Chemical Reactor Modeling using Computational Fluid Dynamics

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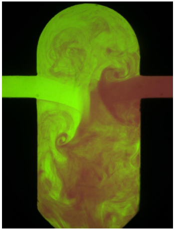
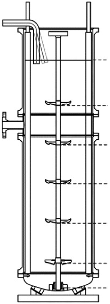
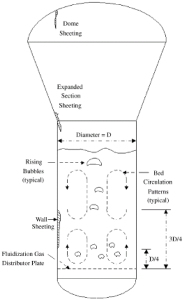
Technologically Relevant Flow Reactors

Turbulent flames

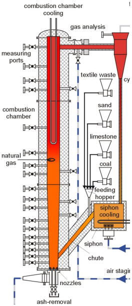
Chemical reactors

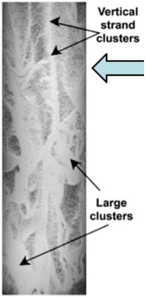






Multiphase flows



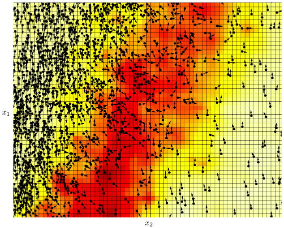


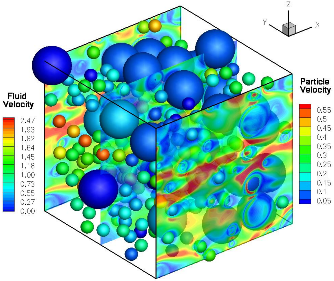
Outline

What is Computational Fluid Dynamics?

Principal steps:

1. Create computational grid
2. Select conservation eqns and closures
   - Mass
   - Chemical species
   - Momentum
   - Energy
   - multiphase, population balance, ...
3. Discretize conservation equations
   - Finite volume for space
   - Stiff ODE solver for reactions
   - Moment method for population balance
   - Lagrangian method for particles, etc.
4. Solve discretized equations
5. Post-process results





Step 1: Computational Grid

Difficulty depends on flow geometry

- Academic cases: simple geometry

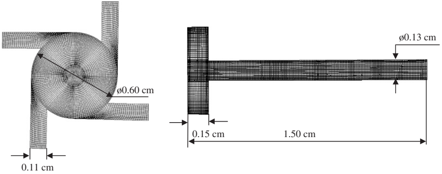
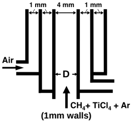
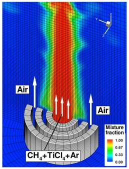
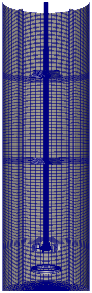


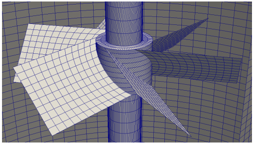
Fig. 4. Geometry and grids of the main mixing chamber.

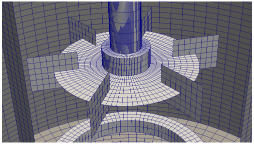




- Industrial cases: complex geometry
  - Grid quality determines solution accuracy
  - Compromise between capturing geometric complexity and solver accuracy
  - Non-negligible time and cost!



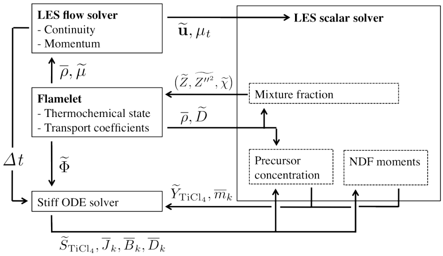


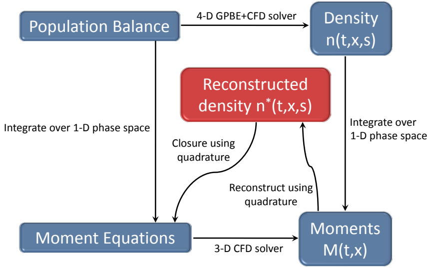


Step 2: Computational Model

Difficulty depends on flow physics

- Single-phase flow
  - Direct numerical simulation (DNS)
  - Turbulence model for unresolved scales (RANS, LES)
- Reacting flow
  - DNS for gas phase
  - RANS or LES for liquid phase
  - Complex chemistry and closures for unresolved scales
- Multiphase flow
  - DNS for "simple" flows
  - Multi-fluid model with closures
  - Multi-scale model key to success



