A lattice Boltzmann method for predicting porous transport layer performance during electrolysis

Ethan Young, Arth Sojitra, Jacob Wrubel, Marc Henry de Frahan

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Background

- Electrolysis, the splitting of water into **oxygen (O_2)** and **hydrogen (H_2)** using electricity, is a sustainable way to produce hydrogen

- In **polymer electrolyte membrane (PEM) water electrolysis**, water is brought into contact with charged catalyst layers and electrochemically separated into O_2 and H_2.
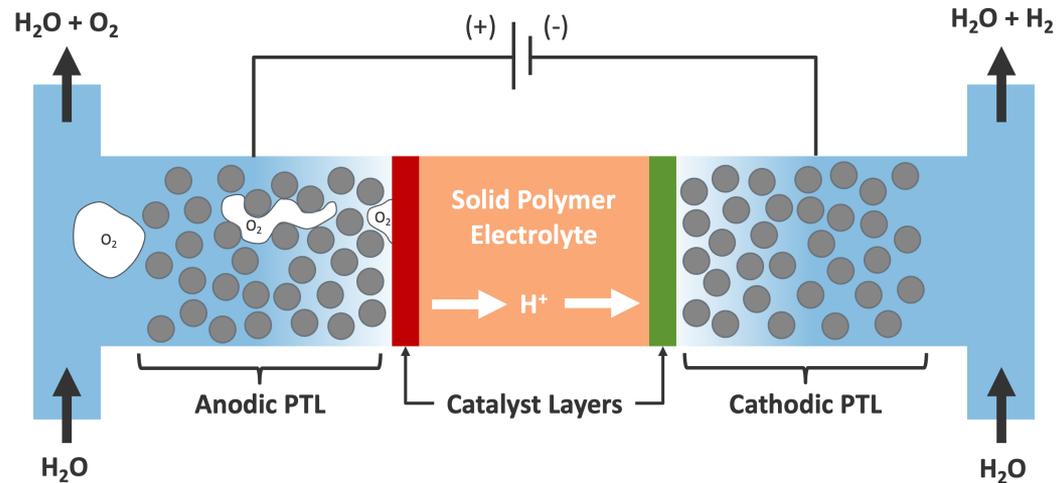
Motivation

- The design of the anodic **porous transport layer (PTL)** aids in the detachment and removal of O_2

- The PTL's ability to remove O_2 from the catalyst surface is a critical factor in overall efficacy


$H_2O + O_2$

O_2

O_2

O_2

Anodic PTL

H_2O
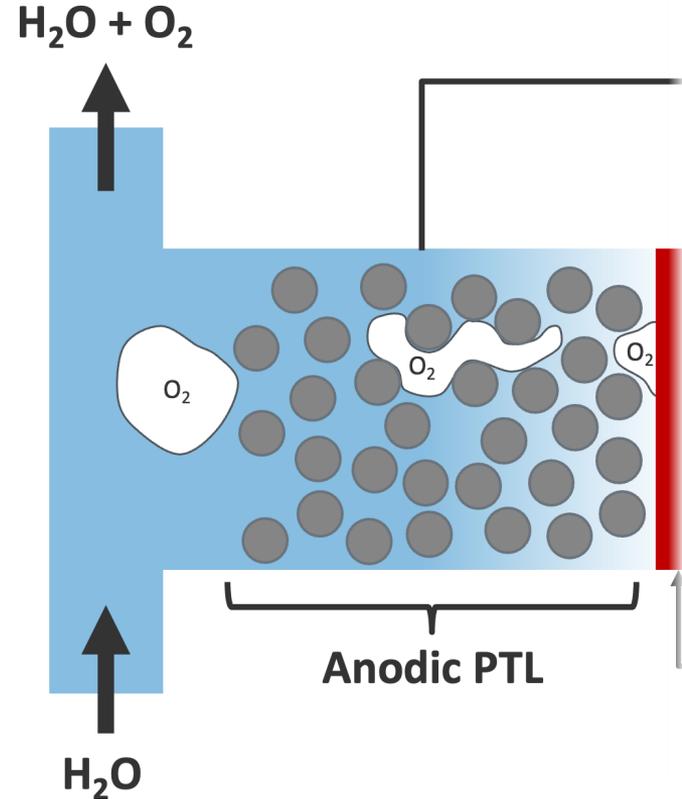
Approach: Lattice Boltzmann

- We use a **d2q9 lattice Boltzmann method (LBM)** to capture the interaction between PTL, H_2O, and O_2
  - Easily capture complex boundaries
  - Relative ease of multiphase algorithm
  - Small Reynolds number

