NREL
NATIONAL RENEWABLE ENERGY LABORATORY

An Adaptive-Mesh-Refinement Based Computational Tool for Simulating Catalysis at Mesoscale

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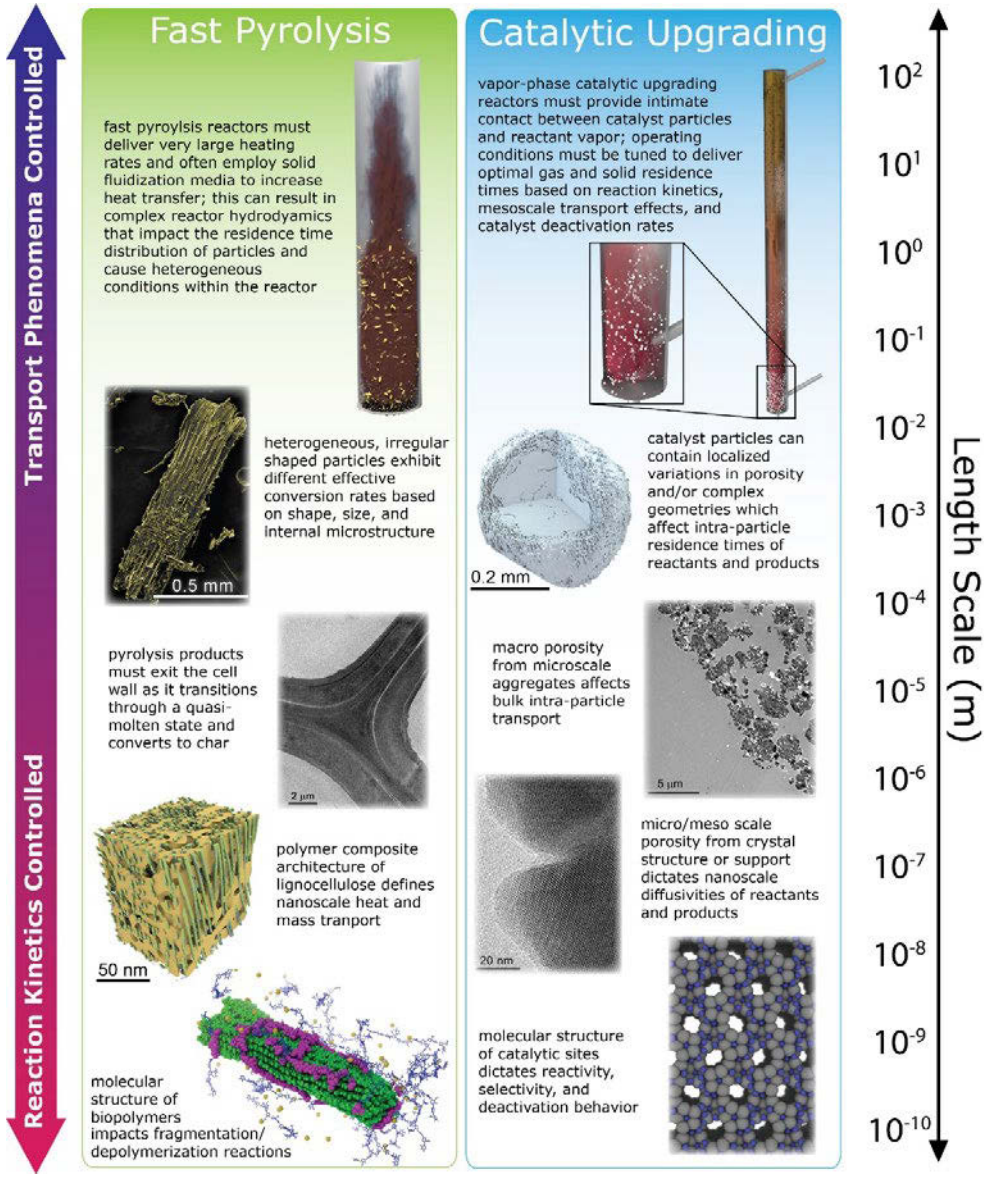
Introduction and Motivation

- Biomass thermochemical conversion and catalytic upgrading are complex multiscale processes.

