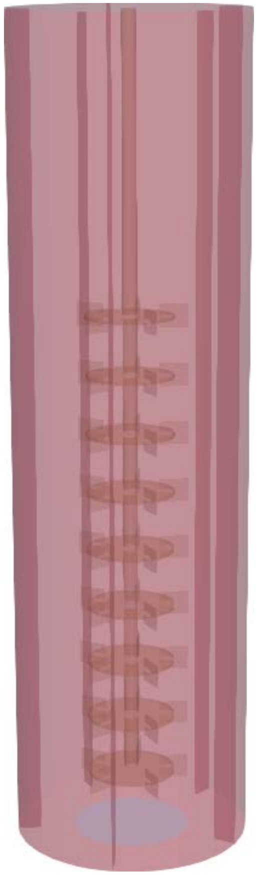
Stir tank

- Streamlines obtained from temporal averaging of liquid velocity at steady-state
- Draft tube allows for better top to bottom mixing
- Impellers in the stir tank form Taylor vortices that aid in better mixing

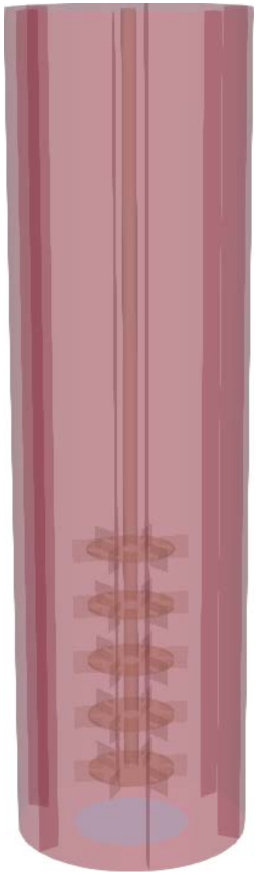
Automated meshing of stir-tank reactor



3 impellers,
10 baffles



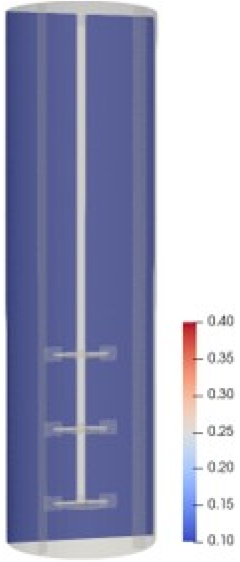
9 impellers,
5 baffles



5 impellers,
6 baffles

- Automated python script allows for a generic design that can be used for optimization

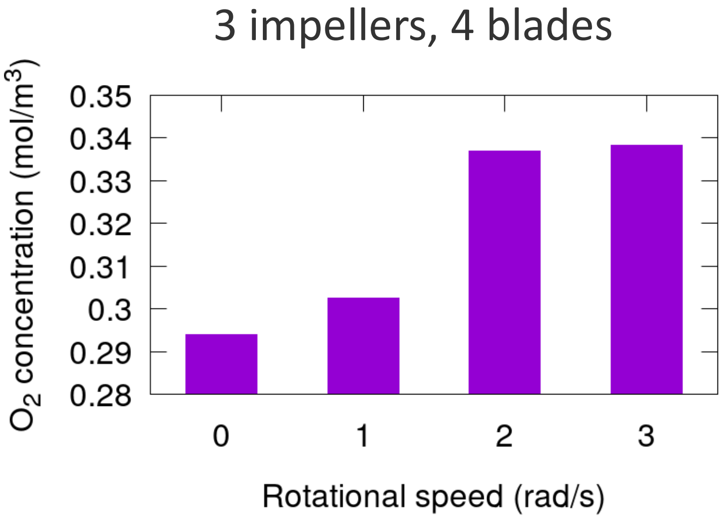
Stir tank optimization

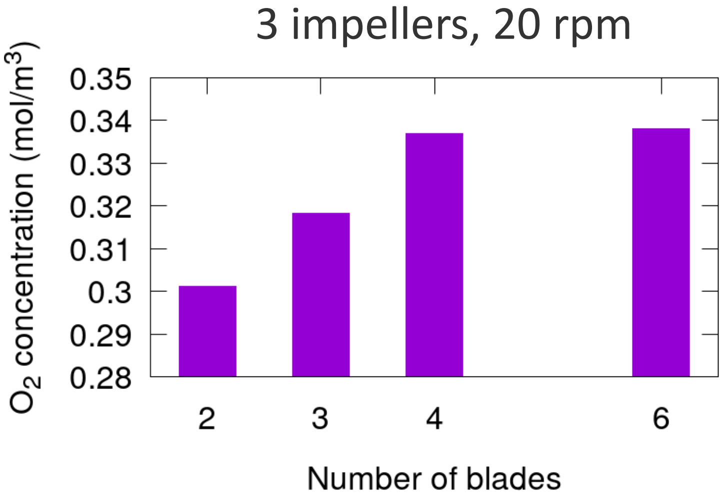

O_2 (mol/m^3)

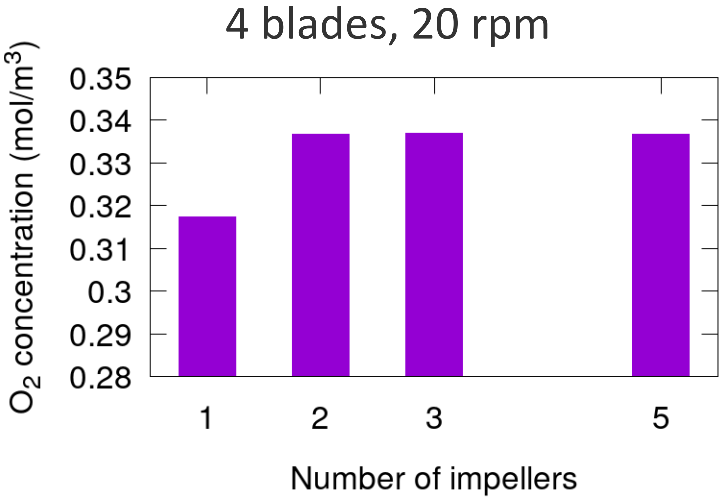
Sensitivity of stir-tank reactor

- 5 m dia, 17 m height
- Vgs=2 cm/s

- average O_2 concentration
  - Rotational speed
  - No: of blades
  - No: of impellers


3 impellers, 4 blades


3 impellers, 20 rpm


4 blades, 20 rpm