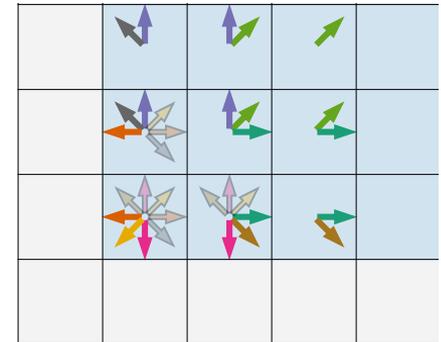LBM: $$\underbrace{f_i(\mathbf{x} + \mathbf{e}_i\Delta t, t + \Delta t) = f_i(\mathbf{x}, t)}_{\text{Streaming}} - \underbrace{\frac{1}{\tau}(f_i(\mathbf{x}, t) - f_i^{eq}(\mathbf{x}, t))}_{\text{Collision}}$$

Macroscopic Variables: $$\rho = \sum_{i=0}^{N} f_i, \quad \mathbf{u} = \frac{1}{\rho}\sum_{i=0}^{N} f_i\mathbf{e}_i, \quad N = 8$$

Equilibrium: $$f_i^{eq} = \rho w_i\left(1 + \frac{3}{c^2}\mathbf{e}_i\cdot\mathbf{u}^{eq} + \frac{9}{2c^4}(\mathbf{e}_i\cdot\mathbf{u}^{eq})^2 - \frac{3}{2c^2}\mathbf{u}^{eq}\cdot\mathbf{u}^{eq}\right)$$
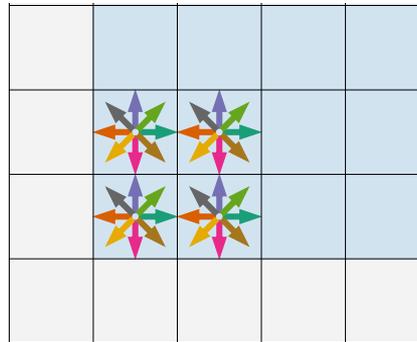
Pre-streaming
$f_i(x, t)$

Post-streaming
$f_i(x + \mathbf{e}_i\Delta t, t + \Delta t)$

Approach: Multiphase

- Use a **single-component multi-phase (SCMP)** Shan-Chen formulation [1, 2]

- Shifted velocity, u^{eq}, in equilibrium distribution calculation captures inter-particle forces
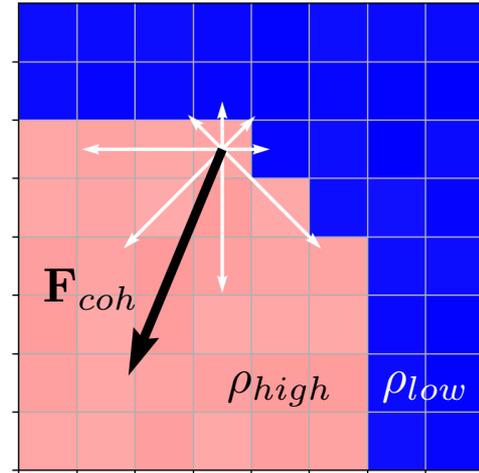
$$\mathbf{u}^{eq} = \mathbf{u} + \frac{\tau \mathbf{F}_{total}}{\rho}$$

- Total force is combined effect of inter-particle **cohesion**, fluid-wall **adhesion**, others

$$\mathbf{F}_{total} = \mathbf{F}_{coh} + \mathbf{F}_{ad} + \mathbf{F}_{grav} + \dots$$

$$\mathbf{F}_{coh} = -G_{coh}\psi(\mathbf{x}, \mathbf{t}) \sum_{i=0}^{N} w_i \psi(\mathbf{x} + \mathbf{e}_i \Delta t, t) \mathbf{e}_i$$

$$\mathbf{F}_{ad} = -G_{ad}\rho(\mathbf{x}, \mathbf{t}) \sum_{i=0}^{N} w_i s(\mathbf{x} + \mathbf{e}_i \Delta t, t) \mathbf{e}_i$$

$\mathbf{F}_{coh}$ ρ_{high} ρ_{low}

Cohesion force near sharp density change maintains this coarse droplet edge

1. X. Shan and H. Chen "Simulation of nonideal gases and liquid-gas phase transitions by the lattice Boltzmann equation", Physical Review E, 1994
2. H. Huang, M. C. Sukop, and X. Lu, "Multiphase lattice Boltzmann methods: Theory and application", 2015

