Novel Geometry Design for Intensified CO_2 Absorbers

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Highlights

Computationally generated TPMS structures on CAD files

CFD models capturing fluid dynamics for countercurrent gas/solvent flow in TPMS geometries

- Model validation using random rings packing

Results on hydrodynamics of TPMS geometries
- Blockage of flow
- Effects of viscosity and liquid flowrate on interfacial and wetted area

Reactive systems for CO_2 absorption on CFD models

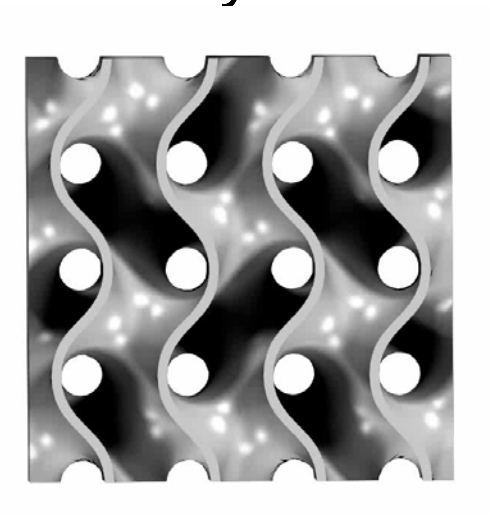
Introduction: Triply Periodic Minimal Surfaces (TPMS Structure)

- **Advantages of TPMS**
  - Improved heat transfer (~10X)
  - Separate independent flow channels

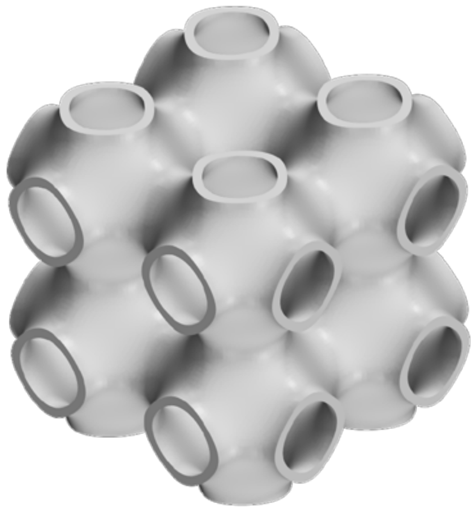
- **For carbon capture**
  - Mass transfer coefficients?
  - Mass transfer areas?
  - Highly viscous solvent?

- **Objective: CFD modeling for the gas/solvent flow in TPMS**

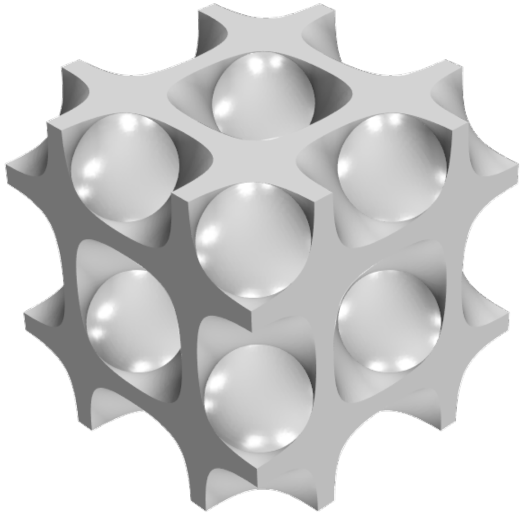
Gyroid

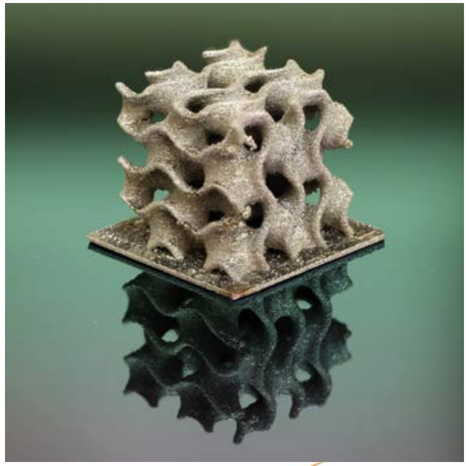


Schwarz-P



Schwarz-D



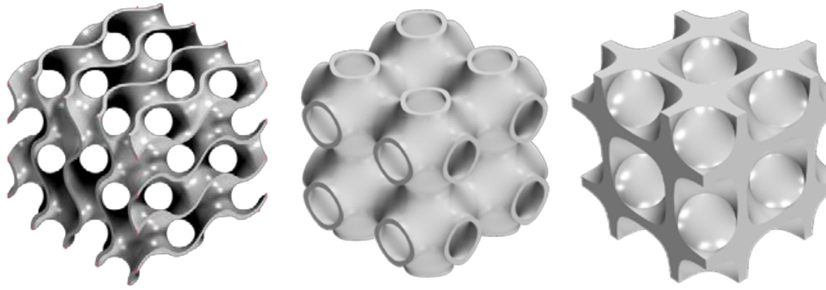


3D printed Gyroid @ LLNL

Introduction: Triply Periodic Minimal Surfaces (TPMS Structure)

- **Mathematical Description**
  - Gyroid: $F(x,y,z) = \cos(x)\sin(y) + \cos(y)\sin(z) + \cos(z)\sin(x) = 0$
  - Schwarz-P: $F(x,y,z) = \cos(x) + \cos(y) + \cos(z) = 0$
  - Schwarz-D: $F(x,y,z) = \cos(x)\cos(y)\cos(x) - \sin(x)\sin(y)\sin(z) = 0$
  - Offset to create wall surface
    - $F_{offset}(x,y,z) = F\left(x \pm a_x, y \pm a_y, z \pm a_z\right)$

$$a_x = t\frac{F_x}{\sqrt{F_x^2+F_y^2+F_z^2}}, a_y = t\frac{F_y}{\sqrt{F_x^2+F_y^2+F_z^2}}, a_z = t\frac{F_z}{\sqrt{F_x^2+F_y^2+F_z^2}}$$



- **Geometrical Parameters**

	Geometry size [cm]	Repeated Units	Wall Thickness [mm]	Porosity ϵ [%]	Surface Area A_p $[\mathrm{mm}^2]$	Specific Area a_p $[\mathrm{m}^2/\mathrm{m}^3]$	Hydraulic Diameter d_h [mm]
Gyroid (**G**)	2x2x2	8	0.45	0.87	4910.08	613.76	5.68
Schwarz-P (**P**)	2x2x2	8	0.49	0.89	3979.04	497.38	7.18
Schwarz-D (**D**)	2x2x2	8	1.30	0.77	2993.04	374.13	8.21

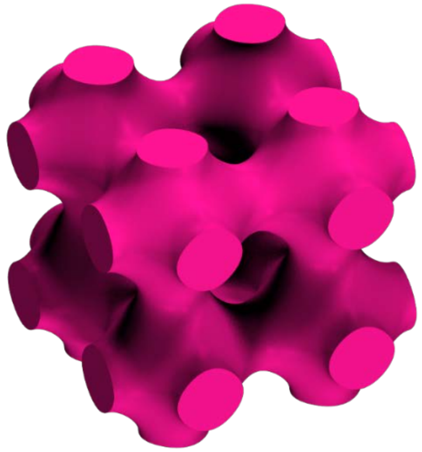
Model Setup: Geometry

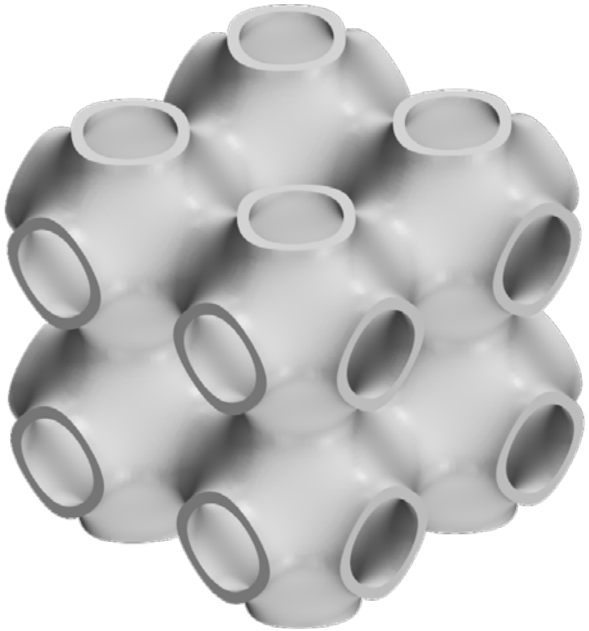
Schwarz-P geometry

- 2cm X 2cm X 2cm
- Periodic in 3 directions

- Two independent channel
- Self-similar structure

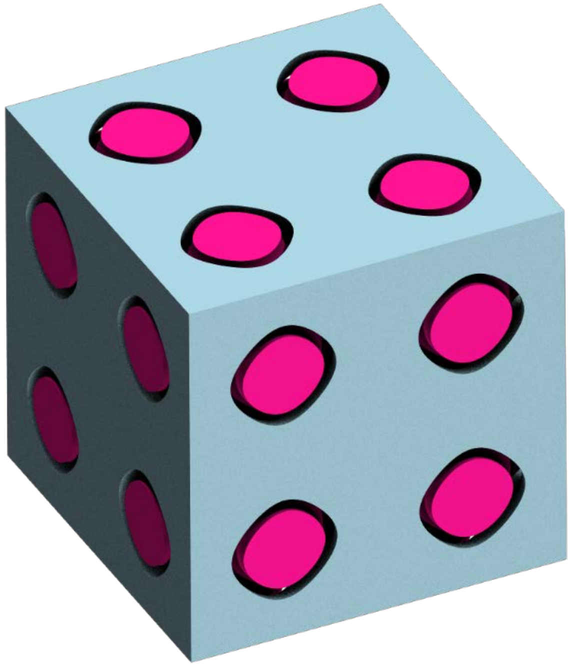
Channel 1





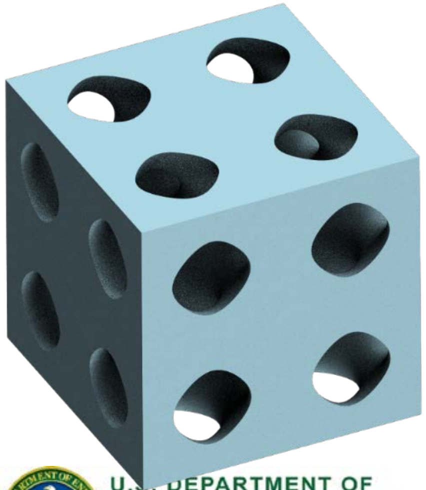
Geometry Wall

Subtract Wall



Flow Region

Channel 2

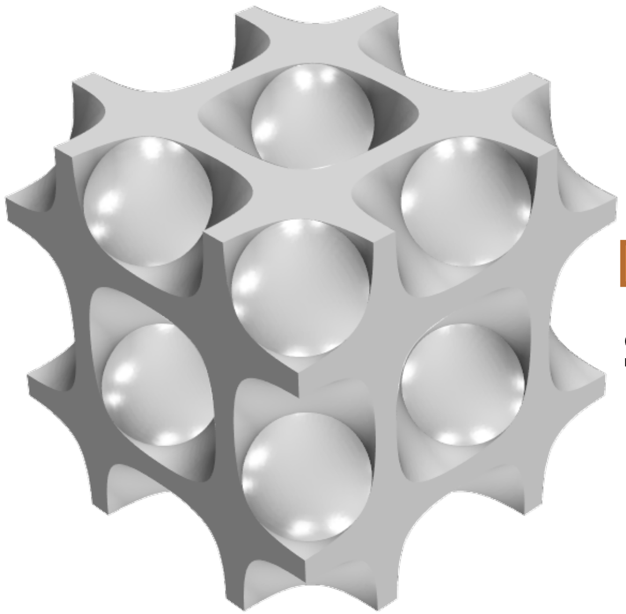


Model Setup: Geometry

Schwarz-D geometry

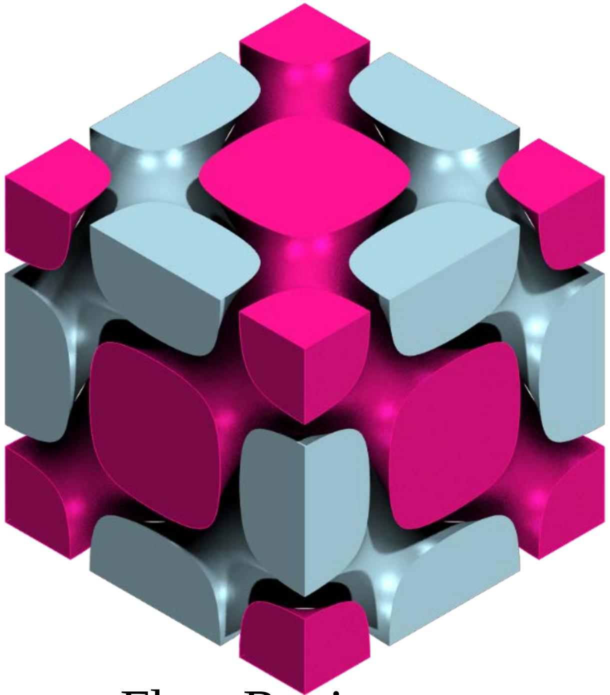
- 2cm X 2cm X 2cm
- Periodic in 3 directions

- Two independent channel
- Self-similar structure



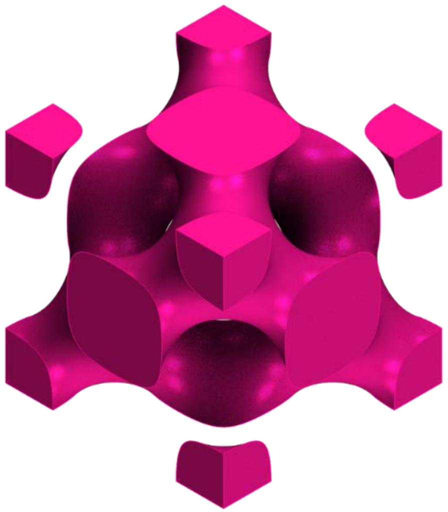
Geometry Wall

Subtract Wall

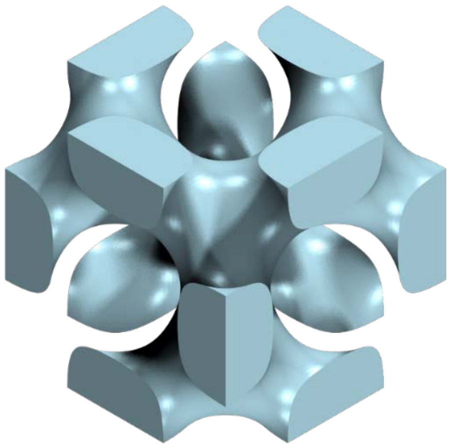


Flow Region

Channel 1



Channel 2

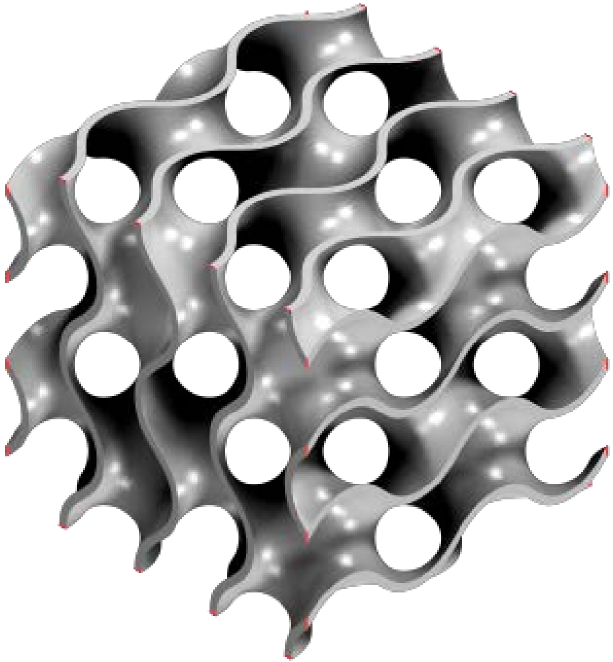


Model Setup: Geometry

Gyroid geometry

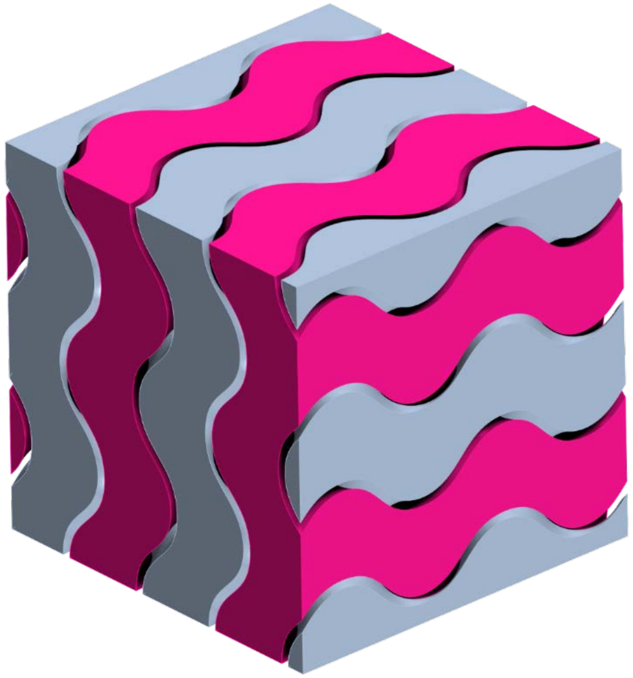
- 2cm X 2cm X 2cm
- Periodic in 3 directions

