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NATIONAL RENEWABLE ENERGY LABORATORY

# Mesoflow: An open-source reacting flow solver for catalysis at mesoscale

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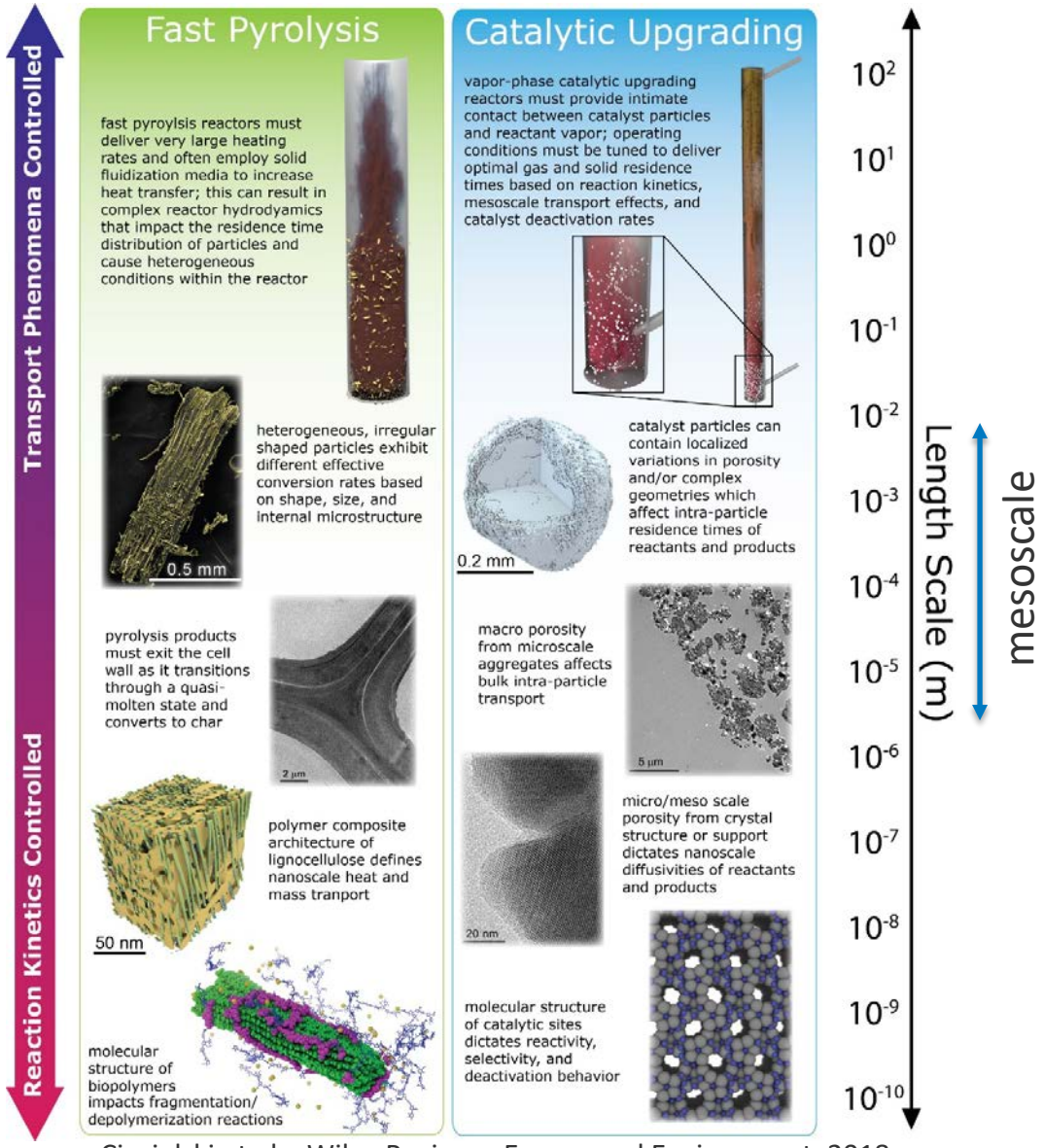
# Introduction and Motivation

- Catalytic and thermochemical conversion are complex multiscale processes.

Mesoscale modeling enables:

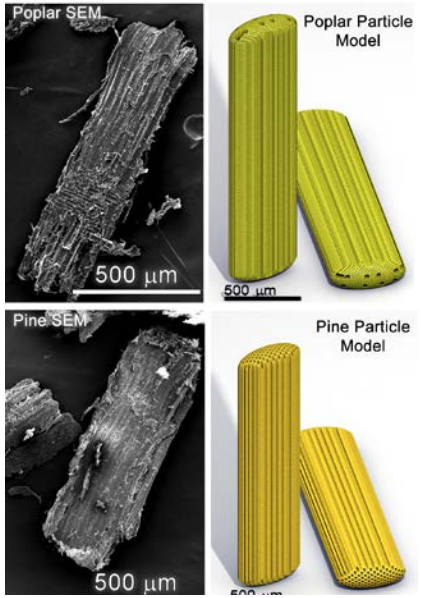
  - Connection between molecular and reactor scales
  - Investigation of heterogenous transport phenomena
  - Complex surface morphologies can significantly affect reactivity
  - Extraction of transport-dependent reaction/deactivation kinetics

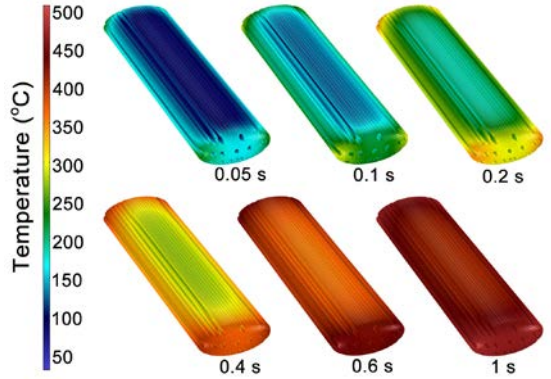



Ciesielski et al. , Wiley Reviews: Energy and Environment, 2018

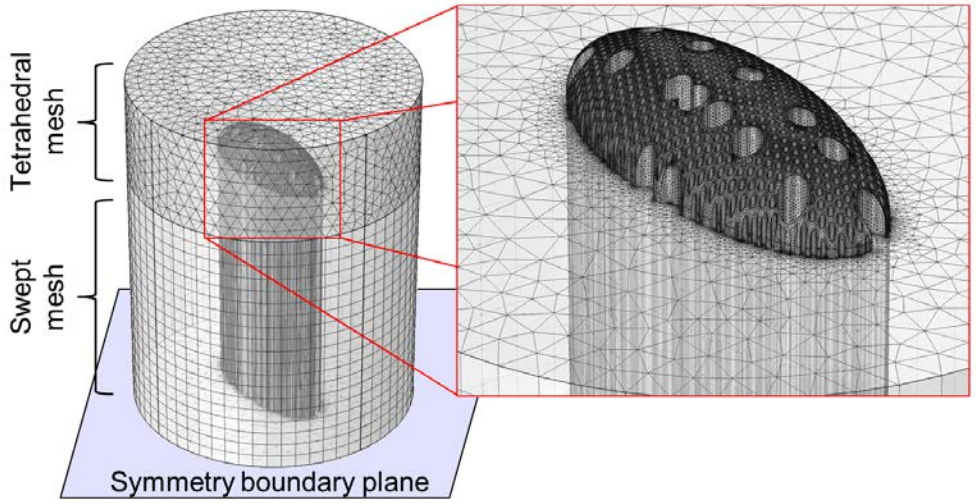
# Current state-of-the-art and limitations

