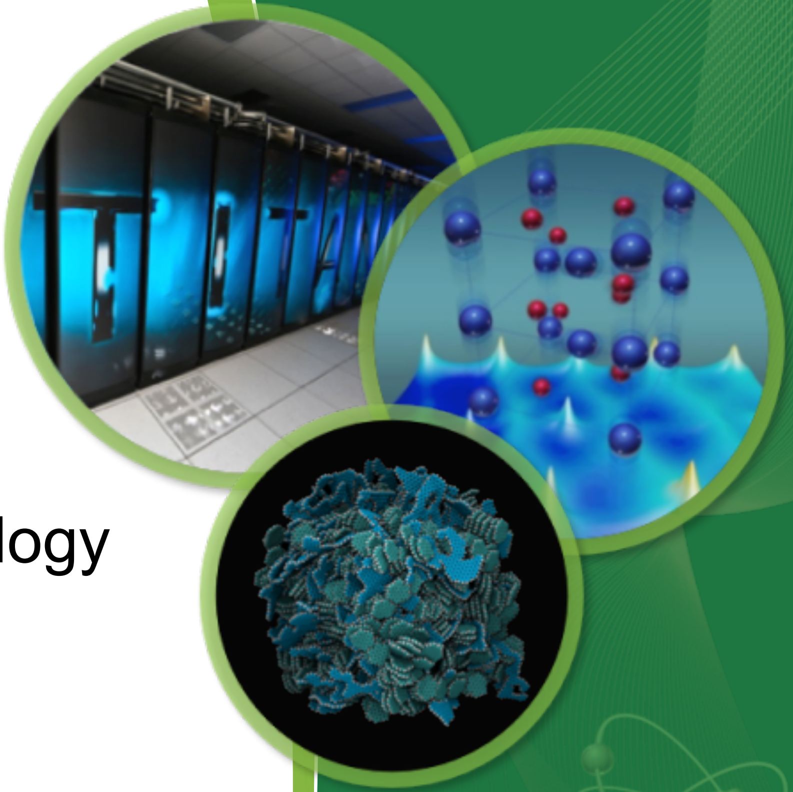
Combining high and low-order computational models to simulate biomass fast pyrolysis reactors

Gavin Wiggins, Emilio Ramirez,
Jonathan Sutton, Jessica Torres,
Charles Finney, and Stuart Daw



AIChE & 8th World Congress on Particle Technology
Monday, April 23, 2018

CCPC

Consortium for Computational
Physics and Chemistry
cpcbiomass.org

ORNL is managed by UT-Battelle
for the US Department of Energy

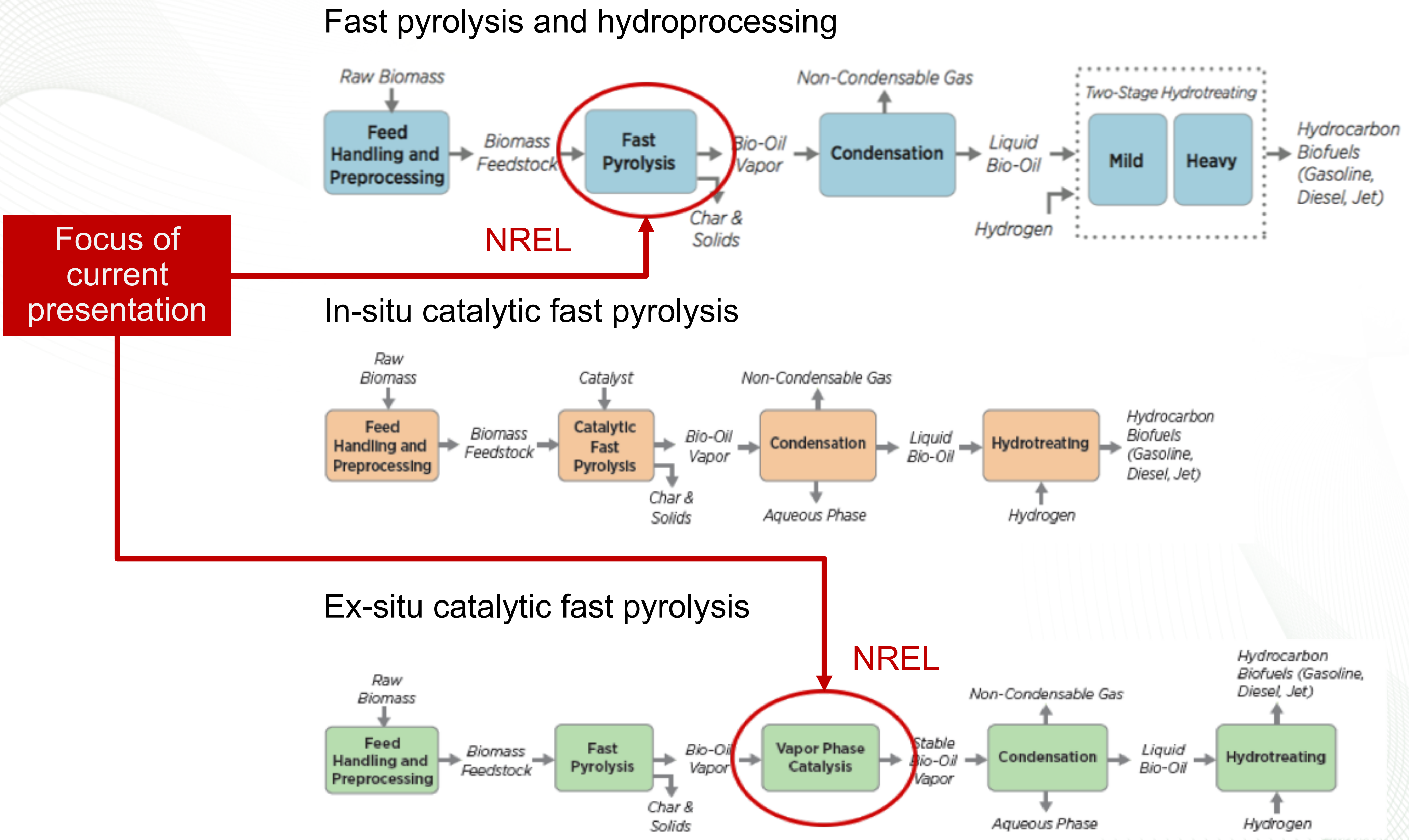
OAK RIDGE National Laboratory | NATIONAL TRANSPORTATION RESEARCH CENTER

Problem Statement

Multiphase reactors for solid biomass thermochemical conversion involve complex mixing and reaction processes that are hard to analyze and optimize.