- Two independent channel
- Self-similar structure



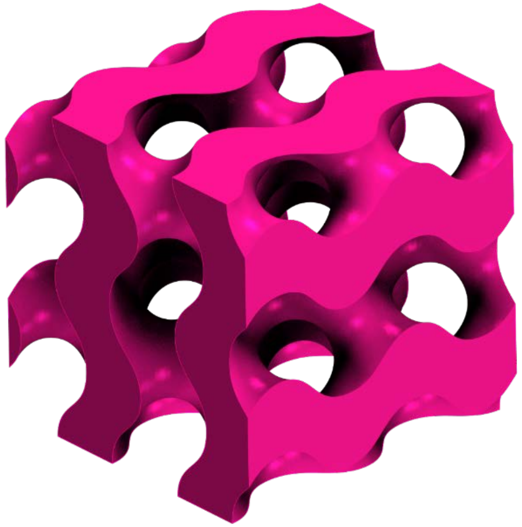
Geometry Wall

Subtract Wall

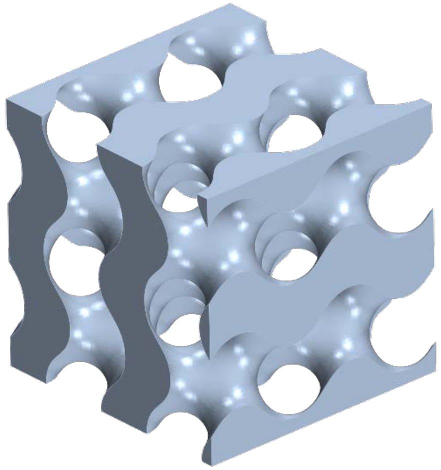


Flow Region

Channel 1



Channel 2



Model Setup: Gas/solvent Countercurrent Flow

Boundary conditions

- Periodic for flow

Initial conditions

- Solid initially wrapped with a thin layer of film (0.5-1.5mm)
- Initial thickness affects the final liquid flow rate

Body force

- Solvent driven by the gravity
- Gas driven by body force

Computational time

- 96 cores on PNNL PIC HPC
- 7-8 CPU hours for every 1s solution

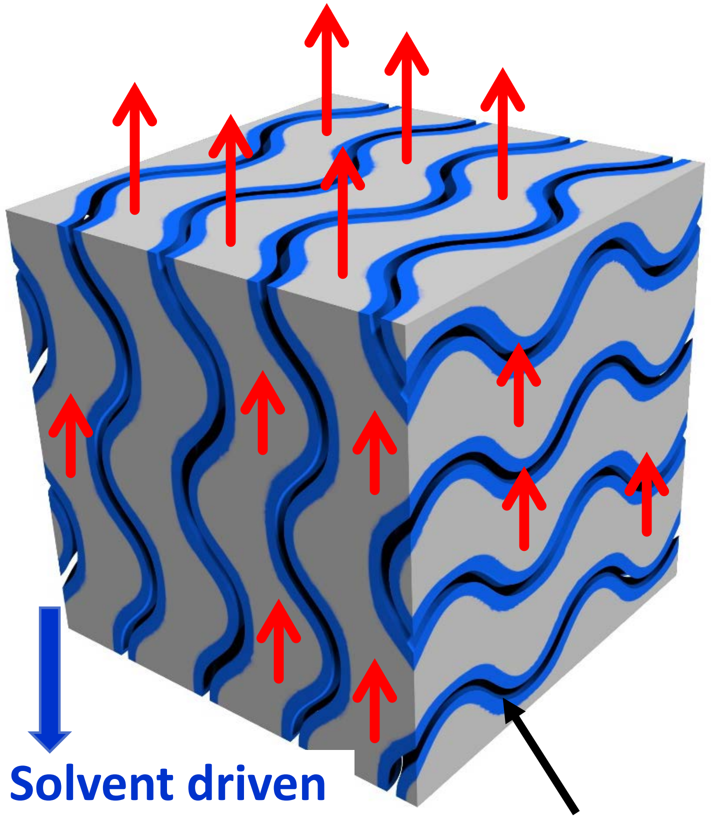
- Gas flow rate ~0.33m/s

Solvent Properties (30% MEA)

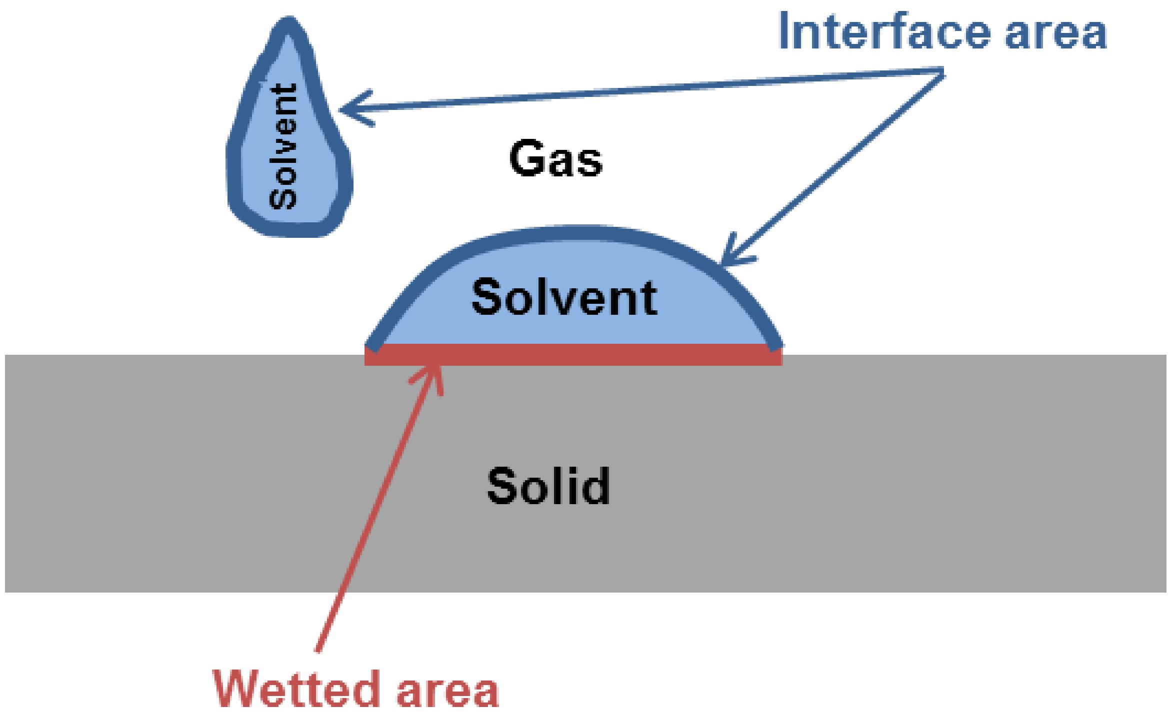
Physical Properties	
Density ρ (kg/m^3)	1000
Viscosity μ (cP)	2.5, 5, 10, 25
$D_{CO_2}[l]$ (m^2/s)	1.0×10^{-9}
Surface Tension (N/m)	0.065
Contact angle (°)	40

Gas Properties (Air)

Physical Properties	
Density ρ (kg/m^3)	1.184
Viscosity μ (cP)	0.0186
$D_{CO_2}[g]$ (m^2/s)	1.0×10^{-5}



Area Definitions



A_i: Interfacial Area. Area between Gas and Liquid
A_w: Wetted Area. Area between Solid and Liquid
A_p: Geometrical Area available between Solid and Liquid

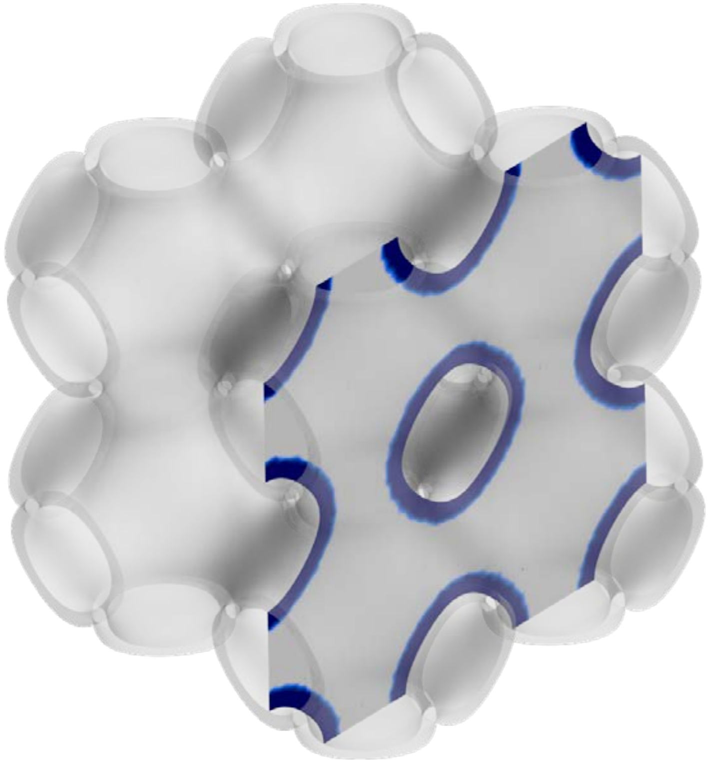
Fractional Area: A_i/A_p

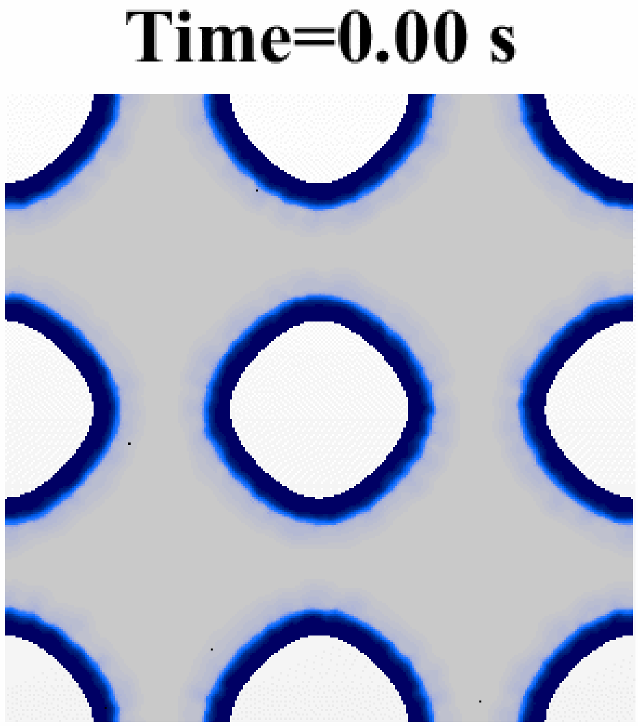
Schwarz-P Results-Flow Visualization

Counter Current Flow Visualization

- Modified Schwarz-P geometry (4 cm)
- Simulation time T=1s
- Liquid Load

Cross Section Visualization





Schwarz-P Results-Flow Visualization

<u>Counter Current Flow Visualization</u>

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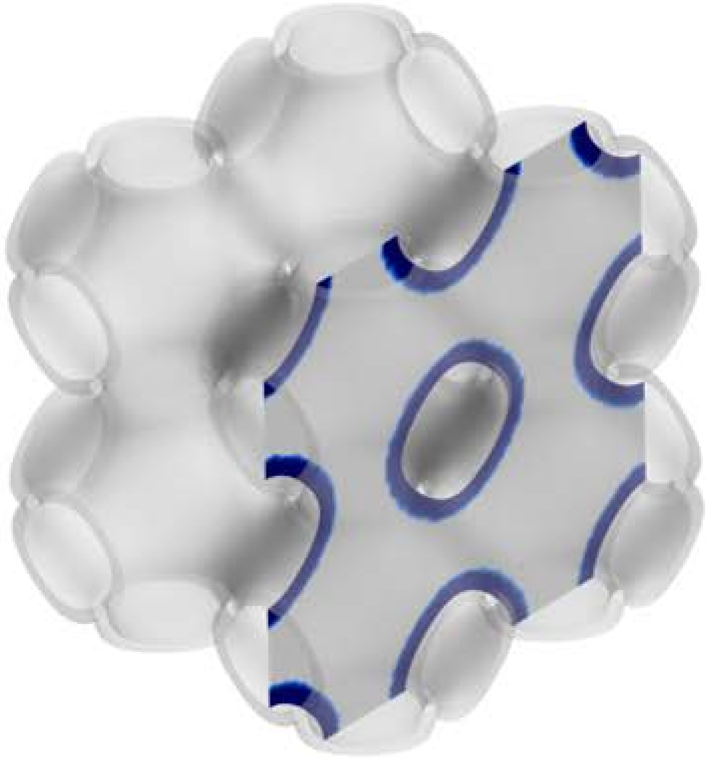
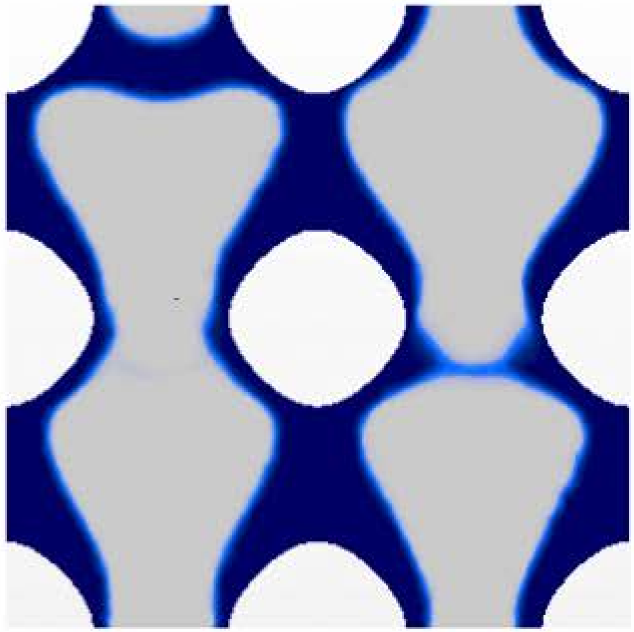
Cross Section Visualization