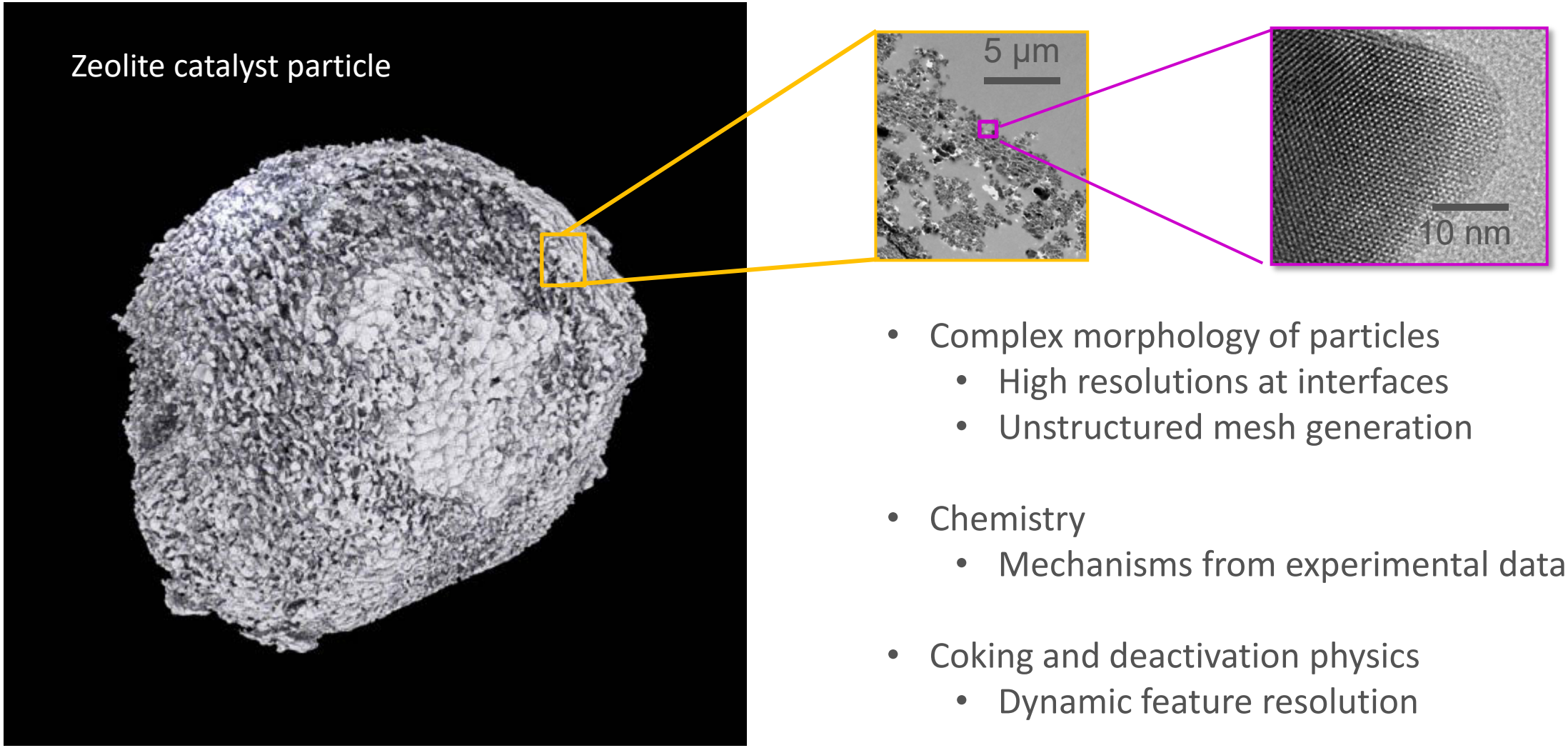
Mesoscale modeling facilitates:

- Connect molecular and reactor scales
- Investigation of intra-particle 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

Mesoscale modeling challenges



- 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

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

Objectives

- Develop a Mesoscale modeling tool
  - Gas-solid flows
    - Catalytic upgrading of biomass vapors
  - Porous media flows
  - Scalable on current and future DOE 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

Summit Supercomputer at Oakridge National Laboratory

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

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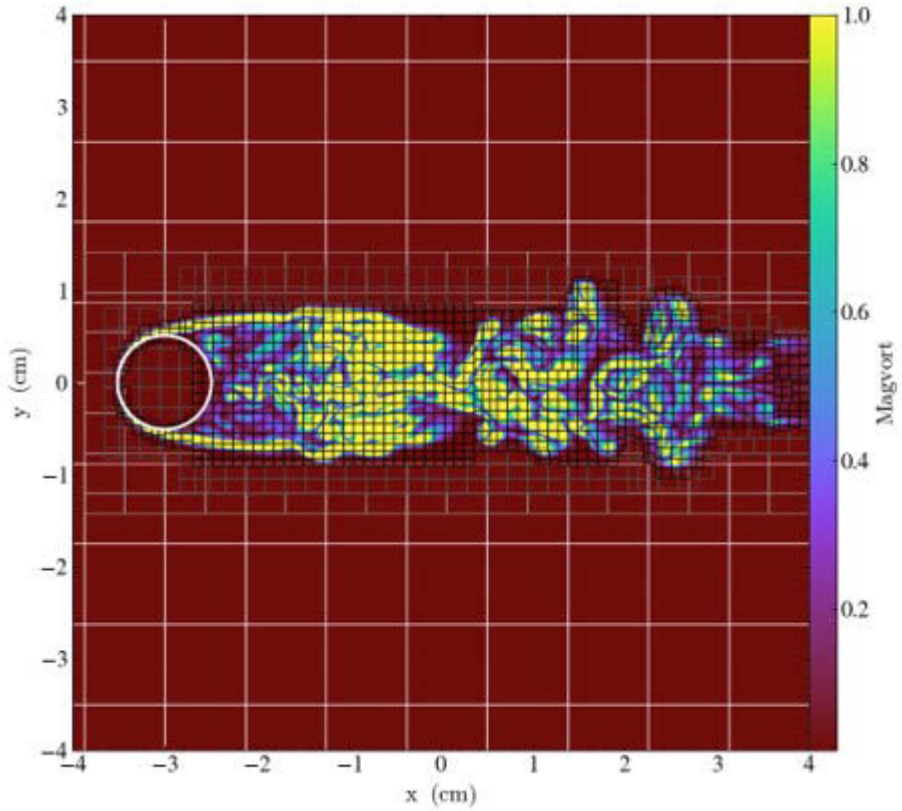
Approach

- 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.