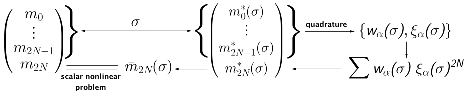


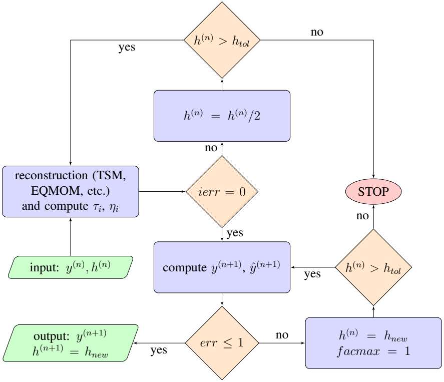
Close moment equations by reconstructing density function

Step 3: Computational Method

Difficulty depends on flow model

- Single-phase flow
  - Finite volume in space
  - Explicit/implicit in time
- Reacting flow
  - Look-up table for chemistry
  - Realizable solver for moments
- Multiphase flow
  - Coupled solver for phase exchanges (mass/momentum)
  - Operator splitting for source terms (reactions, aggregation)
  - Lagrangian and/or Eulerian solver for disperse phase





Step 4: Solver Implementation

Difficulty depends on algorithm

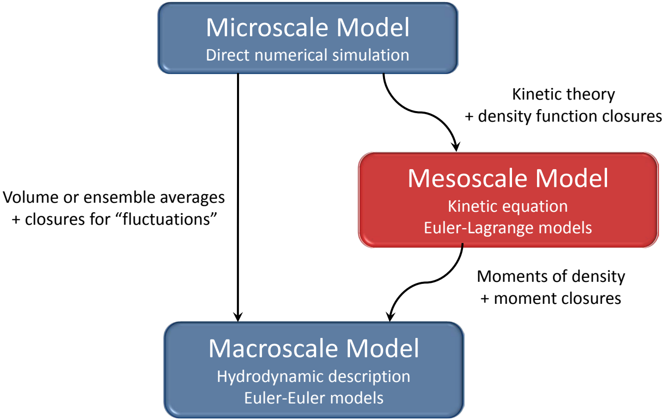
- Multi-CPU workstation
  - Small problems with simple models
  - Single-phase RANS
- Multi-CPU cluster
  - Larger problems with simple models
  - Single-phase LES
  - Simple multiphase flows
- Peta/Exascale computer
  - Very large problems with simple models
  - DNS of canonical single-phase turbulent flows
  - LES of disperse multiphase flows

Step 5: Post-processing

Difficulty depends on data set

- RANS simulations
  - Small data sets
  - Simple plots
- Single-phase DNS/LES
  - Large data sets
  - Time-dependent velocity/scalar fields
  - Simple plots, 3-D animations
- Multiphase DNS/LES
  - Very large data sets
  - Time-dependent velocity/scalar fields for each phase
  - Simple plots, 3-D animations

Multiscale Modeling Approach

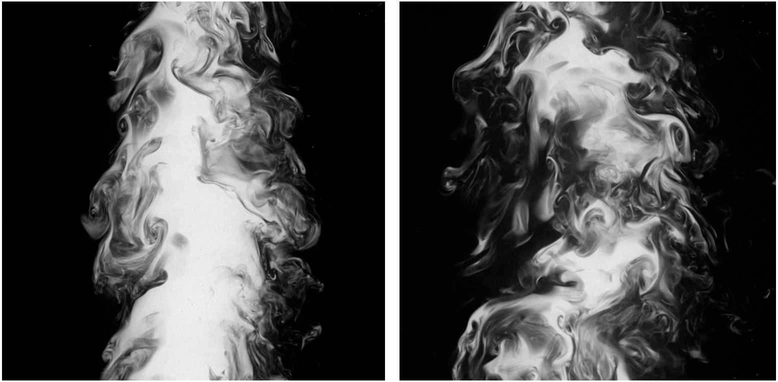


Mesoscale model incorporates more microscale physics in closures!

Outline

Passive Scalar Mixing

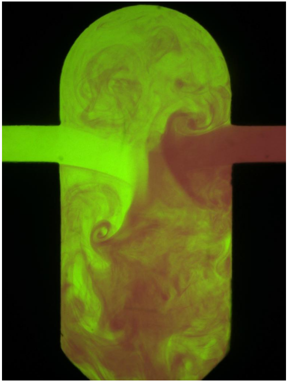
Planar jet in a channel: two downstream locations