Time=0.55 s



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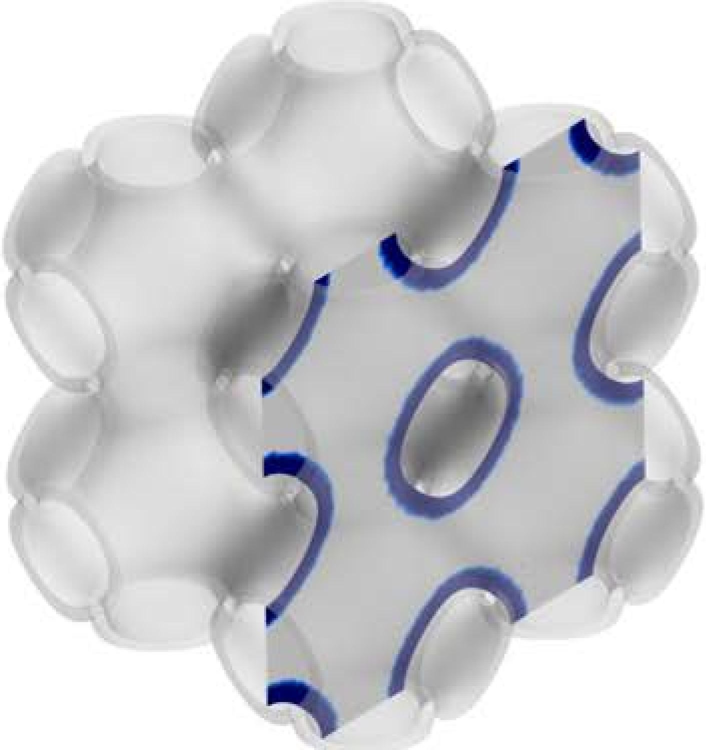
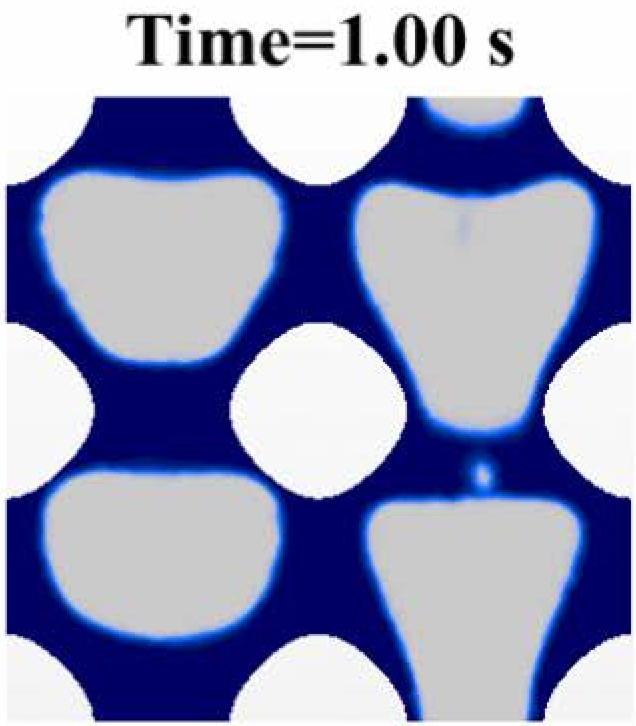


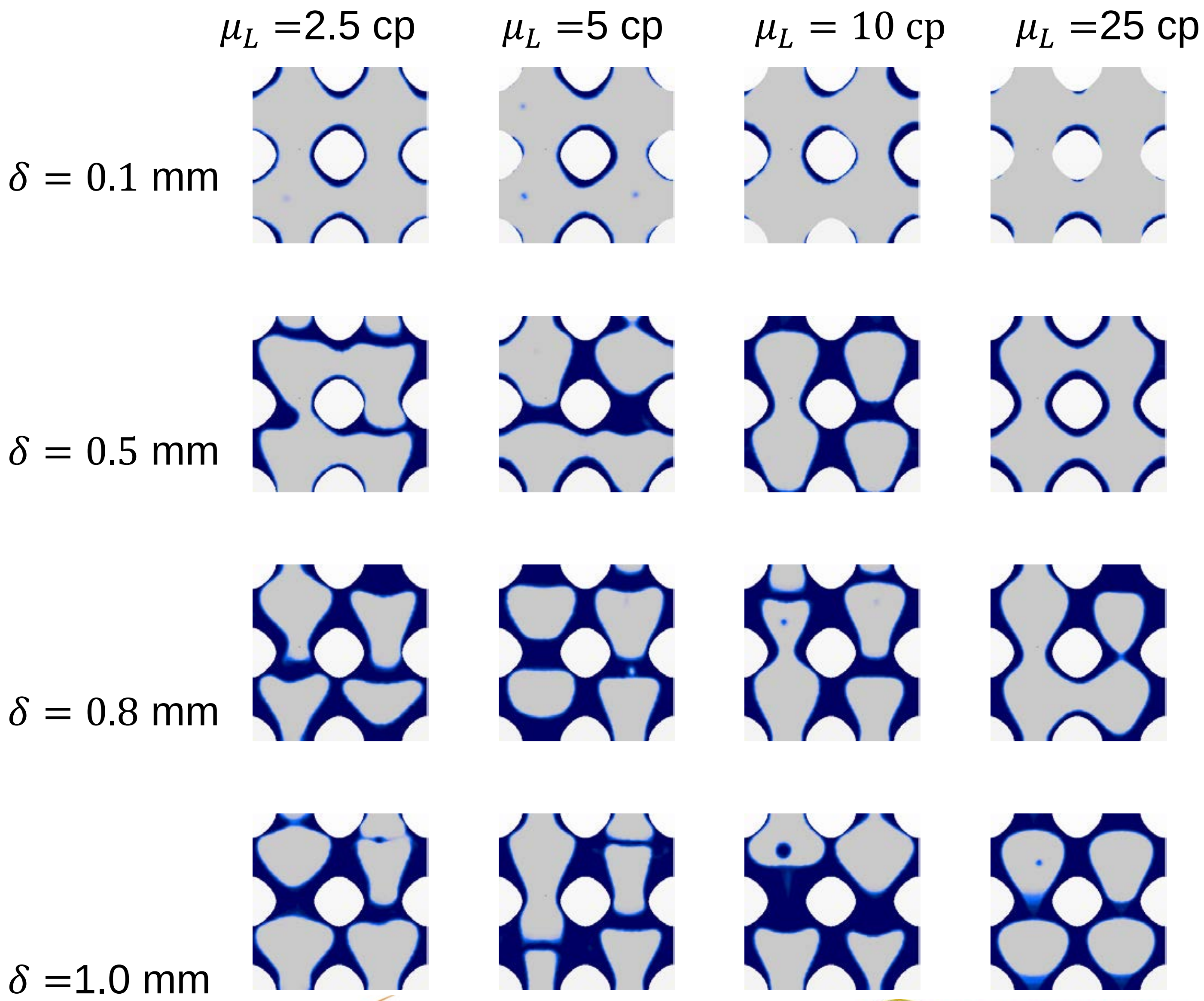


Schwarz-P Flow Visualization

Geometry Size:
2 cm

Initial Liquid Film Thickness δ
0.1, 0.5, 0.8, 1.0 mm

Cross-Section View at T=1s



Schwarz-P Flow Visualization

Geometry Size:
4 cm

Initial Liquid Film Thickness $\boldsymbol{\delta}$
0.2, 0.5, 1, 1.5 1.8 mm

Cross-Section View at T=1s

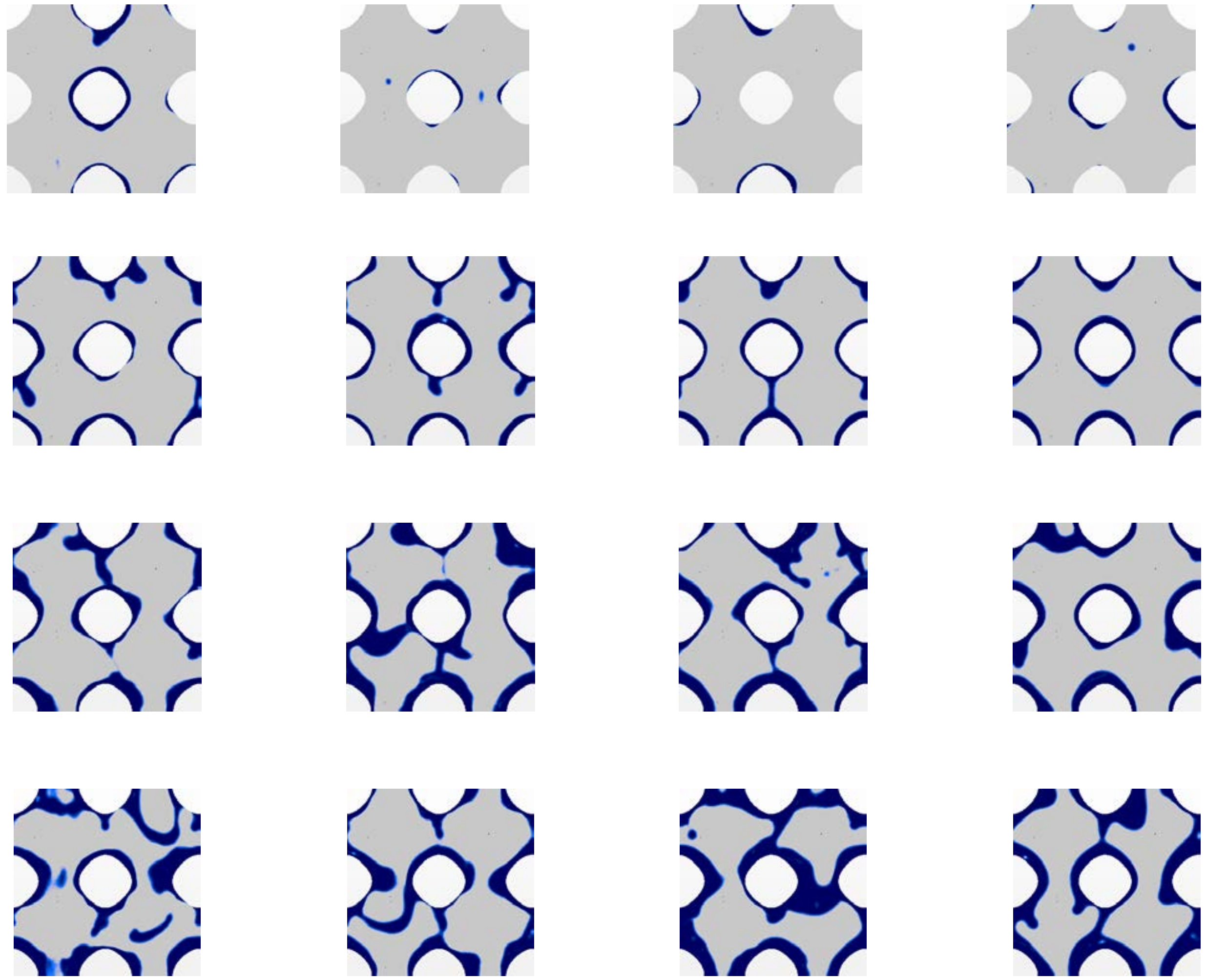
μ_L =2.5 cp | μ_L =5 cp | $\mu_L = 10$ cp | μ_L =25 cp