- Biomass particles are extremely non-spherical in shape and non-homogeneous in material properties
- Explicit computational fluid dynamics (CFD) models are expensive and time consuming to use
- Reaction kinetics (both catalytic and non-catalytic) are usually poorly defined
- Experimental model validations are often unavailable

Pathways to consider for fast pyrolysis of biomass



Fast pyrolysis of biomass involves two key steps

1. Rapid particle heating to decompose the raw biomass into a maximum amount of “tar” (condensable liquid vapors)
   - Depends strongly on particle residence times, gas-solid and intra-solid heat and mass transfer, solid-phase kinetics
2. Transport of tar vapors from their release point while minimizing their further decomposition to light gases and char
   - Depends strongly on gas-solid mass transfer, gas-phase kinetics, and gas residence times

Accurately quantifying the above processes is the primary challenge of fast pyrolysis reactor modeling.

Vapor phase upgrading of bio-oil involves similar issues

1. Complex reactions on catalyst particles to convert raw bio-oil (tars) into maximum yield of target species (transportation fuel or high-value chemicals)
   - Depends strongly on catalyst particle residence times, gas-solid and intra-particle heat and mass transfer, solid-phase kinetics (including catalyst deactivation)
2. Transport of product vapors from their release point while minimizing further reactions to undesirable side products
   - Depends strongly on gas-solid mass transfer, gas-phase kinetics, gas residence times

Accurately quantifying the above processes is the primary challenge of vapor phase upgrading reactor modeling.

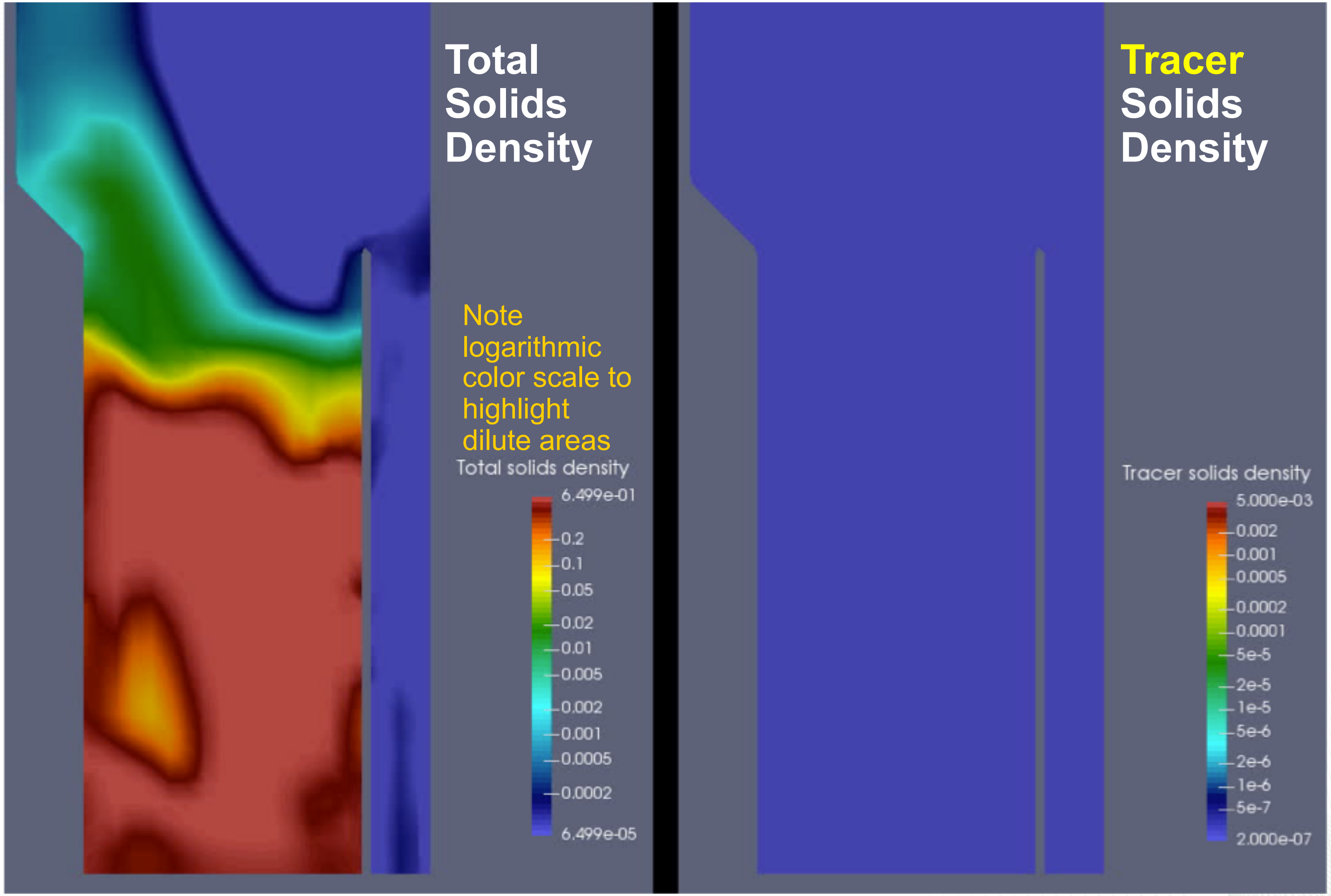
CFD modeling of multi-phase reactors provide insight not available from experiments

- Bench and pilot-scale reactors are typically not equipped to measure crucial model parameters such as gas/solids residence times
- CFD simulations provide insight to reactor conditions not readily available from experiments (residence times, flow regimes)

Issues with CFD simulations

- Complexity and cost of doing detailed simulations limit ability to evaluate parameter sweeps (operating conditions, reactor geometry)
- Low-order models are needed to “compress” the information generated by CFD models to improve physical understanding and allow more rapid predictions