Approach: Multiphase

- Equation of state, ψ, sets inter-particle potentials according to a pressure/temperature relationship

$$\psi = \sqrt{\frac{2(p - c_s^2 \rho)}{c_0 G_{coh}}}$$

- Carnahan-Starling equation of state used to achieve high density ratio typical of liquid gas systems [1, 2]

$$p = \rho RT \frac{1 + b\rho/4 + (b\rho/4)^2 - (b\rho/4)^3}{(1 - b\rho/4)^3} - a\rho^2$$

Values used in current study
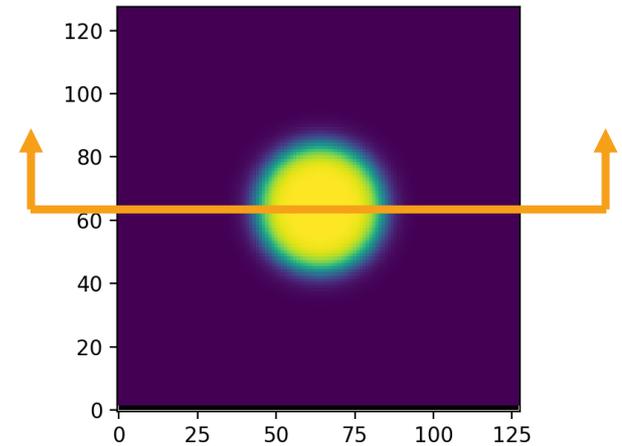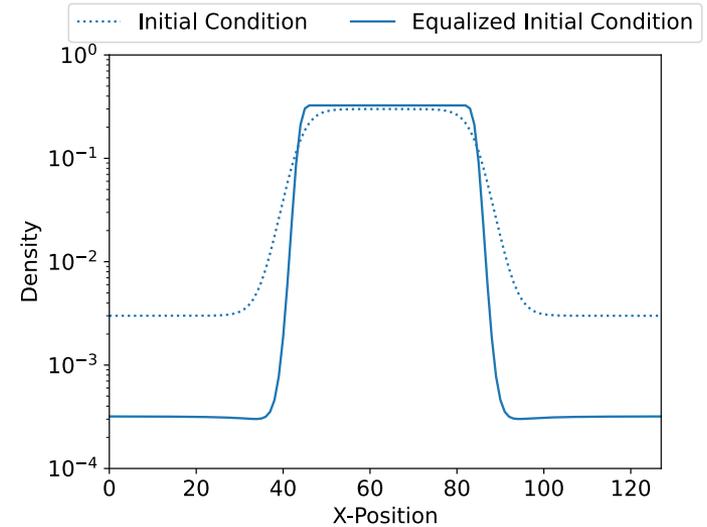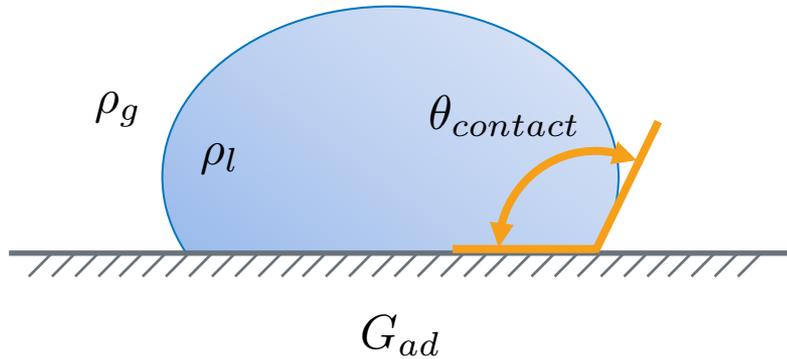a = 1
b = 4
R = 1
T = 0.167
C_0 = 6
$C_s = 1/\sqrt{3}$

1. N. F. Carnahan and K. E. Starling, "Equation of state for nonattracting rigid spheres", The Journal of Chemical Physics, 1969
2. P. Yuan and L. Schaefer, "Equations of state in a lattice Boltzmann model", Physics of Fluids, 2006

