Multiscale/Multiphysics simulations for multiphase gas-solids flow reactors

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July 23, 2009

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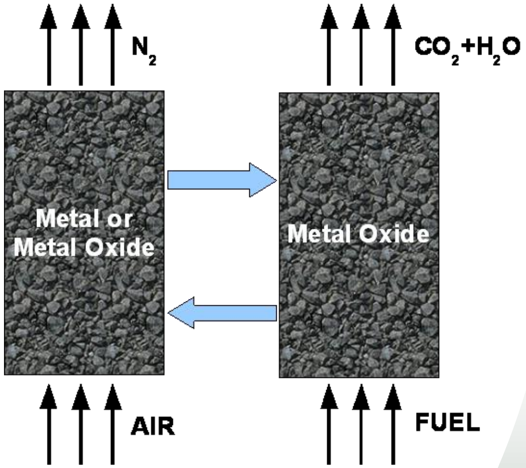
Sponsors: DOE OASCR, FE, NE and EERE

Outline

- Multiscale/multiphysics simulations for multiphase reacting flows
  - Specific issues for biomass pyrolysis/gasification
- Current set of ORNL models used at various scales
- Example simulation results
  - Biomass pyrolysis
  - Fluidized bed CVD coater for nuclear fuel particles
  - Coal gasifier simulation with 1000s of processors
- Importance of multiphysics coupling
- Compound wavelet matrix method (CWM), dynamic CWM, time parallel CWM
- Summary

Role of multiphase flow reactors in Energy Security and Sustainability

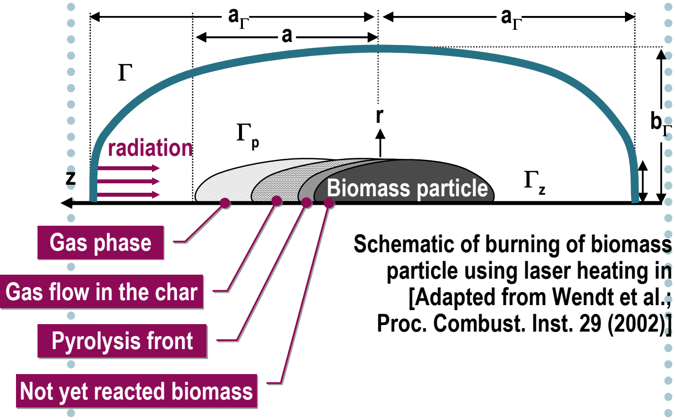
- Fuel Production and Processing
  - Refineries: catalytic crackers, H_2 production, S removal, …
  - Coal gasification, clean-up (SOx, NOx, Hg, CO_2)
  - Biomass (cellulosic) pyrolysis and gasification
  - Nuclear fuel production
- Energy Production
  - Fuel cells
  - Coal and biomass combustion
  - Nuclear reactors, separation etc.
  - Silicon production and coating for photovoltaic applications
  - Novel combustion technologies:
    - Oxycombustion
    - Chemical looping combustion
      - Higher efficiency with lower entropy losses
      - No thermal NOx
      - Separated CO_2 stream for sequestration
      - Potential carbon-negative technology
- Energy Utilization and Efficiency
  - Polymerization reactors
  - Catalytic reactors



Multiphase flows occur in most energy-intensive industrial processes

Biomass gasification

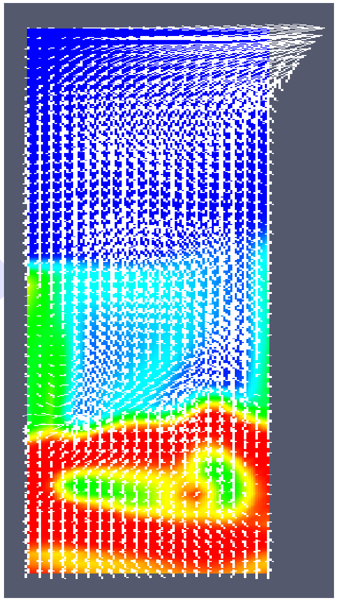
Biomass Particle (small scale)



Schematic of burning of biomass particle using laser heating in [Adapted from Wendt et al.; Proc. Combust. Inst. 29 (2002)]

- ~ mm particles
- Complex flow: gas phase, gas phase in char, pyrolysis front, unreacted biomass
- Wide range of species
- Surface processes at nm length scale and ns time scales

Biomass Gasifier/Pyrolyzer (device scale)



- ~ m in size
- Gasfication/pyrolysis at high temperatures (~1000°C) in reactor with large residence times ~10 s
- Biomass particles cycle thru wide range of conditions where complex chemistry occurs

- Design challenge: Maintain optimal temperatures, species, residence times in each zone to attain right gasification/pyrolysis
- Truly multiscale problem: ~O(13) time scales, ~O(10) length scales
- Materials challenge: Design/understand material properties for the biomass pellets/particles at μm/nm scale
  - Size
  - Porosity
  - Integrity
  - Composition
  - Binders?

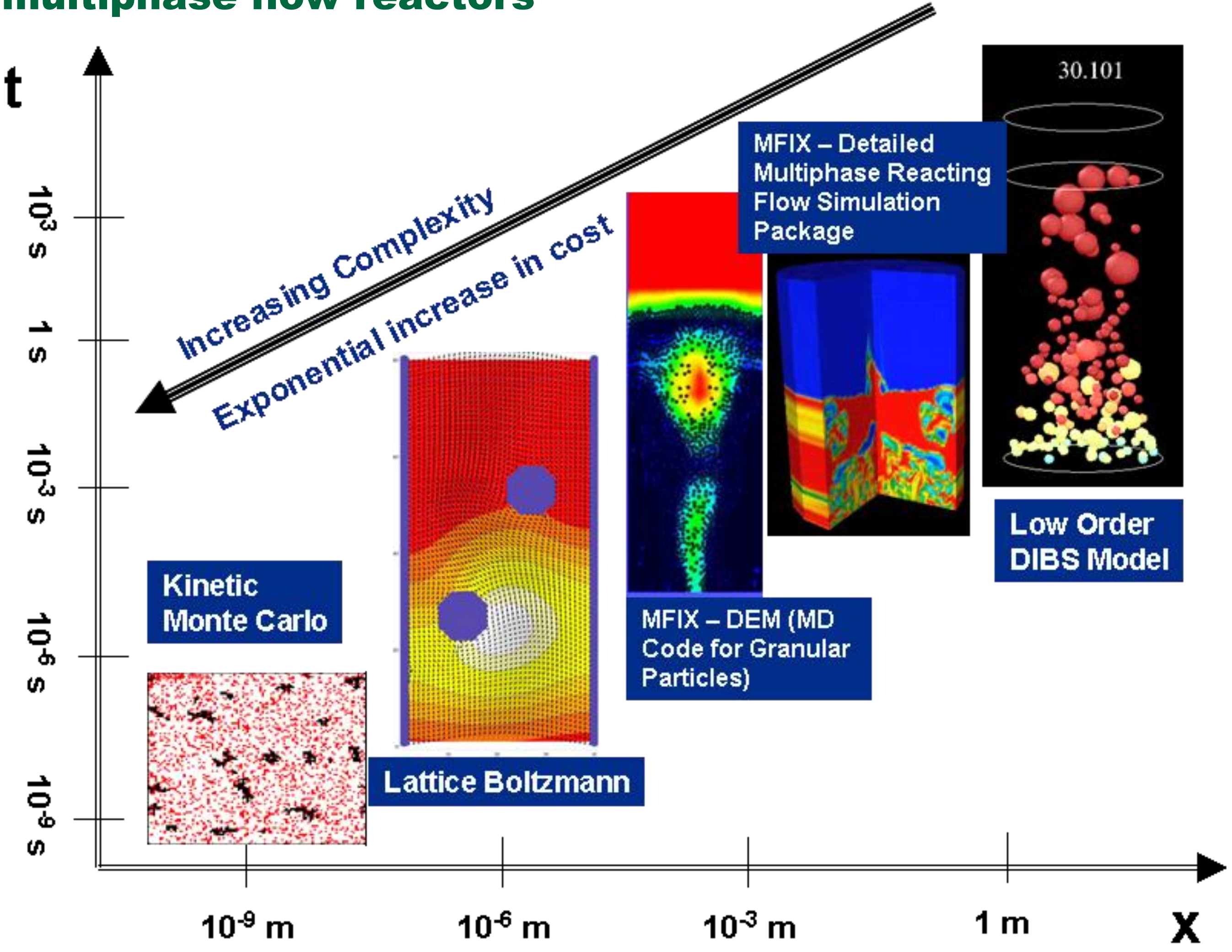
SP et al. (CFD in CRE, 2008; Chapter in a book edited by Mark Nimlos)

What is being done and what can be done differently…

- New technologies take decades
  - Lab scale → pilot scale → production scale
  - Resistance to adopting new ideas
  - Current models have limited quantitative predictability/credibility
  - Cultural barrier
- Why we need to do things differently
  - Energy crisis is current and growing
  - We need tomorrow’s technology today
  - Economic opportunity
- What can be done differently
  - Development of integrated and scalable MSMP predictive models
  - Component and lab-scale experiments targeted to validate computational models
  - Integrate lab scale experiments along with simulations to design new plants and devices

Multiphysics heterogeneous chemically reacting flows for energy systems

Goal: Building a suite of models for unprecedented capability to simulate multiphase flow reactors



- Through support from various DOE offices (FE, EERE, and NE) we have developed suite of models for unprecedented capability to simulate heterogeneous chemically reacting flows
- Hybrid methods to couple two physical models (e.g. MFIX DEM)
- Uncertainty quantification to probe only quantities of interest at smaller scales

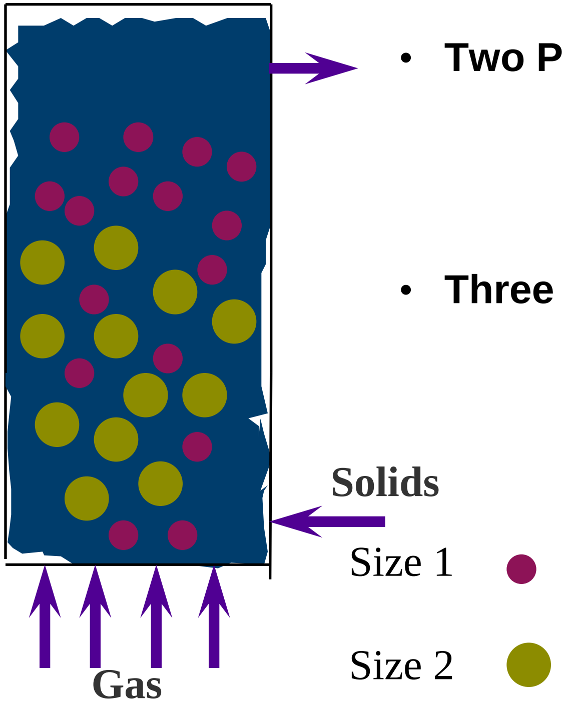
MACROSCALE CFD SIMULATIONS

MFIX simulation package

- **General multiphase flow CFD code which couples hydrodynamics, heat & mass transfer and chemical reactions**
- **SMP, DMP and Hybrid Parallel code which runs on many platforms including Beowulf clusters**
- **Open-source code and collaborative environment (http://www.mfix.org or http://mfix.netl.doe.gov)**
- **Over 1500 researchers from over 500 institutions around the world**

2007

Multiphase formulation



- **Two Phases**

Fluid 🟩

Solids ●●●

- **Three phases**

Fluid 🟩

Solids - 1 ●●

Solids - 2 ●●●

- **Details of flow field and particle interaction have been averaged out.**
- **Account for the information lost due to averaging through the use of *constitutive equations***

Continuity Equation

$$\frac{\partial}{\partial t}(\varepsilon_m \rho_m) + \nabla \cdot (\varepsilon_m \rho_m \vec{v}_m) = \sum_{l=1}^{M} R_{ml}$$