$\delta = 0.2$ mm
$\alpha_L = 2.3\%$

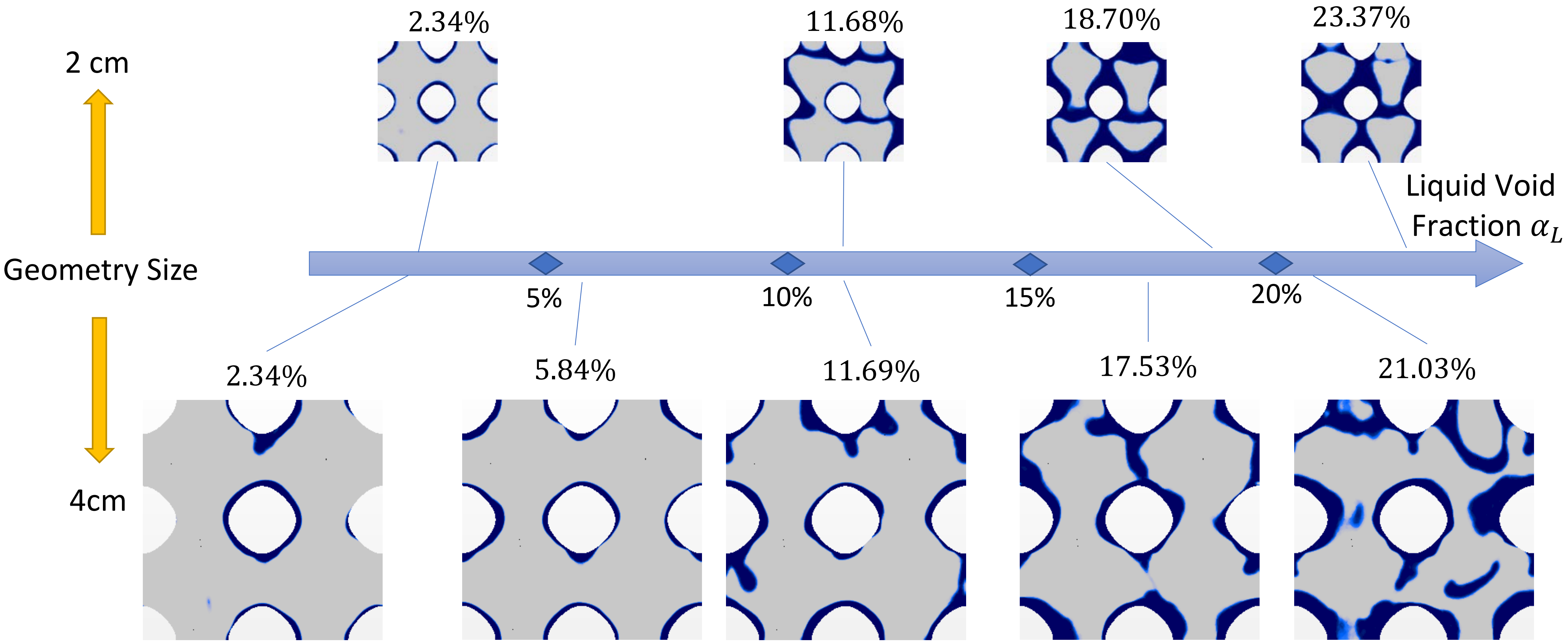
$\delta = 1.0$ mm
$\alpha_L = 11.69\%$

$\delta = 1.5$ mm
$\alpha_L = 17.53\%$

$\delta = 1.8$ mm
$\alpha_L = 21.03\%$



Schwarz-P Geometry Size Effect on Flow

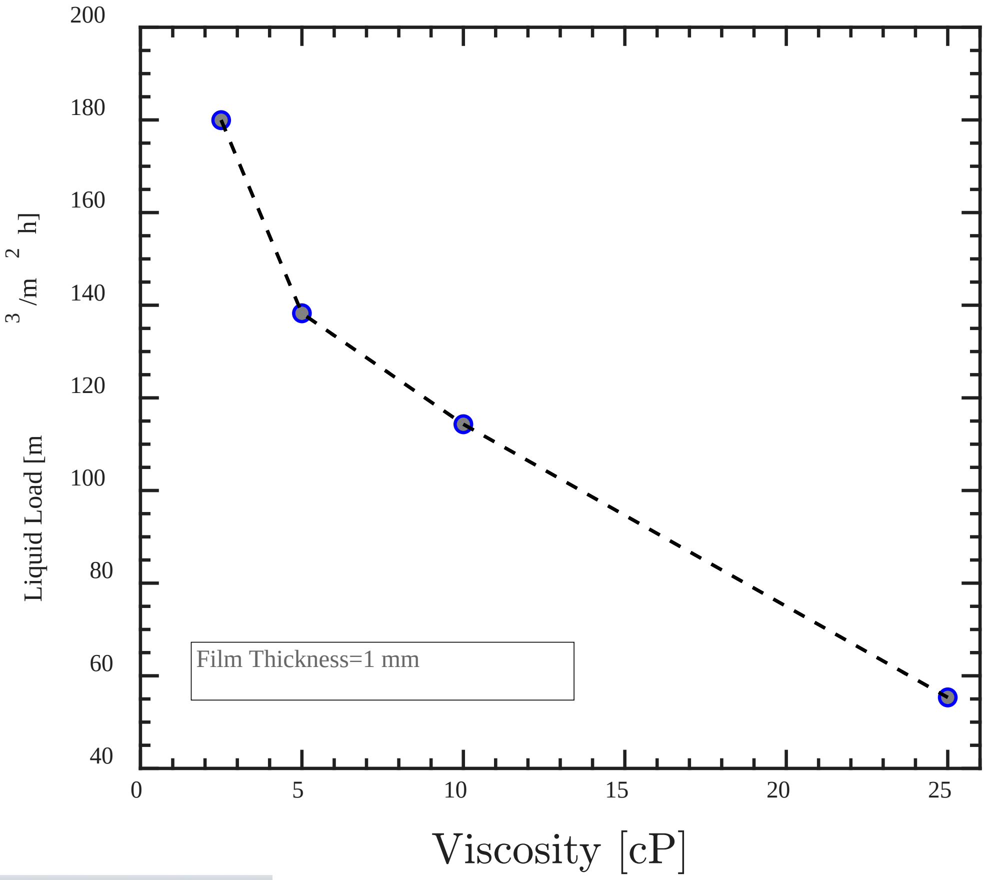


Viscosity: μ_L =2.5 cp, Cross-Section View at T=1s

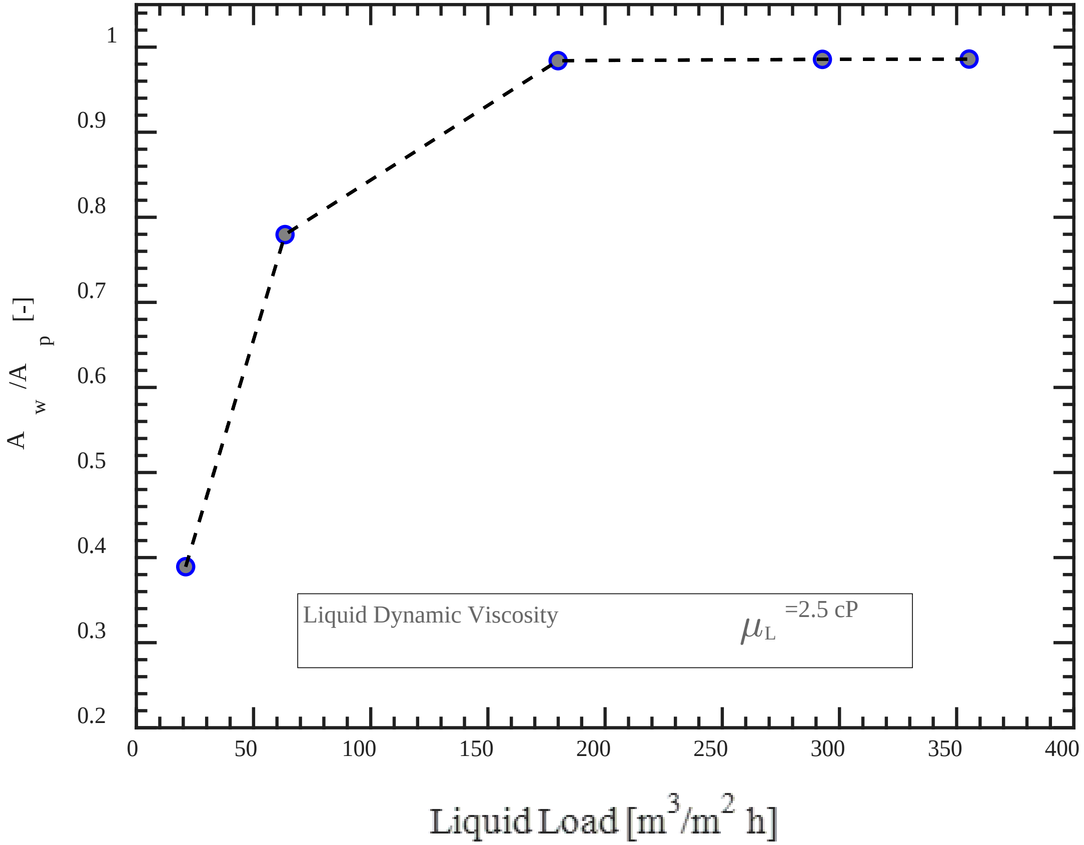
Schwarz-P Results and discussion

Modified Schwarz-P geometry (4 cm)

Flow Rate with Various **Viscosity**



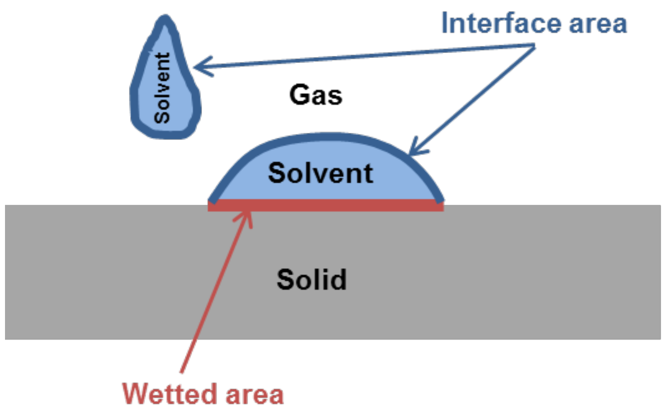
Wetted Area with Various **Liquid Load**

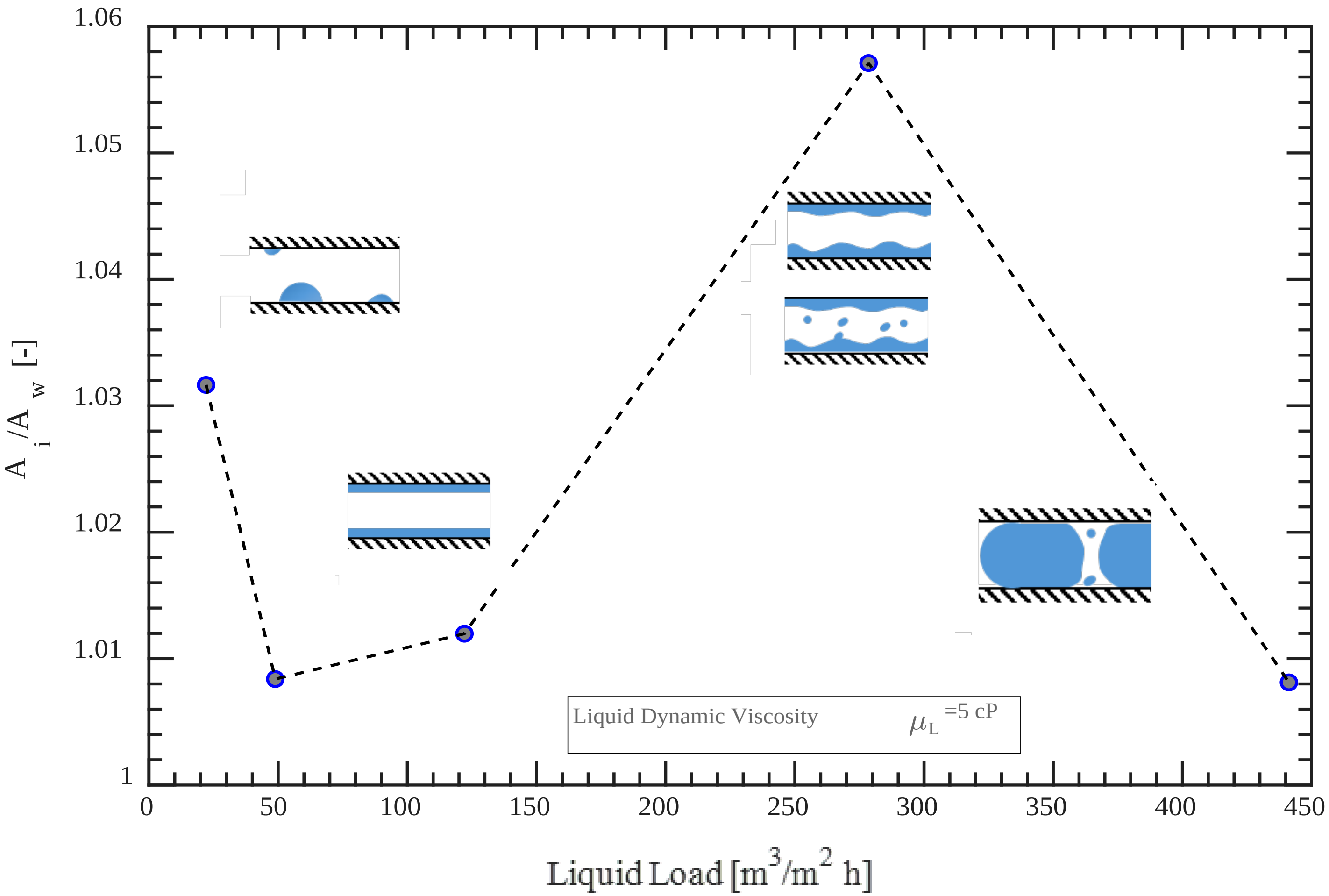


Schwarz-P: Effect of Liquid Load on Regimes of Fluid Flow

Modified Schwarz-P geometry (4 cm)

- Interfacial area and Wetted Area Ratio
  - Rivulet
  - Uniform liquid film
  - Wavy film with free streams
  - Bulky fluids blockage

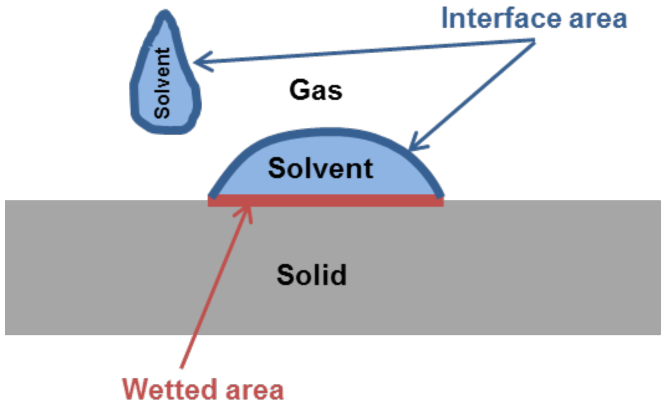


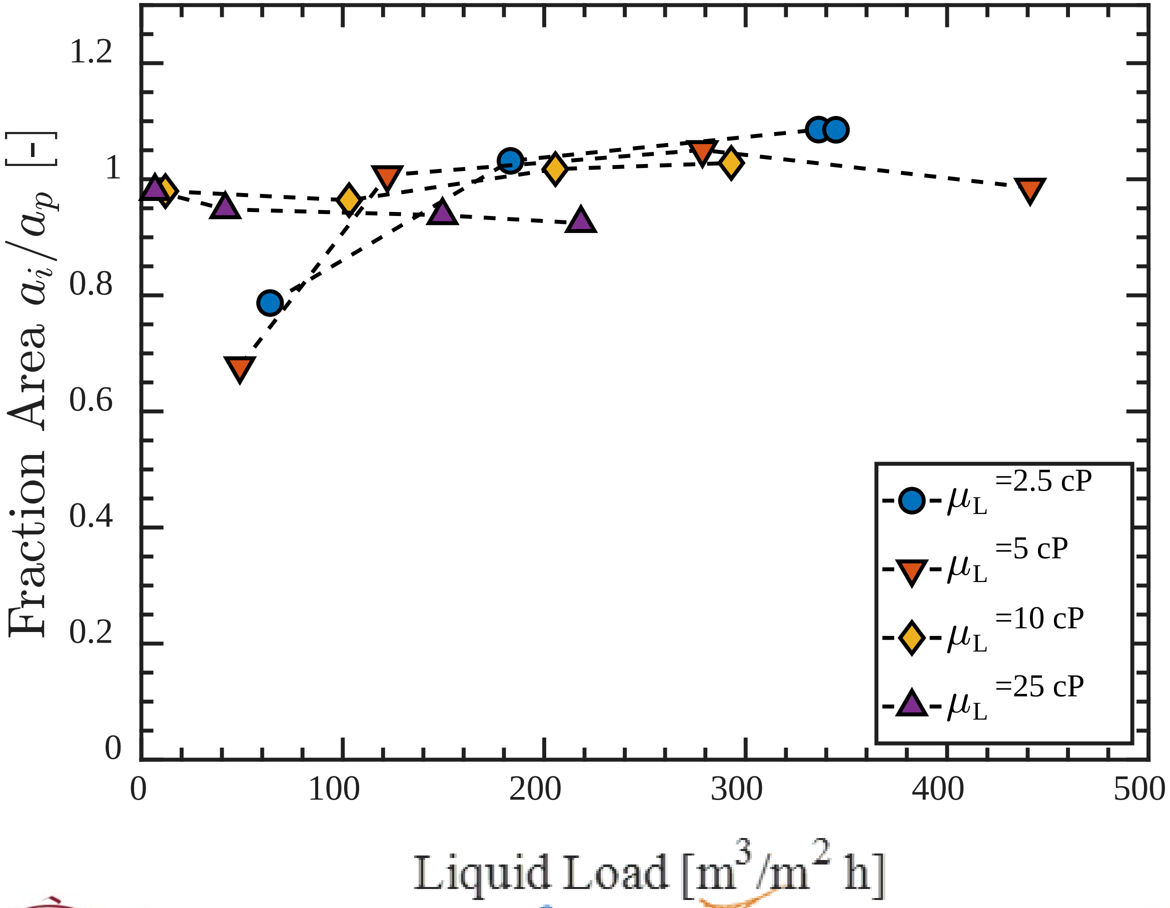


Schwarz-P: Effect of Liquid Load on Fractional Area

Modified Schwarz-P geometry (4 cm)