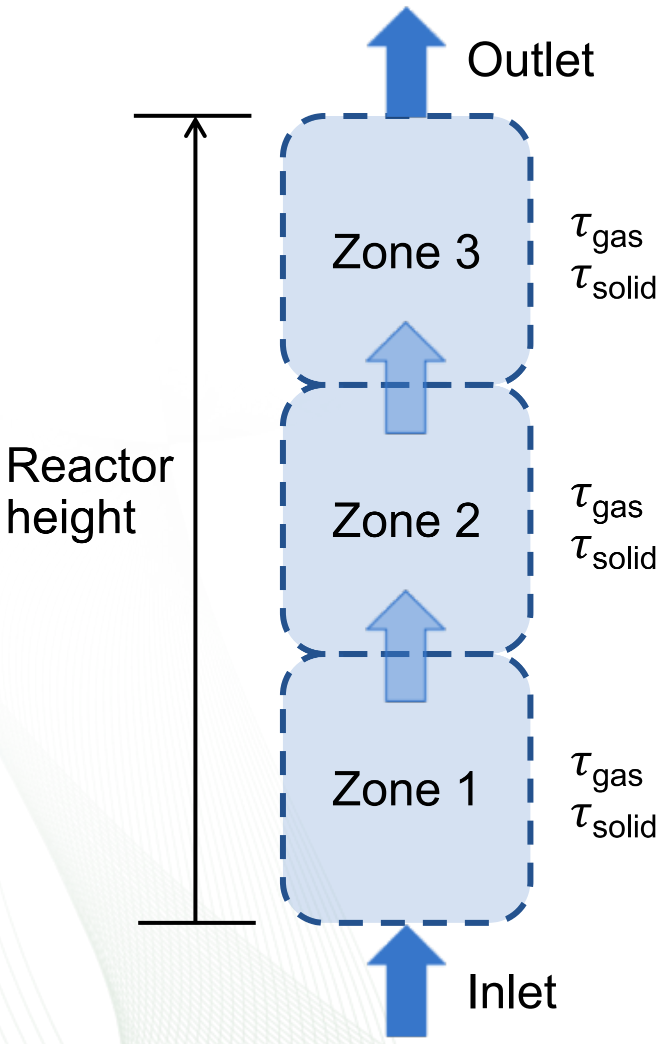
CFD can provide insight on hydrodynamics within reactor



Time dependent void fraction within fluidized catalyst bed for vapor phase upgrading of pyrolysis products. Source: Charles Finney and Jessica Torres.

Mixing zone models can approximate key CFD features

- Characteristic mixing zones can be used to replicate CFD RTDs for solids and gases for selected cases
- Correlations can be used to estimate mean gas and solid phase properties (e.g. void fraction, drag/slip, pressure drop) at steady-state conditions
- Ideal reactor models (CSTR, PFR) can be applied to each zone using appropriate kinetics
- Reaction kinetics can be added explicitly to each zone
- Zone models can be used to interpolate between selected CFD simulations



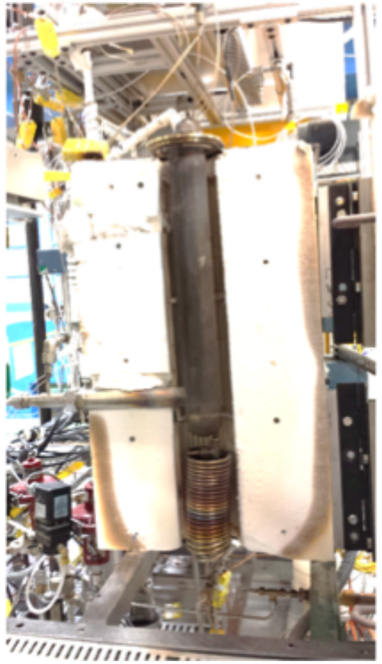
Components of a reactor zone model to account for different regions of mixing in the system.

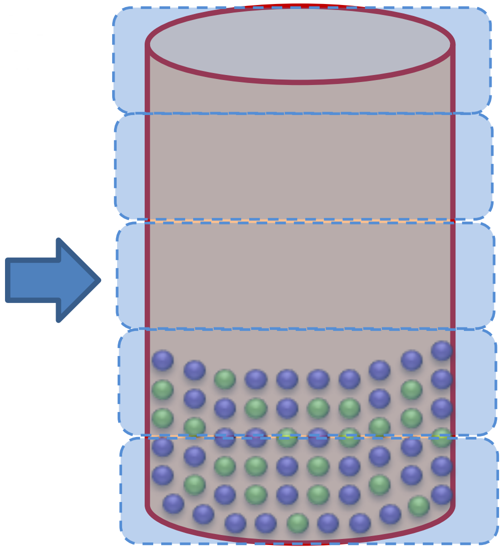
Series CSTR zone models have been useful for pyrolyzers

Bubbling fluidized bed reactor can be represented as a series of CSTRs at steady-state conditions.

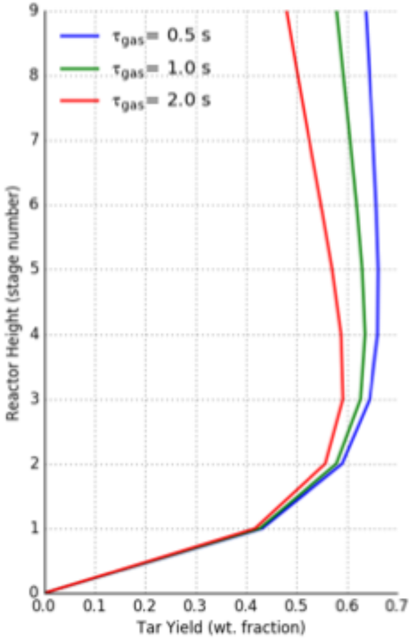
Kinetic parameters from Liden 1988.

Overall gas and solid residence time divided into each stage.

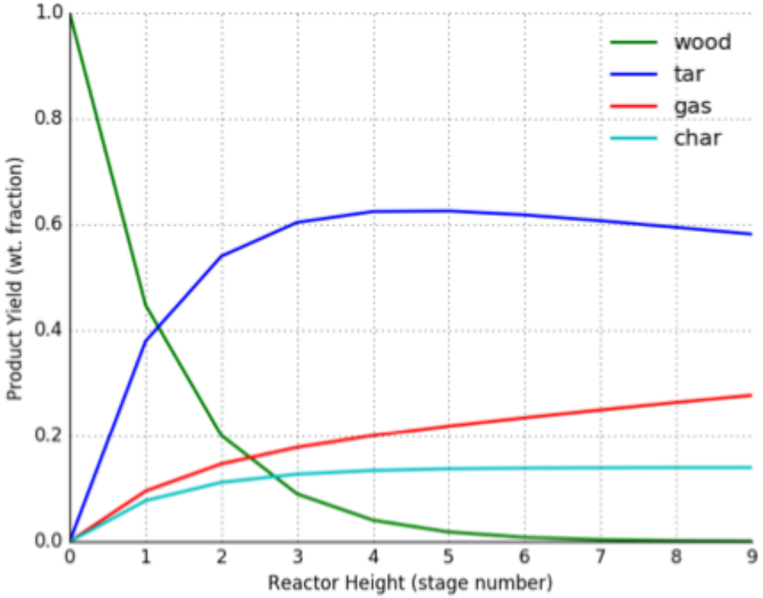




BFB pyrolysis reactor as a series of CSTR reactors. For this example, N = 5 and $\tau_i = \tau_{total} / N$.