- Mesoscale models
  - Use of commercial software – COMSOL, ANSYS
  - Several questions have been addressed
    - Effect of bulk particle morphology
    - Effect of species-specific transport
- Limitations
  - Scaling to higher resolutions and domains
    - Runs done on local high-end workstations
    - Most often accelerators (GPU) are unused
  - Meshing of complex geometries to get accurate solutions difficult and time consuming
  - geometries used are approximations
    - important features may be neglected



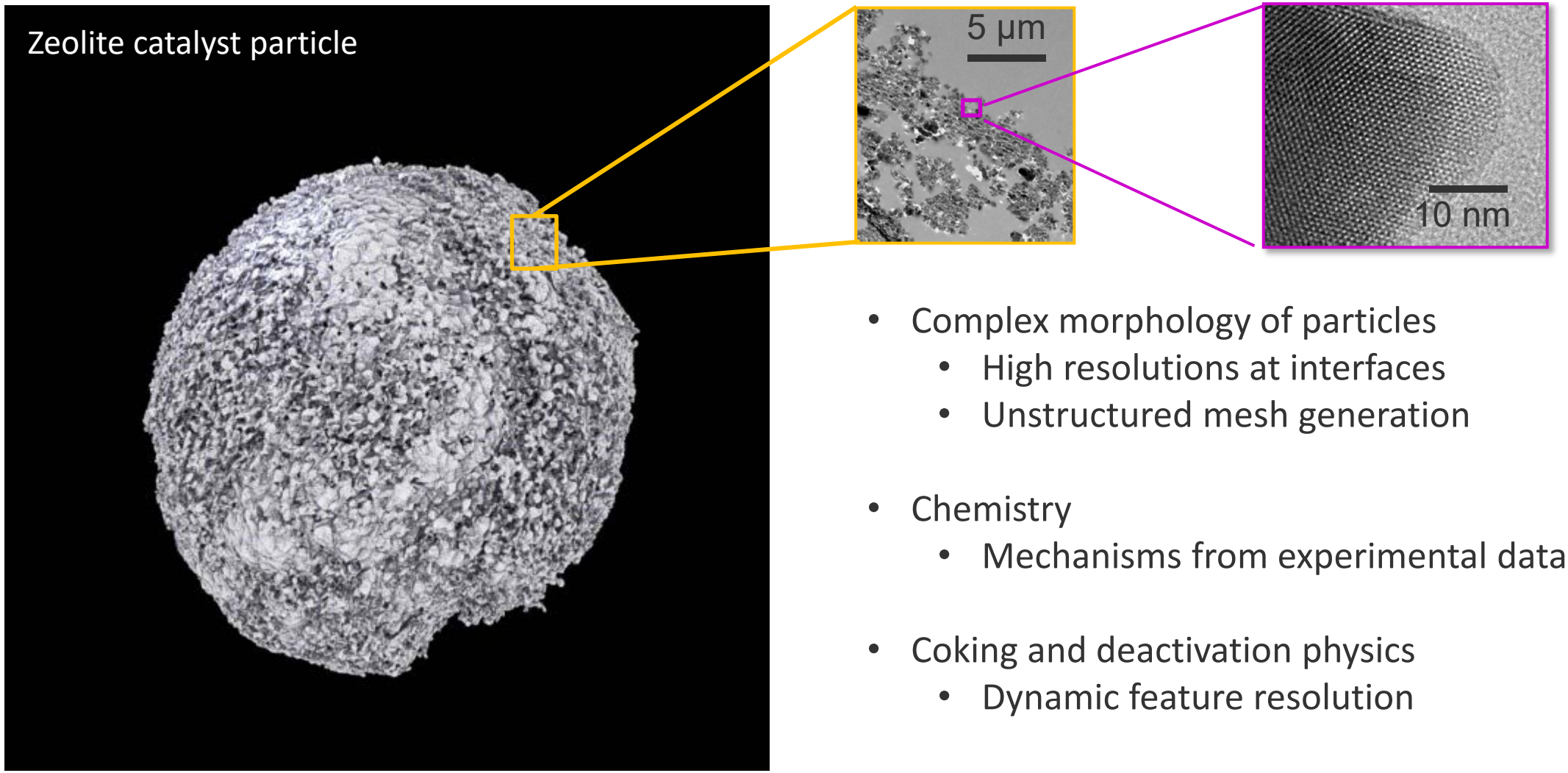




Heat transfer simulation




Ciesielski et al., Energy and Fuels, 2014

# Mesoscale modeling challenges




Ciesielski et al. , Wiley Reviews: Energy and Environment, 2018

- Complex morphology of particles
  - High resolutions at interfaces
  - Unstructured mesh generation
- Chemistry
  - Mechanisms from experimental data
- Coking and deactivation physics
  - Dynamic feature resolution
- HPC is required to answer these physics questions