− Fractional Area with different Viscosity and liquid load





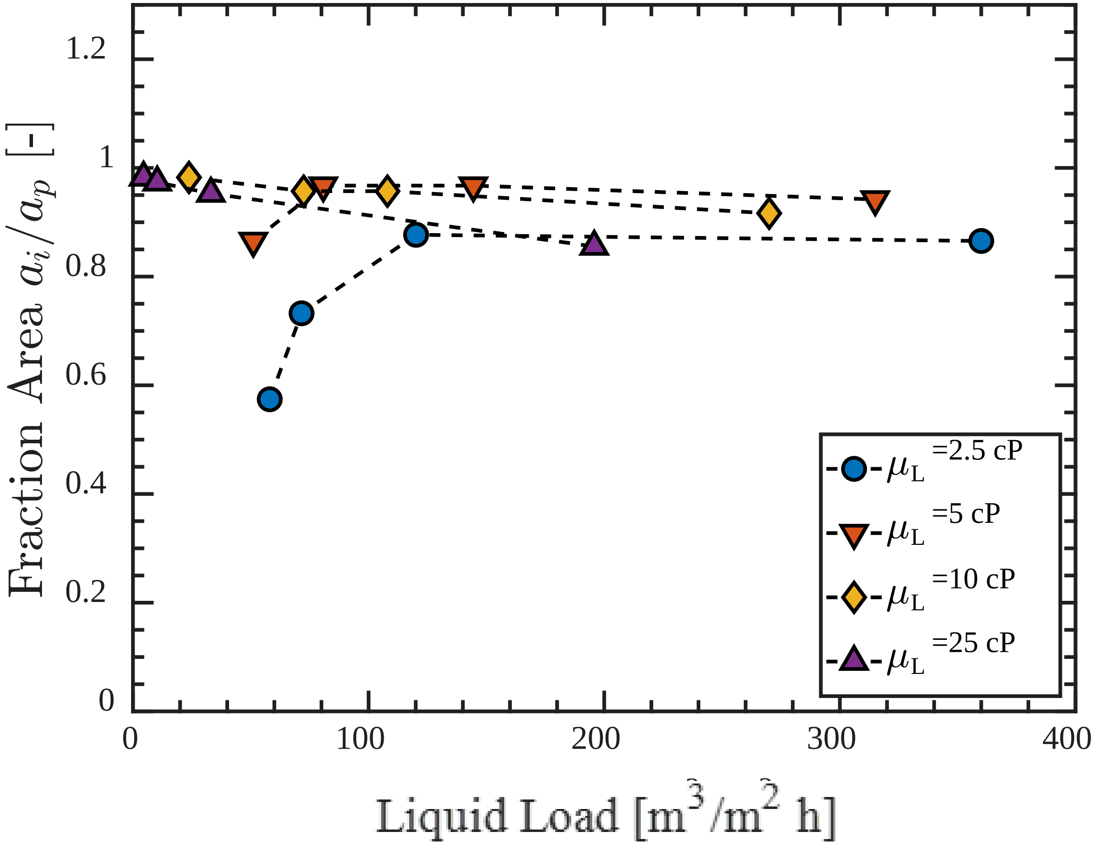
Gyroid: Effect of Liquid Load on Fractional Area

Modified gyroid geometry

- 4cm X 4cm X 4cm
- Channel size: ~9 mm
- Surface to volume ratio: 307 1/m
- Viscosity: 2.5, 5, 10, 25 cp

Findings:

- For viscosity <= 5cp, interface area increases with flow rate (rivulet flow regime)
- For viscosity >=10cp, constant interface area (film flow regime)

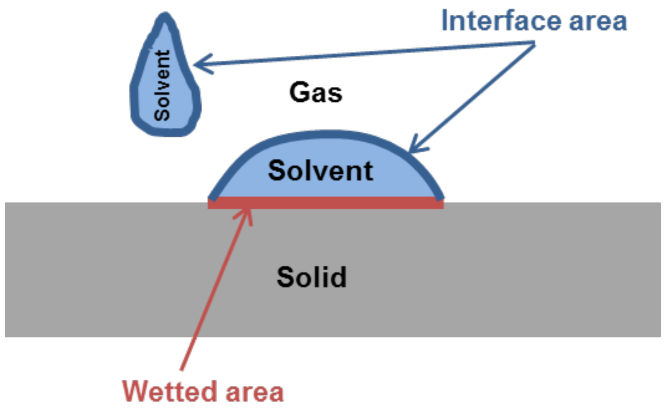


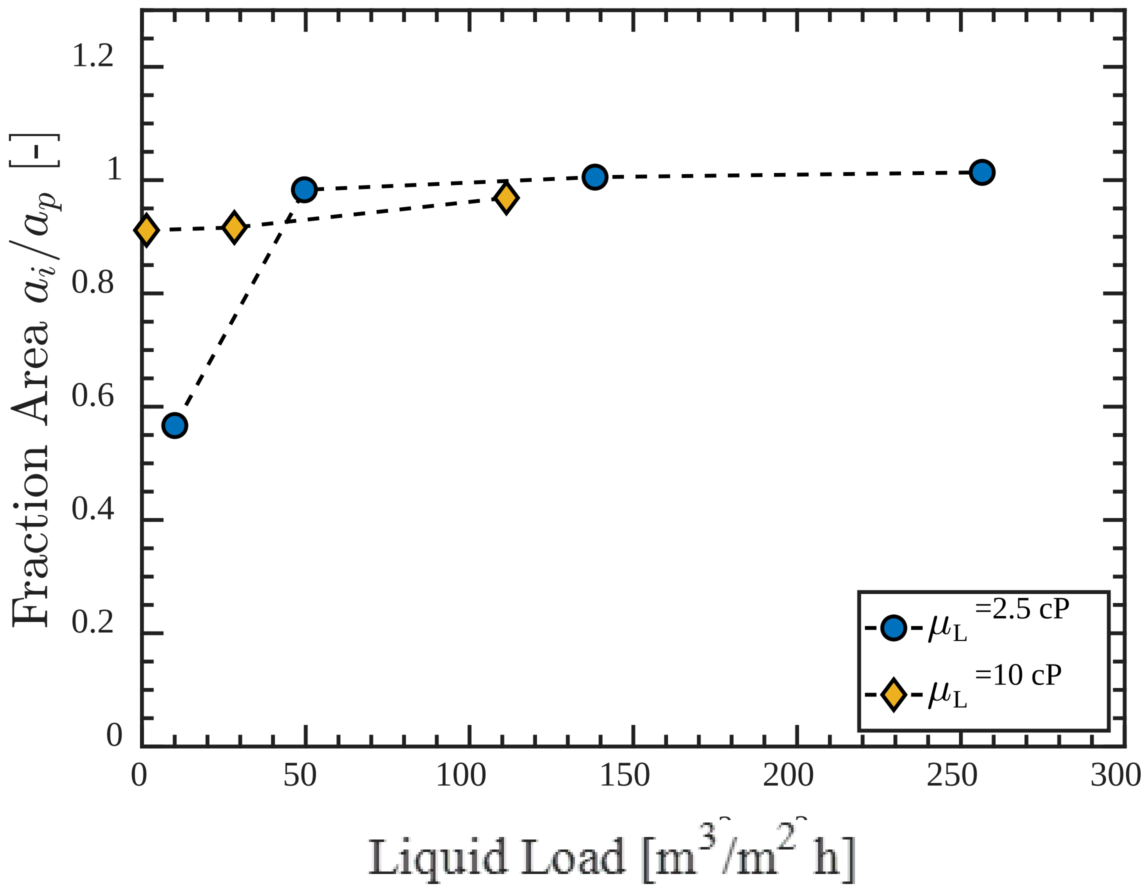
Interface area varying with Solvent flow rate

Schwarz-D: Effect of Liquid Load on Fractional Area

Modified Schwarz-D geometry

- 4cm X 4cm X 4cm
- Surface to volume ratio: 187 1/m
- Viscosity: 2.5, 10cp
- Share similar trends as discovered in Schwarz-P and Gyroid

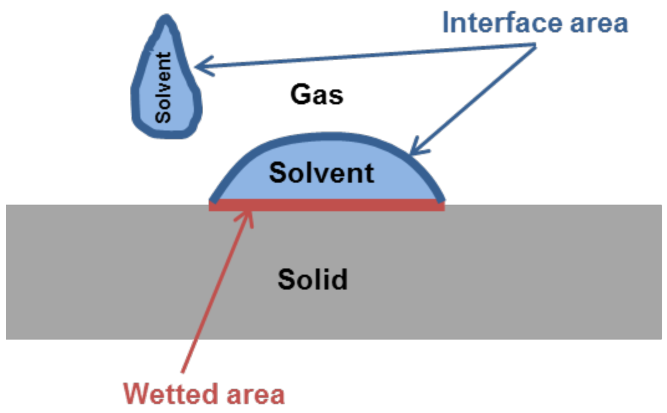


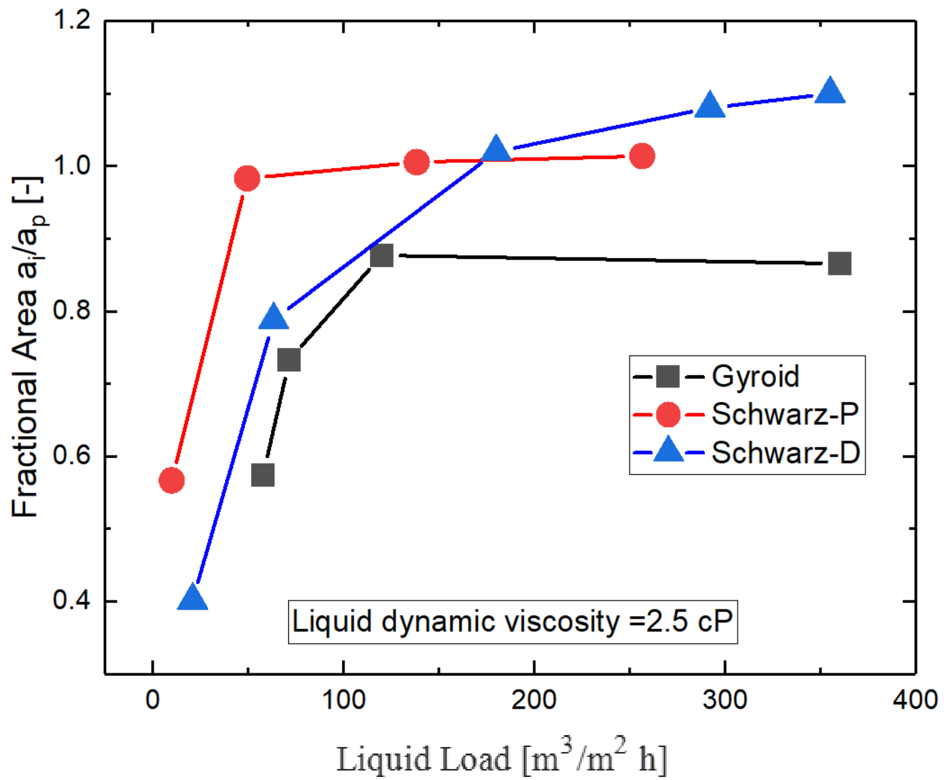


Hydrodynamic Performance for the three geometries

<u>Fractional Area Comparison</u>

- 4cm X 4cm X 4cm
- Viscosity 2.5cp
- Fractional area increases with liquid load and then reaches plateau





Dry Packing Pressure Drop

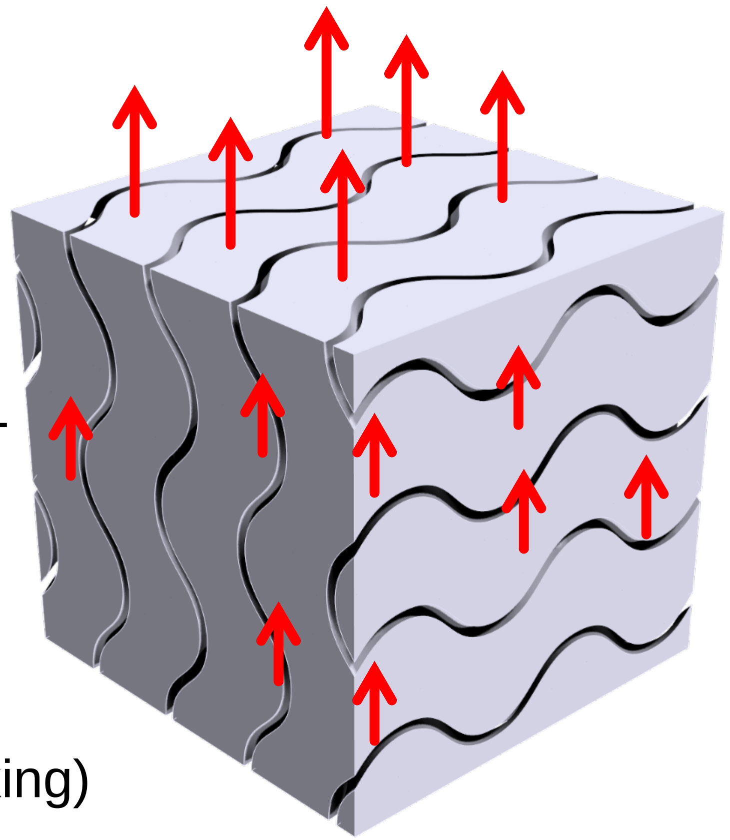
Boundary conditions

- Horizontal: Periodic for flow
- Bottom: Specified Velocity Inlet
- Top: Pressure Outlet

Gas Properties (Air)

Physical Properties	
Density ρ_G (kg/m^3)	1.184
Viscosity μ_G (cP)	0.0186
$D_{CO_2}[g]$ (m^2/s)	1.0×10^{-5}

Single gas phase



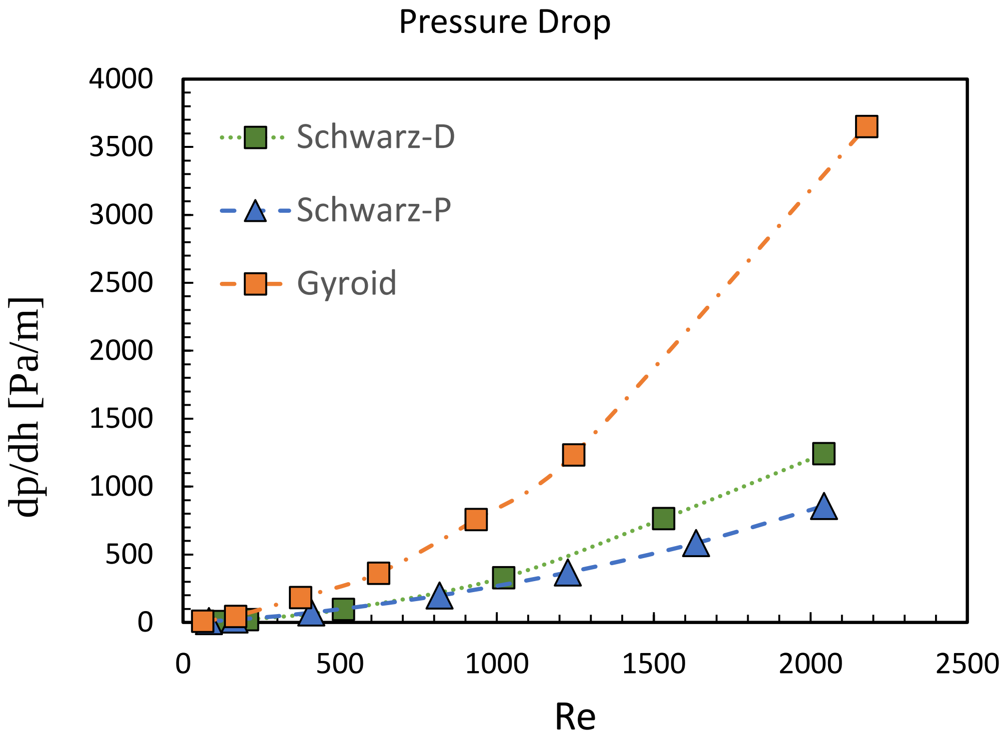
Intial conditions and Dimensionless number