Momentum Equation

$$\frac{\partial}{\partial t}(\varepsilon_m \rho_m \vec{v}_m) + \nabla \cdot (\varepsilon_m \rho_m \vec{v}_m \vec{v}_m) = \nabla \cdot \bar{\bar{S}}_m + \sum_{l=1}^{M} \vec{I}_{ml} + \vec{f}_m$$

Stresses

Interaction Term

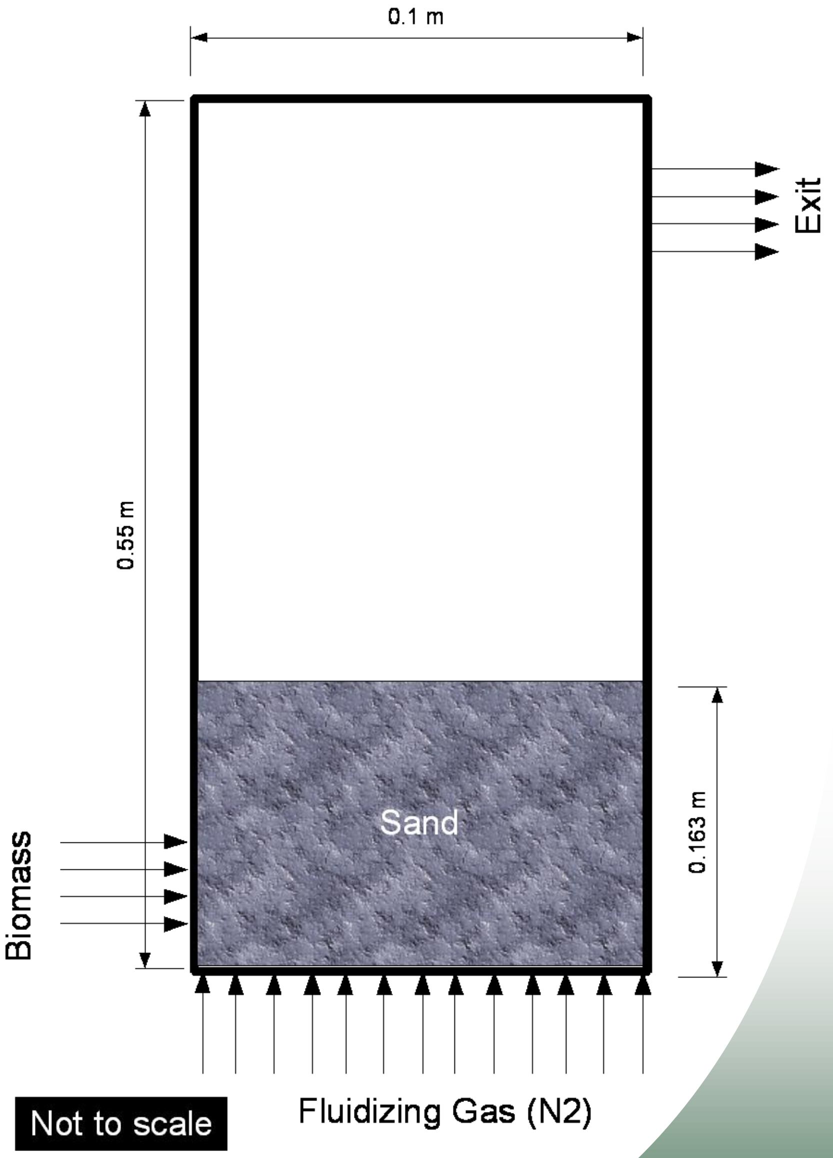
Granular Stresses are modeled by the kinetic theory of granular material in the viscous regime and plasticity theory in the plastic regime

Drag law describes the interaction between the gas and the particles

BIOMASS PYROLYSIS REACTOR

Prototype case

- **Lathouwers and Bellan, IJMF 2001**
- **Good test as the kinetics for a continuum model are available**
- **The geometry and flow conditions simulated are similar to those in this paper for qualitative comparison**
  - **Fluidizing gas velocity: 0.5 m/s**
  - **Biomass feed (0.5 kg/s) – Bagasse – Cellulose (0.36), Hemicellulose (0.47) and Lignin (0.17)**
  - **Gas temperature – 700 K and biomass temperature – 400K**



Chemical thermodynamics and kinetics

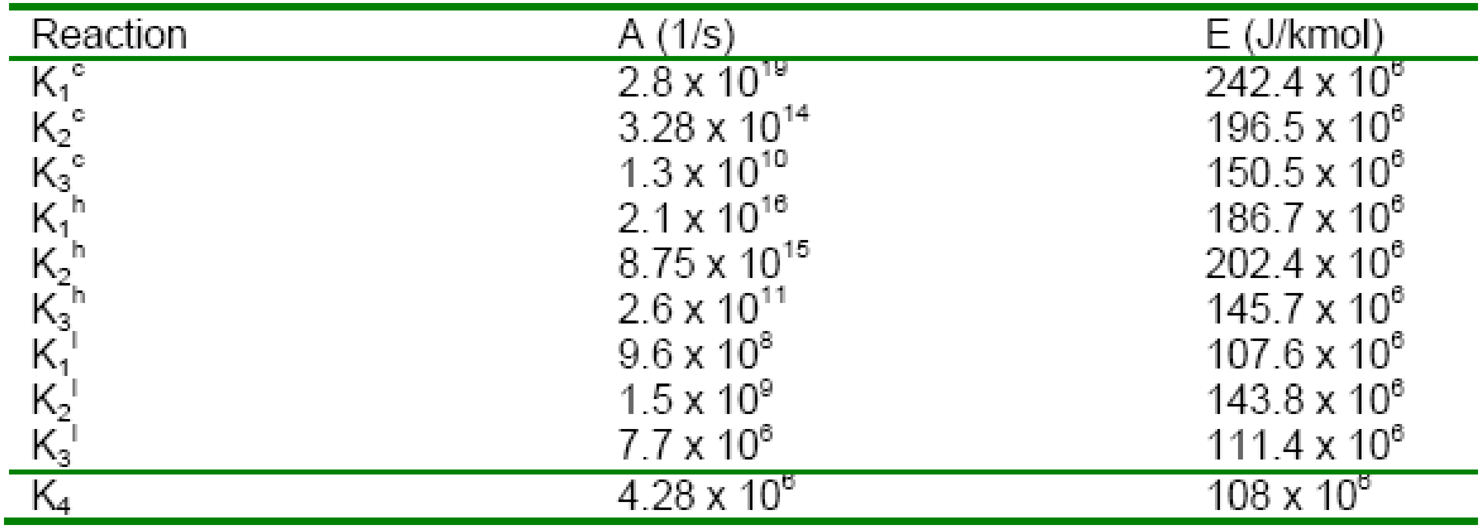
$$virgin\ (s) \xrightarrow{K_1} active\ (s)$$

$$active\ (s) \xrightarrow{K_2} tar\ (g) \xrightarrow{K_4} gas\ (g)$$

$$active\ (s) \xrightarrow{K_3} X\ char\ (s) + (1-X)\ gas\ (g)$$

Δh_1 = 0 kJ/kg
Δh_2 = 255 kJ/kg
Δh_3 = -20 kJ/kg
Δh_4 = -42 kJ/kg

Rate constants and activation energy for the biomass pyrolysis kinetics scheme above:



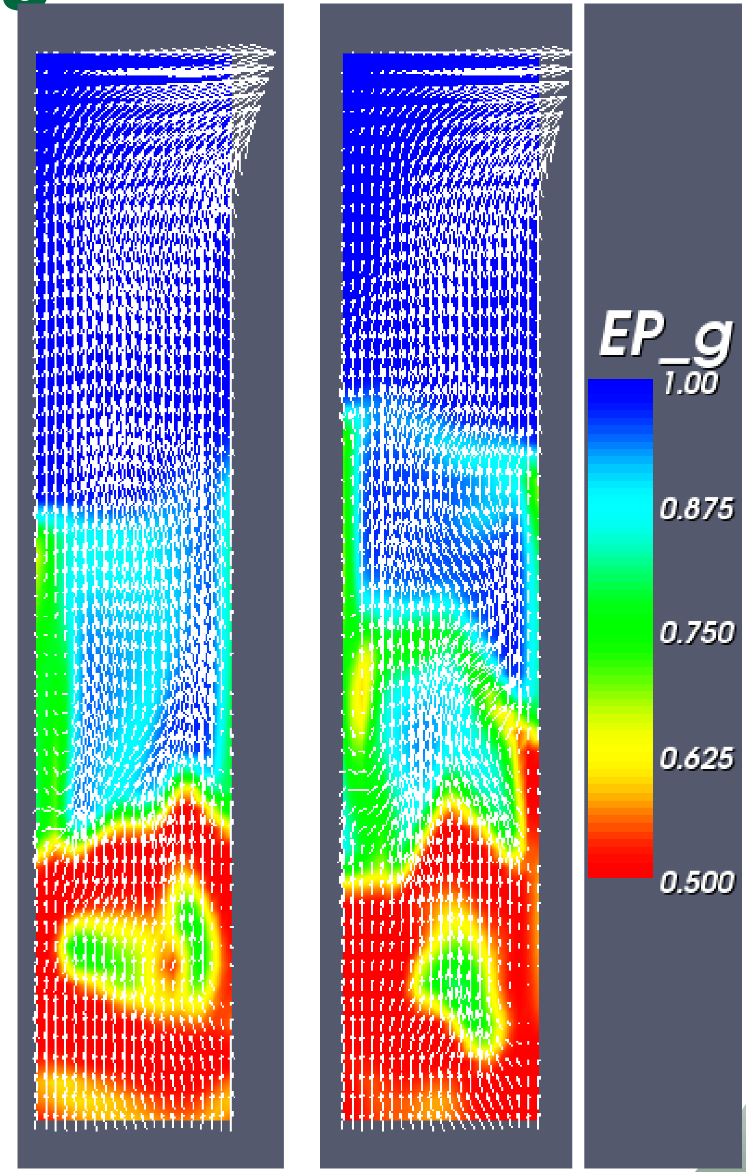
Reaction	A (1/s)	E (J/kmol)
K_1^c	2.8×10^{19}	242.4×10^6
K_2^c	3.28×10^{14}	196.5×10^6
K_3^c	1.3×10^{10}	150.5×10^6
K_1^h	2.1×10^{16}	186.7×10^6
K_2^h	8.75×10^{15}	202.4×10^6
K_3^h	2.6×10^{11}	145.7×10^6
K_1^l	9.6×10^8	107.6×10^6
K_2^l	1.5×10^9	143.8×10^6
K_3^l	7.7×10^6	111.4×10^6
K_4	4.28×10^6	108×10^6

The char formation ratios for reaction K_3 are: $X^c = 0.35$, $X^h = 0.6$, and $X^l = 0.75$