# Objectives

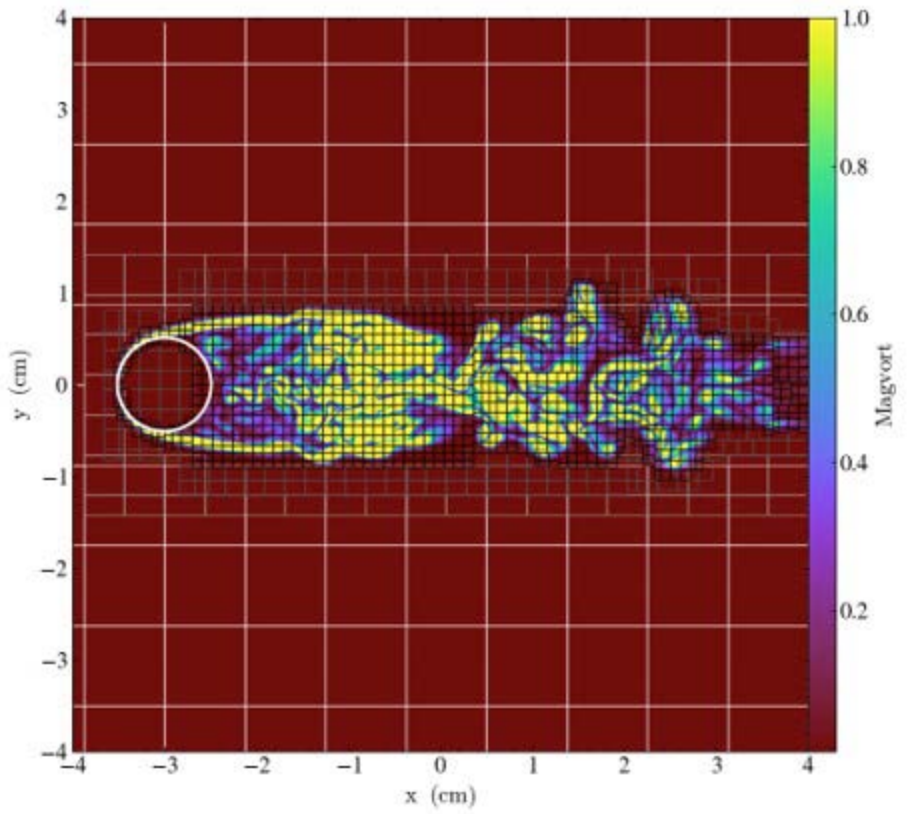
- Develop a Mesoscale modeling tool
  - Gas-solid flows
    - Catalytic upgrading
  - Flow through porous particles
  - Scalable on current and future HPC resources
    - use distributed and shared-memory parallelism
      - Acceleration hardware like GPUs
  - Automated meshing of complex geometries
  - Incorporate chemistry/transport coupling
  - Be able to simulate dynamic geometries
- Mesoflow: **https://github.com/NREL/mesoflow**


*Summit* Supercomputer at Oakridge National Laboratory*

*https://www.ornl.gov/news/ornl-launches-summit-supercomputer

# Mesoflow: numerical methods

- Leverage open-source library – AMReX*
  - developed at LBNL/NREL/ANL
  - Designed for upcoming Exascale machines
- AMReX is parallel adaptive-mesh refinement (AMR) library
  - Mainly provides block-structured AMR data structures and linear solvers
  - Partial Differential Equation solvers need to be developed using AMReX data structures
  - Code development in C++ with GPU kernels




Flow around sphere Re=4000

*Zhang, Weiqun, et al. "AMReX: a framework for block-structured adaptive mesh refinement." *Journal of Open Source Software* 4.37 (2019): 1370-1370.

# Mesoflow: numerical methods

Compressible Navier-Stokes equations (gas phase)

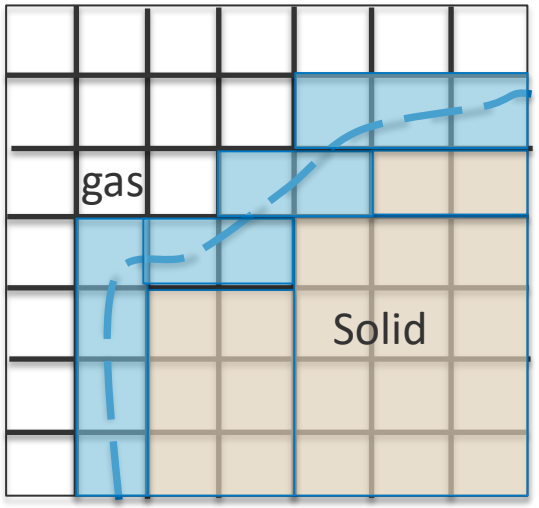
$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{U}) = 0$$

$$\frac{\partial (\rho \mathbf{U})}{\partial t} + \nabla \cdot (\rho \mathbf{U}\mathbf{U}) = -\nabla P + \nabla \cdot \bar{\bar{\tau}}$$

$$\frac{\partial (\rho E)}{\partial t} + \nabla \cdot (\rho E + P) = \nabla \cdot (k \nabla T) + \nabla \cdot (\bar{\bar{\tau}} \cdot \mathbf{U})$$

Species transport and reaction (both gas and solid phase)

$$\frac{\partial c_i}{\partial t} + \nabla \cdot (c_i \mathbf{U}) = \nabla \cdot (D_i \nabla c_i) + \dot{W}_i$$

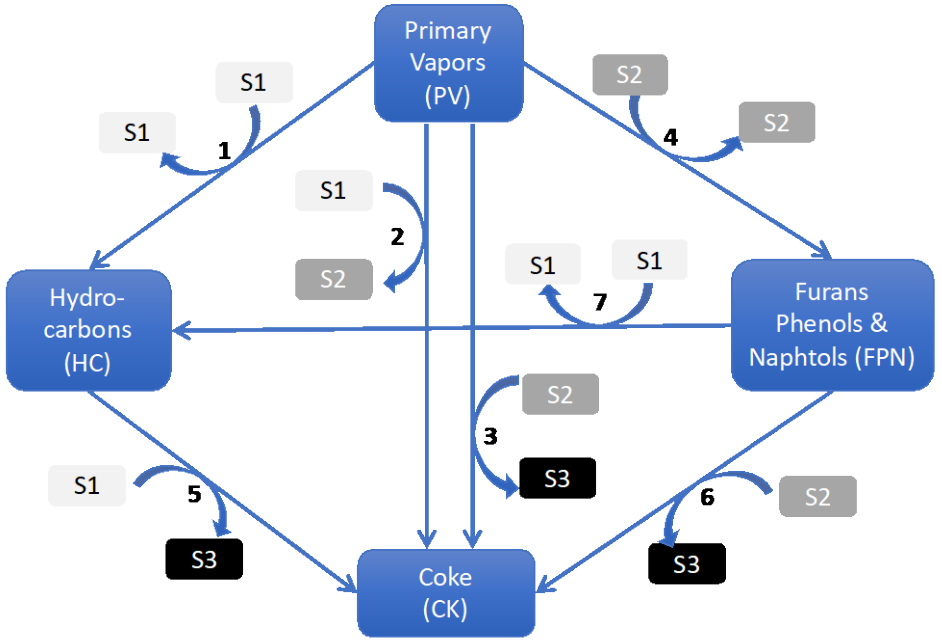



Reflection based boundary conditions for velocity at the interface

- AUSM+up* scheme for low-Mach flows
- Level-set immersed boundary method at the boundaries
- Temperature dependent transport and thermal properties

Liou, Meng-Sing. "A sequel to AUSM, Part II: AUSM+-up for all speeds." *Journal of computational physics* 214.1 (2006): 137-170.