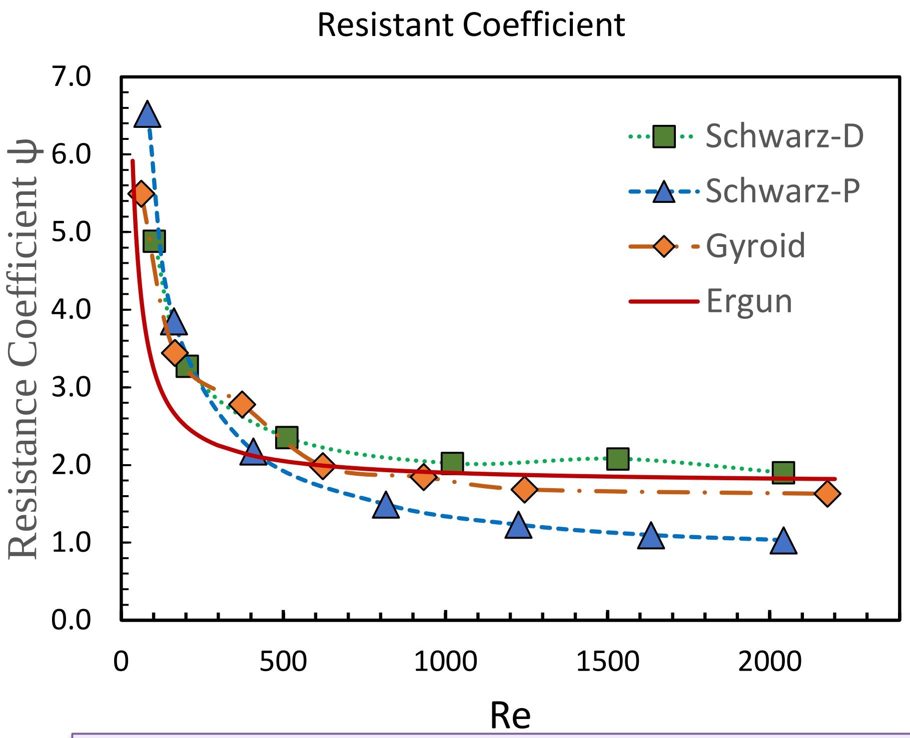
- Modified Reynolds number: $\mathrm{Re} = \frac{u_G d_p \rho_G}{(1-\epsilon)\mu_G} = \frac{3}{2}\frac{u_G d_h \rho_G}{\epsilon \mu_G}$ =[0 2500]
- Superficial gas velocity: $u_G = [0\ 3.5]\ \mathrm{m/s}$
- $d_p = 6(1-\epsilon)/a_p$: particle diameter (analogy to random/structure packing)
- $d_h = 4\epsilon/a_p$: hydraulic dimeter of Packing

Dry Packing Pressure Drop

Pressure Drop and Resistance Coefficient

– Pressure Drop: $\frac{\Delta p}{\Delta H} = \lambda \frac{\overline{u}_G^2 \rho_G}{2 d_h} = \psi \, \frac{1-\epsilon}{\epsilon^3} \frac{F_V^2}{d_p}$,   Resistant Coefficient: $\psi = \frac{d_p \epsilon^3}{1-\epsilon} \frac{\left[\frac{\Delta p}{\Delta H}\right]}{F_V^2}$, $(F_V = u_G \sqrt{\rho_G})$





Ergun Equation:
$\psi = \frac{150}{Re} + 1.75$

Zhijie Xu
Jie Bao,
Chao Wang,
Yucheng Fu

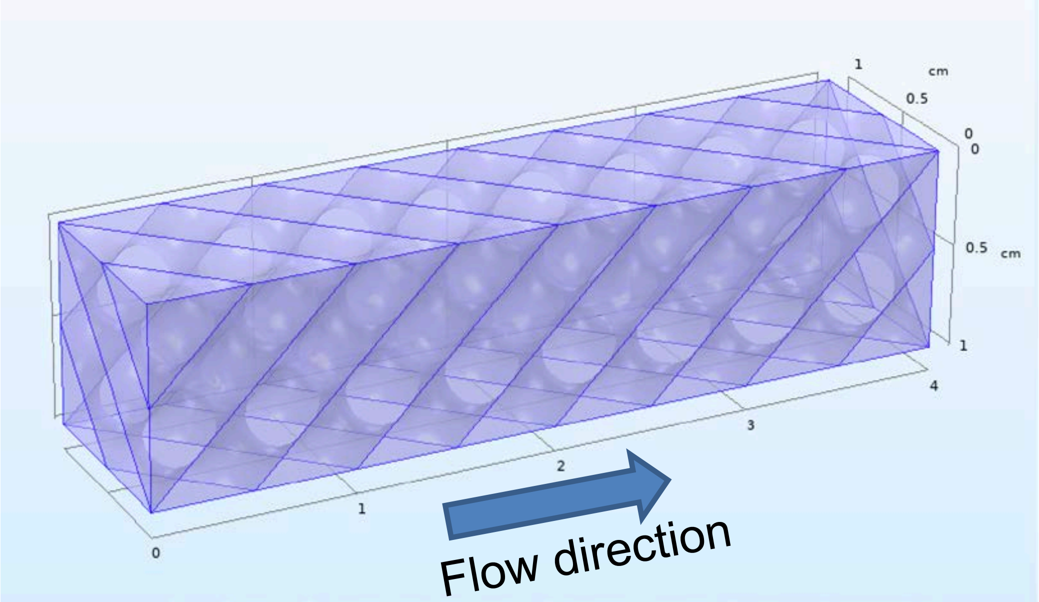
Pressure Drop
Schwarz-P<Schwarz-D<Gyroid

Resistant Coefficient (Re>500)
Schwarz-P< Gyroid<Schwarz-D

Mass transfer in Schwarz-D

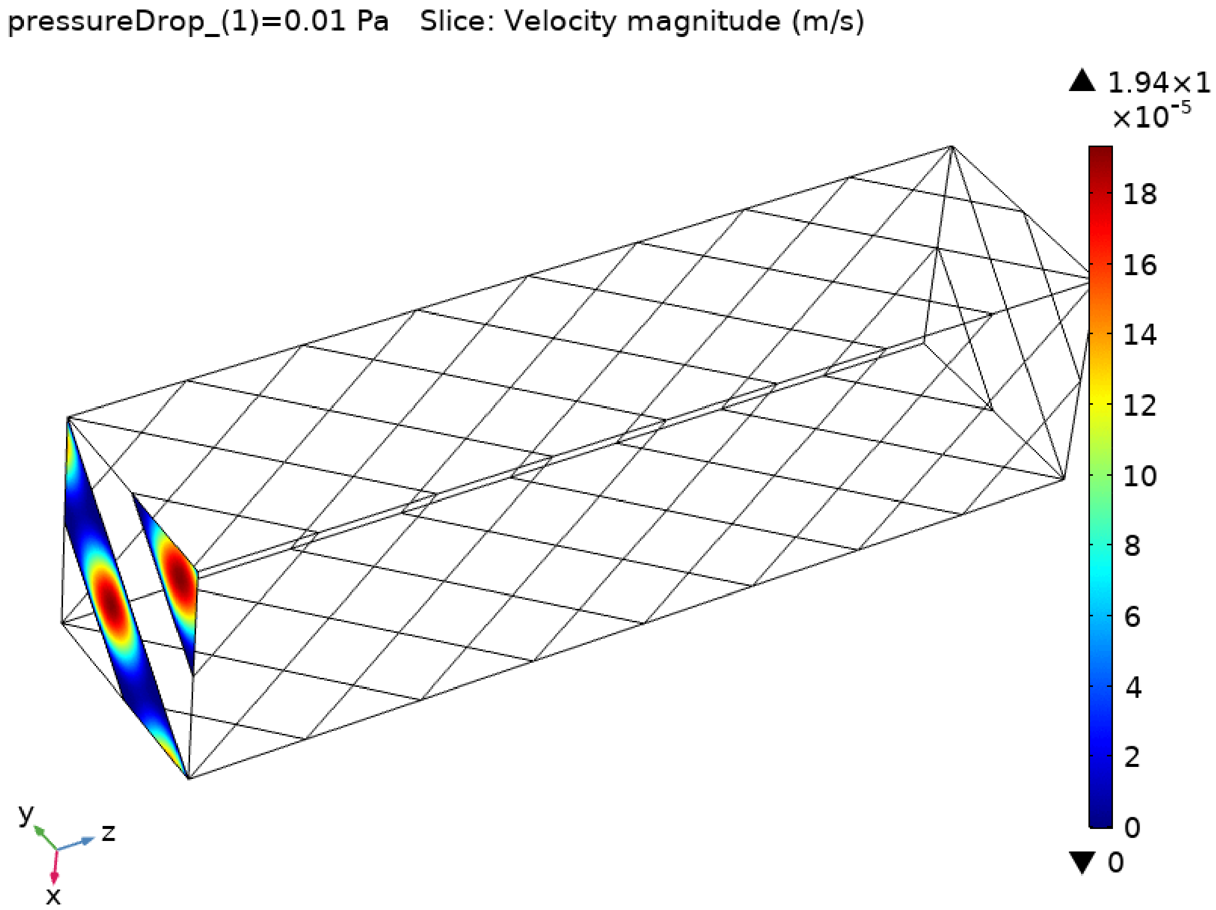
Simulation specifications

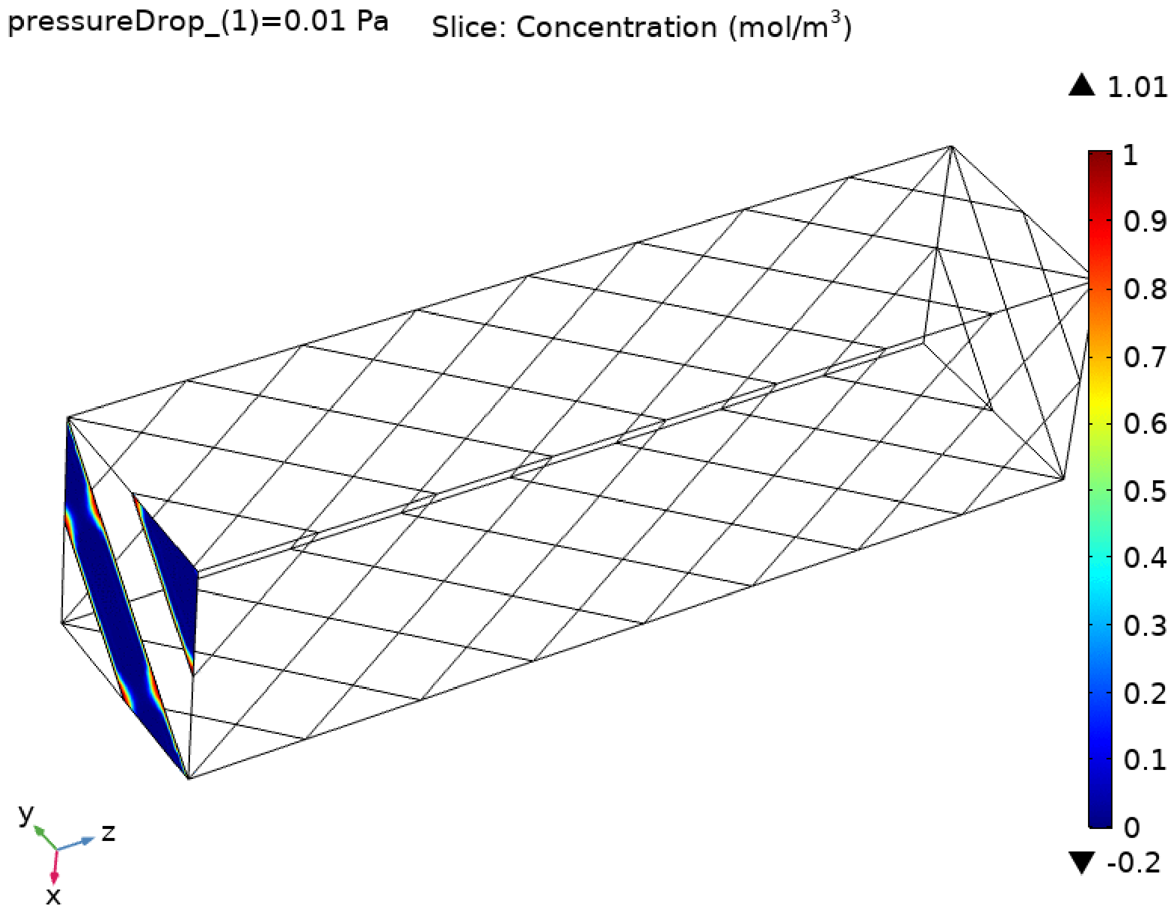
- Domain size: 1 cm x 1 cm x 4 cm
- 1 x 1 x 4 unit cells
- Convection-diffusion problem
- Pressure driven flow along the length of channel
- Periodic boundary conditions in other directions
- Uniform concentration at the walls



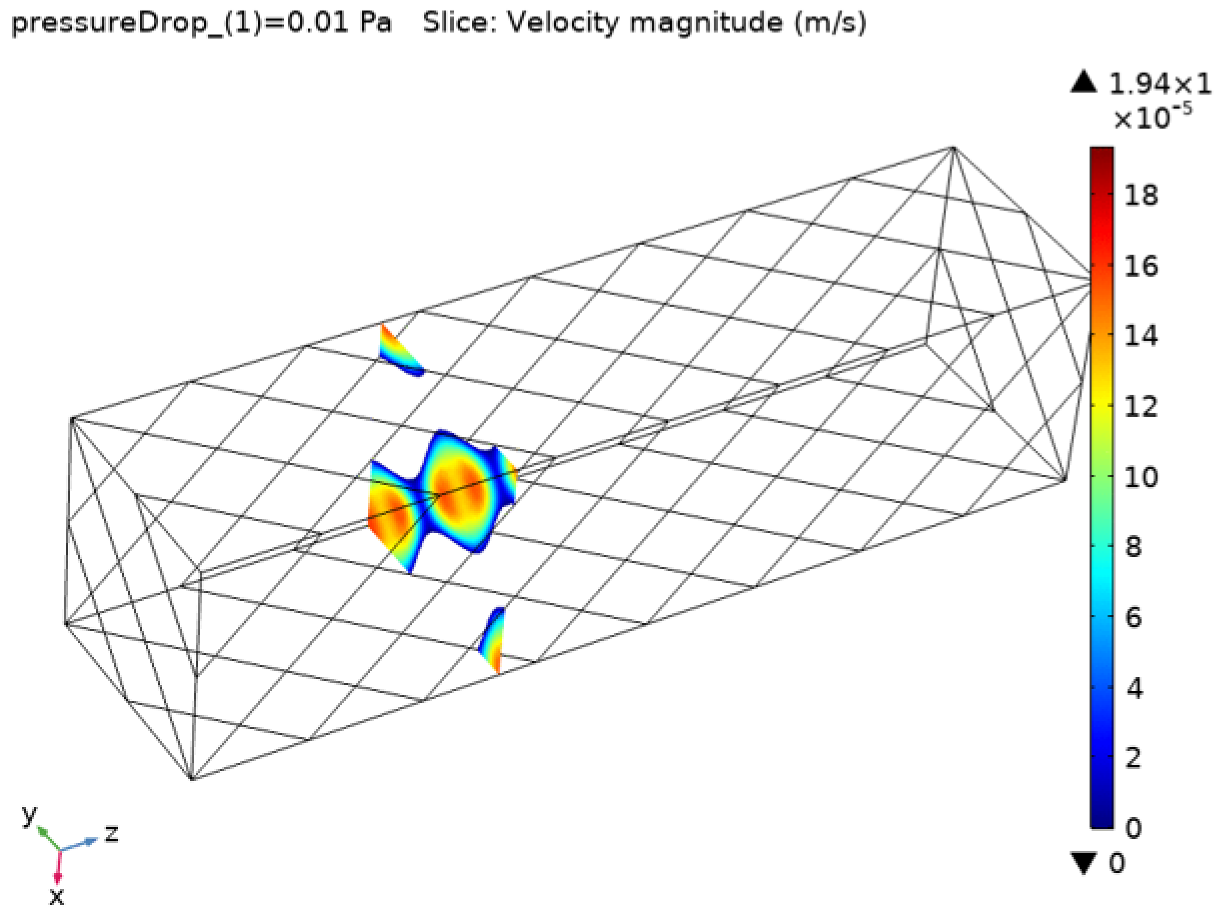
Joshuah Stolaroff
Pratanu Roy
Jaisree Iyer Kannan
Du Thai Nguyen