The species properties are taken from Lathowers and Bellan, 2001

Solids distribution and gas flow

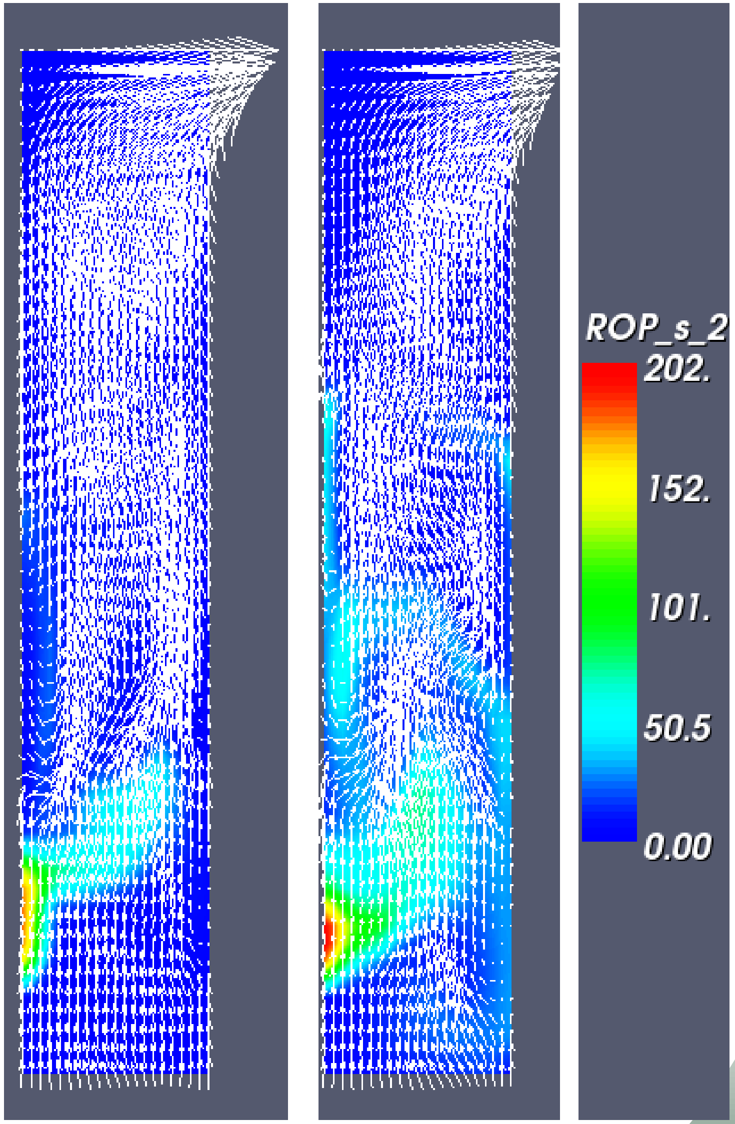
- **Fluidization gas levitates the particles and the biomass**
- **The reactor operates in the bubbling bed regime**
- **The gas undergoes local acceleration and deceleration depending on the flow of the solids**
- **The flow of solids and gas is transient and highly dynamic**
- **The reactor geometry causes recirculation near the top of the reactor**
- **The fluidizing and product gas leave the domain through the exit**



Void fraction with gas velocity vectors at two different instants

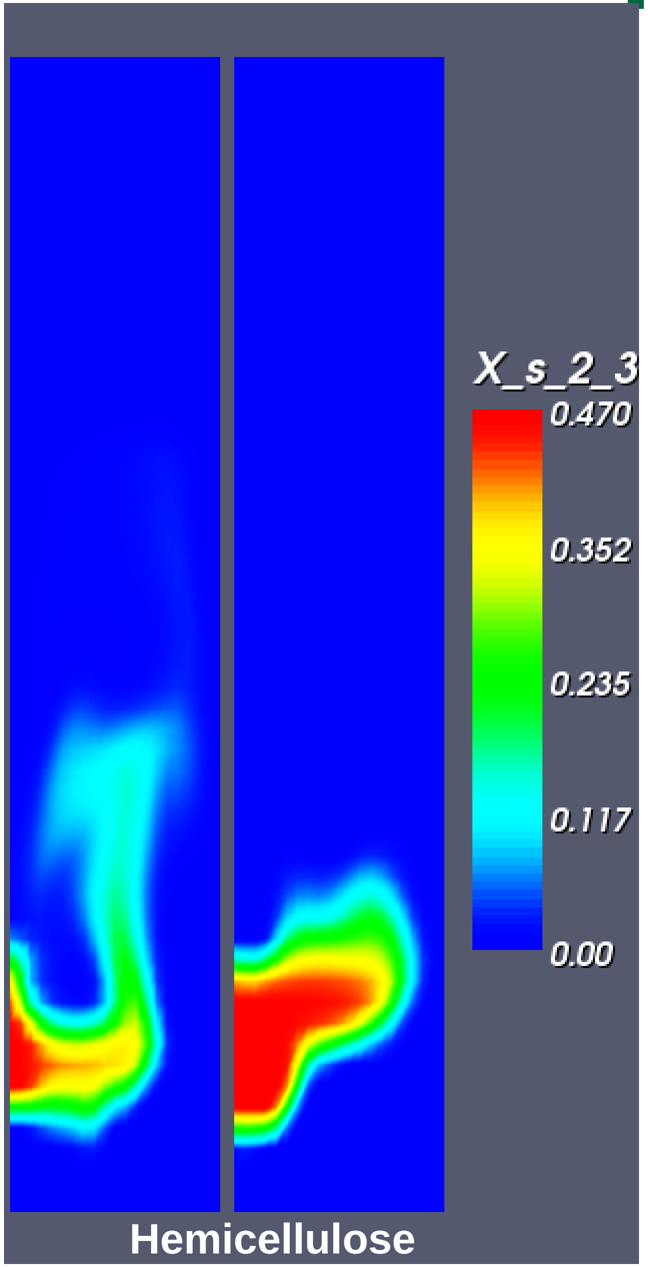
Solids distribution and gas flow

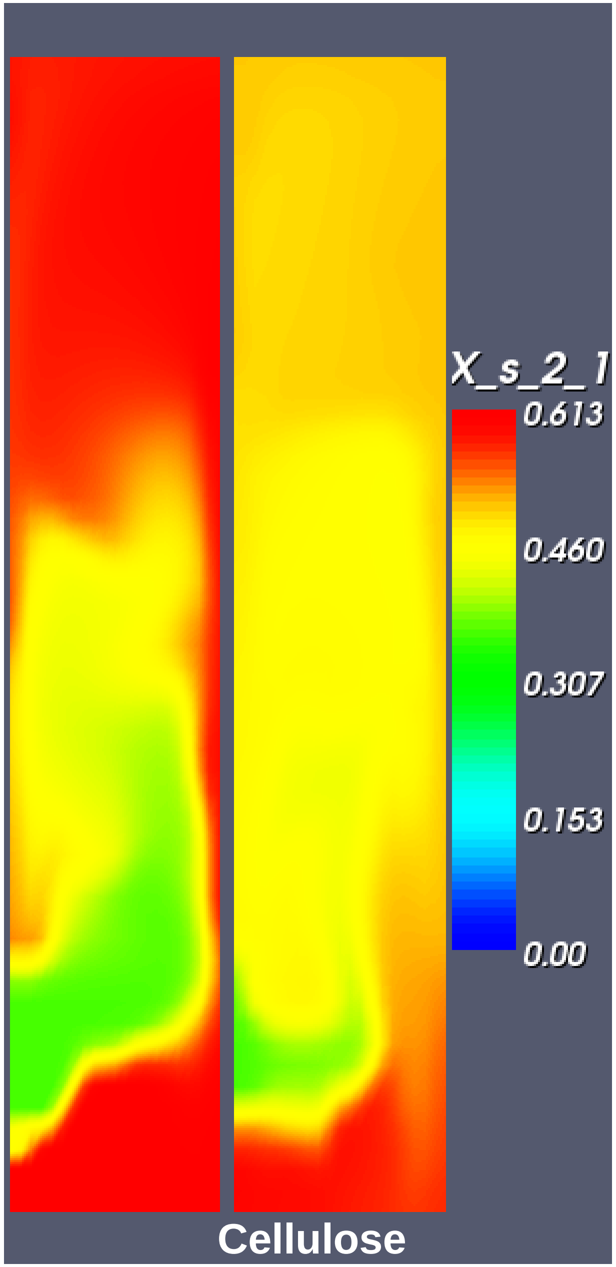
- **The gas flow is perturbed slightly to accommodate the injection of the biomass**
- **Fluidization gas and the bed particles exert axial force on the incoming biomass into the reactor**
- **The biomass accumulates closer to the inlet but quickly disperses and also undergoes pyrolysis in contact with the high temperature gas and solids**



Biomass mass distribution along with gas velocity vectors at two different instants

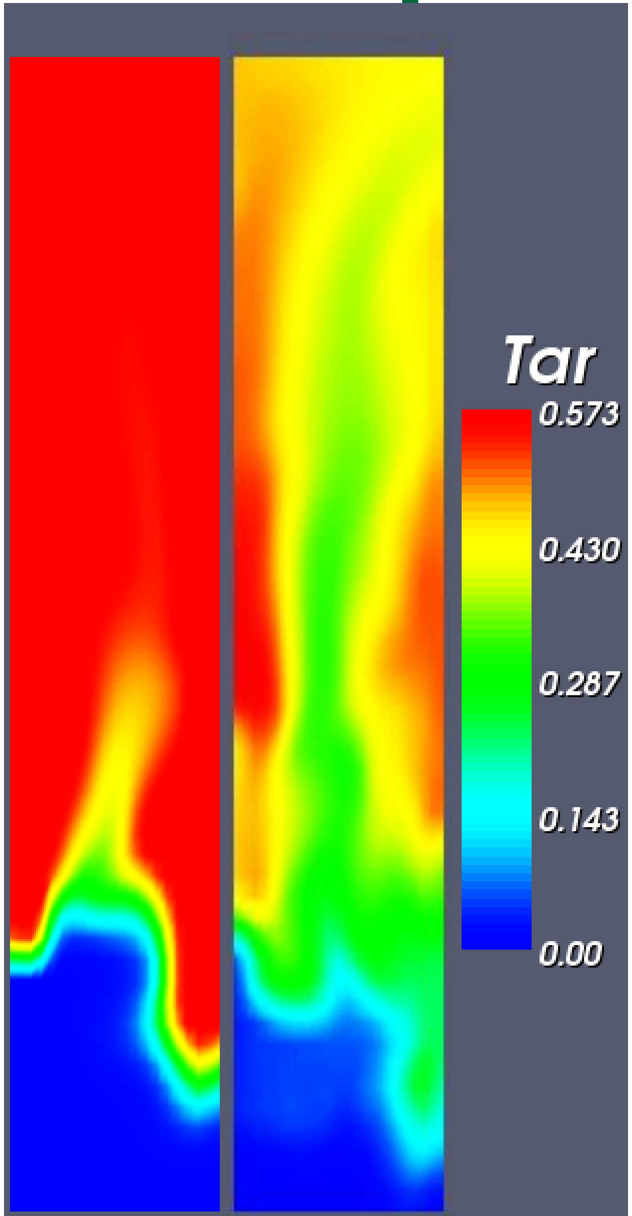
Biomass composition



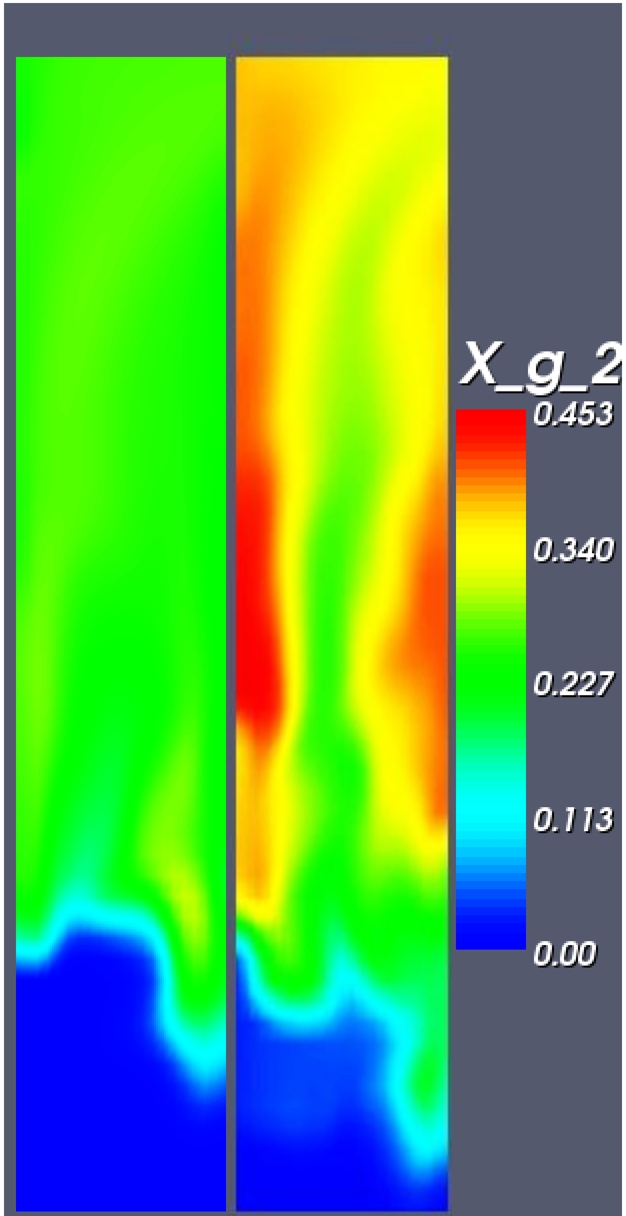


- The fast reacting hemicellulose is primarily found near the inlet contrary to the cellulose
- The distribution of the various species is related to the physical/thermodynamic/transport properties along with the chemical reactivity as functions of local temperature.
- This in turn influences the variation of the product gas both in space and time.