Verification: Drops

Goal: Simulate a high-density drop falling onto a surface

Verification: Drops

Vary hydrophilicity by adjusting adhesion parameter, G_{ad}

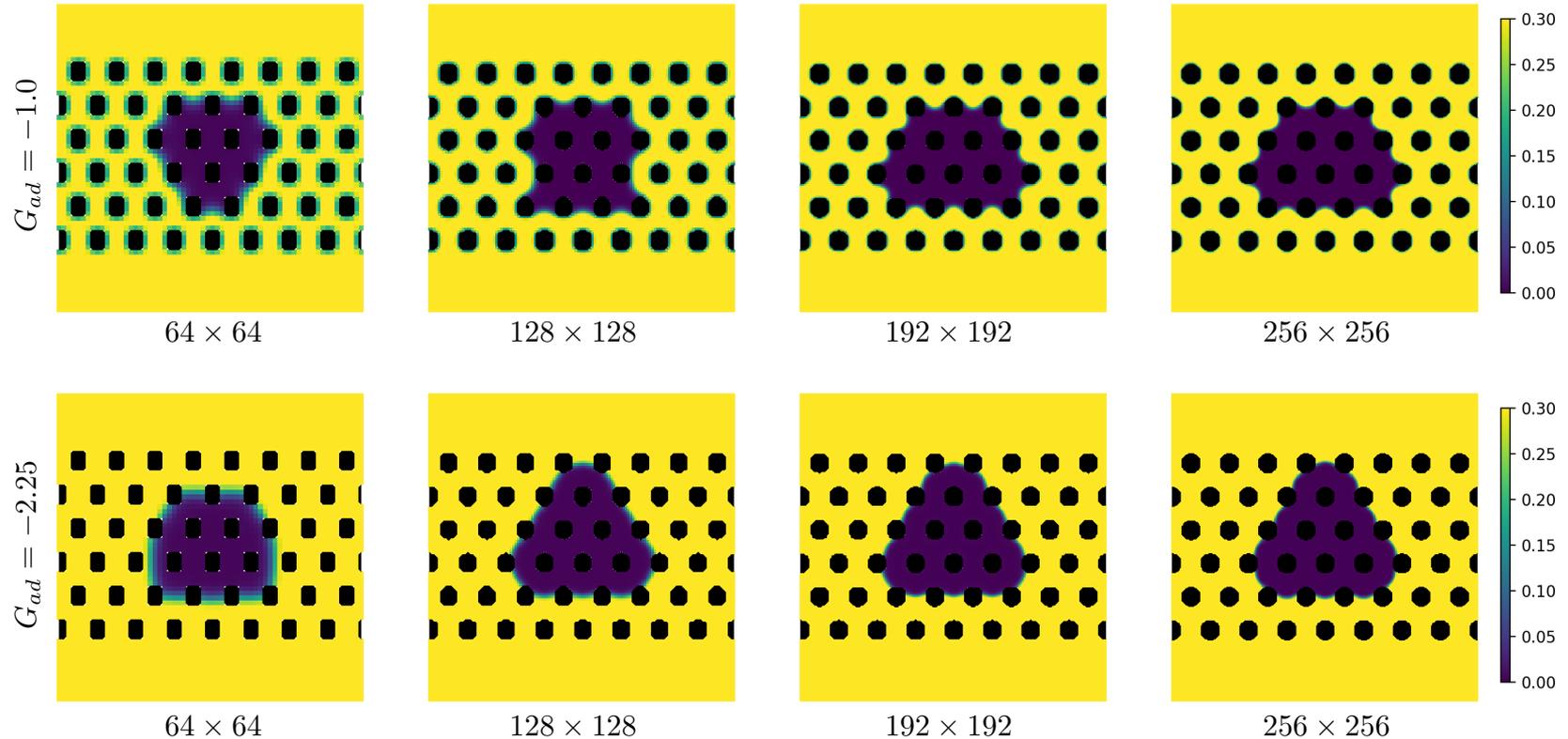$G_{ad} = -1.0$ $G_{ad} = -1.5$ $G_{ad} = -2.0$ $G_{ad} = -2.25$
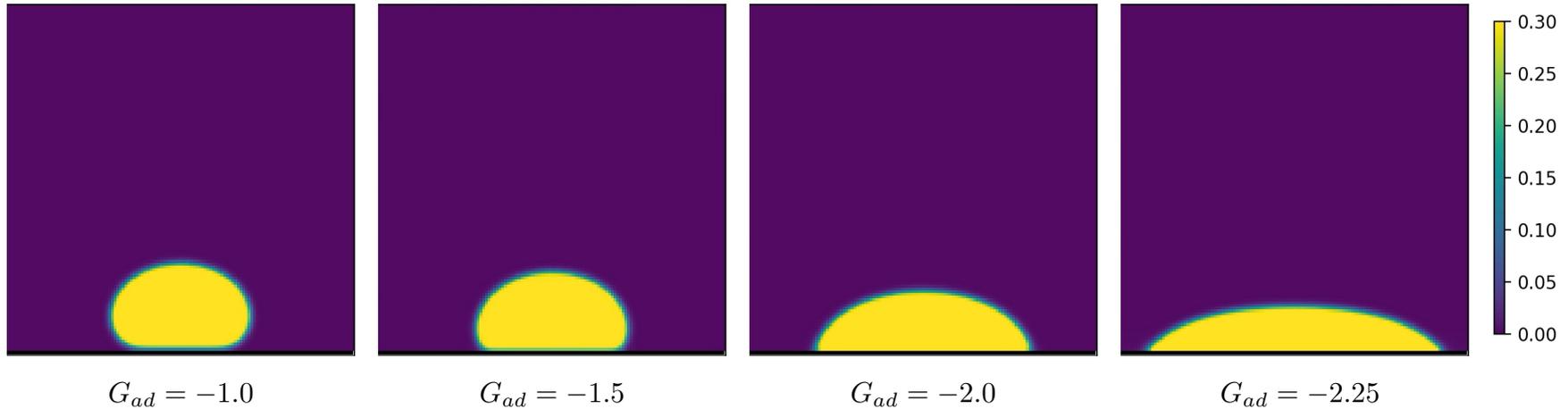
Verification: Vapor Invasion