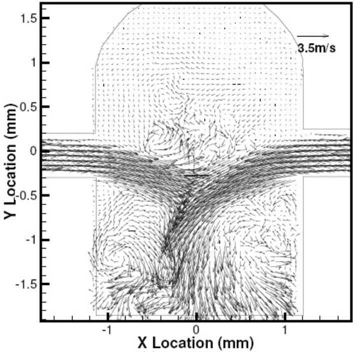


Courtesy of Dr. B Kong

Passive Scalar Mixing

Confined impinging jets



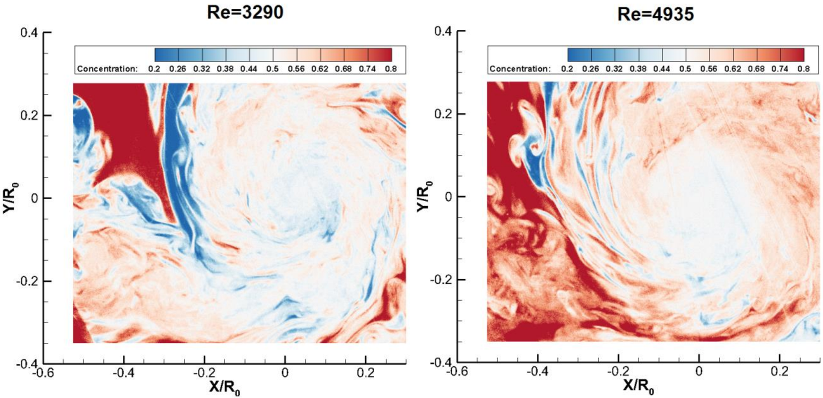


(c) Re 1500

Courtesy of Dr. Y Shi

Passive Scalar Mixing

Multi-inlet vortex mixer

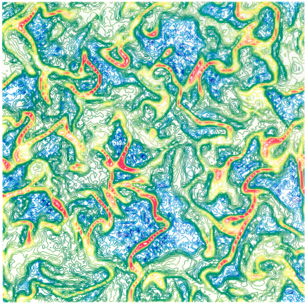


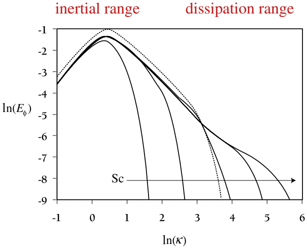
Courtesy of Dr. Z Liu and E Hitimana

Computational Model

Choice depends on *Re* and *Sc*

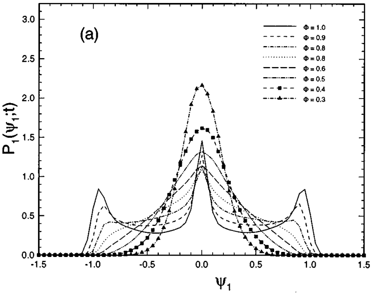
- Laminar flow
  - Direct numerical simulation (DNS)
  - Mixing model for unresolved scales ($Sc \gg 1$)
- Turbulent flow
  - RANS or LES for liquid phase
  - Closures for unresolved scales
- Probability density function (PDF) method
  - PDF transport equation
  - Moments of PDF: mean and variance
  - Closures for transport, mixing



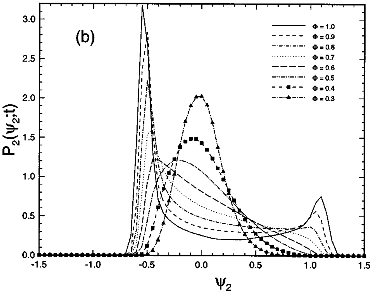


Evolution of PDFs from DNS

Ternary mixing



Binary mixing

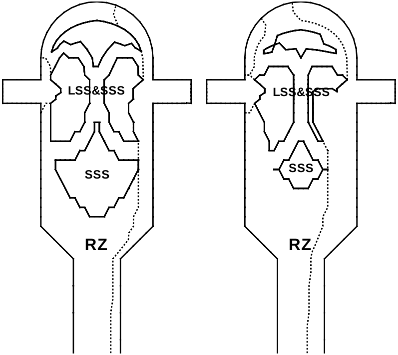


- Means are constant $\langle \phi_1 \rangle$ and $\langle \phi_2 \rangle$
- Variances decrease: $\langle \phi_1'^2 \rangle$ and $\langle \phi_2'^2 \rangle$
- CFD model solves for these variables

CFD Model for Scalar Mixing

$$\frac{\partial \langle \phi \rangle}{\partial t} + \langle \mathbf{u} \rangle \cdot \nabla \langle \phi \rangle = \nabla \cdot (\Gamma_T \nabla \langle \phi \rangle) \quad \Longrightarrow$$ LSS: large-scale segregation

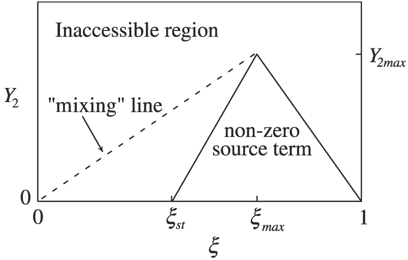
$$\frac{\partial \langle \phi^2 \rangle}{\partial t} + \langle \mathbf{u} \rangle \cdot \nabla \langle \phi^2 \rangle = \nabla \cdot (\Gamma_T \nabla \langle \phi^2 \rangle) - \varepsilon_\phi \quad \Longrightarrow$$ SSS: small-scale segregation