Tar yield along height of reactor at different gas residence times with τ_{solid} = 4s, N = 9 at 500°C.

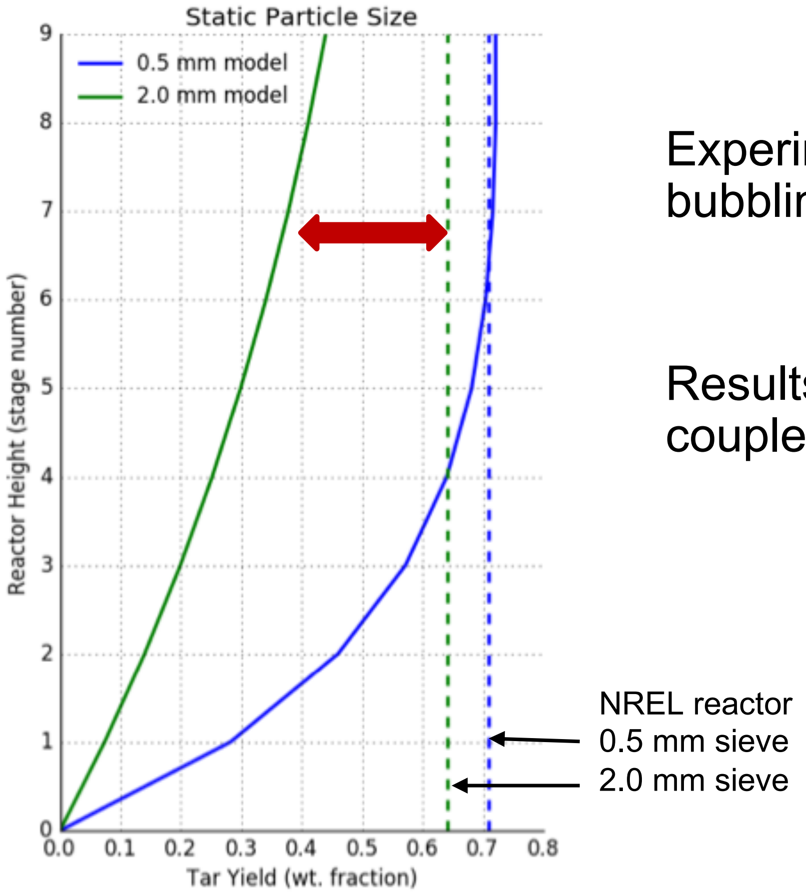


Product yields based on Liden 1988 kinetics at 500°C with τ_{gas} = 1s, τ_{solid} = 3s and N = 9.

Series CSTR pyrolyzer models have produced useful info

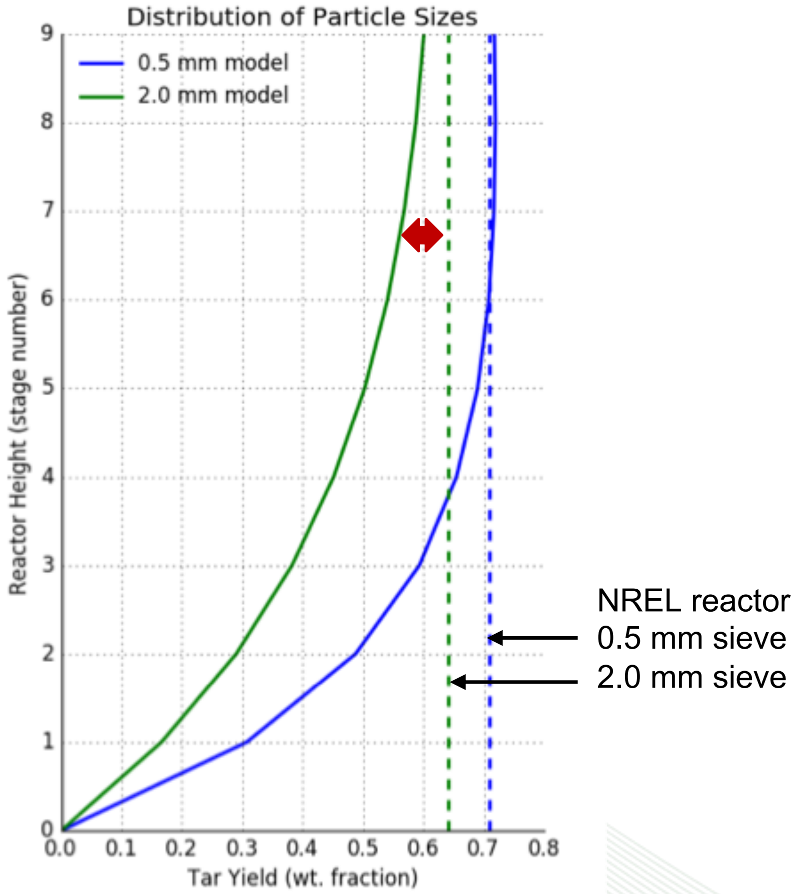
Products (wt. %)	0.5 mm sieve		2.0 mm sieve	
	Experiment	Model	Experiment	Model
Total liquids	**70.8 ± 1.1**	**72.1**	**63.5 ± 1.9**	**44.0**
Char	9.5 ± 0.1	13.7	11.7 ± 1.3	8.2
Gas	15.5 ± 0.6	12.3	18.7 ± 0.8	6.5

Products (wt. %)	0.5 mm sieve		2.0 mm sieve	
	Experiment	Model	Experiment	Model
Total liquids	**70.8 ± 1.1**	**72.1**	**63.5 ± 1.9**	**60.1**
Char	9.5 ± 0.1	13.7	11.7 ± 1.3	11.3
Gas	15.5 ± 0.6	12.3	18.7 ± 0.8	9.6



Experimental data from 2-inch diameter bubbling fluidized bed reactor at NREL.