Governing equations

Compressible Navier-Stokes equations (gas phase)

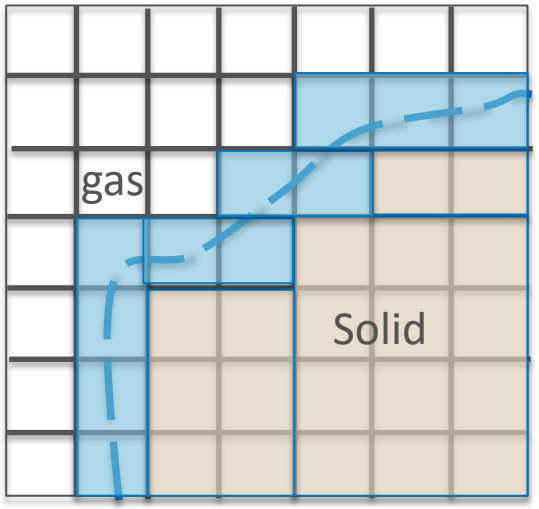
$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{U}) = 0$$

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

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

Species transport and reaction (both gas and solid phase)

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

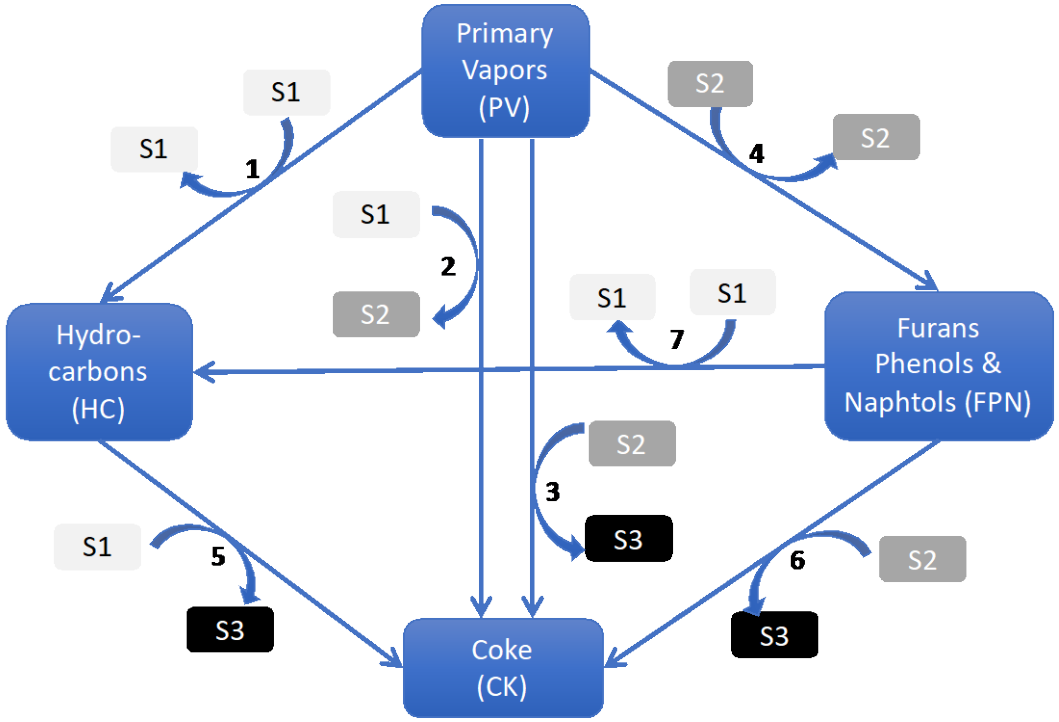


Reflection based boundary conditions for velocity at the interface

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

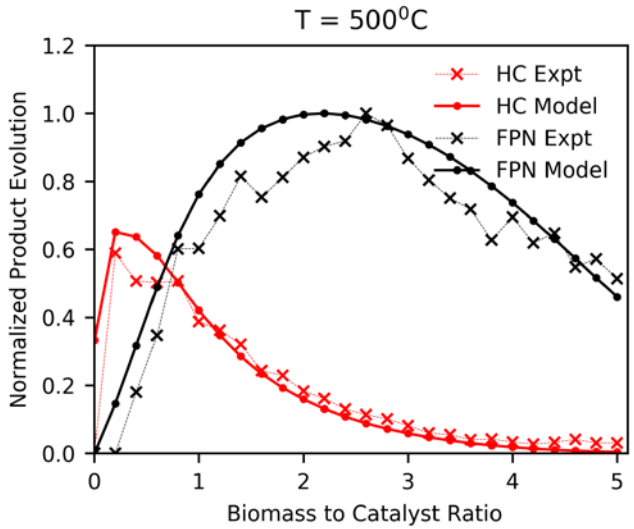
Liou, Meng-Sing. "A sequel to AUSM, Part II: AUSM+-up for all speeds." *Journal of computational physics* 214.1 (2006): 137-170.