# Mesoflow: chemistry modeling

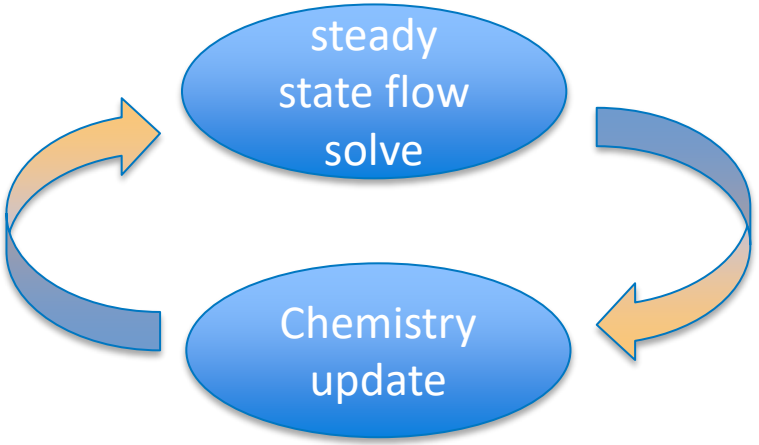



S1- Fresh Active Sites S2- Intermediate Sites S3- Deactivated Sites

| No. | Reaction | k[m³/(mol.s)] @ T=500⁰C |
|---|---|---|
| 1 | PV + S1 → HC + S1 | 2.573 |
| 2 | PV + S1 → CK + S2 | 0.456 |
| 3 | PV + S2 → CK + S3 | 0.152 |
| 4 | PV + S2 → FPN + S2 | 2.904 |
| 5 | HC + S1 → CK + S3 | 0.507 |
| 6 | FPN + S2 → CK + S3 | 0.006 |
| 7 | FPN + S2 → HC + S2 | 0.051 |

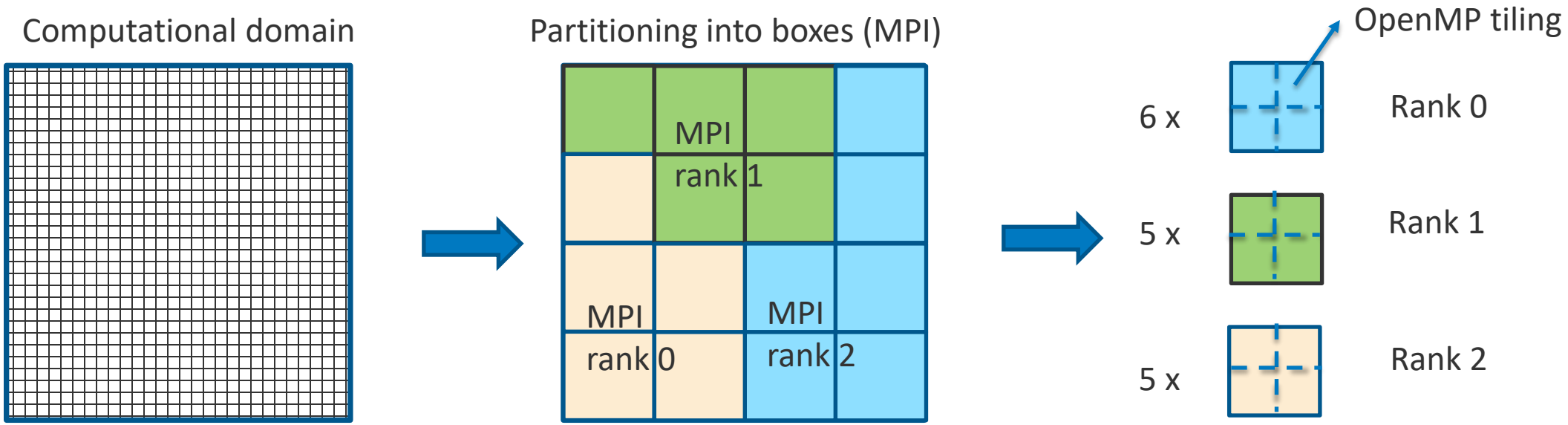
Bharadwaj et al. *In preparation*

- Finite rate chemical mechanism evaluation
  - Implemented as matrix-product evaluations
- Example: 7 species chemistry for Zeolite catalyst
  - Gas phase - primary vapors, products
  - Solid phase - Coke and catalyst sites
- Operator splitting approach to couple with transport




Operator-split update

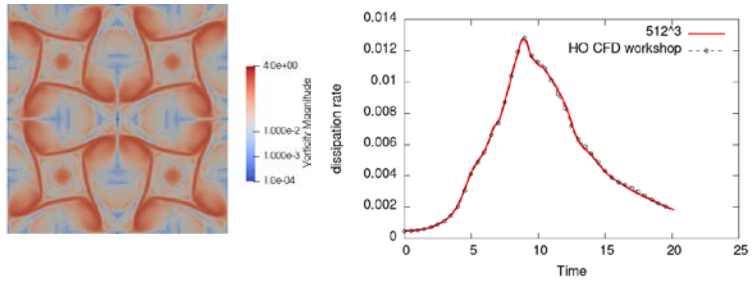
# Mesoflow: parallelization strategy



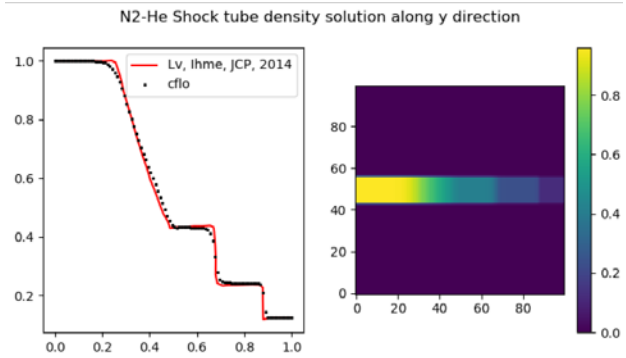

- Boxes ($32^3$ or $64^3$ size) per rank run using either
  - Openmp threads on smaller tiles
  - multiple GPU streams
- Stencil operations executed through asynchronous GPU kernels
  - Inlined functions called from within C++ lambda
- Data movement managed through AMReX
  - Both Gpu managed and dedicated device memory
- Currently setup with 1 GPU per MPI rank configuration