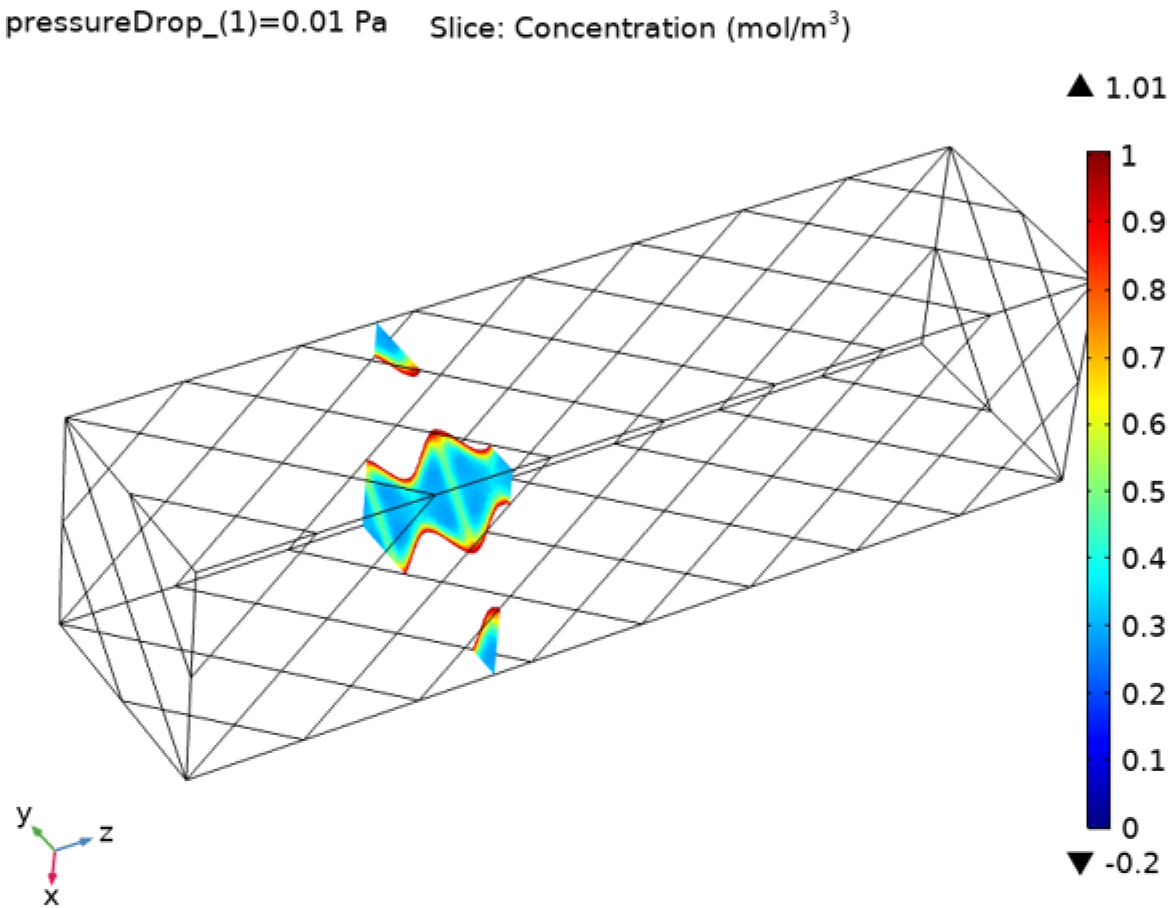
Velocity and concentration profiles in Schwarz-D structure



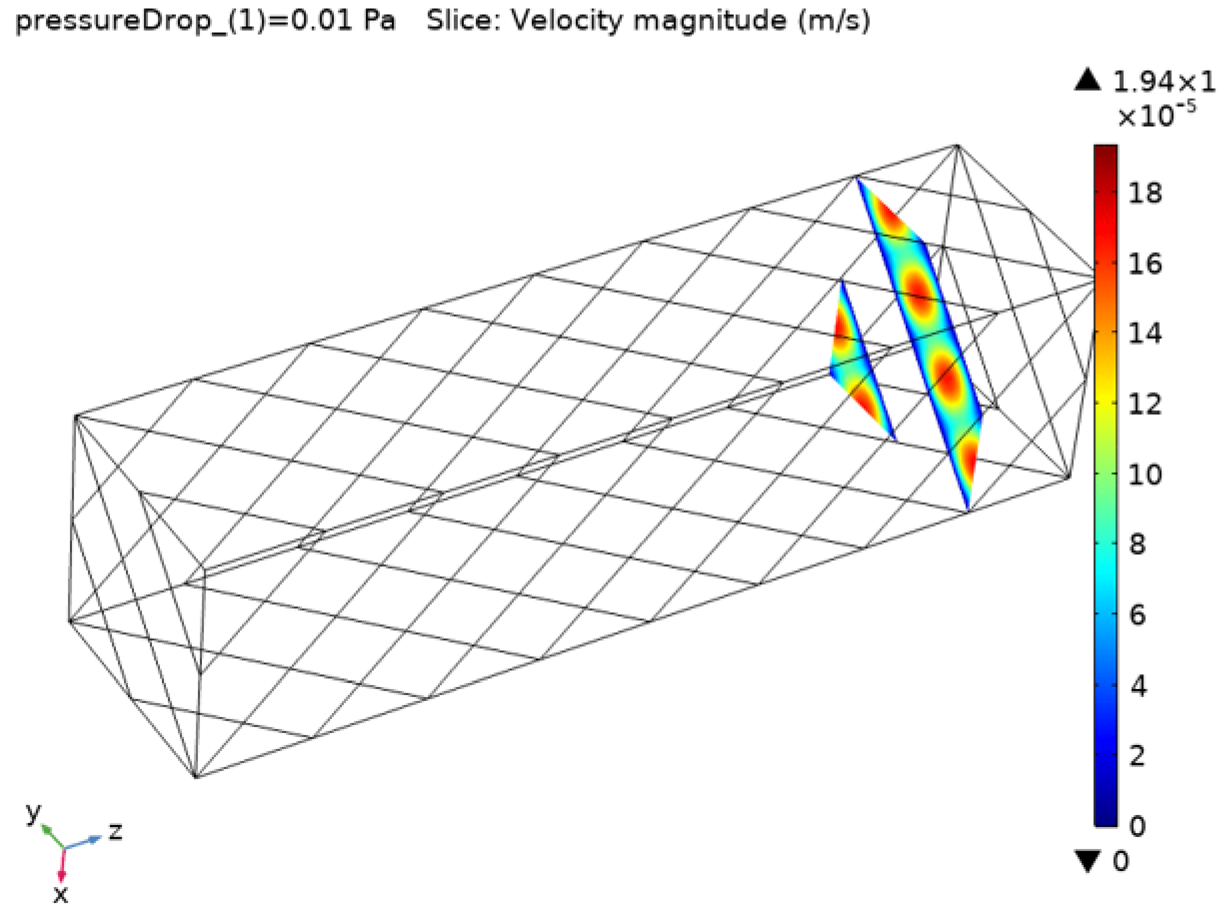


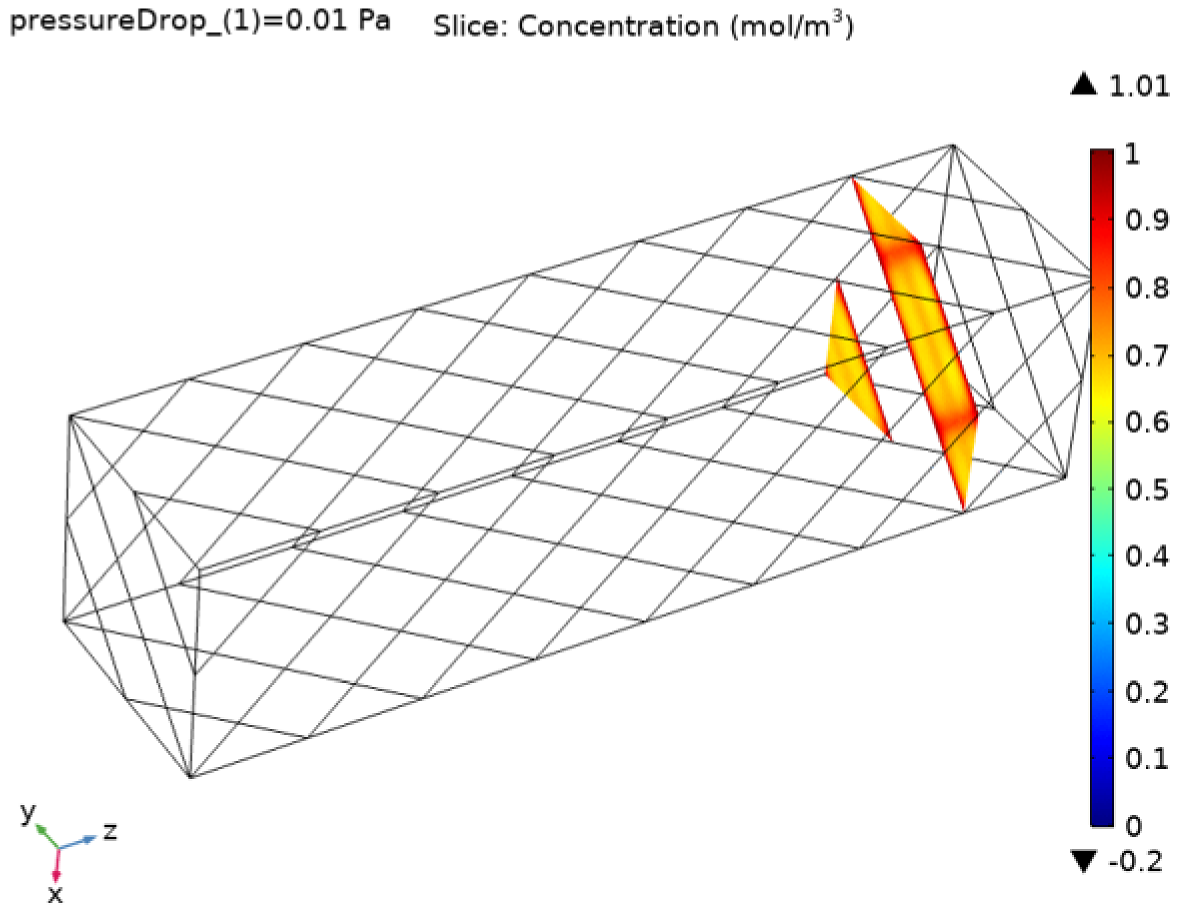
Velocity and concentration profiles in Schwarz-D structure





Velocity and concentration profiles in Schwarz-D structure



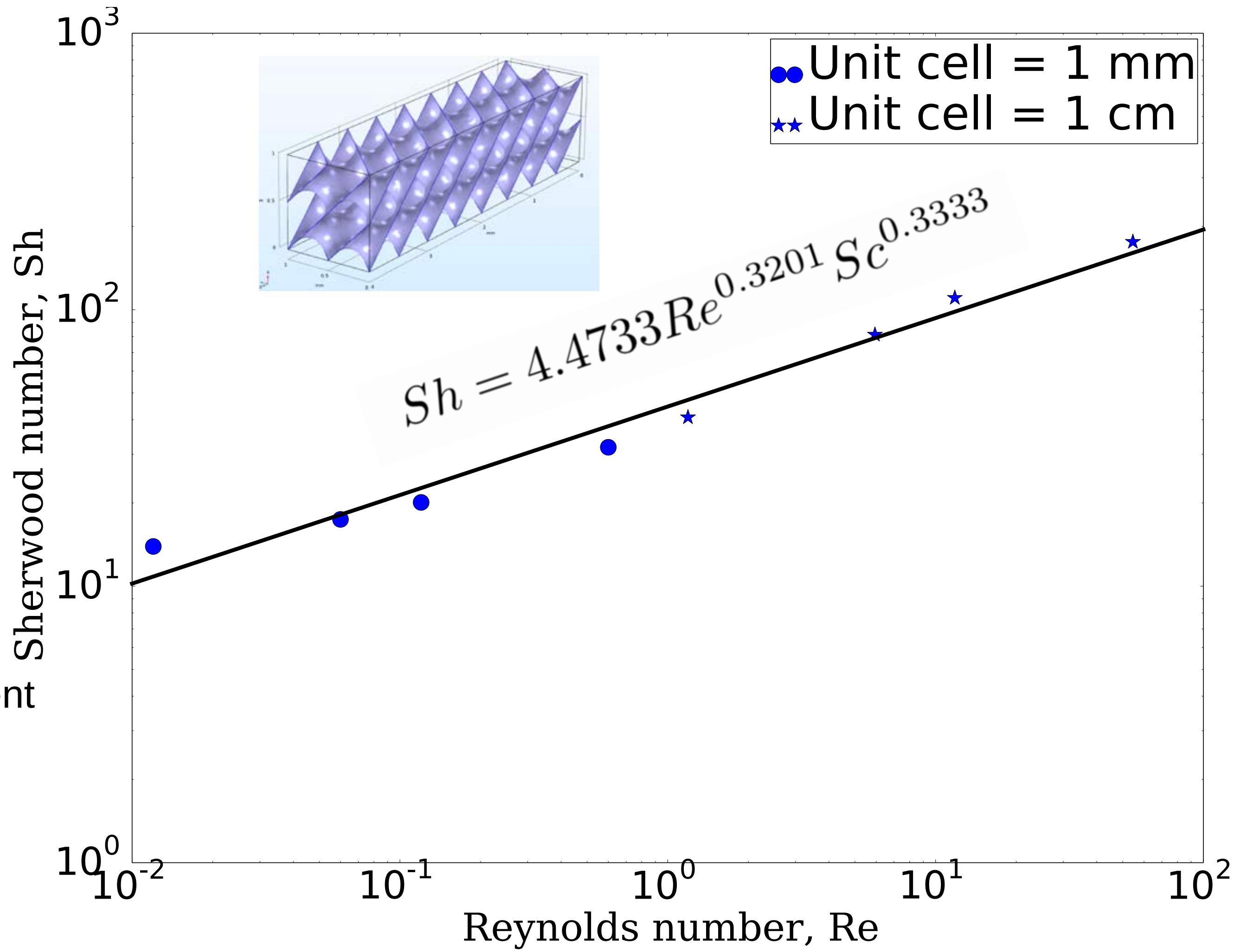


SchwarzD: Sh and Re correlations

$$Sh = kD_h/D$$

$$Sc = \nu/D$$

Sh = Sherwood Number
Sc = Schmidt Number
D = Diffusivity
D_h = Hydraulic diameter
ν = Kinematic viscosity
k = Mass transfer coefficient



For uniform wall concentration in a pipe flow Sh is constant (=3.66), and is not a function of Re.

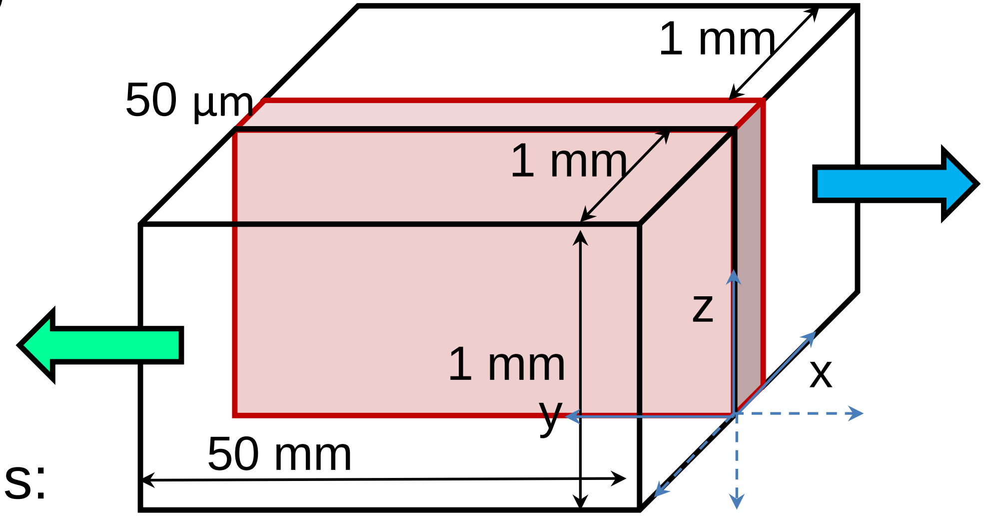
Since the boundary layer is disrupted continuously in Schwarz-D structure, the Sh changes with Re and Sc.