Gas composition



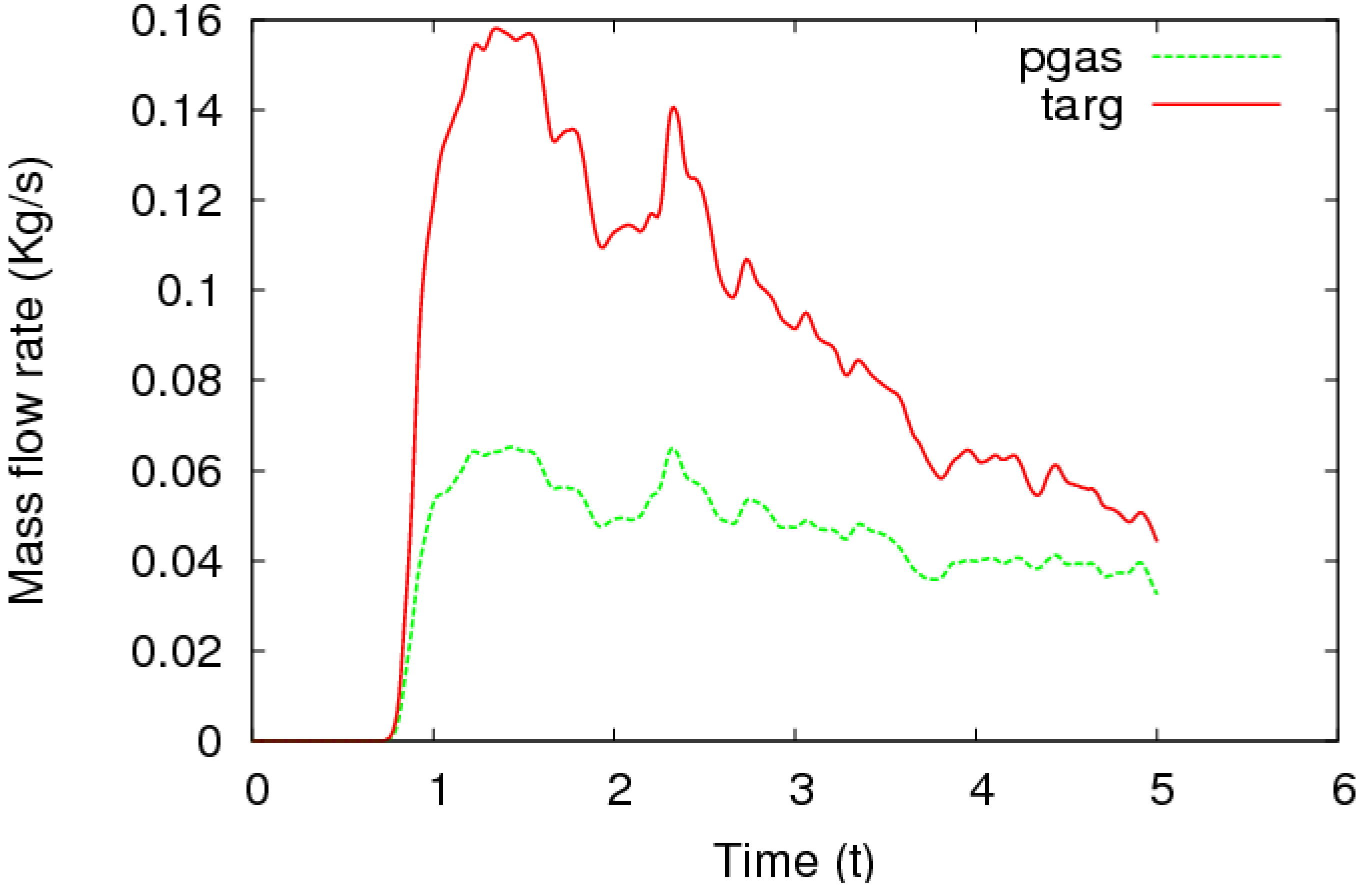
Tar



Product Gas

- **Fluidizing gas bypasses through the bed to the outlet**
- **Product species have higher residence time in the reactor.**

Temporal variation of exit gases



The product gas and tar gas are still in the transient regime after 5 seconds of the biomass injection → function of the reaction kinetics, the temperature distribution, the solids and gas contacting etc.

Conclusions from biomass study

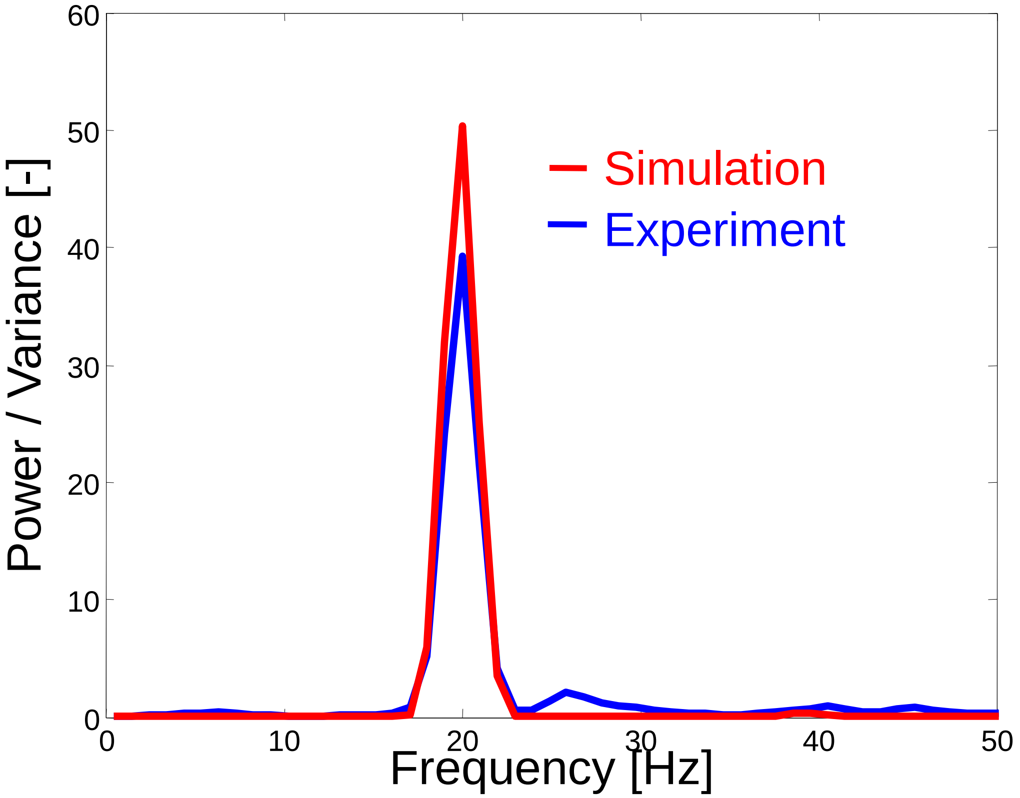
- The detailed CFD simulations can provide spatio-temporal variations of the solids, biomass, gas, gas and solid species, reaction rates etc.
- The MFIX simulation software used here can run on 1000s of processors and can be employed for detailed 3D simulations of realistic pyrolysers and gasifiers
- The model is quite well validated for various fluidized bed reactors
- However, reaction kinetics and thermodynamics conducive to continuum mulitphase flows need to be determined through experiments
- The models still need to be systematically validated to improve confidence levels
- Once validated models are available, the reactors can be optimized by simulations and reduce the design space for experiments

FLUIDIZED BED CVD COATER FOR NUCLEAR FUEL PARTICLES

Simulation objectives:

- Demonstrate simulations with *sufficient detail* to capture known effects of coater operation and design on quality
- Develop analytical tools that aid coater scale-up and design
- Develop improved nuclear fuel coaters with unprecedented levels of product quality
- Develop improved fundamental understanding of the controlling mechanisms for both C and SiC chemical vapor deposition
- Develop improved fundamental understanding of the dynamics of spouted bed reactors

Observation (1): MFIX can predict correct dynamic time scales



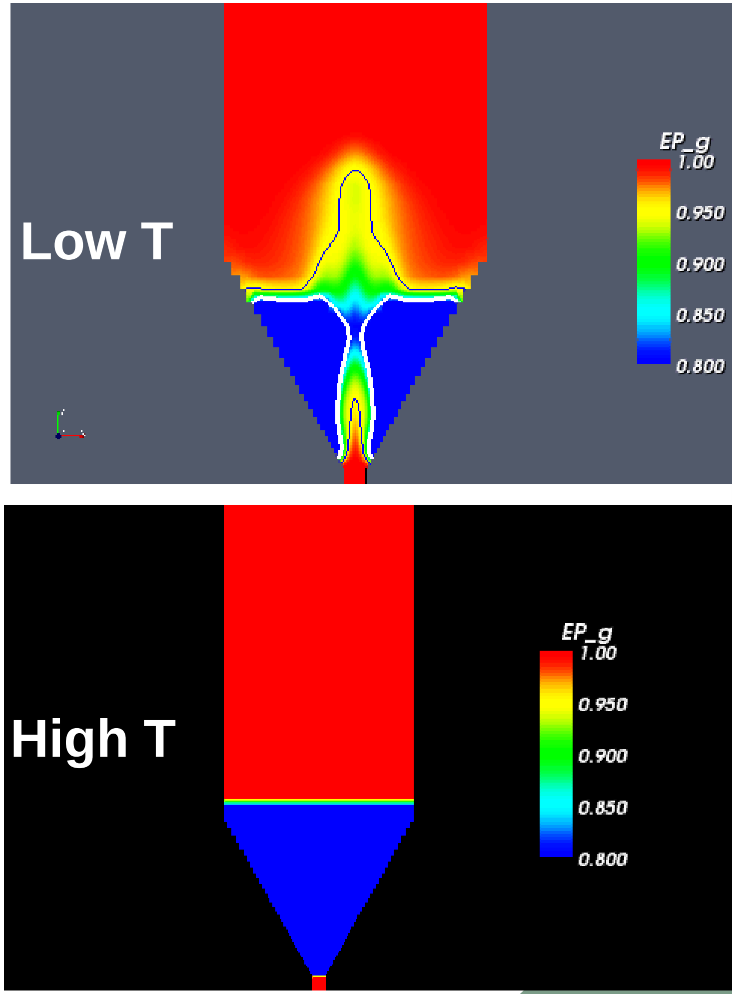
500 µm ZrO_2 at 300 K in air for UTK 2-inch mockup

- Gas pulsations are directly measurable
- Pulsations contain important information on solids circulation
- The circulation times also relate to particle-coating gas contact time

Observation (2): Standard heat transfer correlations in MFIX appear to work well for this application