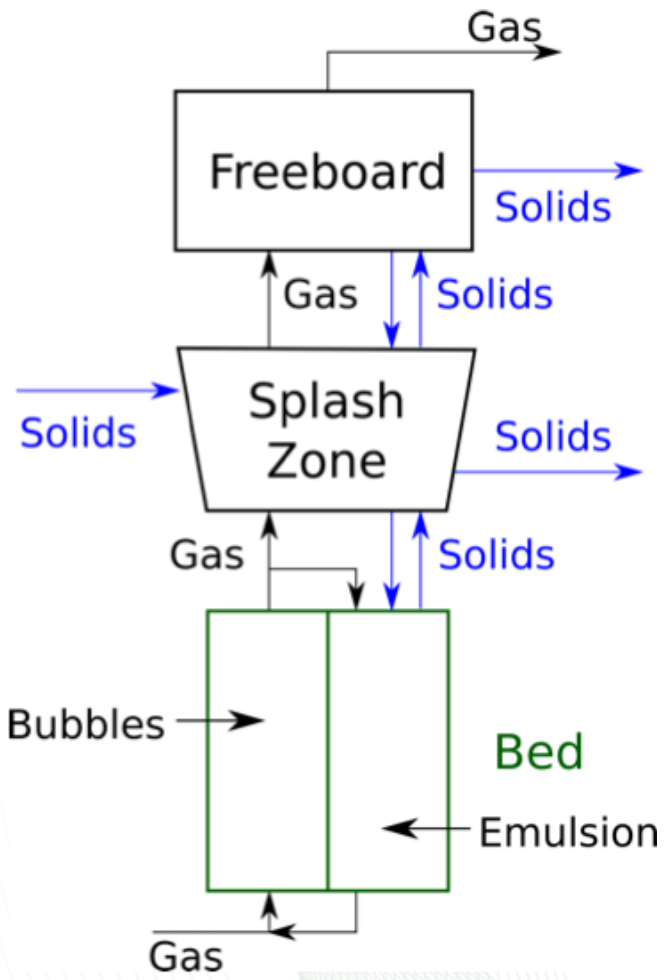
Results are from Dsv particle model [2] coupled to a low-order reactor model.



[2] Wiggins, Gavin M., Peter N. Ciesielski, and C. Stuart Daw. **Low-Order Modeling of Internal Heat Transfer in Biomass Particle Pyrolysis**. *Energy & Fuels* 30, no. 6 (2016): 4960-4969.

We are applying a similar approach to VPU reactors

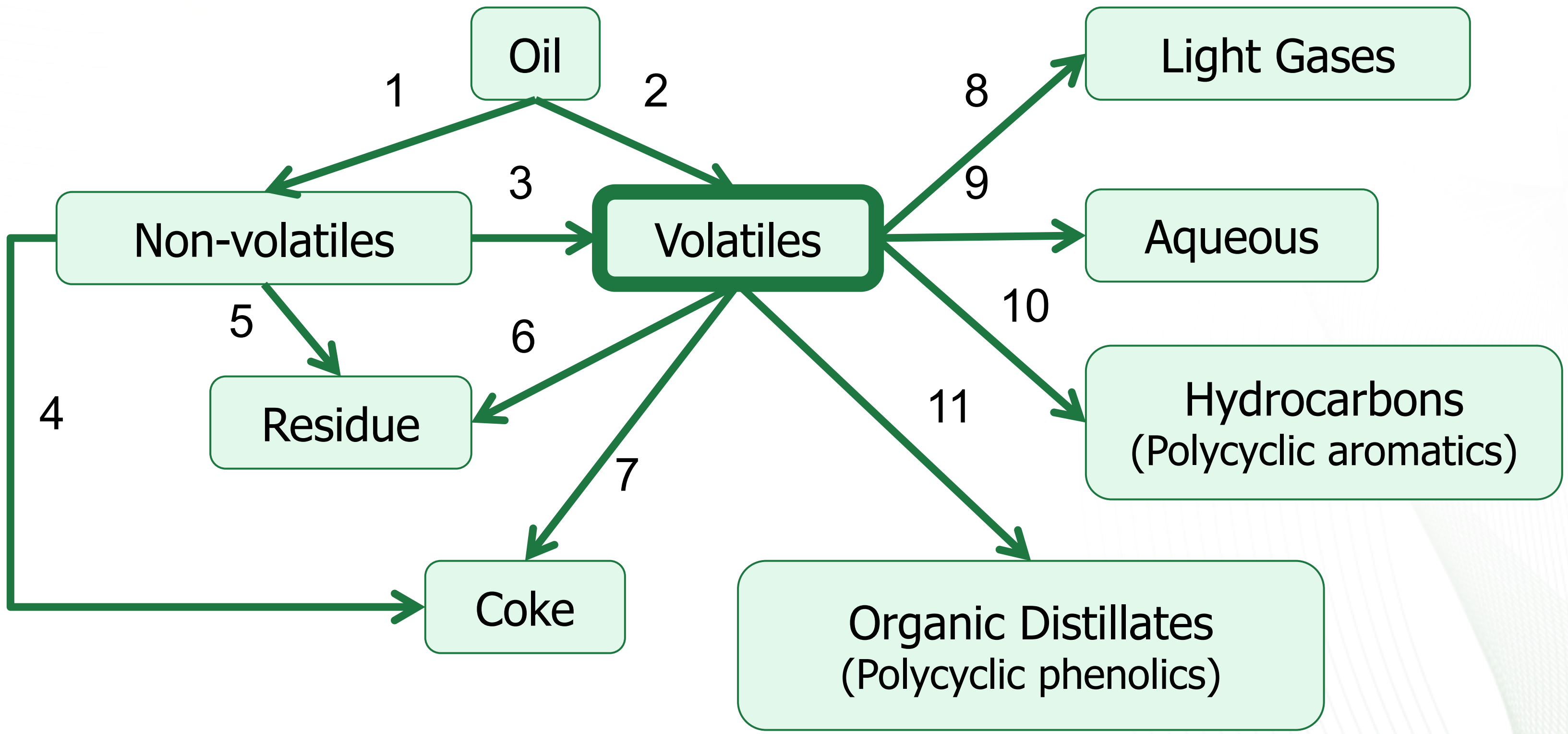
- Identify characteristic zones in the VPU bubbling bed and assign ideal mixing approximations to each zone
- Hydrodynamics
  - recirculation of gas/solids in bed
  - circulation of solids between splash zone and freeboard
  - unidirectional of gas flow from splash zone to freeboard
- Solids elutriation rates from transport disengagement theory



Divide vapor phase upgrader into multiple zones based on mixing characteristics observed from simulations. Source: Jonathan Sutton.