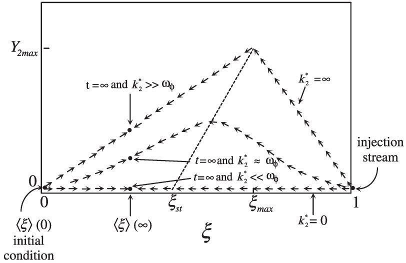


Reactive Scalars: $A + B \to R$, $B + R \to S$

Joint PDF $f(\xi, Y_2)$ for mixture fraction ξ and progress variable Y_2

Reaction source term for Y_2

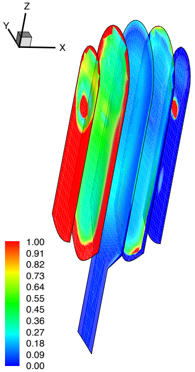




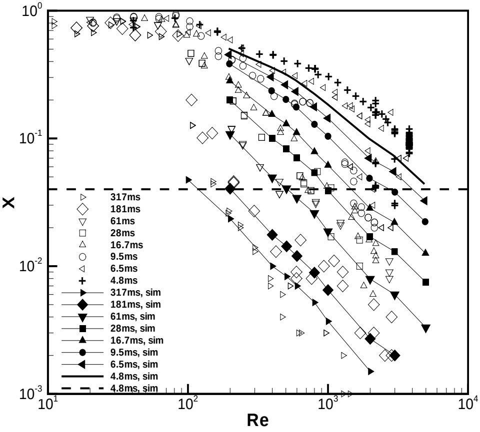
- PDF is “discretized” using moment methods
- CFD code solves moment transport equations

Confined Impinging-Jets Reactor

Damköhler number

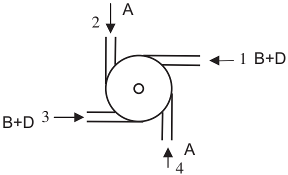


Conversion of undesired product



lines: CFD; symbols: Johnson & Prud'homme (2003)

Multi-Inlet Vortex Reactor



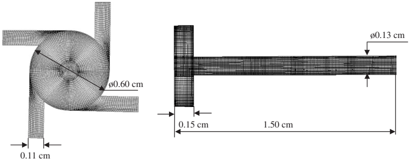


Fig. 4. Geometry and grids of the main mixing chamber.

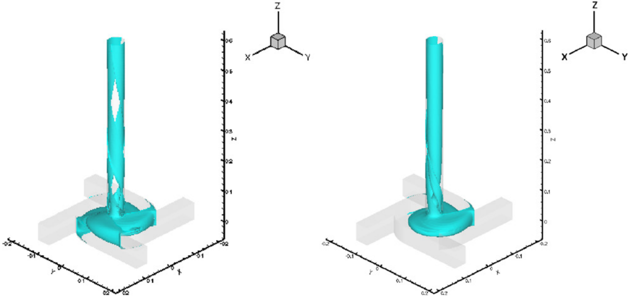


Fig. 12. Acid concentration at $Re = 1371$ for symmetric arrangement of the inlet streams (a) and asymmetric arrangement of the inlet streams (b).

CFD Validation for MIVR

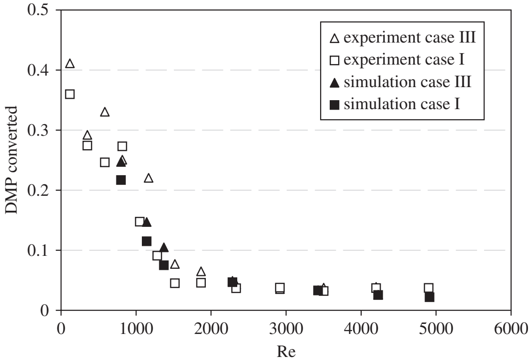
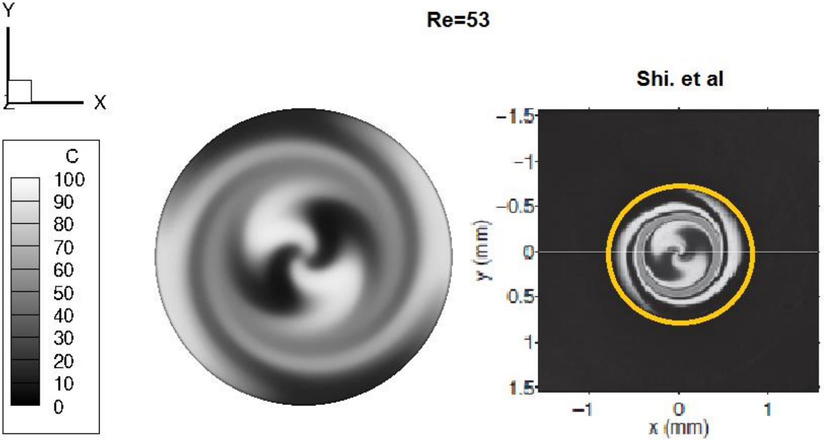


Fig. 10. Conversion of DMP vs. *Re* in the MIVM (■, numerical simulation data for case I; ▲, numerical simulation data for case III; □, experimental measurement for case I; △, experimental measurement for case III).