Chemistry



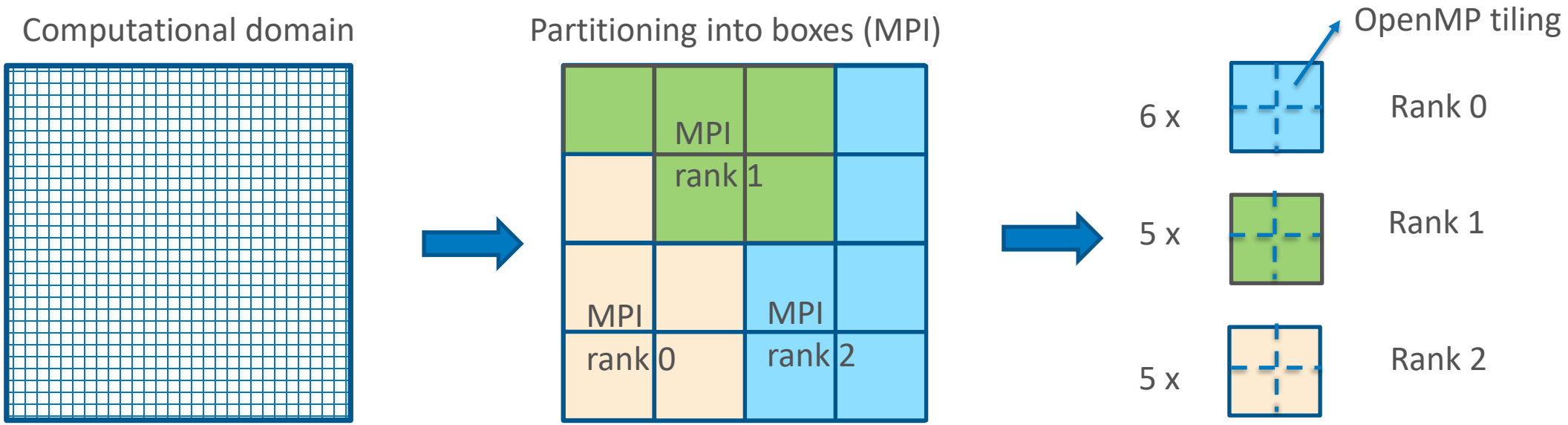
S1- Fresh Active Sites S2- Intermediate Sites S3- Deactivated Sites

No.	Reaction	k[m^3/(mol.s)] @ T=500^{0}C
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



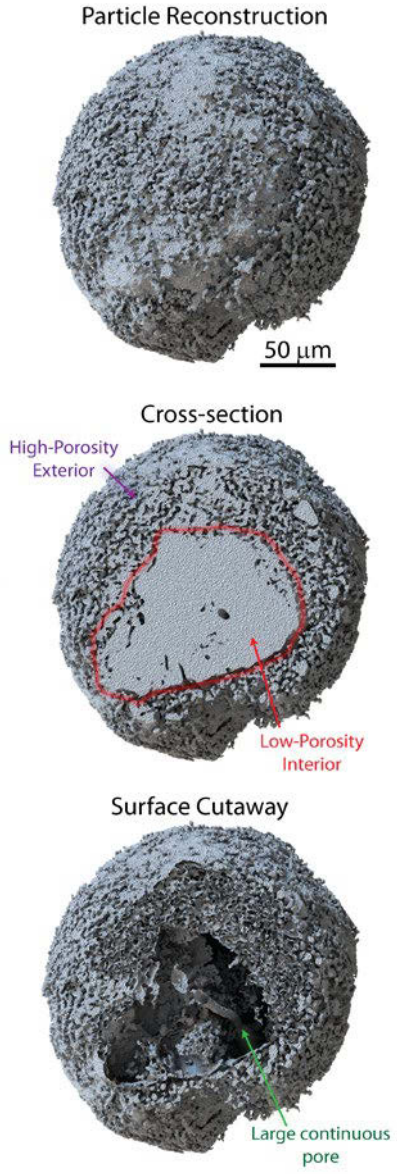
- Validated 7 species chemistry for Zeolite catalyst
  - Gas phase - primary vapors, products
  - Solid phase - Coke and catalyst sites
- Operator splitting approach to couple with transport