A major challenge is getting reliable VPU catalytic kinetics

- Adjaye kinetics developed for upgrading of liquefaction bio-oil
- Had to increase rates by 10^4 to get yields comparable to experiment



Modified catalytic conversion scheme from Adjaye, J.D., and N.N. Bakhshi. 1995.

With improved kinetics, we expect to investigate trends in VPU conversion performance with operation

Low-order VPU Model

Model inputs

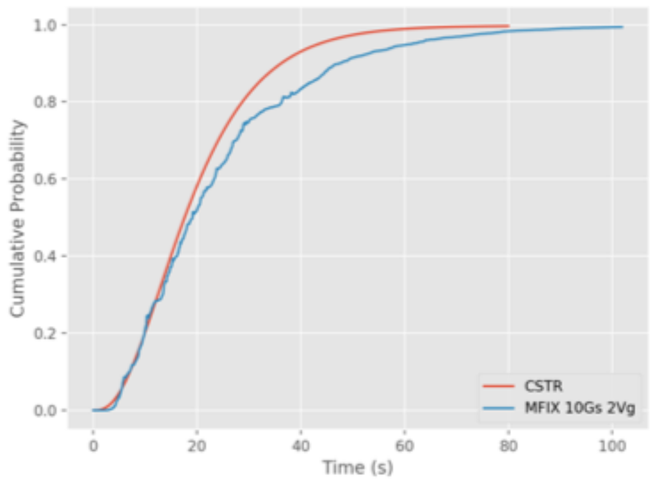
- feedstock/catalyst type
- particle size distribution
- particle shape
- reactor geometry
- kinetic mechanism

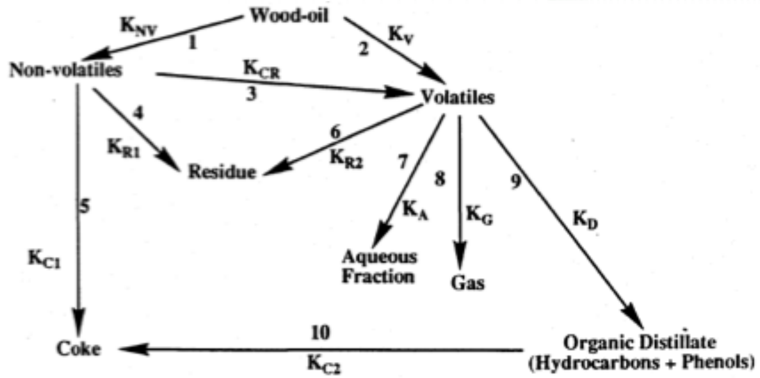
Hydrodynamics: RTD, pressure profile, velocity profile

Vapor Phase Upgrading: catalyst coking, vapor conversion

Model outputs

- solids and gas RTD
- conversion (yields)
- extent of coking
- axial pressure profile
- choking velocity
- operating regimes





Current status

- Working with experimental setups at NREL and NETL to validate and improve reactor models
- Validating low-order models with detailed CFD simulations at ORNL and NETL
- Developing catalyst kinetics at NREL to improve VPU models

Next steps

- Investigate scale-up issues from bench to pilot scale
- Open source Python package for developing reactor models

Upcoming talks by CCPC members

Computational Fluid Dynamic Study of Biomass Vapor-Phase Upgrading Process. Xi Gao, Tingwen Li, William Rogers, Rupen Panday, Jonathan Higham, Greggory Breault, and Jonathan Tucker.

Wednesday, 1:30pm, Crystal Ballroom K

Computational Study on Biomass Fast Pyrolysis: Hydrodynamic Effects in a Laboratory-Scale Fluidized Bed. Emilio Ramirez, Tingwen Li, Mehrdad Shahnam, and Stuart Daw.

Thursday, 2:06 pm, Crystal Ballroom E

Questions?

Consortium for Computational Physics and Chemistry
cpcbiomass.org

Gavin Wiggins
Email wigginsg@ornl.gov
Website gavinw.me

Open source tools developed by the CCPC available online
github.com/ccpcode

Experimental and simulation data for model development
zenodo.org

Acknowledgements

- **Jeremy Leong**, **Trevor Smith** at U.S. Department of Energy (DOE), Bioenergy Technologies Office (BETO)
- **Rick French**, **Kristiina Iisa**, **Danny Carpenter**, **Katie Gaston**, **Kristin Smith**, **Christopher Golubieski** at NREL
- **William (Bill) Rogers, Tingwen Li, and Xi Gao** at NETL