# Mesoflow: code verification and correctness

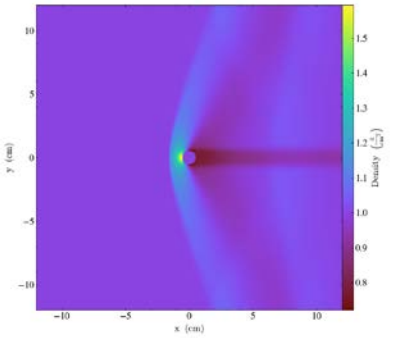
## Taylor green vortex



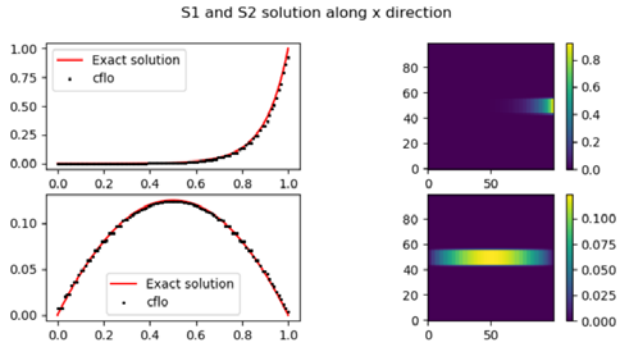

## Multispecies shock tube



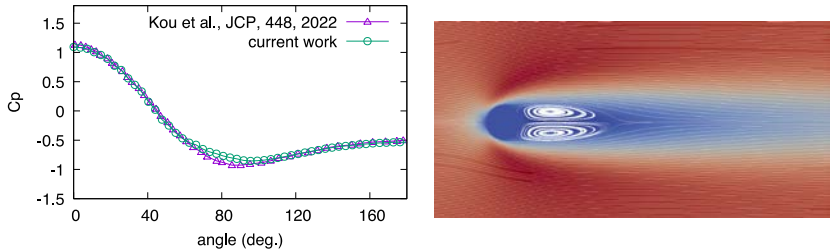

## Blunt body flow

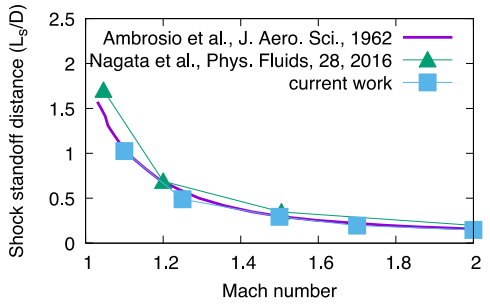


## Species transport tests

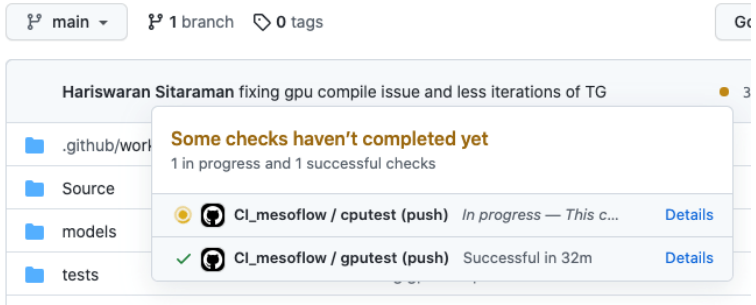



## Cylinder flow at Re=40

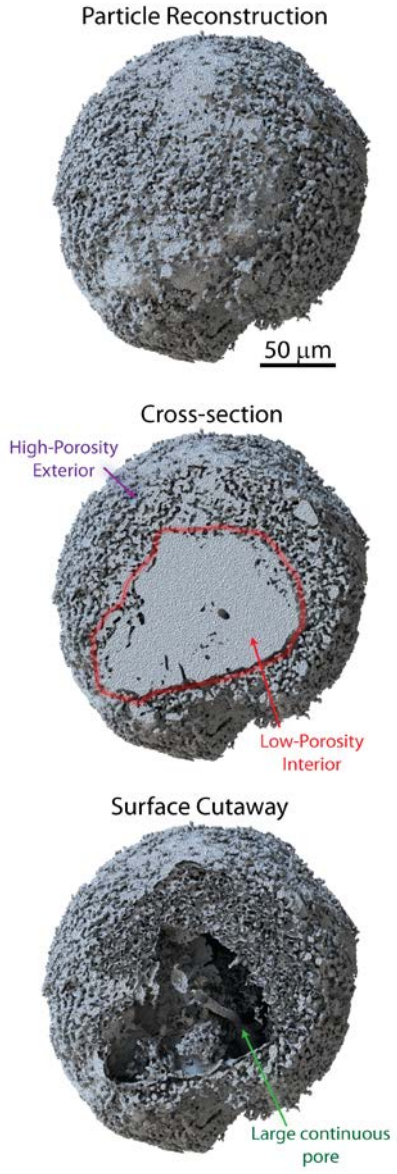


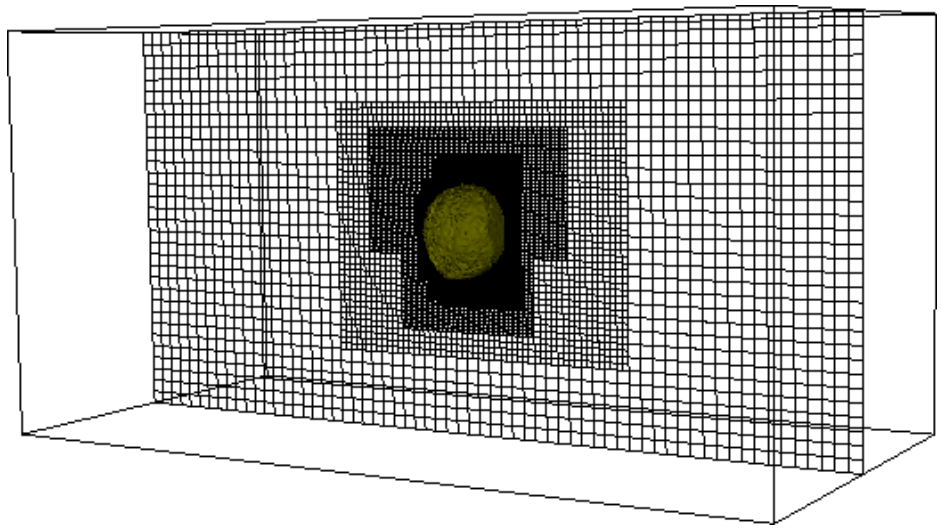



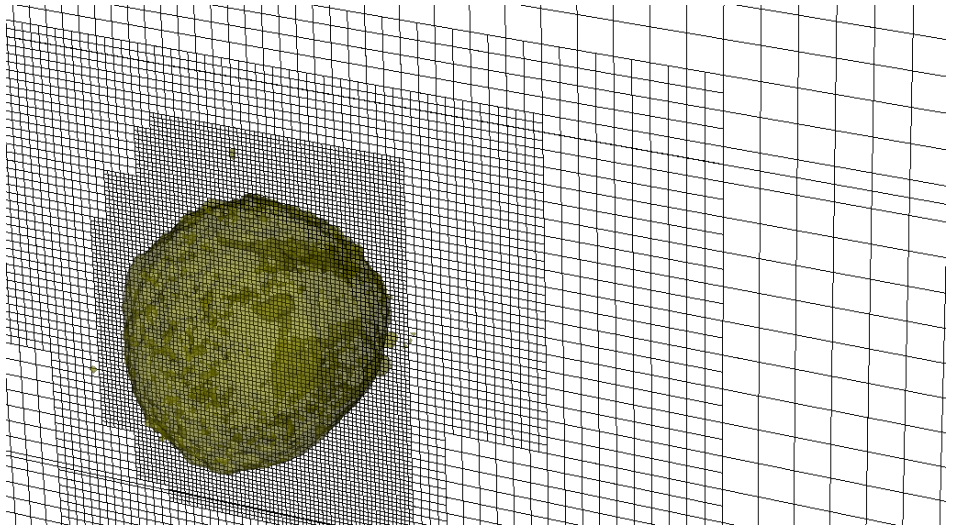

- Automated Regression testing
- Code changes tested using github actions
- Testing done on both CPU and GPU



# Example case: Zeolite catalyst particle

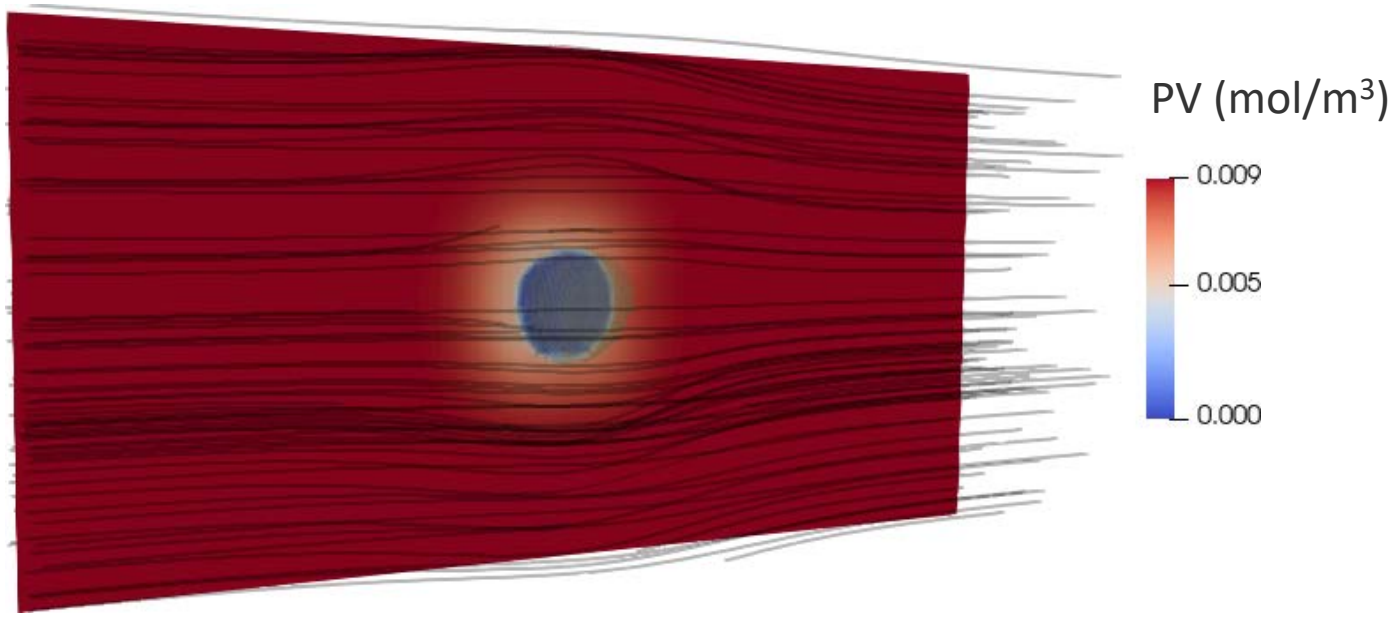




