CO2 Reduction and Upgrading for e-Fuels Consortium
U.S. DEPARTMENT OF ENERGY

NREL/PO-2C00-86729

Multiphysics computational fluid dynamics for design and scale-up of CO_2/syngas bioreactors

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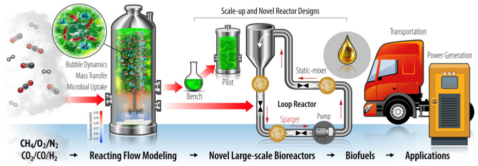
Overview

What are we trying to do: Develop validated, high fidelity computational models for gas fermentation using CO_2, CO and H_2.
How is it done today?: microbiological advances are made at the lab scale in well mixed reactors. Process scale-up remains an open question when considering novel microorganisms/reaction systems.
Why is this important?: lab-to-production scale transition is required to achieve CO_2 conversion at Mton/Gton scale and computational models can de-risk/accelerate this transition.



Goal: enable scaled-up bioreactor design and optimization for CO2 conversion through validated high fidelity Multiphysics simulations

Milestone	Criteria	Date	Status
Survey different methodologies to simulate bubble size distributions (BSD)	Literature on novel approaches to modeling and simulation of bubble breakage, aggregation, and mass transfer using population balances	3/3/2022	Completed
Kinetic mechanisms on bioreaction pathways	Selection/construction of lumped bioreaction kinetic models	6/30/2022	Completed
Influence of BSD on gas composition	Using high-fidelity Eulerian methods, determine differences in BSD and mass transfer for varying gas mixtures	9/30/2022	Completed
Hydrodynamics validation set-up and preliminary simulation towards go/no go decision	Quantify validation parameters and set-up preliminary simulations to compare against experiments reported in literature	12/31/2022	Completed
Meshing and preliminary simulation of sparger geometries	Create high resolution meshes for ring/porous plate sparger designs	03/31/2023	Completed
Novel design of sparging systems	Use multiphase flow models to explore various sparger designs such as ring/porous plates used at scale and explore variations in geometric parameters.	06/30/2023	85% completion

Table 1. Technical approach

Computational models

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

$$\frac{\partial n_v}{\partial t} + \nabla\cdot(v_{ph} n_v) = h_v$$
Bubble size distribution transport (population balances)

$$h_v = \underbrace{\frac{1}{2}\int_0^v n_{v'} n_{v-v'} C_{v',v-v'} dv'}_{\mathrm{coalescence}(+)} - \underbrace{n_v \int_0^\infty n_{v'} C_{v,v'} dv'}_{\mathrm{coalescence}(-)} + \underbrace{\int_v^\infty n'_v B_{v'} \beta_{v,v'} dv'}_{\mathrm{breakup}(+)} - \underbrace{n_v B_v}_{\mathrm{breakup}(-)} - \underbrace{\frac{\partial(\dot{v} n_v)}{\partial v}}_{\mathrm{drift}} + \underbrace{\dot{n}_v}_{\mathrm{nucleation}}$$
Bubble dynamics

Bibliography
1. Mozumder et al., (2015). Poly(3-hydroxybutyrate) (PHB) production from CO2: Model development and process optimization. *Biochemical Engineering Journal* 98,107–116
2. Ngu et al., (2022). Spatio-temporal 1D gas–liquid model for biological methanation in lab scale and industrial bubble column. *Chemical Engineering Science*. 251, 117478
3. Decwer et al., (1978). A Comprehensive Study on CO-Interphase Mass Transfer in Vertical Co-current and Counter-current Gas-Liquid Flow. *The Canadian Journal of Chemical Engineering*, Vol. 56 43-55
4. Hissanaga et al., (2020). Mass transfer modeling and simulation of a transient homogeneous bubbly flow in a bubble column. Chemical Engineering Science 218, 115531.
5. Hassanaly Malik, Sparger mesh generation for OpenFOAM. https://github.com/NREL/spargerDesign

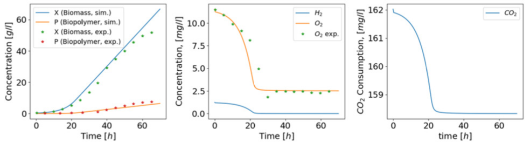
Lumped bioreaction kinetics

$$\frac{d\rho_{H_2}}{dt} = k_L a_{H_2}\left(\rho^*_{H_2} - \rho_{H_2}\right) - \left(\frac{\mu_{Xs}}{Y_{X,H_2}} + \frac{\mu_{Ps}}{Y_{P,H_2}}\right)\rho_X$$
$$\frac{d\rho_{O_2}}{dt} = k_L a_{O_2}\left(\rho^*_{O_2} - \rho_{O_2}\right) - \left(\frac{\mu_{Xs}}{Y_{X,O_2}} + \frac{\mu_{Ps}}{Y_{P,O_2}}\right)\rho_X$$
$$\frac{d\rho_{CO_2}}{dt} = k_L a_{CO_2}\left(\rho^*_{CO_2} - \rho_{CO_2}\right) - \left(\frac{\mu_{Xs}}{Y_{X,CO_2}} + \frac{\mu_{Ps}}{Y_{P,CO_2}}\right)\rho_X$$

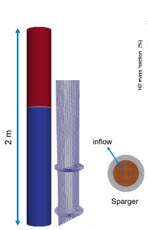
$$\mu_X = \mu_X^{max}\left(\frac{\rho_{H_2}}{K_{X,H_2} + \rho_{H_2}}\right) \times \left(\frac{\rho_{O_2}}{K_{X,O_2} + \rho_{O_2}}\right) \times \left(\frac{\rho_{CO_2}}{K_{X,CO_2} + \rho_{CO_2}}\right) \times \left(\frac{\rho_N}{K_N + \rho_N + \frac{\rho_N^2}{K_{IN}}}\right)$$