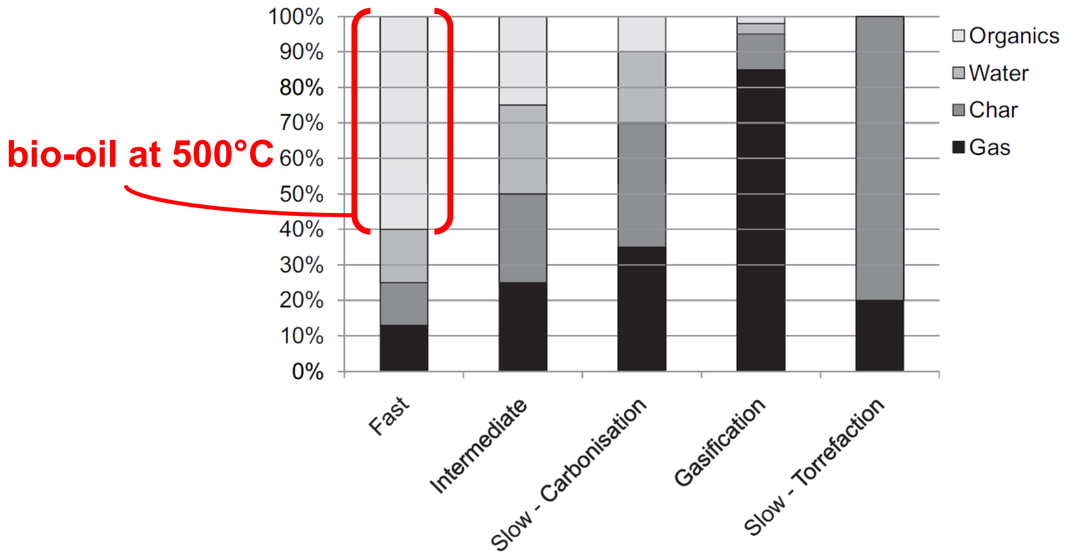
Supplemental Material

Fast pyrolysis of biomass involves 2 key steps

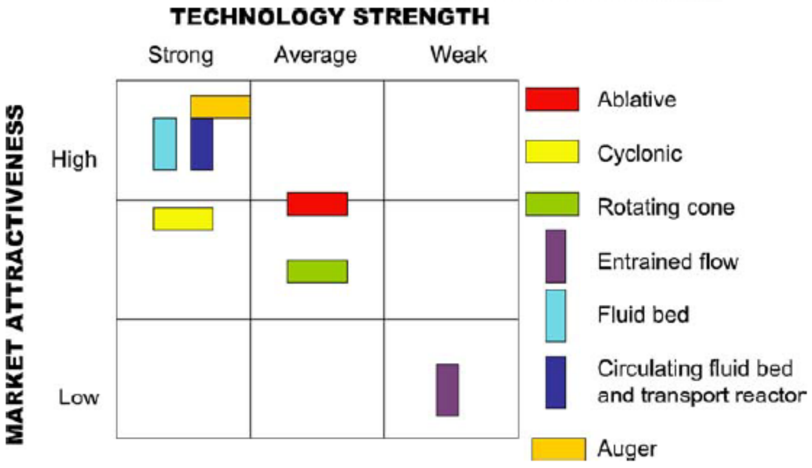
Anaerobic, rapid heating of biomass particles to produce non-condensable gases, solid char, and liquid (condensable vapors).

Goal is to maximize the liquid yield (i.e. bio-oil or tar).

The liquid can be stored and transported, and used for energy, chemicals or as an energy carrier. [Bridgwater 2012]



Product distribution from different types of pyrolysis. Source: Bridgwater 2012.

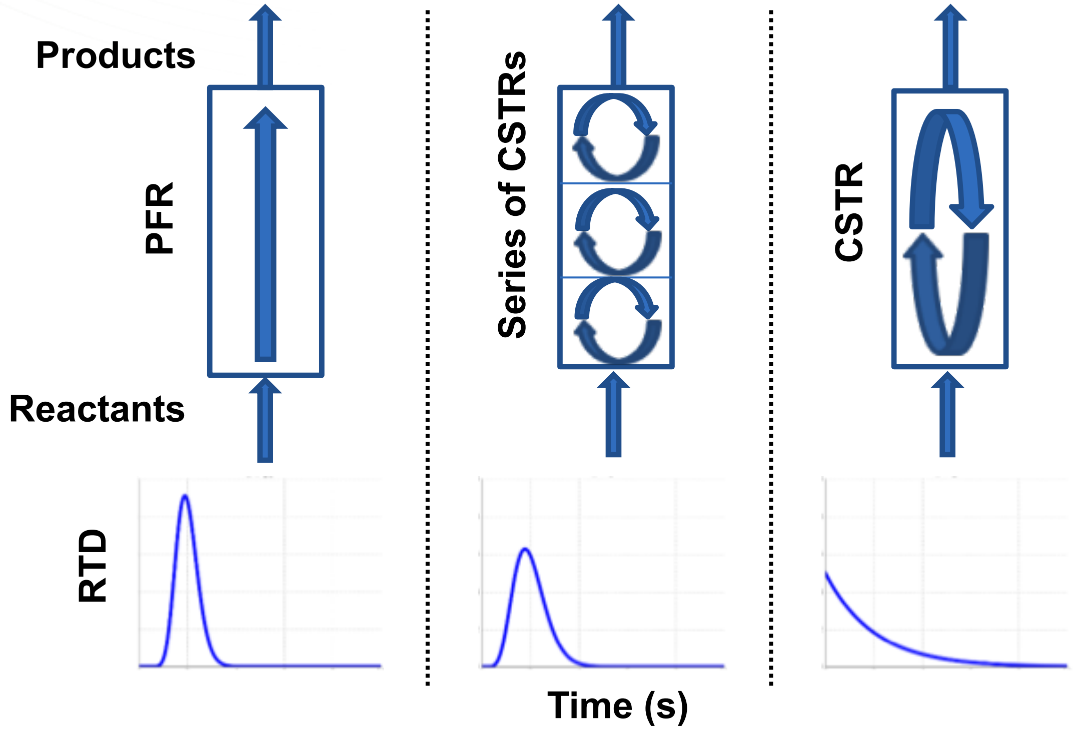


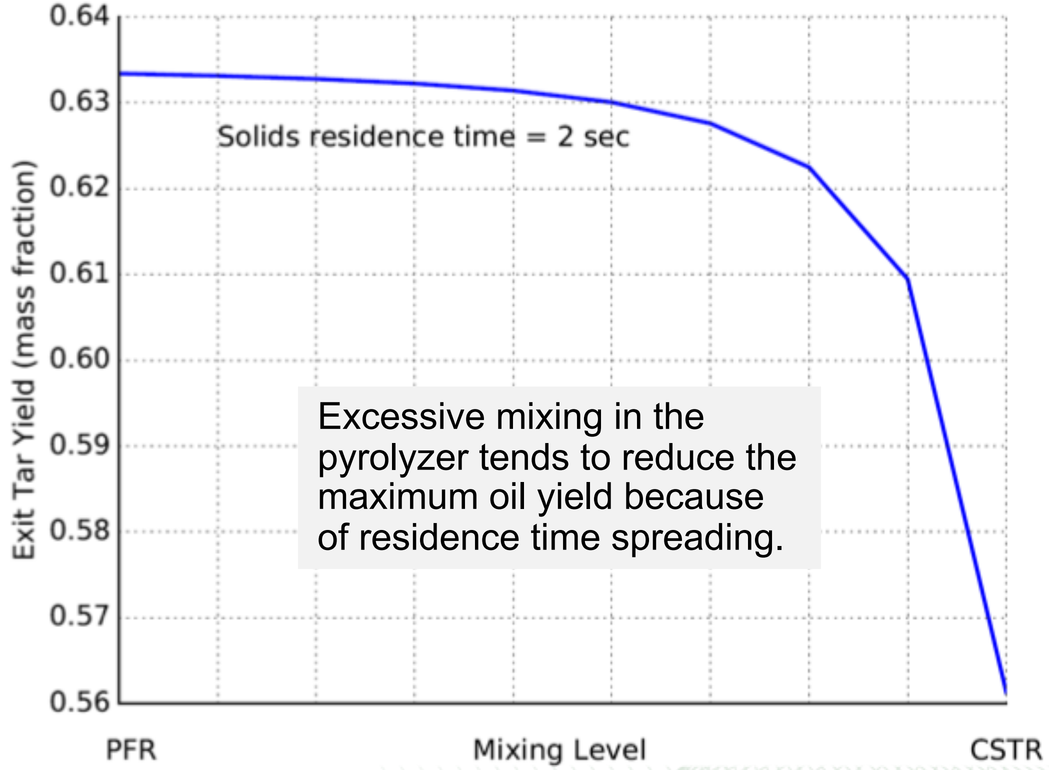
Commercial potential of fast pyrolysis technologies. Source: Butler 2011.