- Directly reconstruct volume fraction field in an AMR mesh
  - X-ray tomography data in the form of MRC/TIFF files
    - Pixel data with a threshold is used to identify solid domain
  - Stereolithography (STL) surface files
    - Ray tracing/Plucker coordinates method to identify solid region

# Catalyst particle simulations

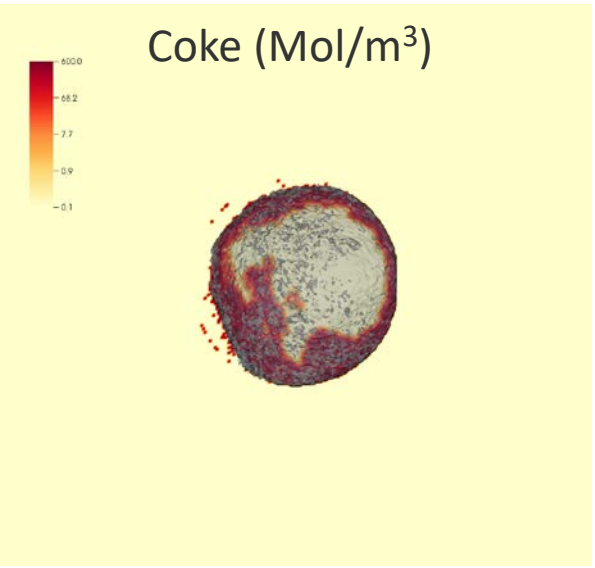



Davison Circulating Riser* at NREL

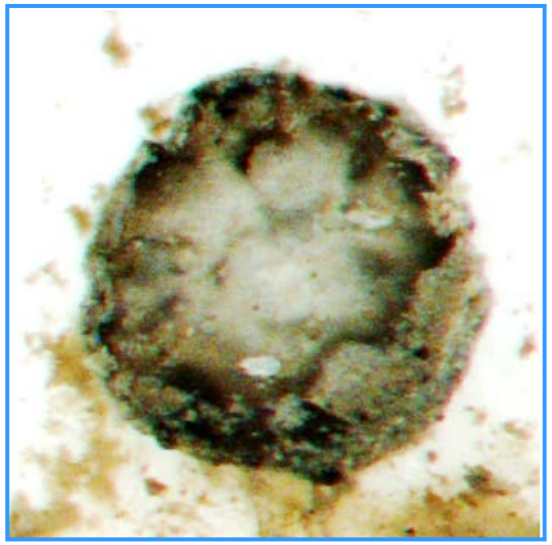
- Setup is similar to experiments done at NREL
  - catalytic upgrading of biomass pyrolysis vapors in a DCR reactor
  - Primary vapors in helium background gas
  - 100 $\mu$m size particle
  - Free stream conditions: P=1 atm., T = 700 K, V = 0.25 m/s