Large-Eddy Simulation of MIVR

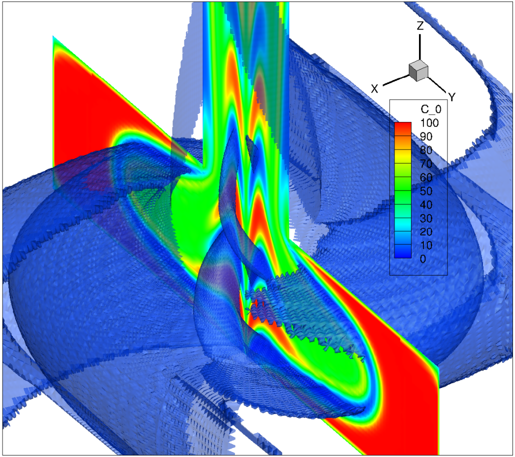
LES PLIF



Courtesy of Dr. Z Liu

LES of MIVR

Bypassing fluid arrives from bottom of mixer



Courtesy of Dr. Z Liu

Scale Up of MIVR

Excellent predictions compared to SPIV

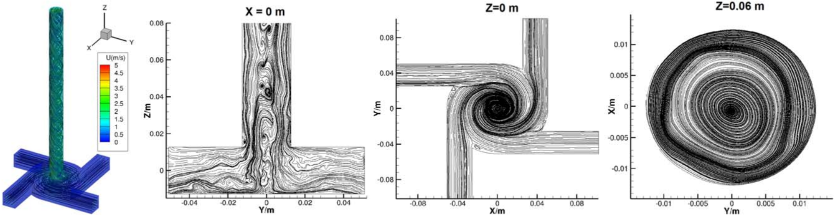
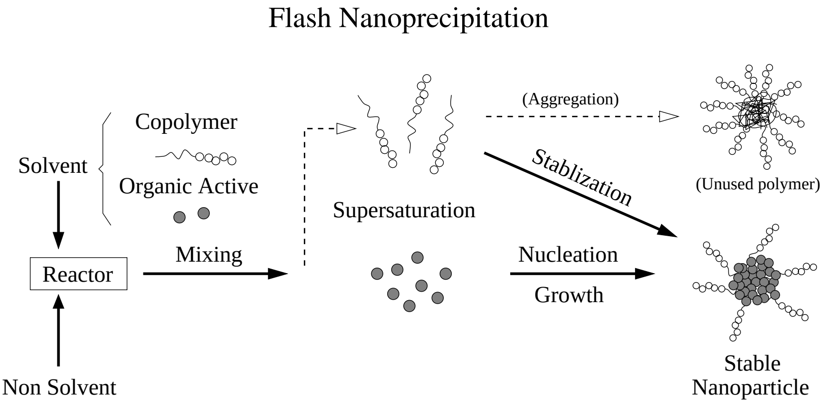


Figure 6. Instantaneous velocity field (left: contour plot of velocity magnitude, middle left: vertical crosssection through center of outlet, middle right: horizontal cross-section through center of inlets, right: horizontal cross-section at outlet).

Outline

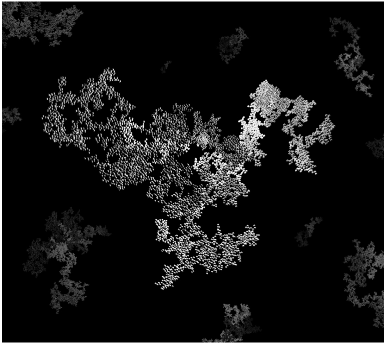
Nanoparticles in Turbulent Flow Reactors



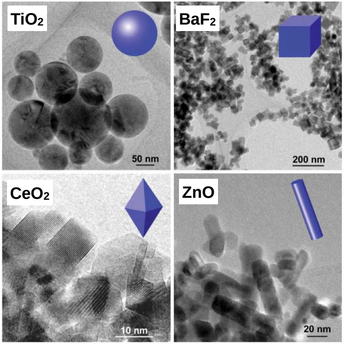
Produced in CIJR & MIVR

Nanoparticles in Turbulent Flames

Soot



Metal oxides



(Strobel & Pratsinis 2007)