- Gunn (1978) heat & mass transfer correlations used
- Large effects of temperature due to
  - Density and viscosity change
  - Sudden radial and axial expansion
- Two different example cases
  - 500 μm ZrO2 in 30.06 m/s air at 298 K
  - 536 μm buffer coated UCO in 14.6 m/s Argon/Acetylene/Propylene mixture at 1523K
- Jet expansion is much more dramatic at higher T
- At higher T, spouting also becomes more vigorous and pulsation frequency drops by ~1/2
- Consistent with experiments
  - indicates proper coupling between heat transfer and hydrodynamics

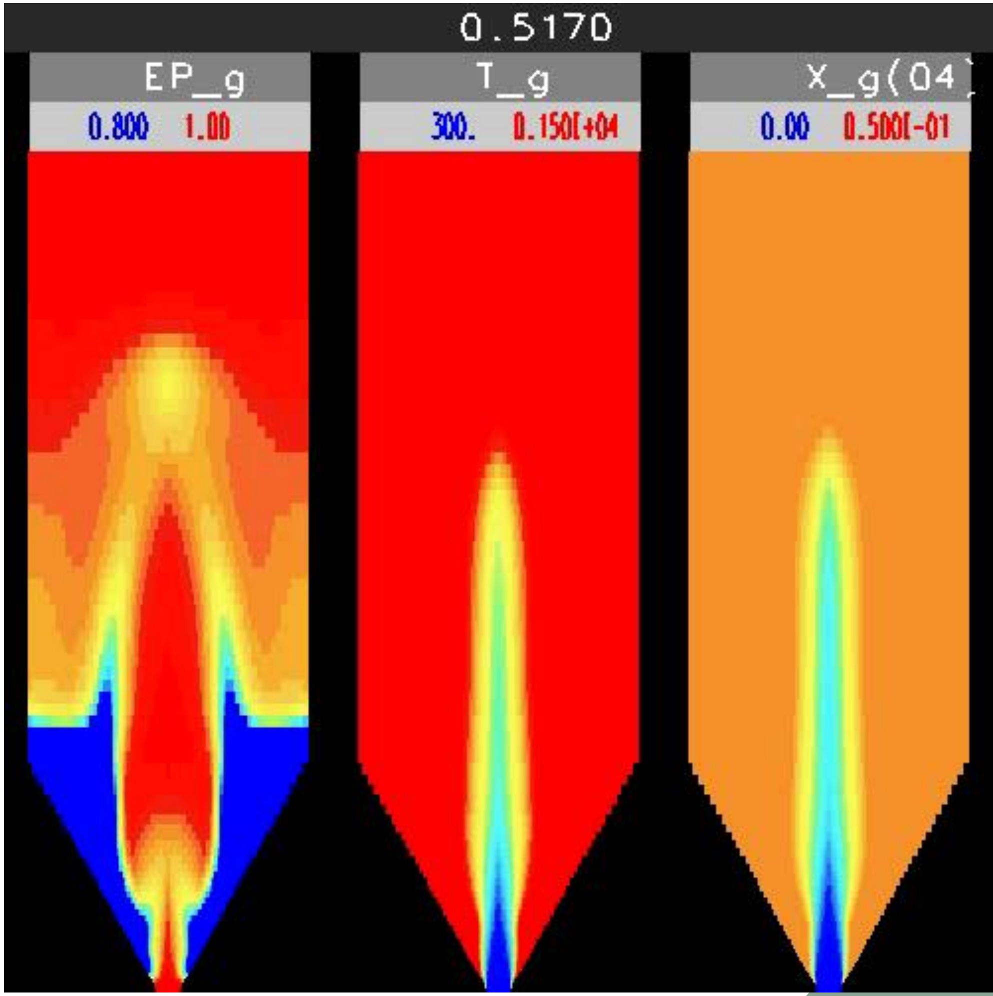
Void Fraction



Observation (3): We see very high spatial & temporal gradients at coating conditions

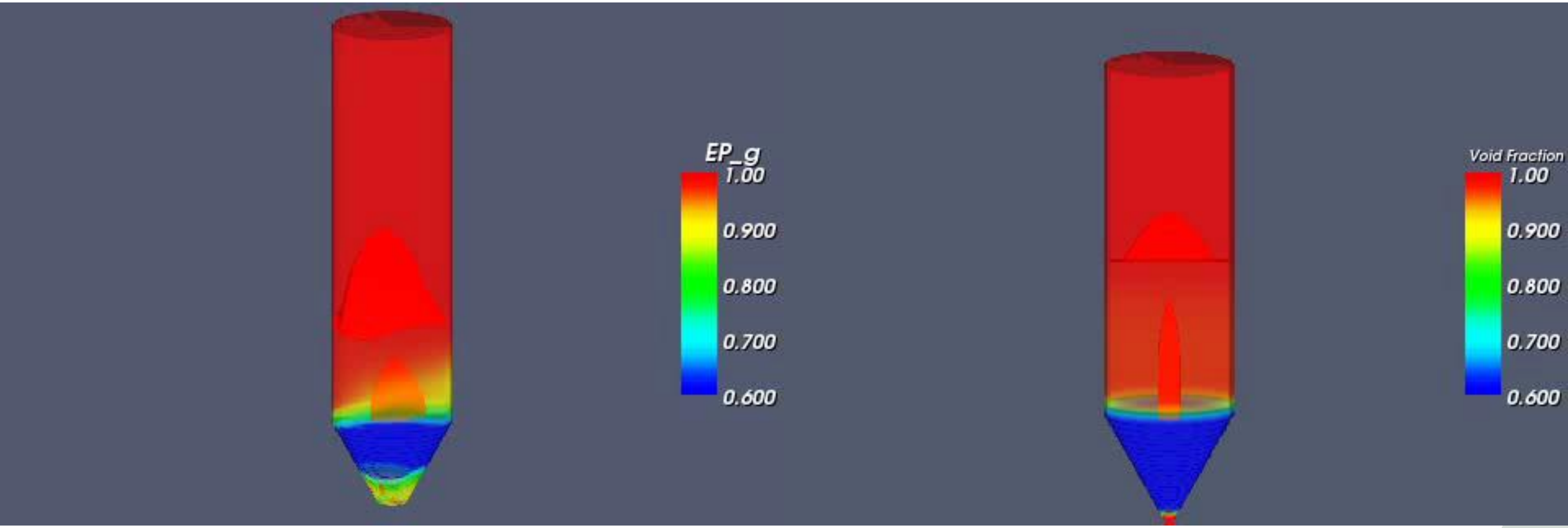
- **Experimental observations indicate core zone is the most important (location of 'snow' formation during C deposition)**
- **Inlet gas heats very quickly to furnace temperature with solids (unlike pure gas flow)**
- **Very large absolute fluctuations in velocities, temperatures & concentrations during pulsation cycle**
- **Characteristics of these gradients, fluctuations expected to be major factors for design, scaling**

Void Fraction, Gas Temperature, H_2 mass fraction



Observation (4): Injector design very critical to overall spouting behavior

Side View (Translucent)
[Contour surface corresponds to 0.99 void fraction]
Similar to peering into a glass bed with marbles



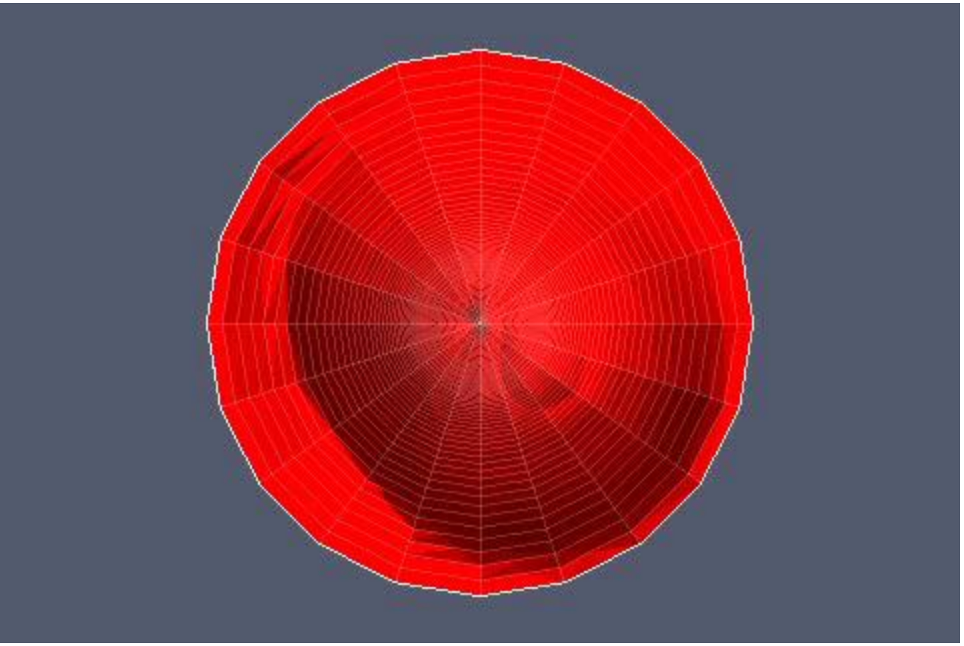
3D Multi-hole (6 holes: 1 + 5)

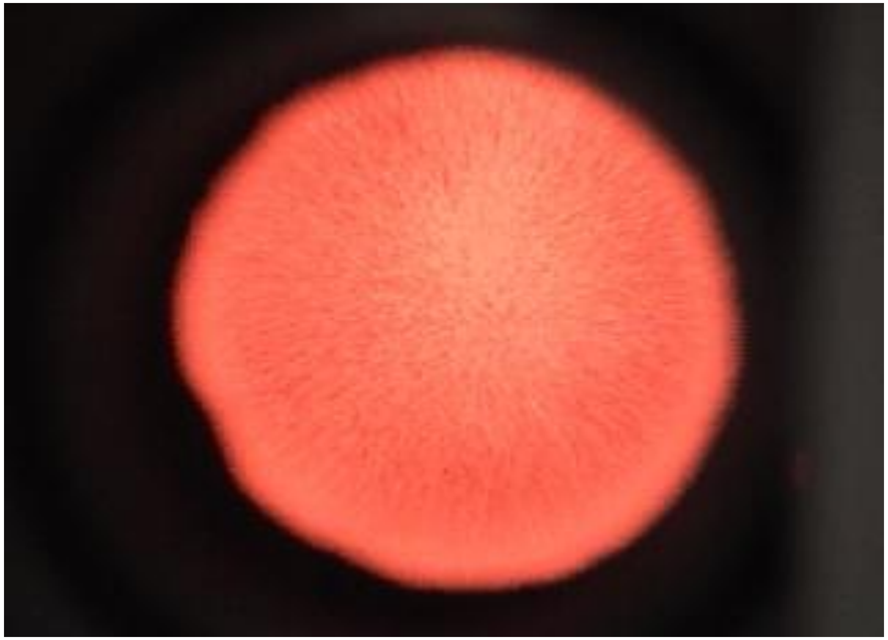
Reference NUCO IPyC condition

3D Single-hole

Observation (5): MFIX simulation predicts surface ‘sloshing’ observed in experiments

Top View





Simulations (Void Fraction = 0.99)

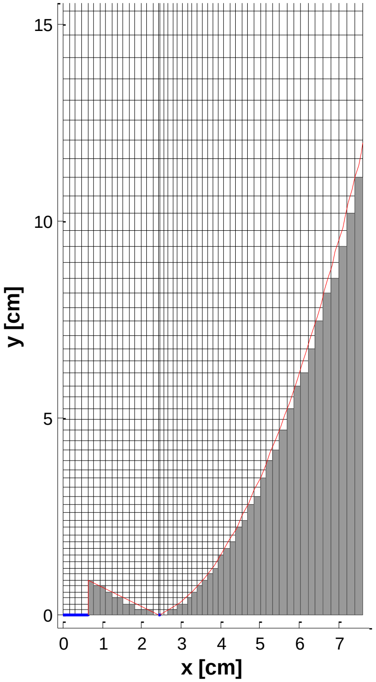
Experiments

Discriminating characteristics (DCs) have been proposed as generic quantitative indicators