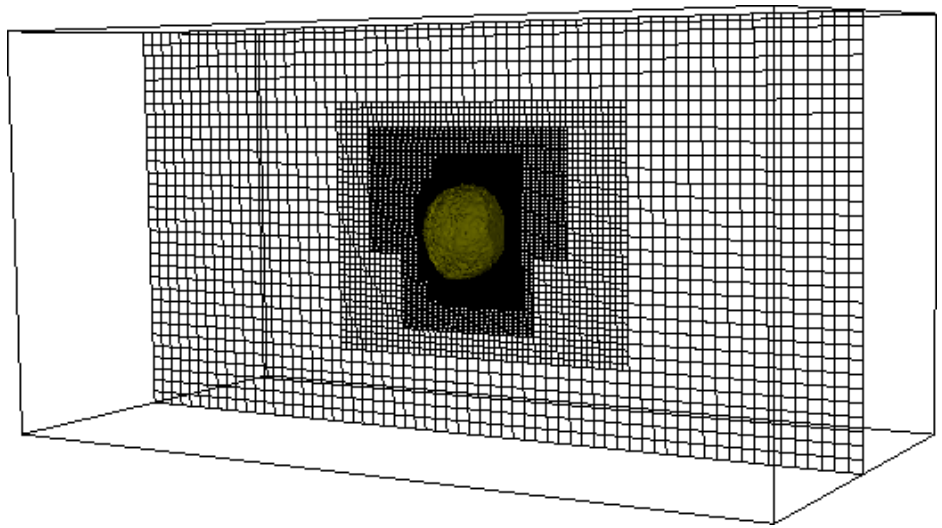
Parallelization strategy

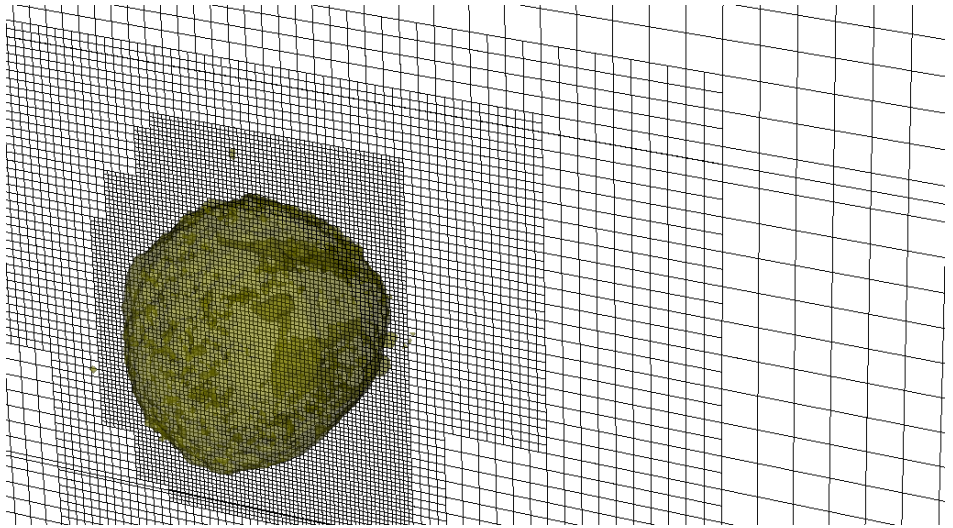


- Boxes (32^3 or 64^3 size) per rank run using either
  - Openmp threads on smaller tiles
  - multiple GPU streams
- Stencil operations executed through asynchronous GPU kernels
  - Inlined functions called from within C++ lambda
- Data movement managed through AMReX
  - Large chunk of unified memory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

Catalyst particle simulations

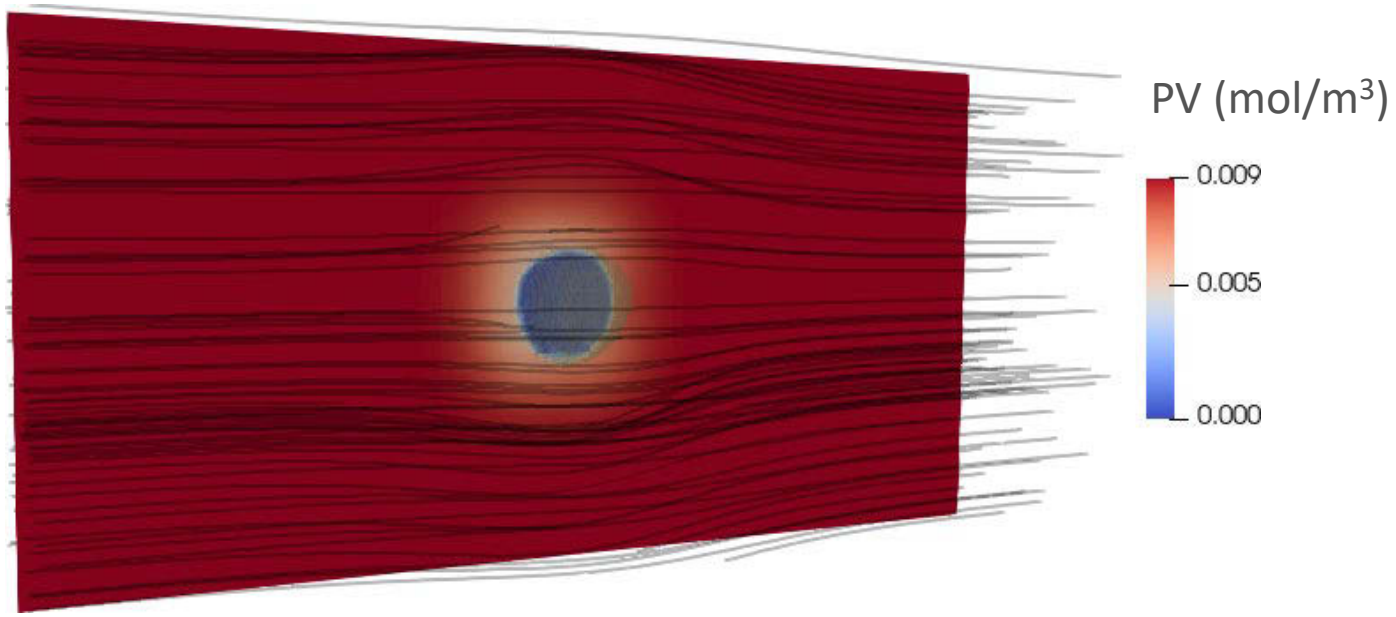






- Directly reconstruct volume fraction field in an AMR mesh
  - X-ray tomography data in the form of MRC/TIFF files
    - Pixel data with a threshold is used to identify solid domain
  - Stereolithography (STL) surface files
    - Ray tracing/Plucker coordinates method to identify solid region