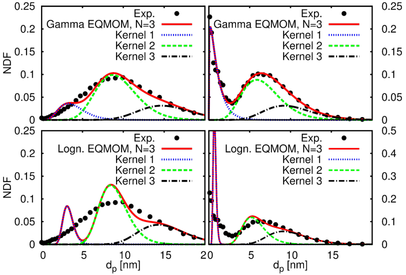
Produced in turbulent flames

Computational Model

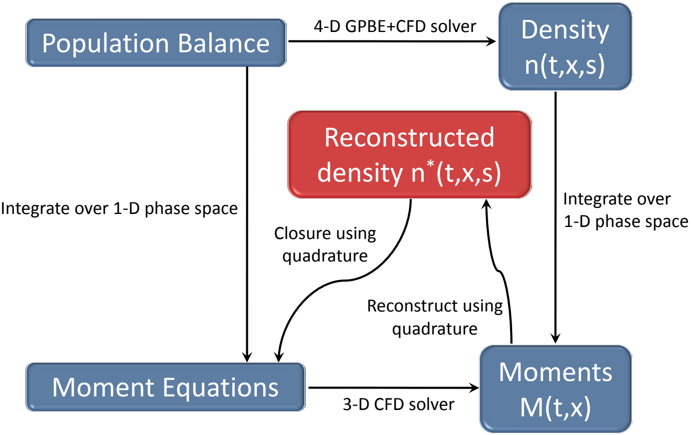
Particle size distribution (PSD)

- Population balance equation
  - Solve for NDF $n(v)$
  - Coupled to velocity and reactive scalars
  - Non-linear integro-PDE
- Moment methods
  - Solve for moments of NDF
  - Close by reconstructing $n(v)$

$$\underbrace{\frac{\partial n(v)}{\partial t}}_{\text{(Rate of change)}} + \underbrace{\nabla \cdot \left(\mathbf{u}\, n(v)\right)}_{\text{(Convection)}} = \underbrace{\nabla \cdot \left(\Gamma(v,\Phi)\, \nabla n(v)\right)}_{\text{(Particle diffusion)}} + \underbrace{J(\Phi)\, f(v_0)}_{\text{(Nucelation)}}$$
$$+ \underbrace{\int_0^\infty \beta(v-v',v')\, n(v-v')\, n(v')\, dv'}_{\text{(Aggregation-birth)}}$$
$$- \underbrace{\int_0^\infty \beta(v,v') n(v) n(v')\, dv'}_{\text{(Aggregation-death)}} ,$$

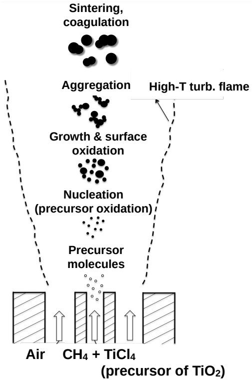


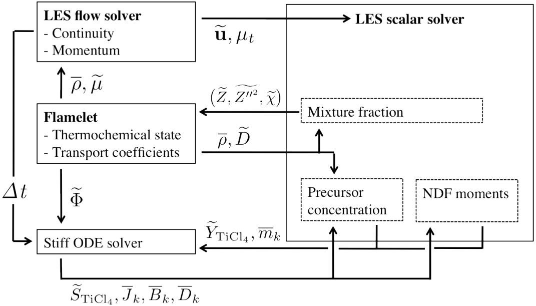
Quadrature-Based Moment Methods



Close moment equations by reconstructing density function

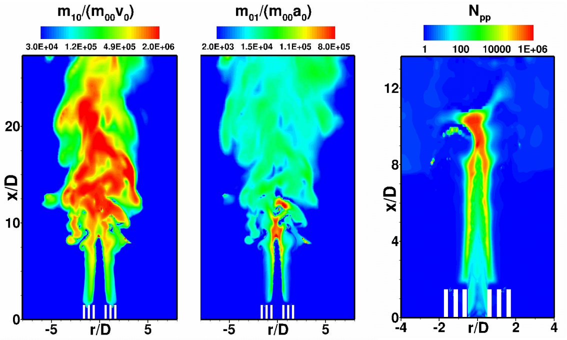
Flame Synthesis of Metal Oxide Nanoparticles





- Flamelet model for combustion
- Moment closure for volume–surface NDF $n(v, a)$
- Account for nucleation, growth, aggregation, and sintering