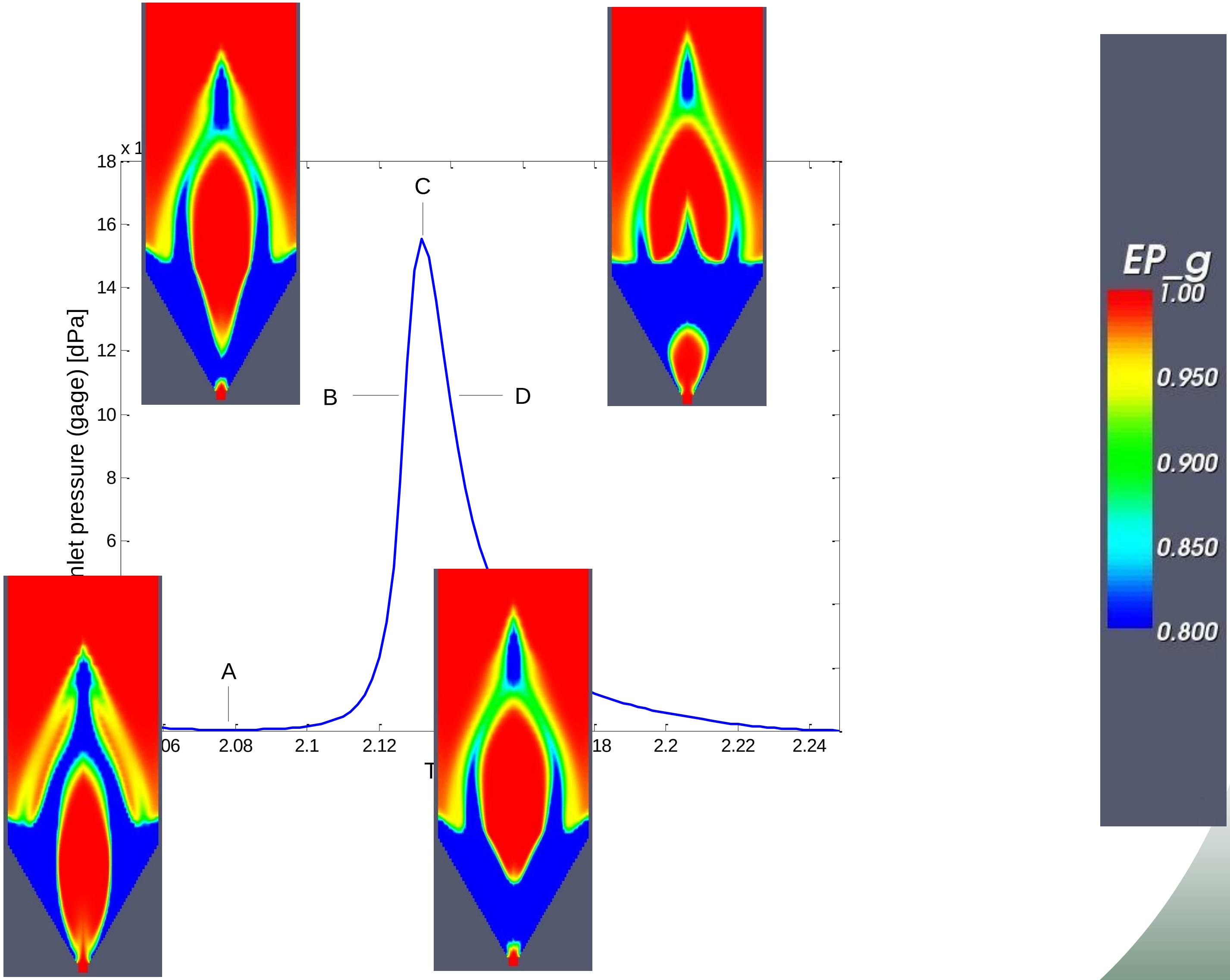
Discriminating Characteristic	Multi-hole	Single-hole	Comments
Dimensionless Solids Circulation Time (DSCT)	20.75	35.34	40% reduction
Ballistic Particle Profile (BPP) (10%) (cm)	8.56	10.97	Significant reduction in fountain height
Net Solids Impact Rate (NSIR)(g/s)	32.77	17.83	90% increase in wall impacts
Core Diameter (CD) (cm)	2.96	2.18	Significant increase in core diameter
Gas Velocity at center line at initial bed height (VG@CH) (cm/s)	346	949	Significant decrease in gas velocity
Solids Velocity at center line at initial bed height VS@CH (cm/s)	39.8	80.4	
Gas T at center line at initial bed height TG@CH (K)	1371	1000	Gas heats up quickly
Solids T at center line at initial bed height TS@CH (K)	1516	1486	
H2 concentration at center line at initial bed height H2@CH	0.0393	0.0243	Significant product formation at bed height
Acetylene concentration at center line at initial bed height C2H2@CH	0.0221	0.0788	Significant decrease in the reactant species
Propylene concentration at center line at initial bed height C3H6@CH	0.0179	0.0639	Significant decrease in the reactant species
GRADT (K/cm)	857.87	249.37	Huge difference in the gas heat-up rate
T@GRADT (K)	434.5	461.9	
Y@GRADT (cm)	0.25	1.382	Gas heats up very close to the inlet

Geometries and gas distributors

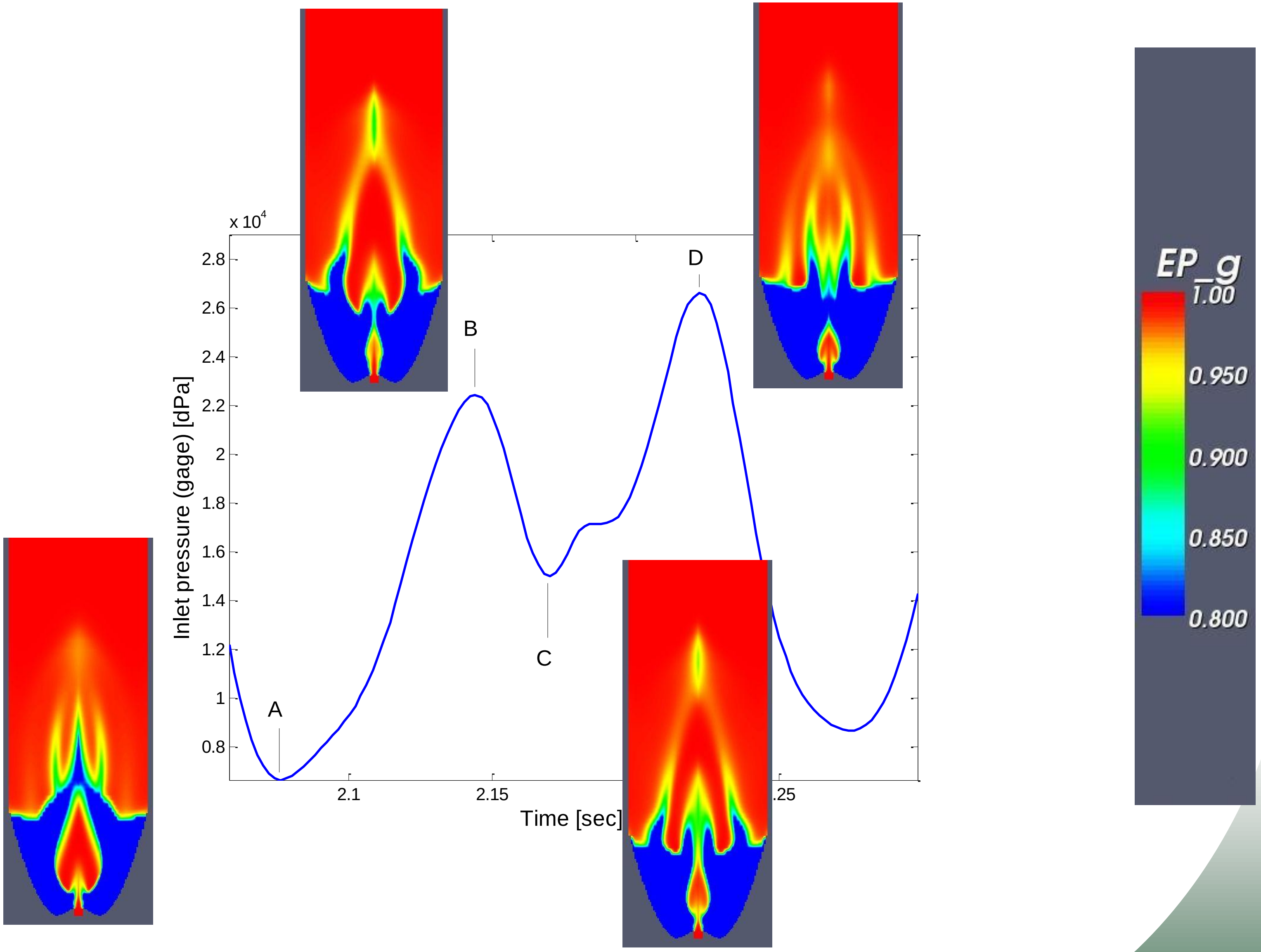
Run	Geometry	Distributor	Diluent gas partition Center | ring	Total gas mass flow rate [kg/hr]
1	Cardioid	Ring+center	0.908 | 0.092	10.7
2	Cardioid	Ring+center	0.938 | 0.062	16.1
3A	Cardioid	Multiport	0.908 | 0.092	10.7
3B	Cardioid	Multiport	0.908 | 0.092	10.7
4	Cardioid	Multiport	0.908 | 0.092	10.7
5	Cone	Center	N/A	10.7