Catalyst particle simulations

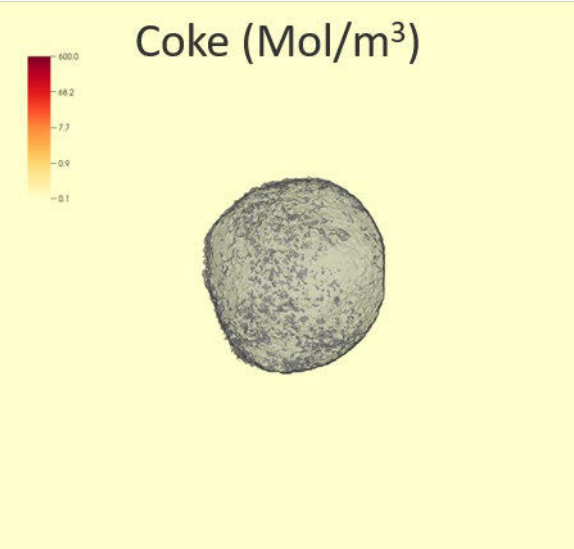


Davison Circulating Riser* at NREL

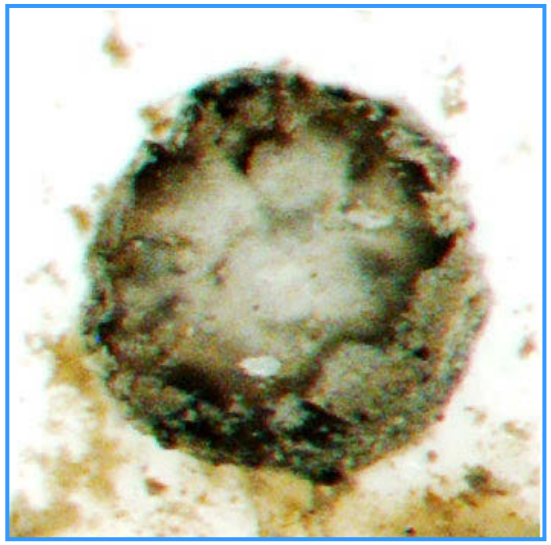
- Setup is similar to experiments done at NREL
  - catalytic upgrading of biomass pyrolysis vapors in a DCR reactor
  - Primary vapors in helium background gas
  - 100 μm size particle
  - Free stream conditions: P=1 atm., T = 700 K, V = 0.25 m/s
- First, we solve non-reacting flow to steady state

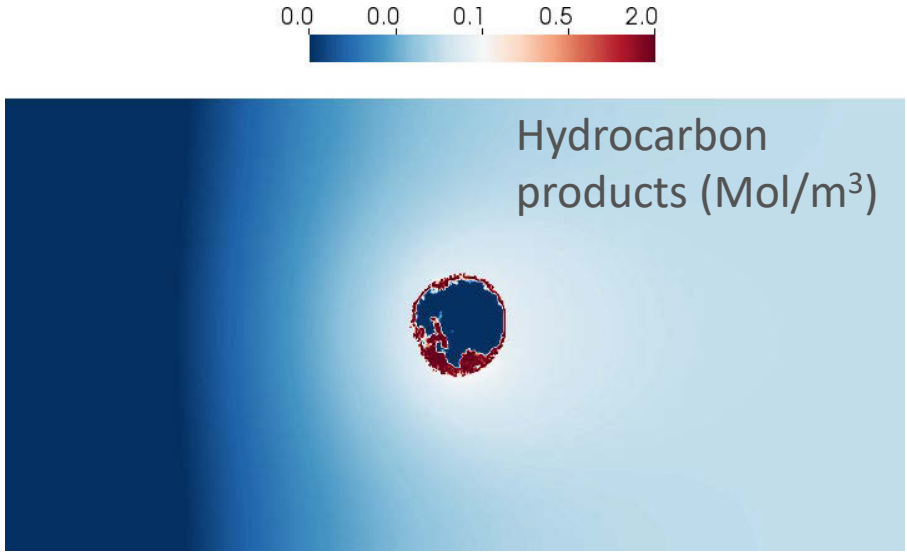
*Jarvis, Mark W., et al. "Catalytic upgrading of biomass pyrolysis oxygenates with vacuum gas oil using a Davison circulating riser reactor." *Energy & Fuels* 32.2 (2018): 1733-1743.