LES Results

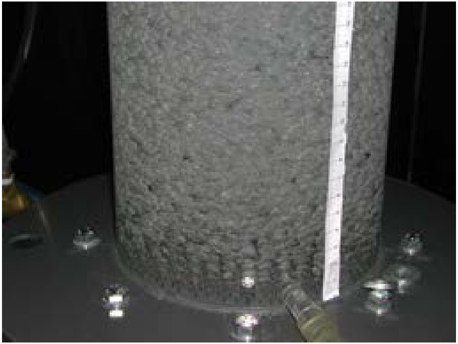


- m_{10} mean particle volume
- m_{01} mean particle surface area
- N_{pp} primary particles (nuclei)

Outline

1. Introduction
2. CFD for Turbulent Flows
3. Turbulent Flow Reactors
4. **Bubbly Flow Reactors**
5. Particle-Laden Flow Reactors
6. Conclusions

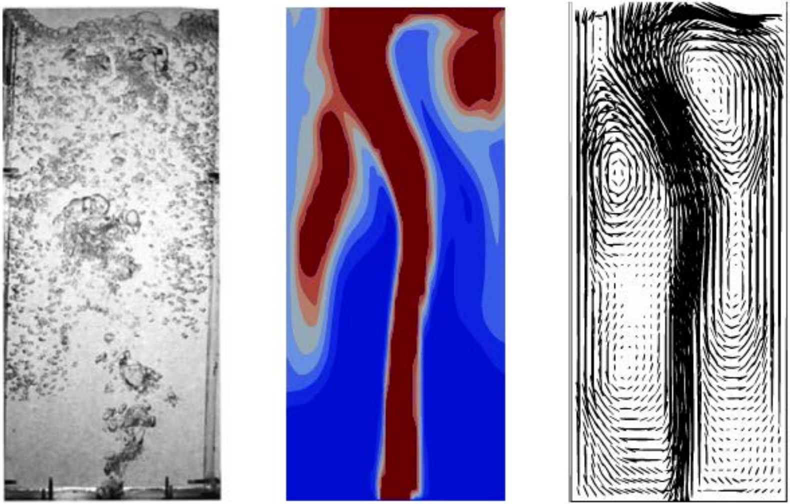
Bubbly Flow Reactors



Homogeneous

Heterogeneous

Polydisperse Bubbly Flows



Computational Model

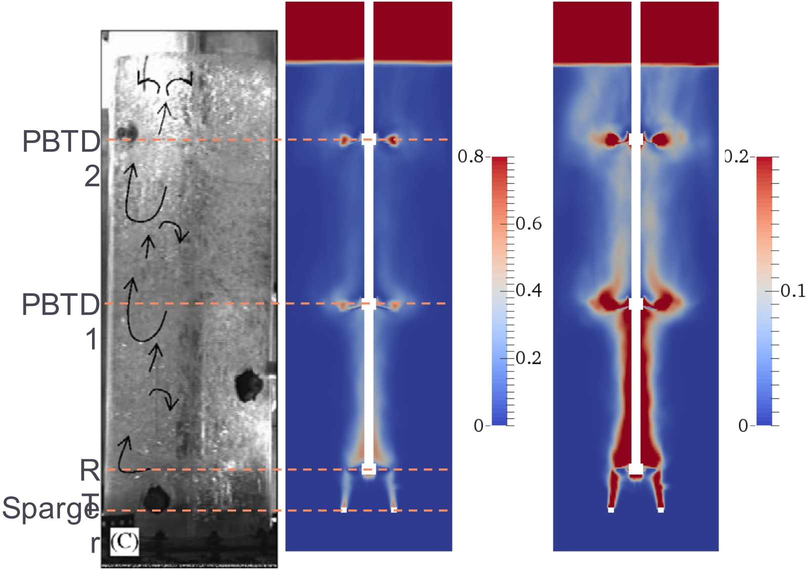
Two phases: gas and liquid

- Mass & momentum balances
  - Buoyancy & drag forces
  - Added mass & lift forces
  - Strong phase coupling
  - Flow regime depends on forces

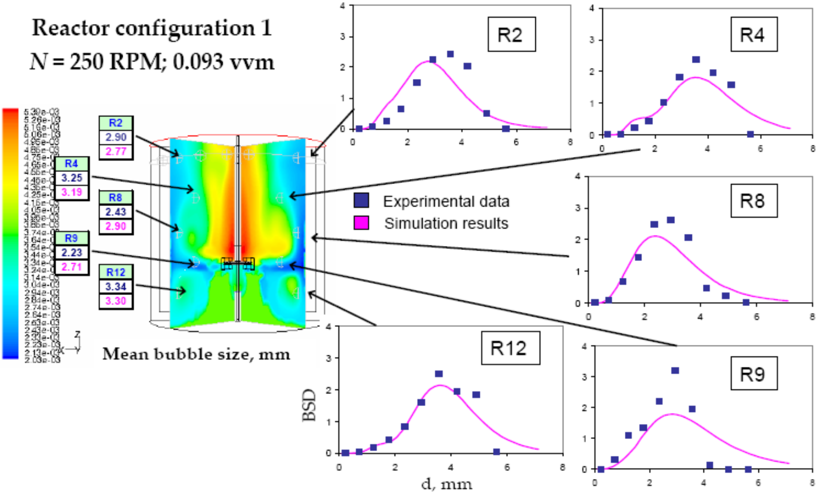
Bubble size distribution (BSD)