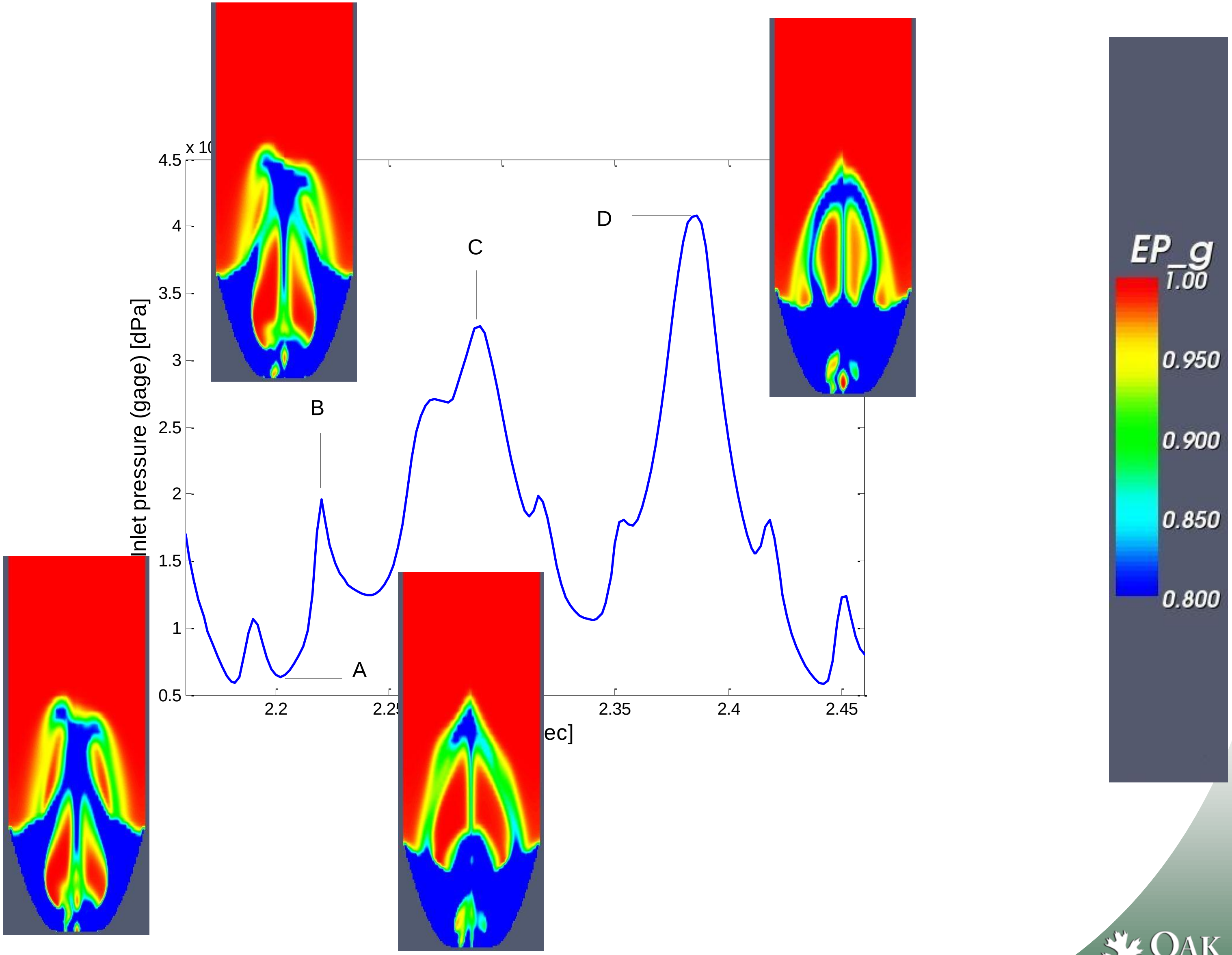
Reference cone design



Ring + center (Design #1)



Multiport injector (Design #3)



Conclusions from FBCVD scale-up studies

- Cardioid chalice with the multi-port design appears to have the best gas-solids mixing and heat transfer rates
  - Conical spouted beds cannot be scaled from 2” to 6”
  - Impact of swirl is minimal
  - Coater hydrodynamics and heat transfer are only minimally affected by mass transfer and chemical reactions.
- Design time reduced by order of magnitude at a fraction of cost

COAL GASIFIER

Carbonaceous Chemistry for Continuum Modeling (C3M)

Excellence in Technology Transfer Award 2008

- **Devolatilization**

  HF: Volatile Matter $\rightarrow \alpha_d$ Tar + $\beta_d^{CO}CO + \beta_d^{CO2}CO_2 + \beta_d^{CH4}CH_4 + \beta_d^{H2}H_2 + \beta_d^{H2O}H_2O$

- **Cracking** *IF:* Tar $\rightarrow \alpha_c C + \beta_c^{CO}CO + \beta_c^{CO2}CO_2 + \beta_c^{CH4}CH_4 + \beta_c^{H2}H_2 + \beta_c^{H2O}H_2O$

- **Drying** *GF:* Moisture (coal) → H_2O

- **Water-gas shift reaction** *EF:* $CO + H_2O \leftrightarrow CO_2 + H_2$

- **Gasification**

  BF: $C + H_2O \leftrightarrow CO + H_2$

  CF: $C + CO_2 \leftrightarrow 2CO$

  DF: $\frac{1}{2}C + H_2 \leftrightarrow \frac{1}{2}CH_4$

Combustion

0F: $H_2 + 2O_2 \rightarrow H_2O$

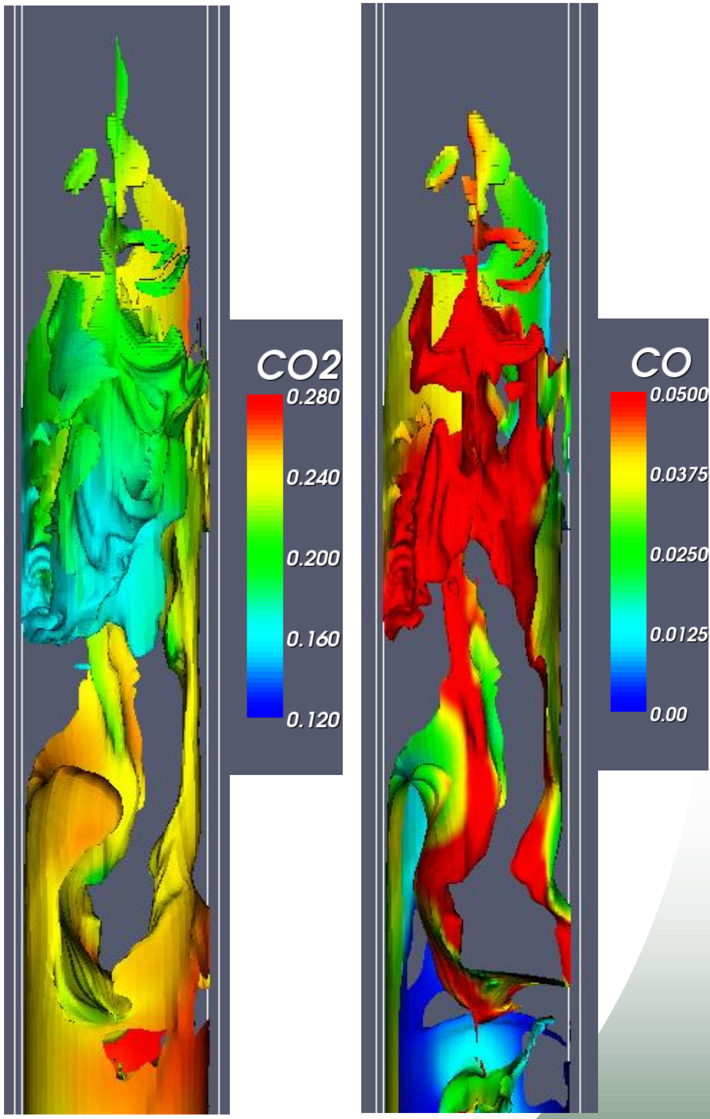
1F: $CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O$

2F: $CO + \frac{1}{2}O_2 \rightarrow CO_2$

AF: $2C + O_2 \rightarrow 2CO$

Coal gasification simulations

- **We are conducting high resolution gasifier simulations with collaborators at NETL**
  - **2 week run using 2048 processors for a 10M grid**
- **Earlier simulations led to design modifications**
  - **Lower riser gas and solids velocities**
  - **Down flow at the wall (clusters are present)**
  - **Improved mixing in the riser**
- **Coal gasifiers are integral part of current clean coal technologies**



PSDF reactor at Wilsonville: Validation

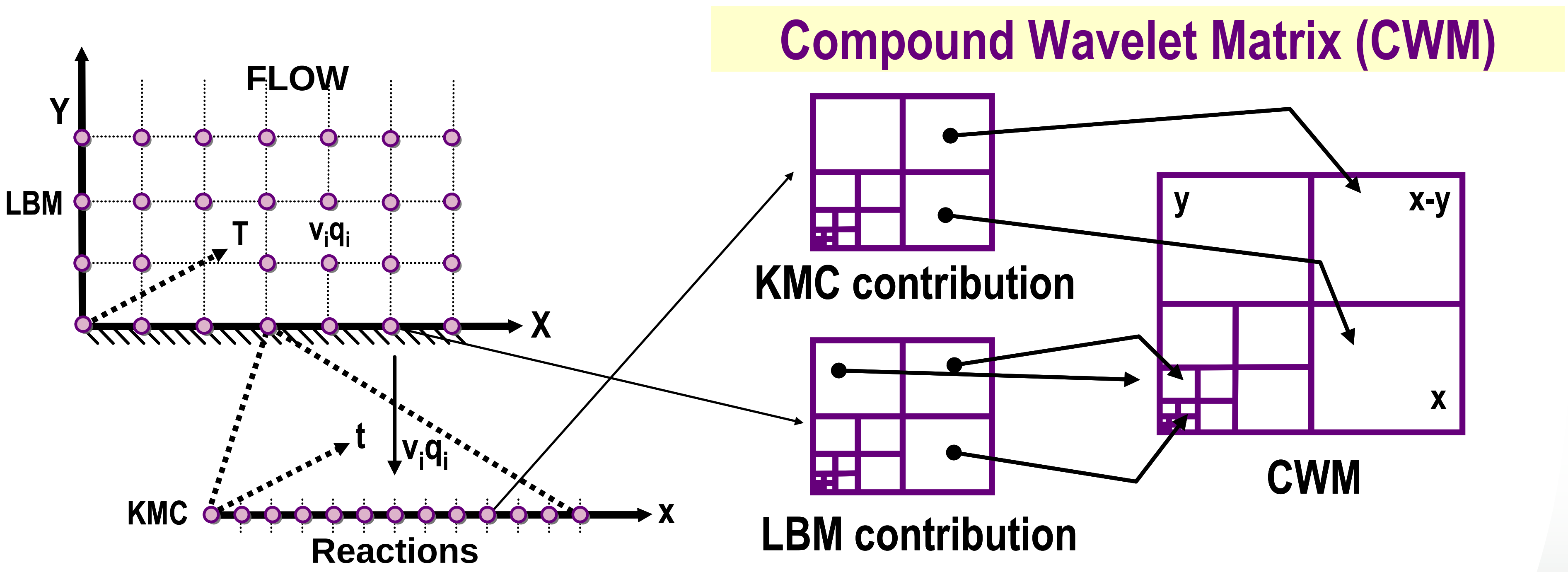
MFIX/Experiment	PRB Oxygen Blown	PRB Air Blown	Hiawatha Air Blown	Hiawatha Oxygen Blown
CO	12.7/11.7	14/11	3.4/3.5	8.5/6.4
CO2	11.2/14.1	5.1/7.4	11/9.3	13.8/12.6
CH4	2.2/2.8	1.9/1	2.6/1.3	3.5/2.3
H2	18/14.7	3/6.2	4.3/4.8	11/9.4
H2O	28/22.9	7/8.3	17/23	37/33.8
CO/CO2	1.1/.8	2.7/1.5	.3/.4	.62/.5
Exit Temp (F)	1668/1674	1749/1757	1763/1779	1783/1714
Percentage of Carbon Conversion	66%/87%	98%/98%	99%/97%	100%/96%
Syngas Rate in Lbs/hr	13400/16000	19800/21000	19400/21000	13200/14600

Courtesy: Chris Guenther @ NETL

MULTISCALE/MULTIPHYSICS COUPLING

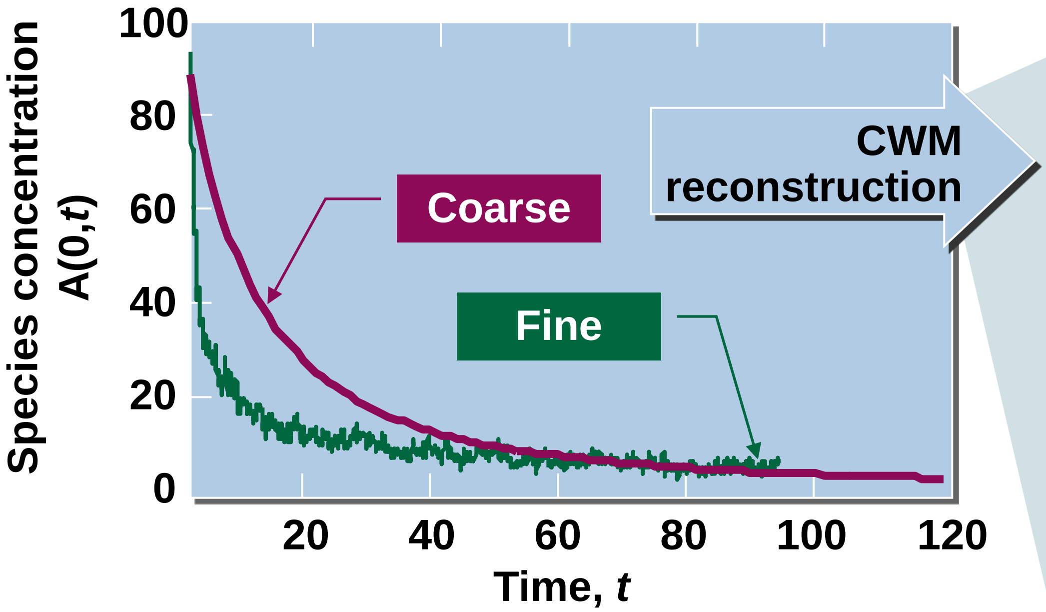
MSMP modeling of heterogeneous chemically reacting flows