Low-order reactor model approach

We use low-order reactor mixing models to estimate residence times of biomass particles and the resulting oil yield.

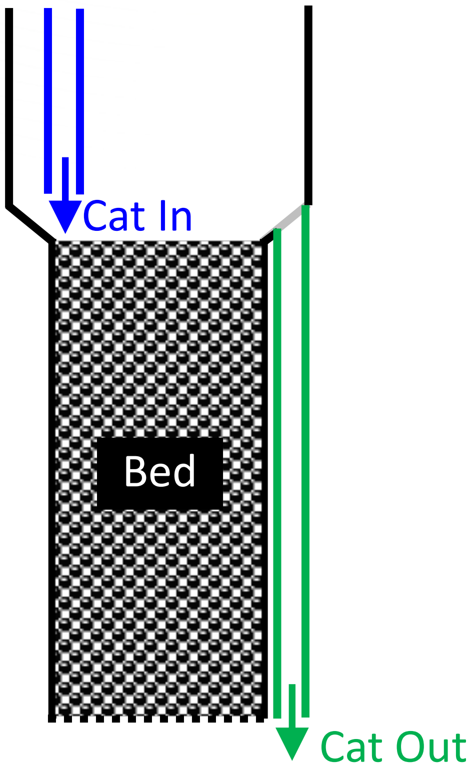
These models account for particle and gas circulation in the pyrolyzer to link particle models with overall performance.

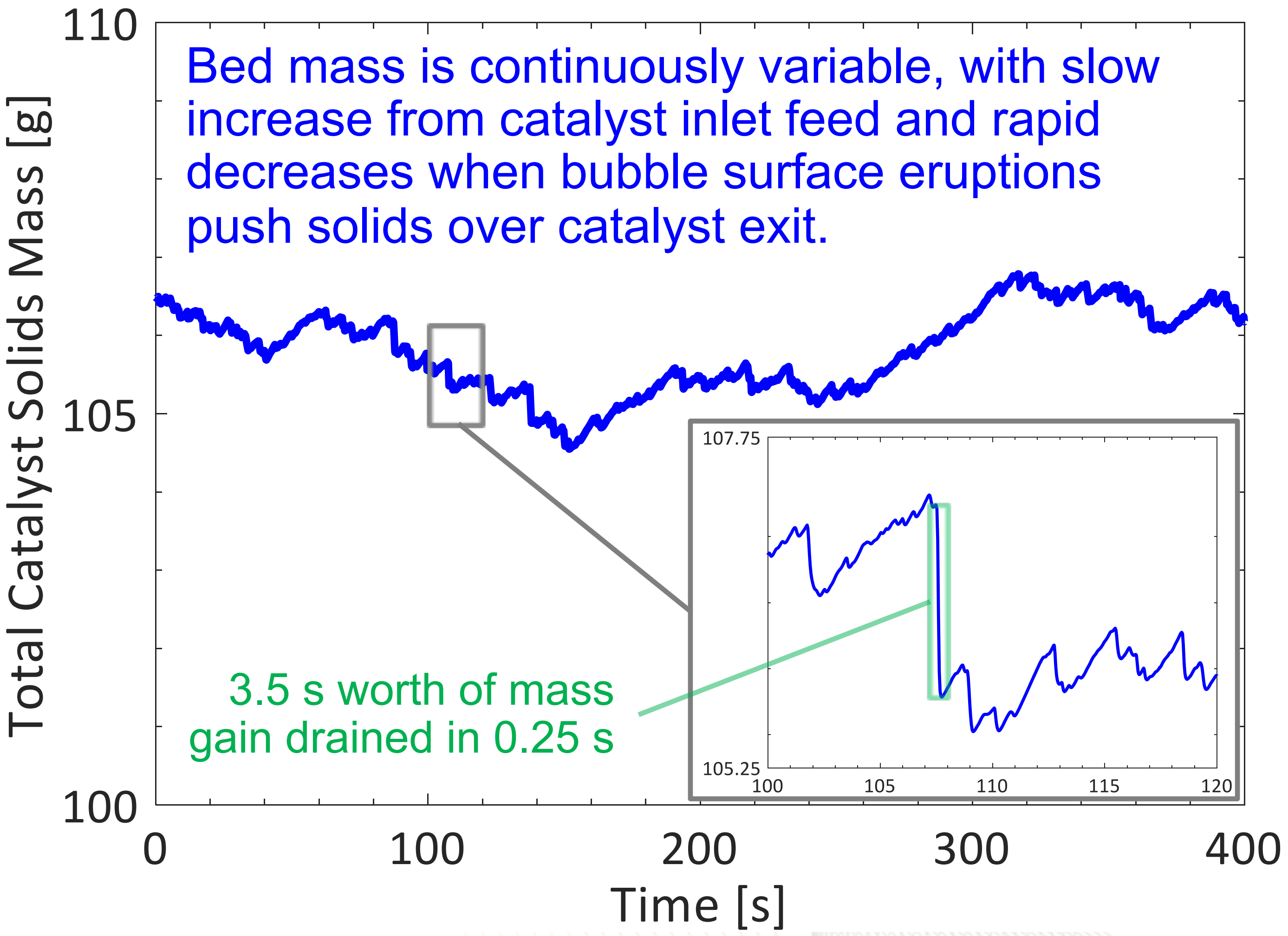




CFD modeling for solids residence time in bed

- MFiX two fluid model
- Continuous inflow of catalyst
- Hydrodynamics only





A combination of 2 CSTR zones and 1 PFR zone appear to replicate the MFiX RTDs in the R-cubed riser