Development of COMSOL model for carbonate-based carbon capture system

- Benchmark model for comparison against experiment

$$CO_2 + OH^- \leftrightarrow HCO_3^-$$
$$HCO_3^- + OH^- \leftrightarrow CO_3^{2-} + H_2O$$
$$H_2O \leftrightarrow H^+ + OH^-$$

2.87 mol/m³ K_2CO_3 solution (v = 0.01 m/s)

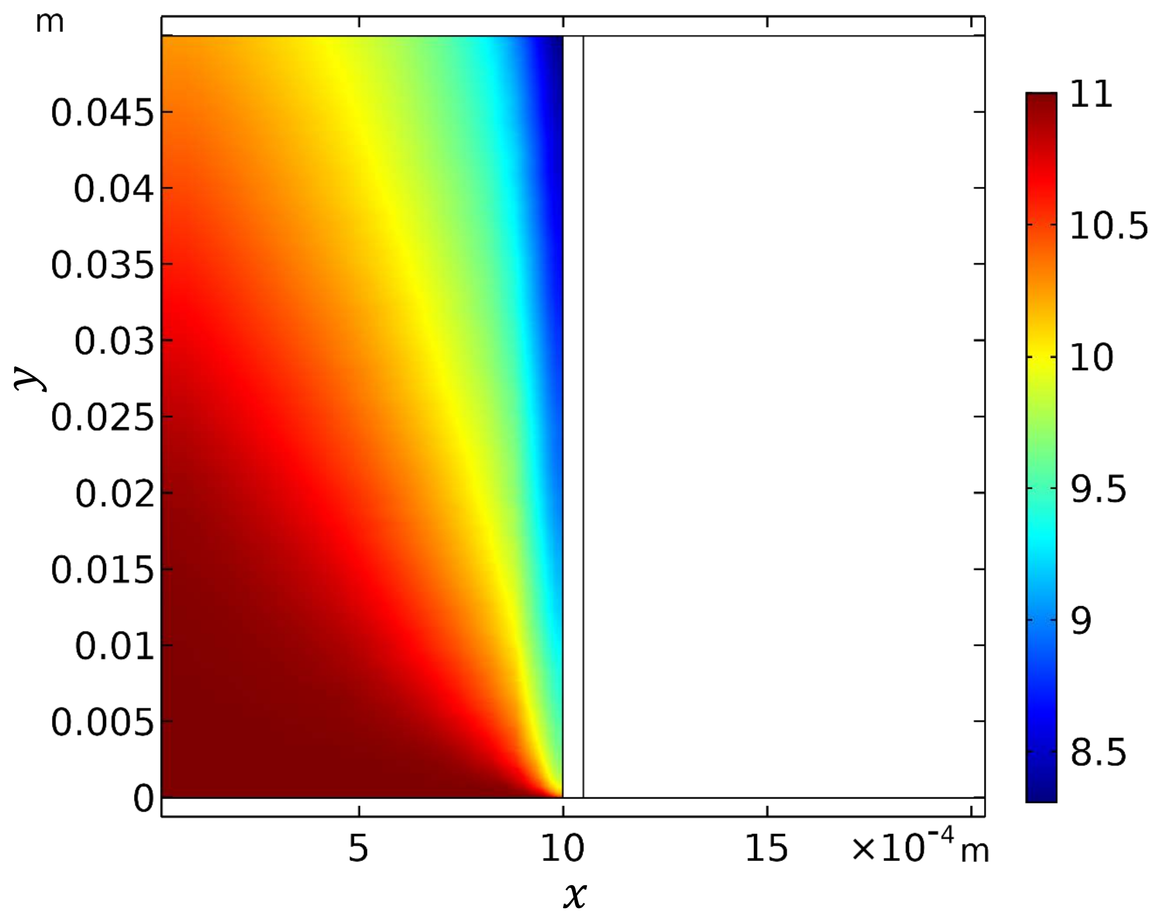


40.034 mol/m³ CO_2 gas (v=1.7 m/s)

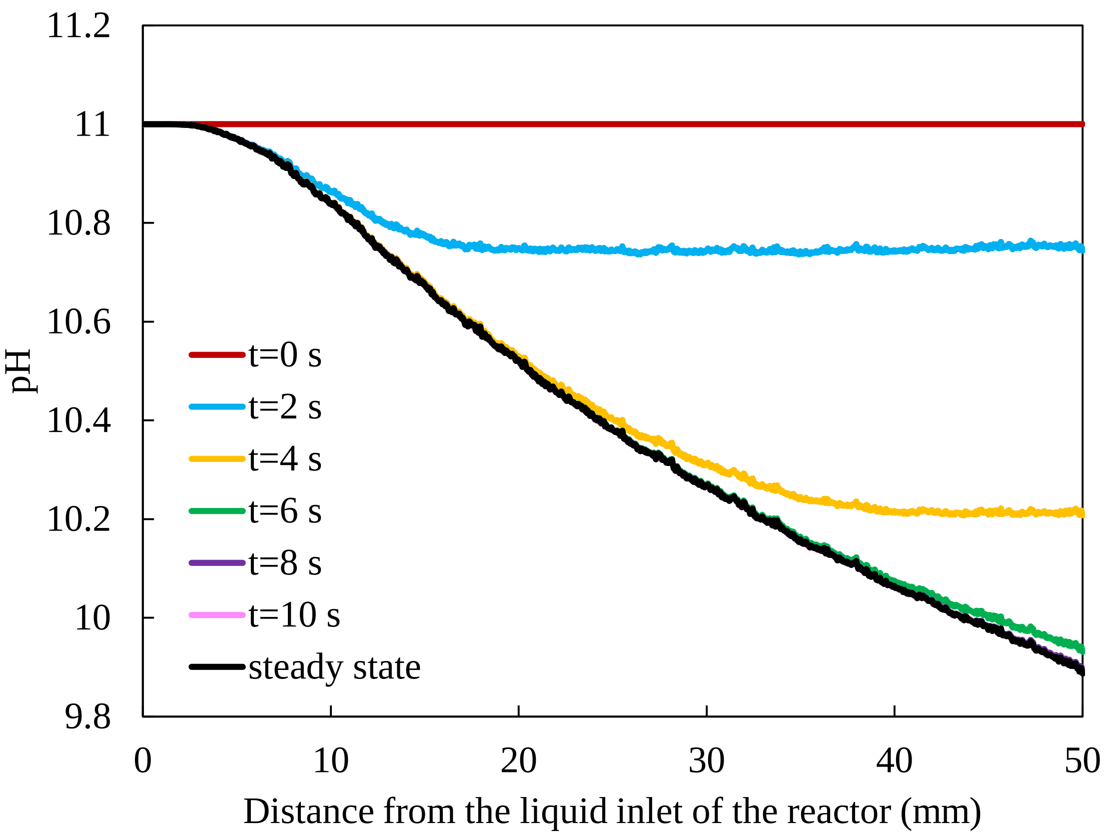
- Model simplifications:
  - 2-D instead of 3-D
  - No flux at all external faces except for the flow faces
  - Gas average velocity reduced to 1.7 m/s from 16.67 m/s (1 L/min) to maintain laminar flow

Spatial and temporal variation of the pH

Steady state pH in the liquid domain



pH along the centerline of the liquid domain

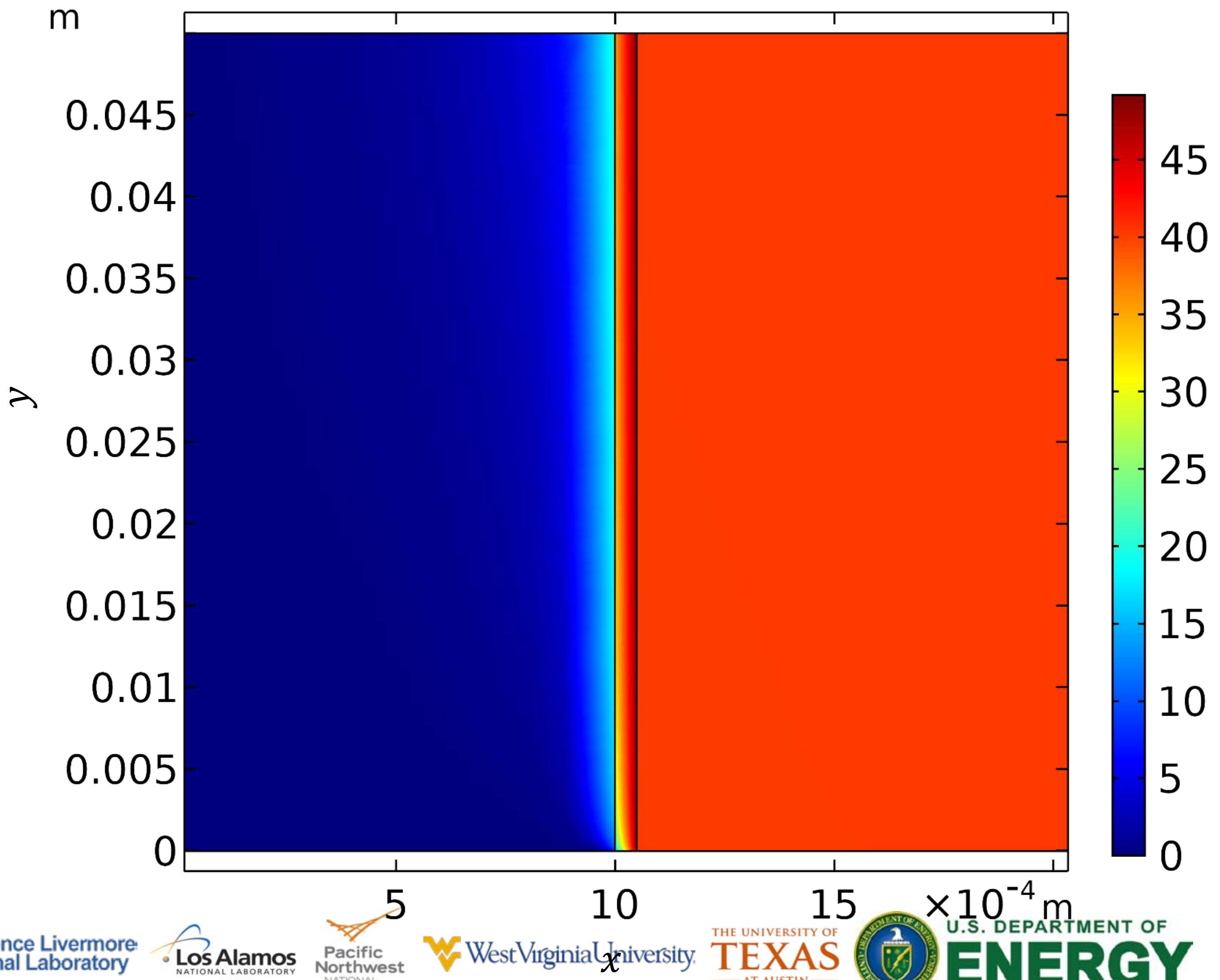


- It takes about 10 s to reach steady state with a pH change of approximately 1
- Fast reactions had to be slowed down to obtain convergence
    - Could have resulted in inaccuracies

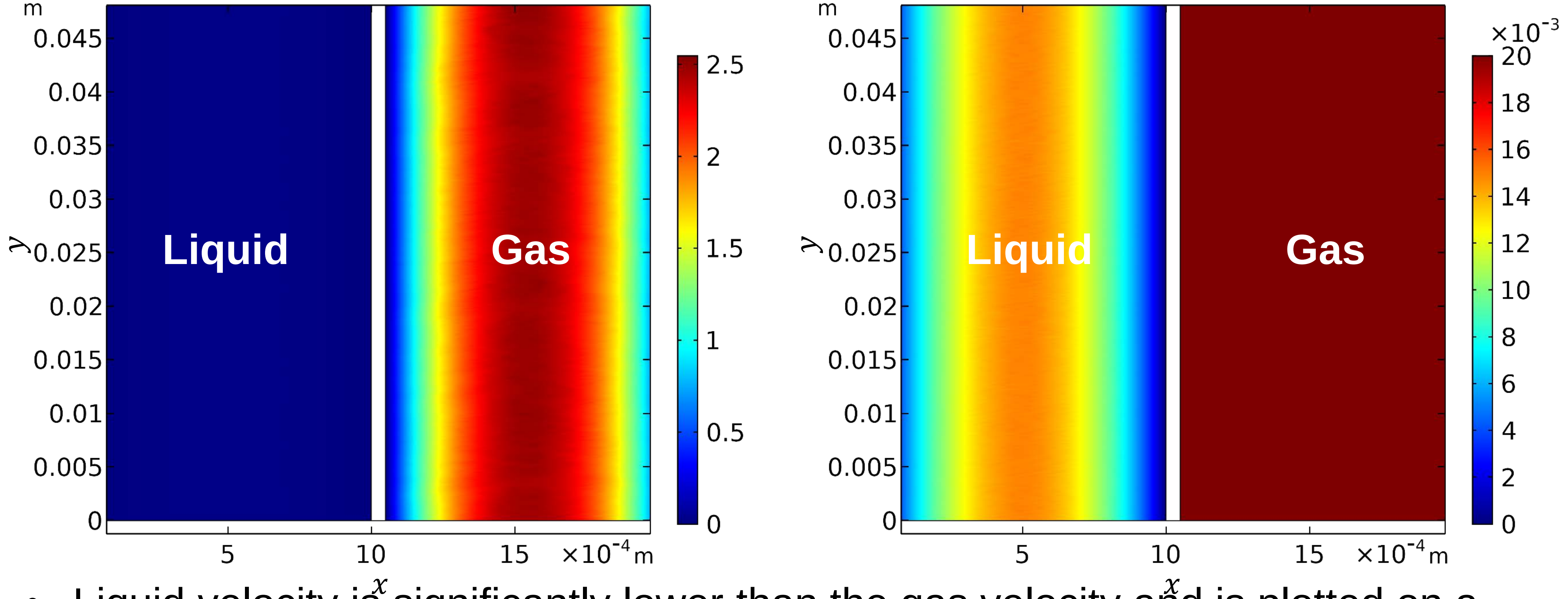
Steady state CO_2 concentration (mol/m^3)

- Concentration in the gas domain doesn't change significantly due to the high velocity

- CO_2 concentration in the liquid domain sees large changes near the membrane but remains unchanged away from it



Velocity in the gas and liquid domain (m/s)



- Liquid velocity is significantly lower than the gas velocity and is plotted on a different color scale

Moving Forward

Perturb the gyroid geometry in a mathematically driven way in order to improve performance

Develop a CFD framework that can be efficiently integrated with the ML, optimization and CAD software tools

Develop a CFD framework that can cope with the complexities of the setup in terms of:

- Multiphase flow with varying physics. i.e Turbulence, rivulet/waves, blockage
- Chemical reaction of absorption
- Heat transfer
- Parametrization of the geometry

Machine learning and optimization approaches to design computational experiments for data generation

Manufacture through 3D printing a geometry to calibrate model and validate computational and theoretical work

Acknowledgments

Carbon Capture Simulation for Industry Impact ($CCSI^2$)

PNNL: Zhijie Xu, Charlie Freeman, David Heldebrant, Richard Zheng, Rajesh Singh, Jie Bao, Chao Wang, Yucheng Fu

LLNL: Joshuah Stolaroff, Pratanu Roy, Du Thai Nguyen, Jaisree Iyer Kannan, Brenda Ng, Charles Tong, Wenqin Li

NETL: Michael Matuszewski, Benjamin Omell, Joshua Morgan, Grigorios Panagakos

UT Austin: Gary Rochelle

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Carbon Capture Simulation for Industry Impact

For more information

https://www.acceleratecarboncapture.org/

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