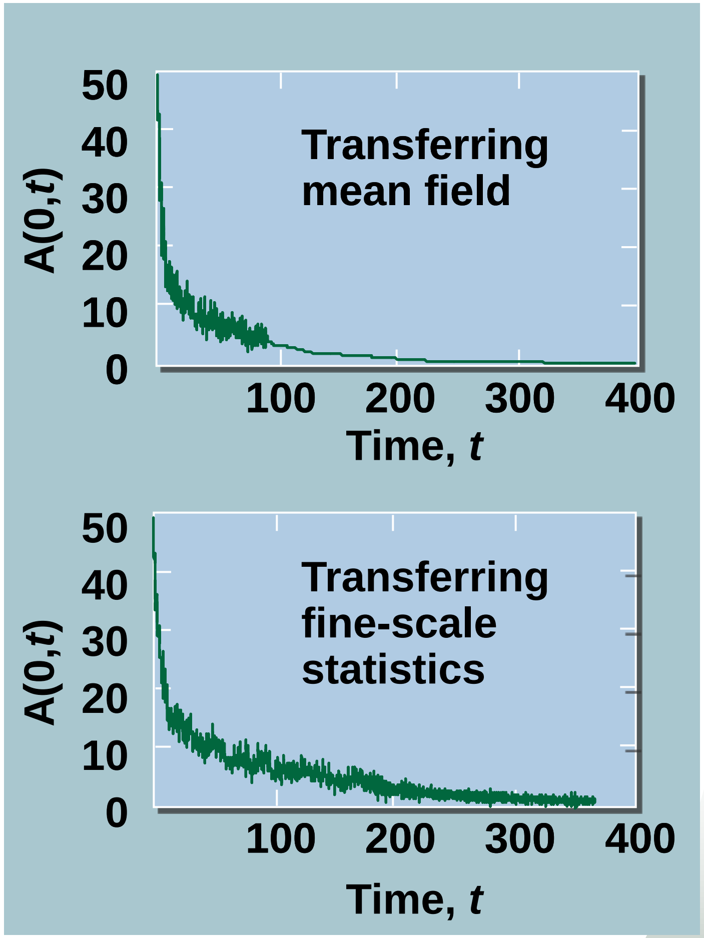
Goal: Develop a MSMP framework for accurate modeling of heterogeneous reacting flows over catalytic surfaces



Procedure: Perform upscaling and downscaling using CWM

Results from a prototype reaction diffusion problem





- **Successfully applied CWM strategy for coupling reaction/diffusion system**
- **An unique way to bridge temporal and spatial scales for MSMP simulations**

*Frantziskonis et al., *International Journal for Multiscale Computational Engineering*, **5-6**, 755, 2006
Muralidharan et al., *Phy. Rev. E*, **77**, 2, 026714, 2008
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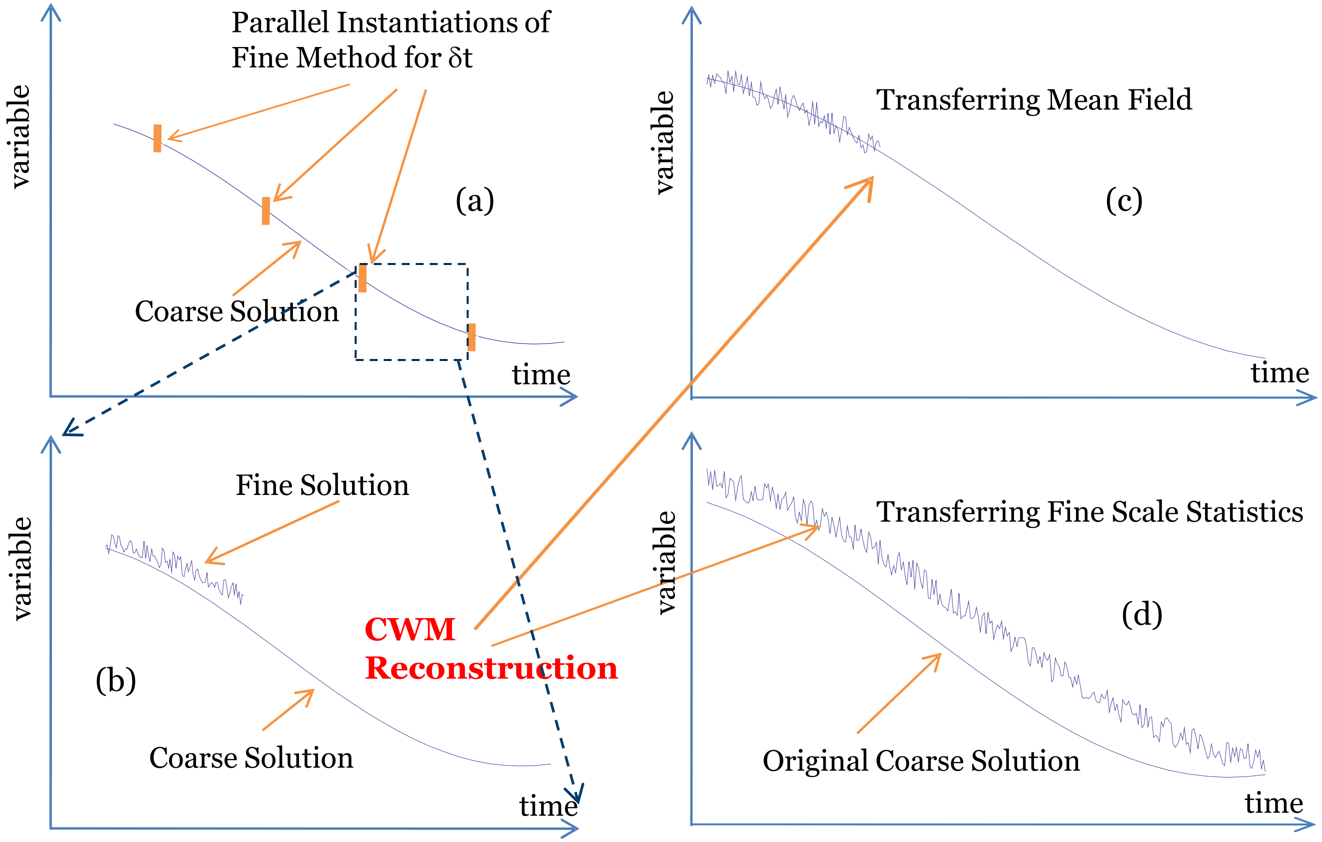
Frantziskonis et al., 3rd IC-SCCE conference, Athens, 2008.

tpCWM (Time Parallel CWM)



Schematic of the TP and CWM methods. (a) The TP method. The fine method instantiates at several temporal "nodes" typically for a period δt that covers time until the next node. (b) The temporal CWM. The fine method is employed for a fraction of the coarse method. (c) The CWM reconstruction updates the mean field. (d) The CWM reconstruction updates the temporal statistics.

tpCWM applied to Lotka-Volterra predator-prey equations

$$X + Y_1 \xrightarrow{\alpha} 2Y_1$$

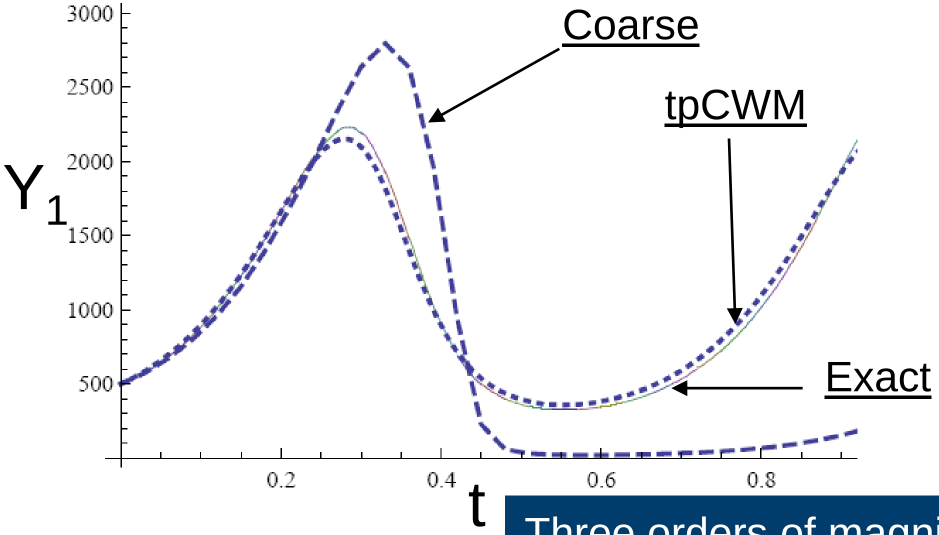
$$Y_1 + Y_2 \xrightarrow{\beta} 2Y_2$$

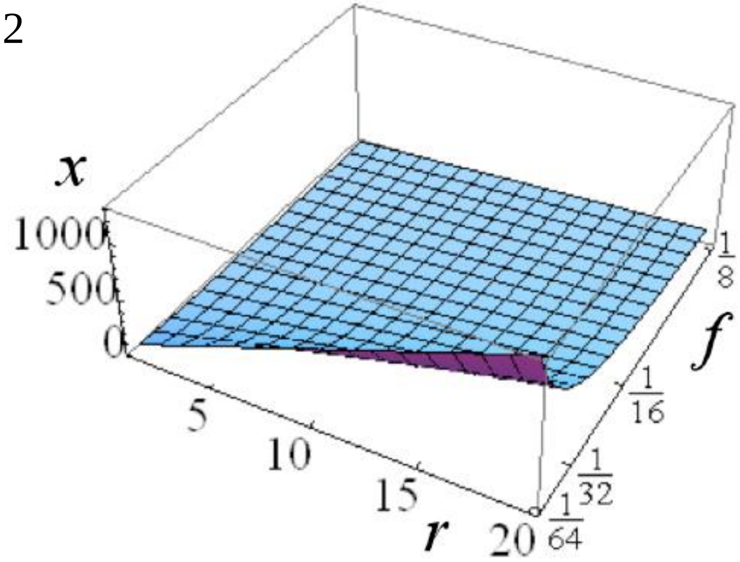
$$Y_2 \xrightarrow{\gamma} Z$$

$$\frac{dY_1}{dt} = \alpha X Y_1 - \beta Y_1 Y_2$$

$$\frac{dY_2}{dt} = \beta Y_1 Y_2 - \gamma Y_2$$

Lotka-Volterra System





Factor of computational savings, X as a function of the ratio r (number of processors/number of iterations) and the fraction f (fraction of KMC time used in each assigned time interval).

Three orders of magnitude savings can be achieved by r in the range of 20 and f in the order of 1/64.

Summary

- **Integrated experiments and simulations at scale can revolutionize the design of energy devices**
  - **Include all relevant scales so that molecular scale interactions are included when designing device scale**
  - **Cut down the current 20-30 year design cycle**
  - **Break cultural barriers**
- **Develop computations based feedback control systems to run devices in most optimal fashion**
  - **Adjust for feedstock etc. online rather than offline adjustments with huge safety margins**
- **Simulation science can and *has to* play a catalytic and important role in bringing innovation to the energy market place**

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