Goal: Predict the final state of a bubble trapped by an array of solids

Verification: Vapor Invasion

$G_{ad} = -1.0$

64×64 128×128 192×192 256×256

$G_{ad} = -2.25$

64×64 128×128 192×192 256×256

Verification: Vapor Invasion

$G_{ad} = -1.0$
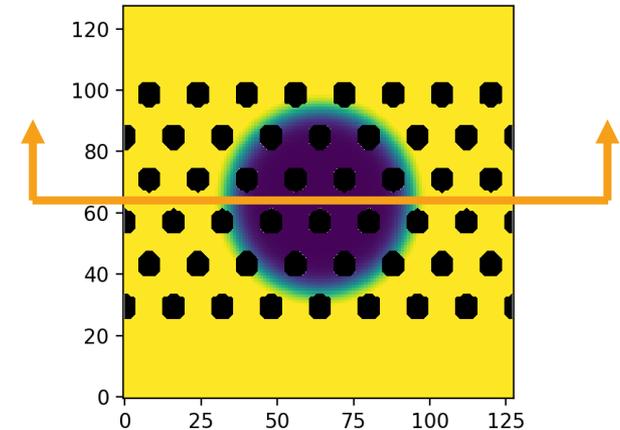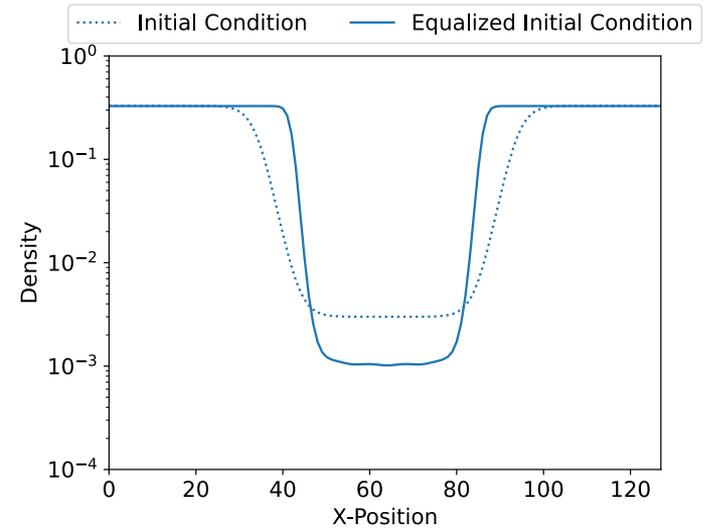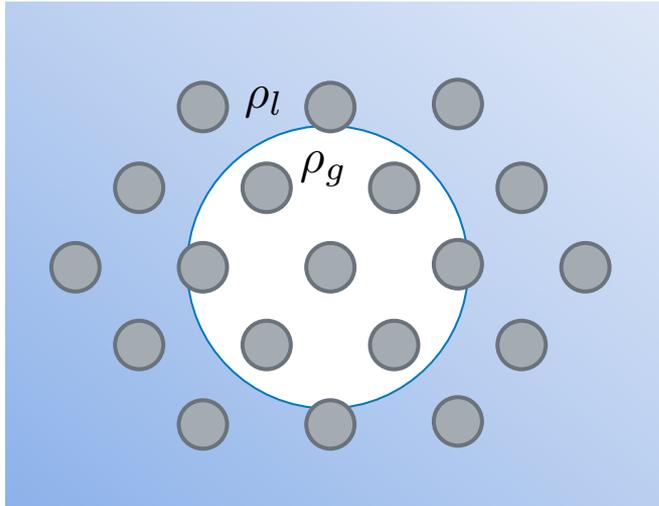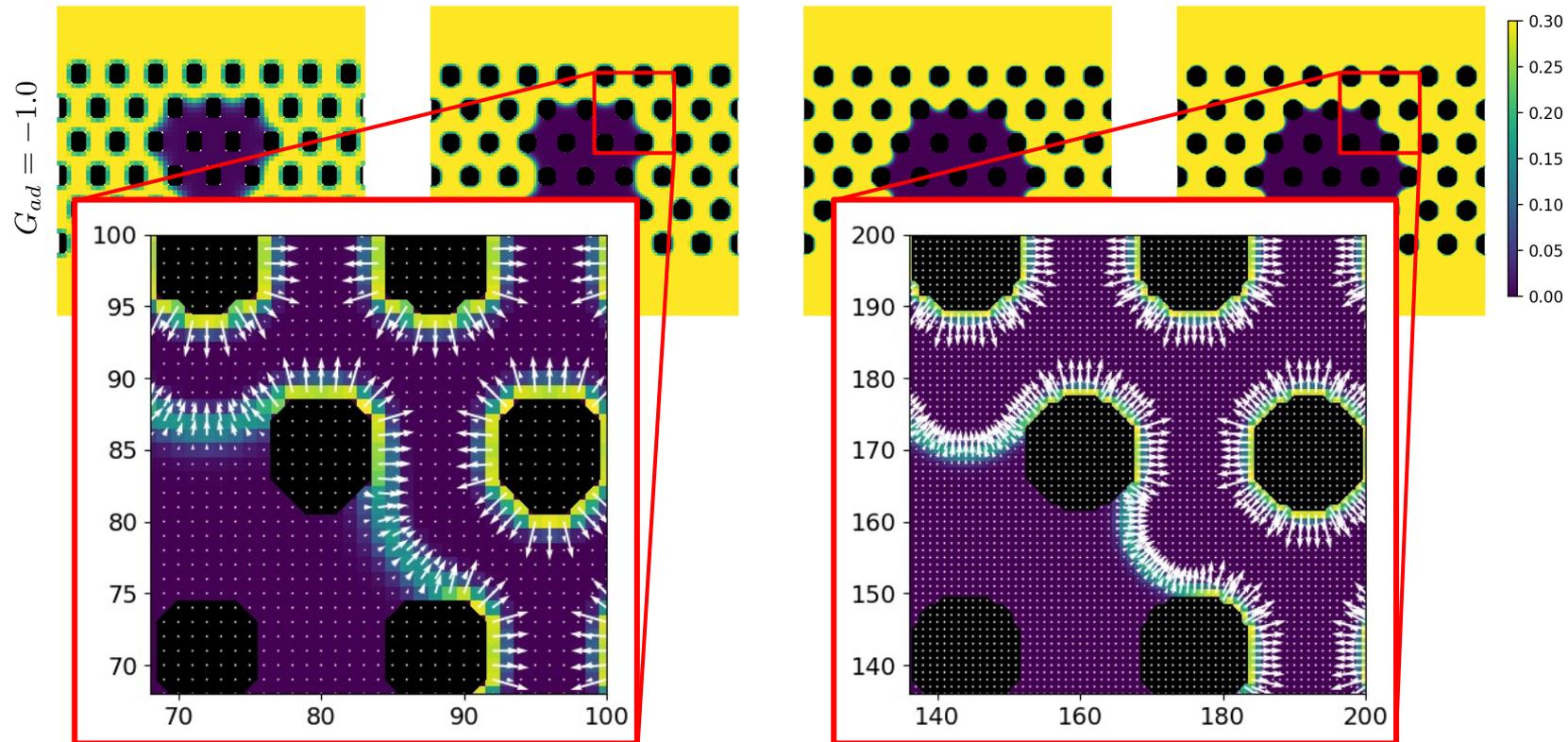
Progress on PTL Result

Goal: Preliminary simulation of buoyancy-driven O_2 bubbles interacting with PTL and water