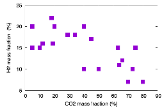
$$\mu_P = \mu_P^{max}\left(\frac{\rho_{H_2}}{K_{P,H_2} + \rho_{H2}}\right) \times \left(\frac{\rho_{O_2}}{K_{P,O_2} + \rho_{O_2} + \frac{\rho_{O_2}^2}{K_{IO_2}}}\right) \times \left(\frac{\rho_{CO_2}}{K_{X,CO_2} + \rho_{CO2}}\right) \times \left[1 - \left(\frac{f_P}{f_{P,max}}\right)^\beta\right] \times \frac{K_{IN}}{\rho_N + K_{IN}}$$

- We verify a mixture **Monod model** for a case of PHB production from the fermentation of CO_2/H_2/O_2 gas mixtures in nitrogenous fermentation medium described by Mozumder et al.
- The kinetic model is general enough because it considers the effect of all gases with possible nutrient limitations as well as the effect of the biopolymer in the rates of growth and production.
- The ODEs represent the term R_{ij} in the species transport equation.

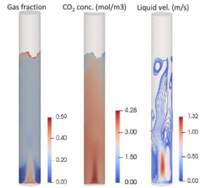


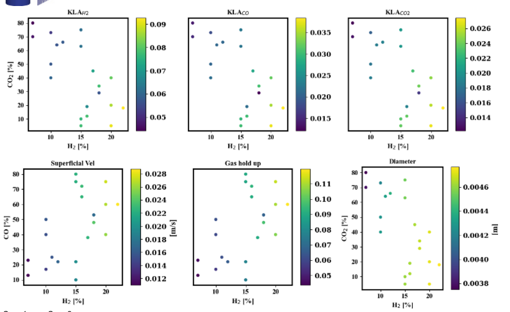
Influence of bubble size distribution (BSD) and gas composition

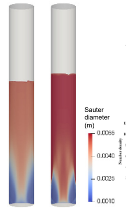


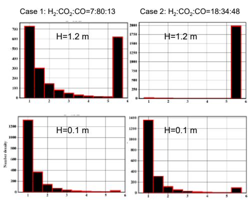


- Mass flow rate and gas fractions are specified at bottom inlet.
- Gas fraction (H_2:CO_2:CO) was varied while mass flow rate was kept constant (0.45 g/s).





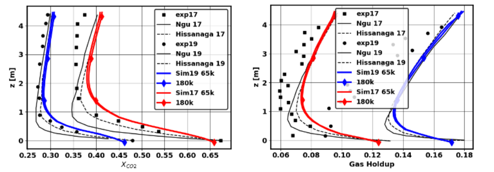




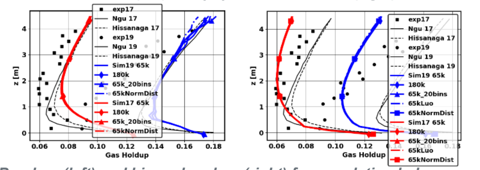
- Higher H_2 fractions results in higher:
  - superficial velocities
  - gas holdup
  - Mean diameter
- specifically, for 2 cases, one with higher and one with lower H_2 content, there seems to be faster bubble coalescence and less breakage.
- Higher H_2 fractions also results in greater mass transfer for all gases at a constant mass flow rate.

Validation setup

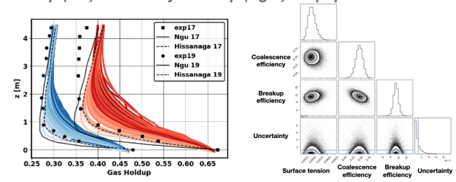
- As first step towards validation, we have compared our CFD modeling against computational/experimental work presented in Ngu et al., Decwer et al., and Hissanaga et al. The work represents dissolution of air/CO2 mixtures in a bubble column.
- The comparison was carried out assuming **constant bubble** size and modeling the bubble size distribution via **population balances** where we use **breakup** and **binary breakup** models to represent changes in bubble size due to breakup events.
- Additionally, we set up a Bayesian calibration methodology to answer the question: How should one modify hydrodynamics models to capture experiments? this involves hundreds of runs varying different modeling parameters for population balances



Constant diameter (left) and population balances (right) validation

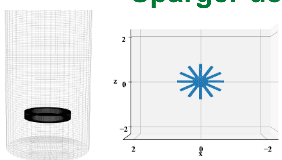


Breakup (left) and binary breakup (right) for population balances

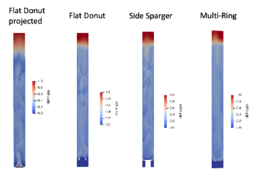


Bayesian calibration for breakup and coalescence efficiencies

Sparger design



- Sparger design is influential in the control of the bubble size distribution. Additionally, uniformity in gas sparging, affects gas-hold up, interfacial area, mixing time and mass transfer coefficient.
- We have created tools to generate the computational meshes for the spargers:
  - Automated STL generation
  - Automated block-cylindrical meshing
- These tools are publicly available: https://github.com/NREL/spargerDesign



- We are looking for differences in performance for different types of spargers: multiring, single ring, ladder, as well as computational approximations of these configurations.
- So far multi-ring sparger has the largest effect on interphase diffusion.
- We are working to make spatial (height dependent) and temporal comparisons between al configurations