Catalyst particle simulations

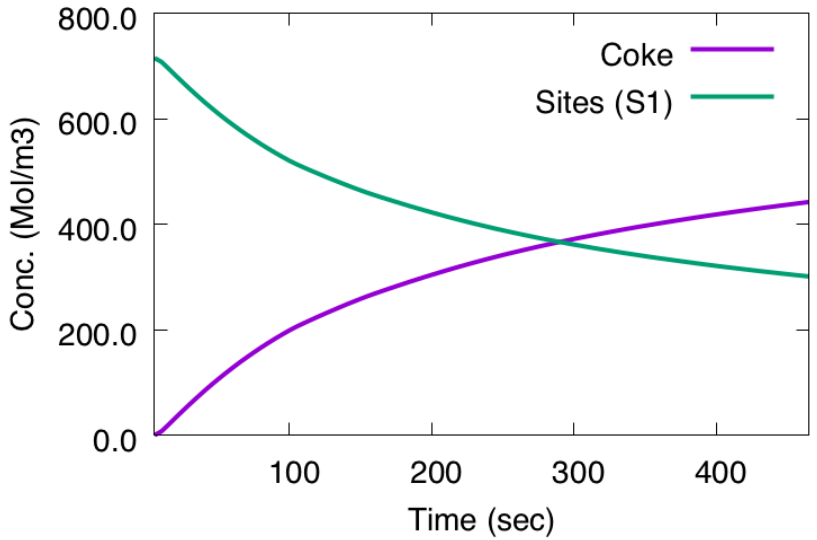


Partially deactivated particle

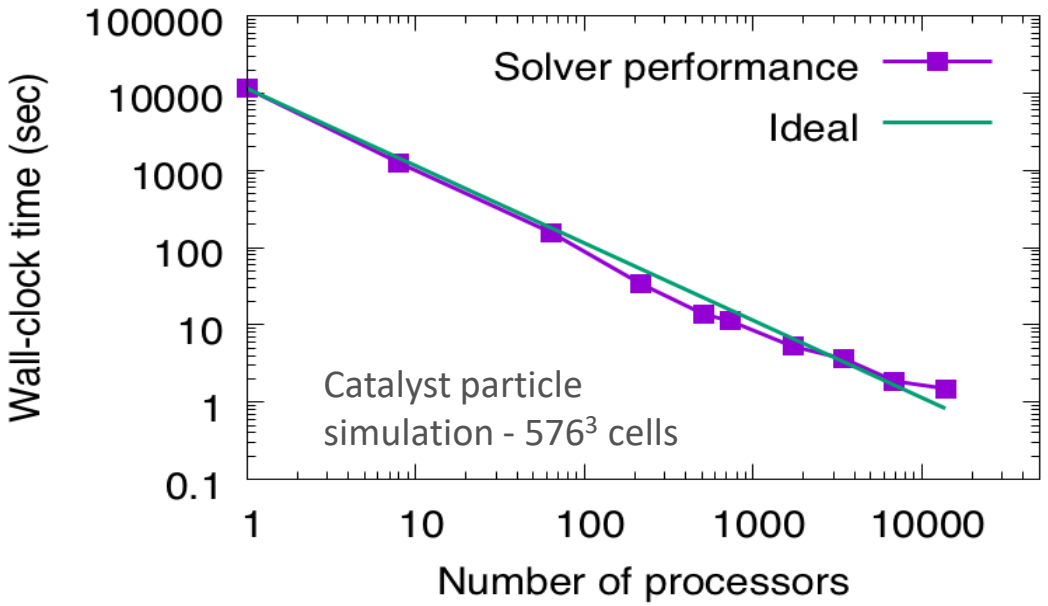


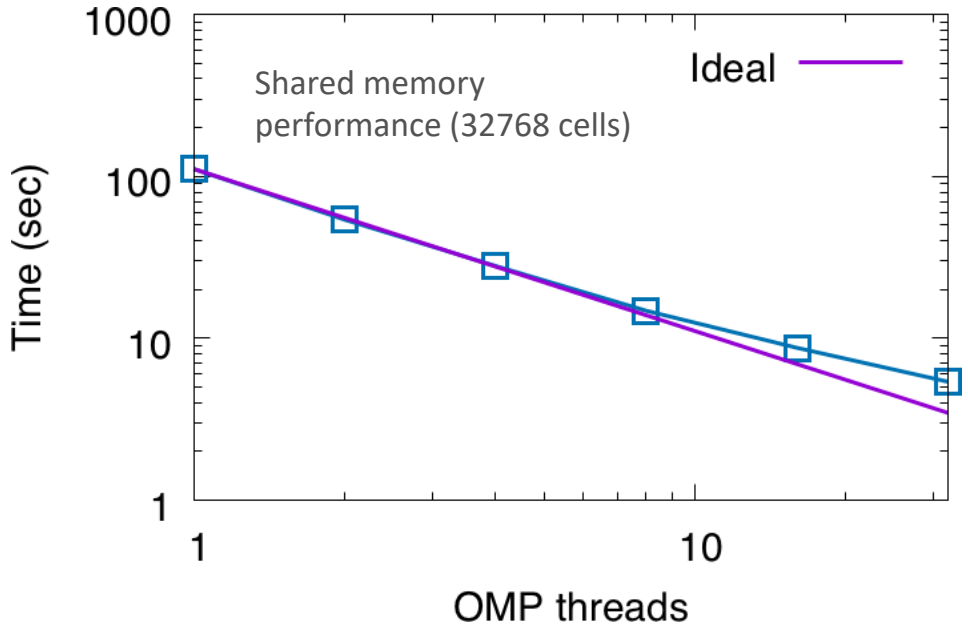


- Coke formation in the outer porous regions
- Qualitatively matches coked catalyst images
- Hydrocarbon products are transported out of porous region
- Site concentration reduces as coking happens
- Deactivation time-scale ~ 10s of mins
  - Matches experimental data