Physical Details

$$D = 250 \ \mu m$$

$$V = 1 \ mm/s$$

$$\rho_l \approx 1000 \ kg/m^3$$

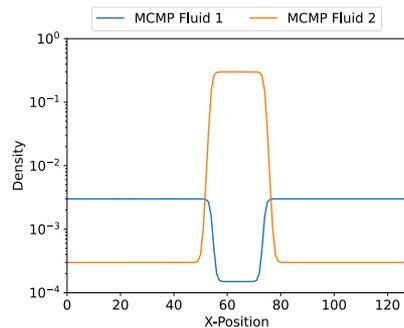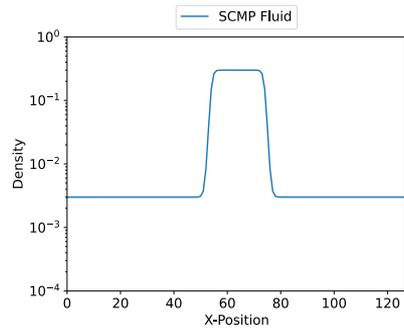$$\rho_g \approx 1 \ kg/m^3$$

$$\text{Re} \approx 0.25$$
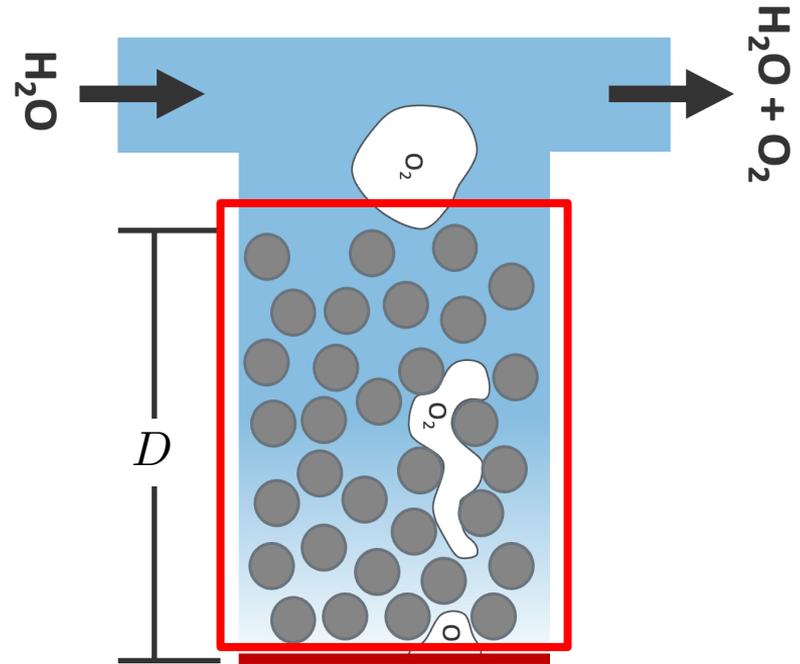
Progress on PTL Result

Current simulation captures buoyancy effect, uses periodic boundaries to "generate" new bubble at catalyst
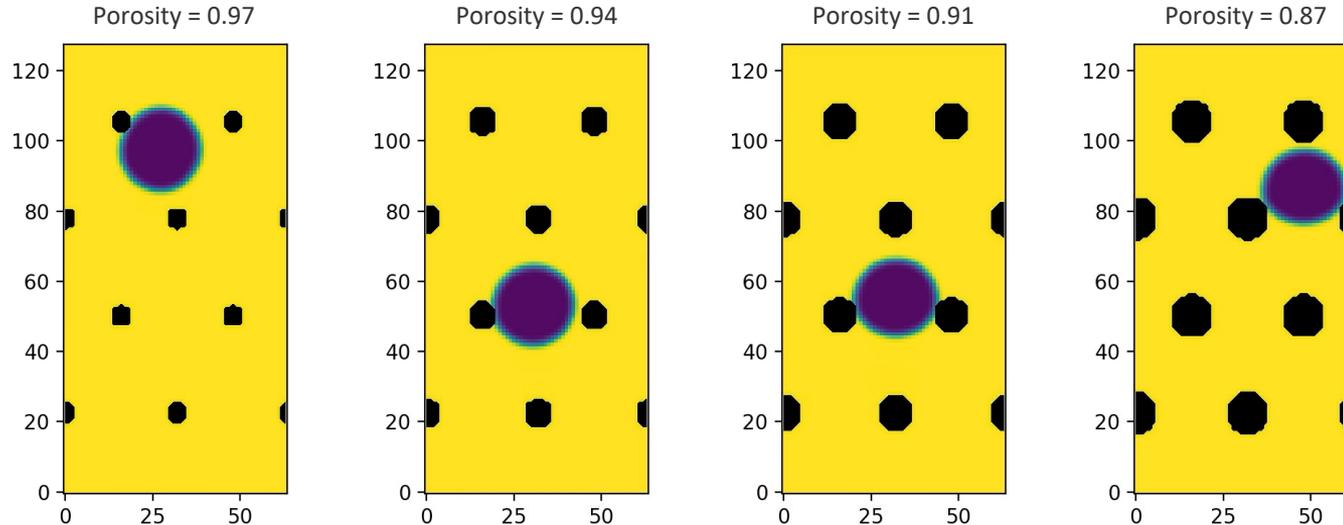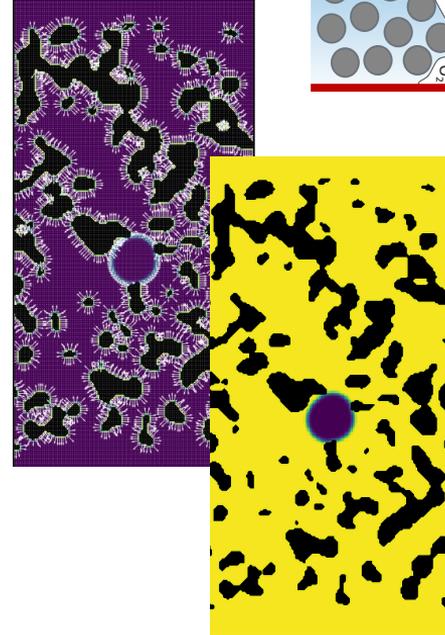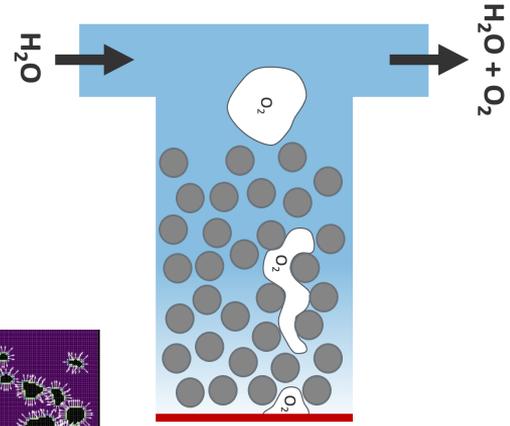
Progress on PTL Result