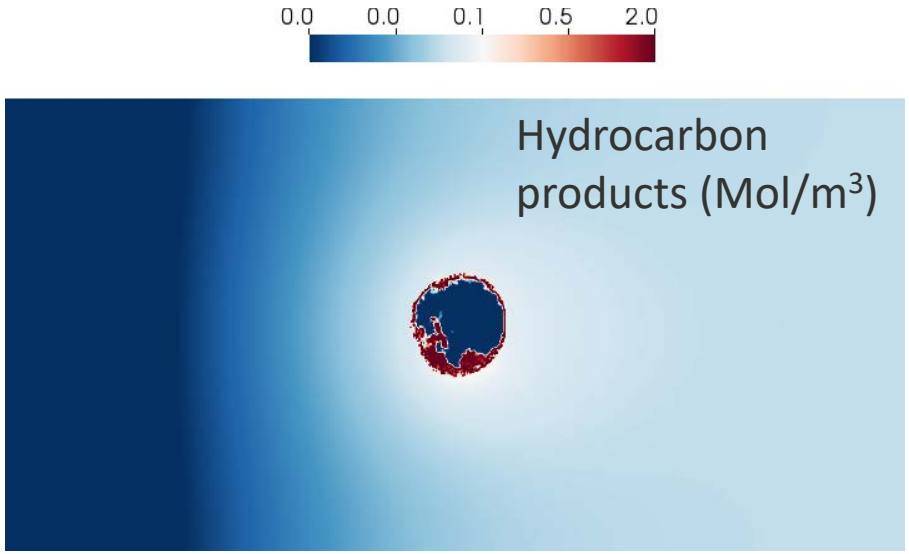
*Jarvis, Mark W., et al. "Catalytic upgrading of biomass pyrolysis oxygenates with vacuum gas oil using a Davison circulating riser reactor." *Energy & Fuels* 32.2 (2018): 1733-1743.

# Catalyst particle simulations

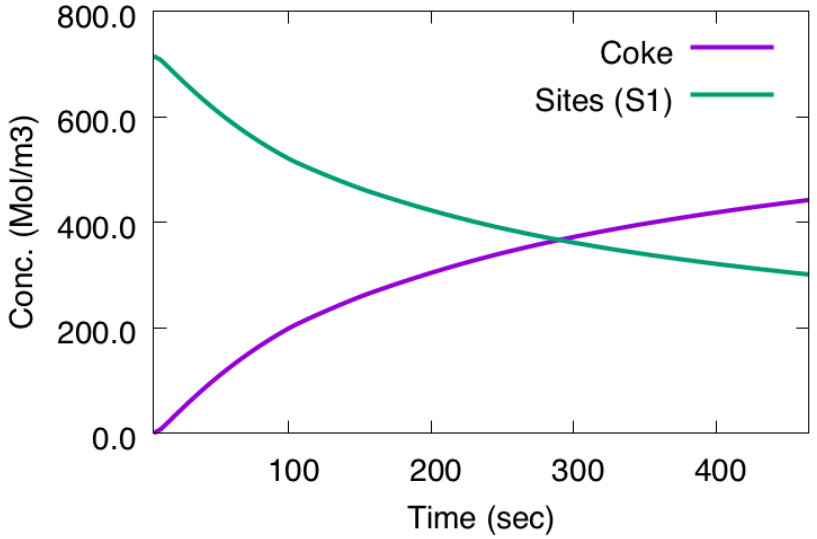



Partially deactivated particle

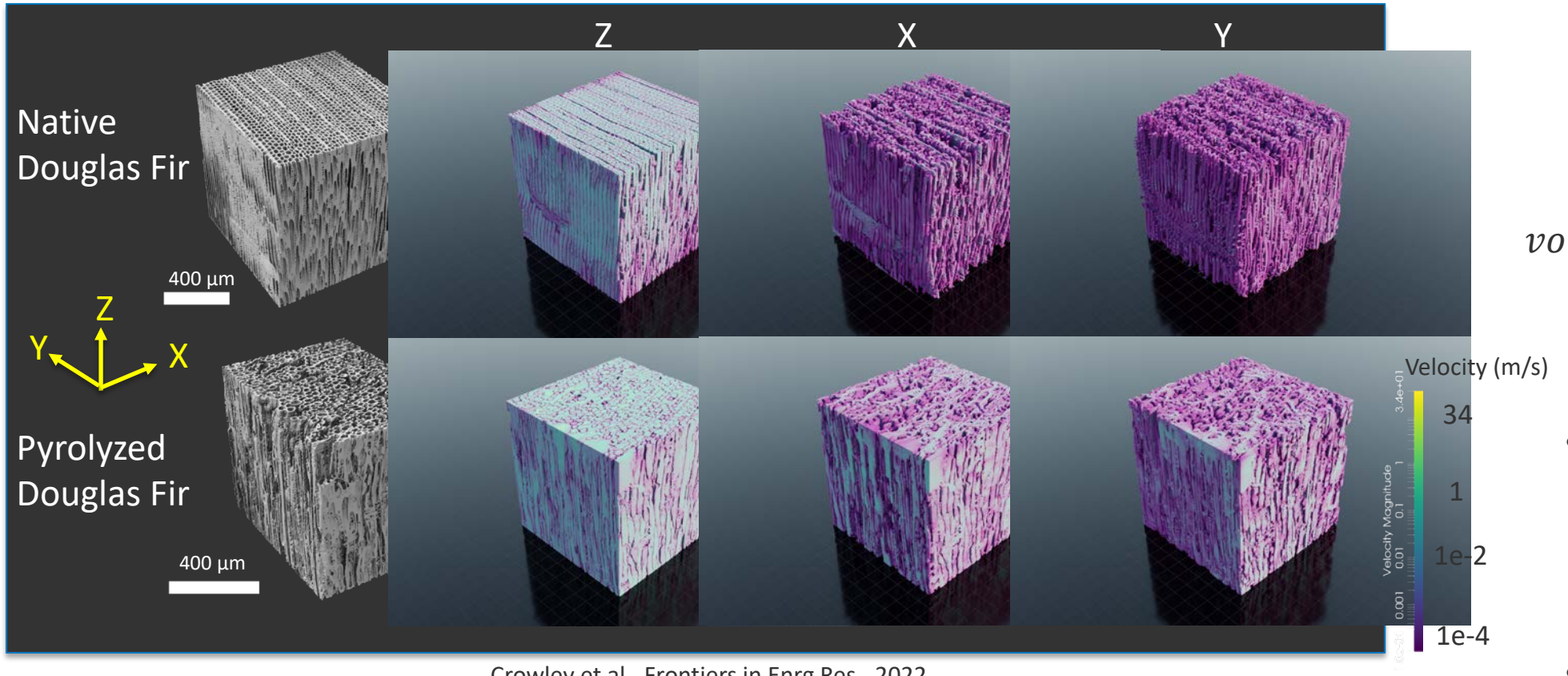





- Coke formation in the outer porous regions
- Qualitatively matches coked catalyst images
- Hydrocarbon products are transported out of porous region
- Site concentration reduces as coking happens
- Deactivation time-scale ~ 10s of mins
  - Matches experimental data