Computational performance

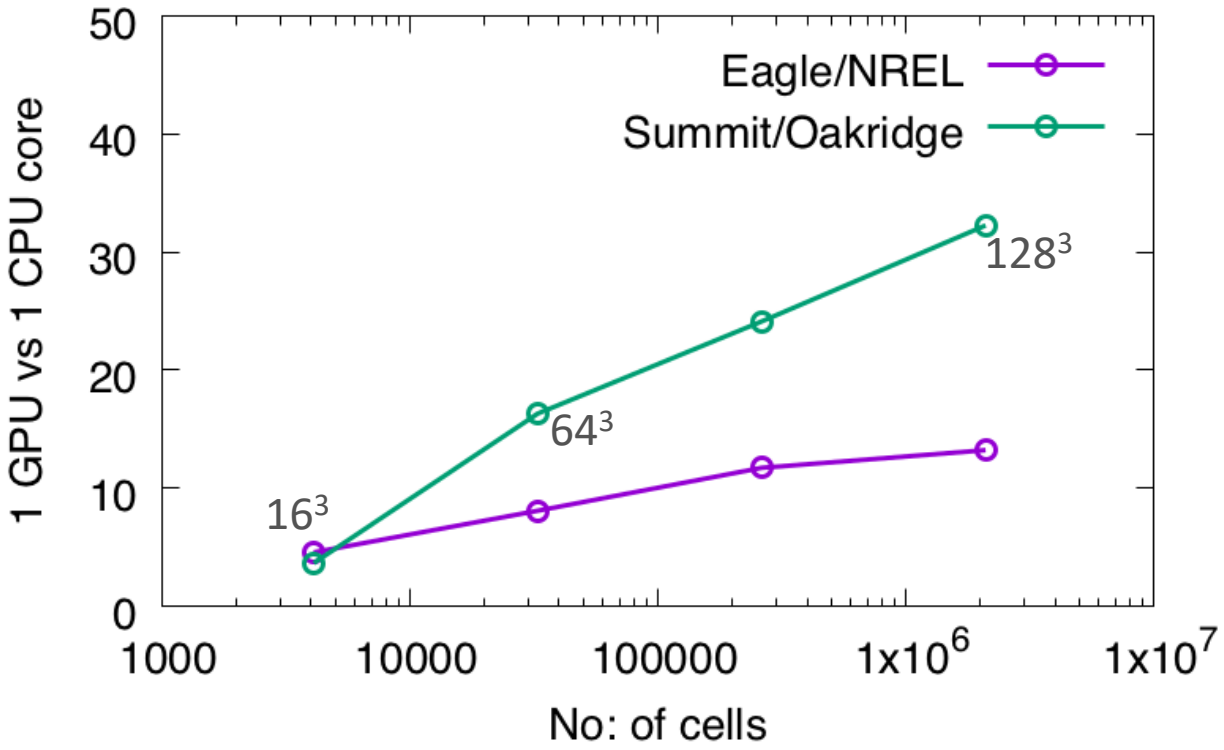




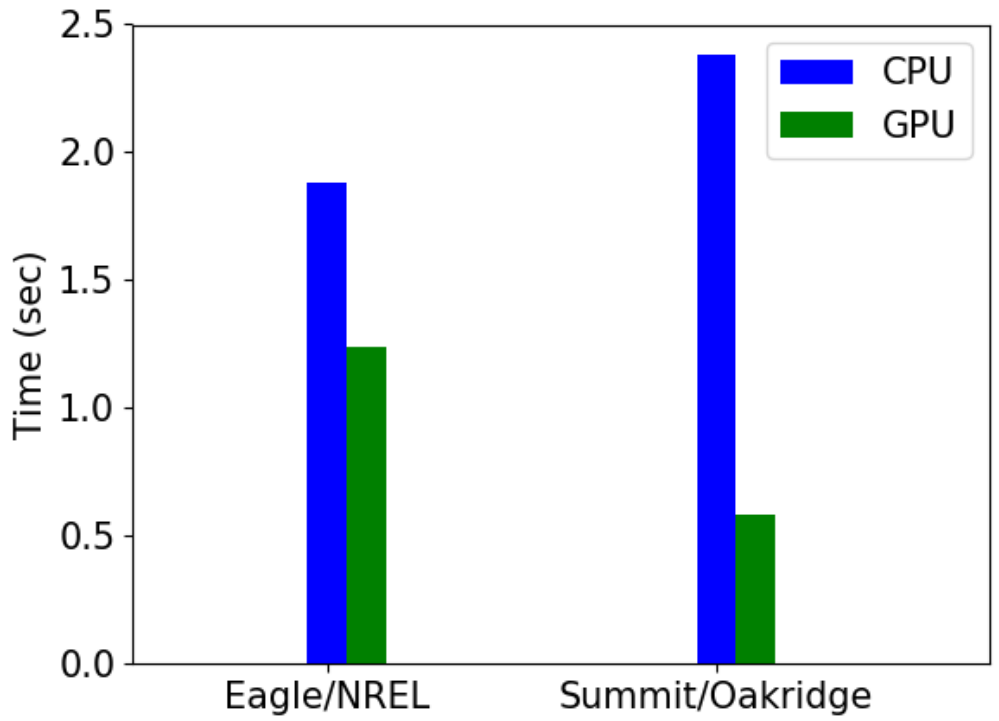
- Strong scaling observed over 10000 MPI ranks
  - 20% of NREL Supercomputer, Eagle
- OpenMP also shows good scaling on 32x32x32 boxes

GPU performance

Serial performance



Single node performance



- 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 a mesoscale modeling tool with
    - Using AMReX library
    - Adaptive mesh refinement
    - Complex geometry representation
    - Hybrid parallelization on HPC
  - Studied catalysis of biomass pyrolysis vapors over zeolite particle
    - Coking phenomena qualitatively matched imaging
- Future work
  - Performance optimizations on GPU
  - Incorporate heat-transfer effects
  - Other catalysis problems
- Questions/comments – email me at Hariswaran.Sitaraman@nrel.gov

www.nrel.gov

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