To enable setting fluid properties for water and air separately, we are testing a **multi-component multi-phase (MCMP)** implementation
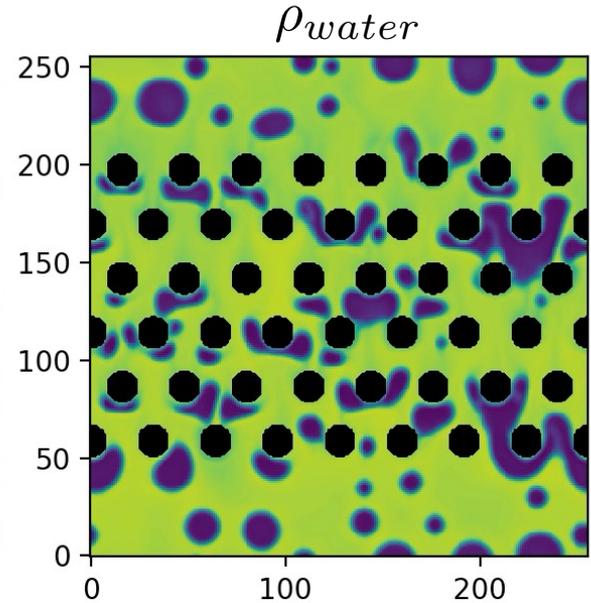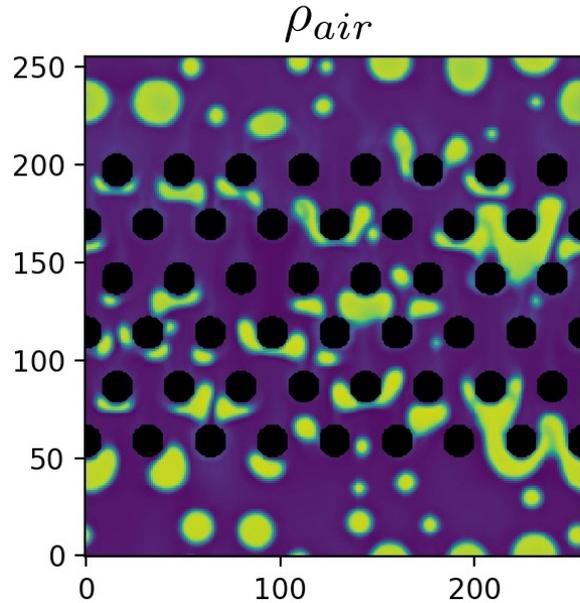
Future Work

- Early in project, but we have the **fundamental pieces to solve the PTL problem in place**

- Must develop better **bubble nucleation, growth, release BCs** at anodic catalyst and outflow

- Include experimentally-imaged PTL geometries

- Move simulations to **adaptive mesh refinement, d3q27 LBM code**

Thank You

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