# Permeability of porous structures




Crowley et al., Frontiers in Enrg Res., 2022

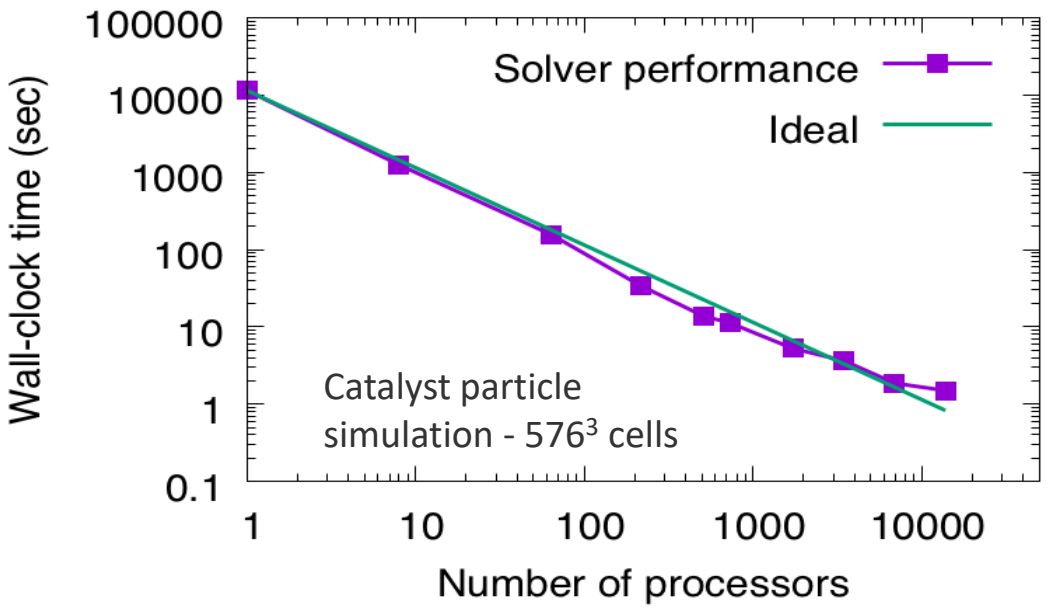
permeability

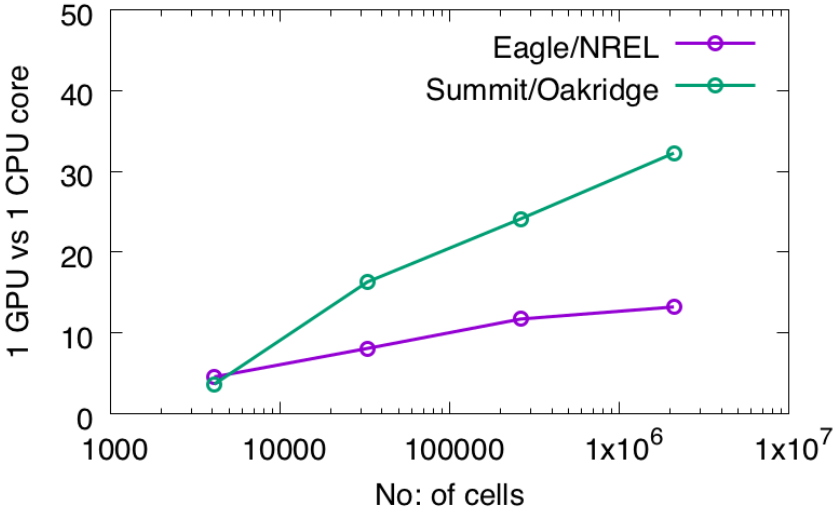
$$volume\ flux\ q = \frac{k}{\mu}\frac{dp}{dx}$$

- Study permeability of porous catalyst/biomass structures
- Anisotropy in permeability
- Pressure gradient is imposed along each direction

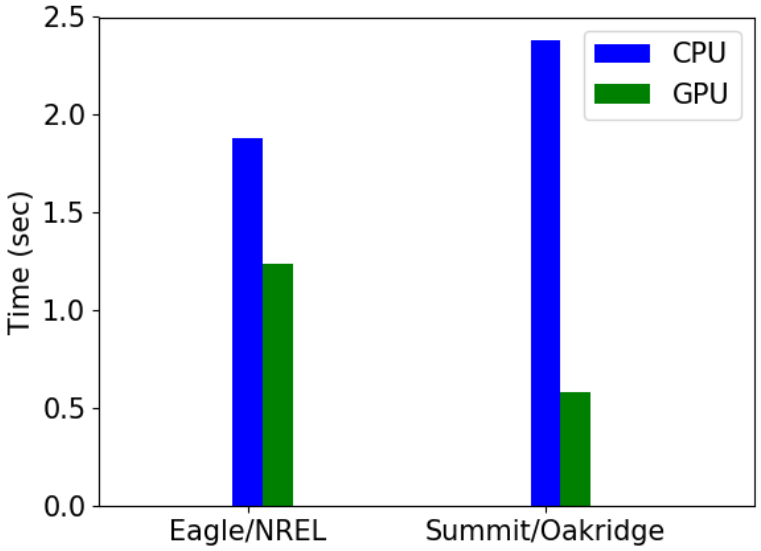
| Direction | Native Douglas fir Calculated (darcy) | Literature range for Douglas fir (darcy) | Pyrolyzed Douglas fir Calculated |
|---|---|---|---|
| **Longitudinal (Z)** | 1.36 | 0.9 to 30 | 2.81 |
| **Radial (X)** | 0.0181 | 8e-5 to 0.002 | 0.229 |
| **Tangential (Y)** | 0.000346 | 1e-5 to 0.001 | 0.133 |

# Computational performance




- Strong scaling observed over 10000 MPI ranks
- A single GPU provides ~ 10X or more performance compared to a single CPU
- ORNL summit shows superior performance compared to Eagle GPUs
- Summit has 6 GPUs per node compared to 2 GPUs on eagle



# Conclusions and future work

- Conclusions
  - Developed an open-source mesoscale modeling tool
    - Adaptive mesh refinement (through AMReX library)
    - Complex geometry representation through immersed boundary method
    - Hybrid parallelization on large scale computing resources
- Future work
  - Incorporate conjugate heat-transfer effects
  - Robust chemistry integration schemes
  - Code documentation

# Questions/comments