- Population balance equation
  - Solve for NDF $n(v)$
  - Size-conditioned bubble velocity
  - Non-linear integro-PDE
- Moment methods
  - Solve for moments of NDF
  - Close by reconstructing $n(v)$

Stirred Reactor with Gas Sparger



Stirred Reactor with Bubble Size Distribution



Courtesy of Dr. DL Marchisio

Outline

Particle-Laden Flow Reactors

Volume fraction

Fluid velocity

Computational Model

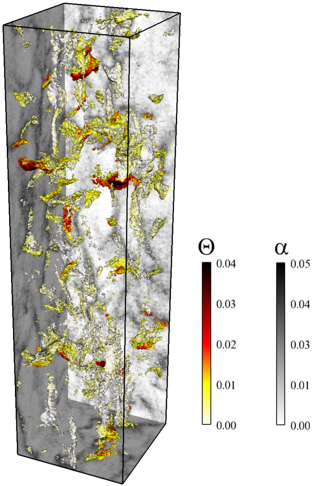
Two phases: solid and gas

- Mass, momentum & energy balances from Kinetic Theory of Granular Flow
  - Buoyancy & drag forces
  - Granular temperature
  - Strong phase coupling due to drag

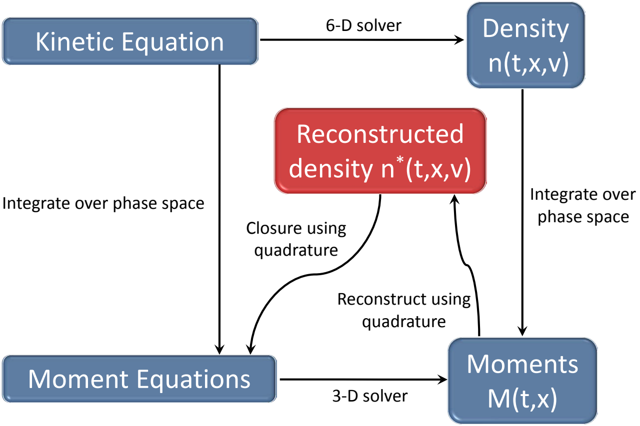
Euler-Euler (EE) & Euler-Lagrange (EL)

- Kinetic equation
  - NDF of particle velocity
  - EL solver tracks individual particles

  - EE solver finds moments of NDF
  - Use more moments to capture high Knudsen number flows



Quadrature-Based Moment Method



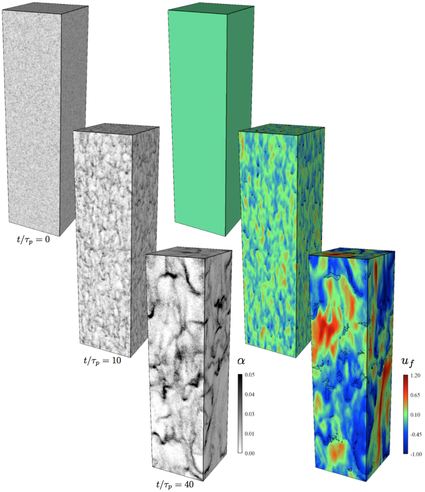
Close moment equations by reconstructing density function

Particle Trajectory Crossing

Kinetic equation without collisions

Courtesy of R Patel

Cluster-Induced Turbulence



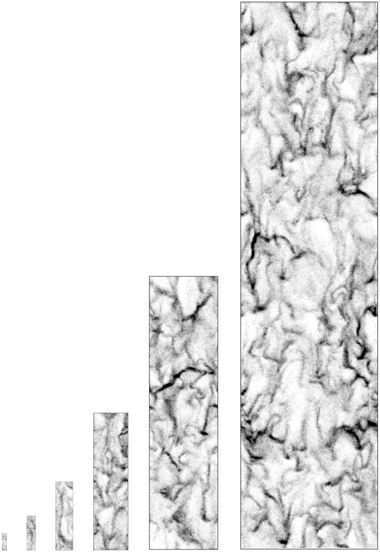
- Particles fall due to gravity
- Fluid velocity in zero due to pressure gradient
- Clusters form spontaneously
- Clusters drag fluid downward, create wakes
- Fluid turbulence breaks up clusters

Do Eulerian and Lagrangian Models Agree?

Cluster-induced turbulence

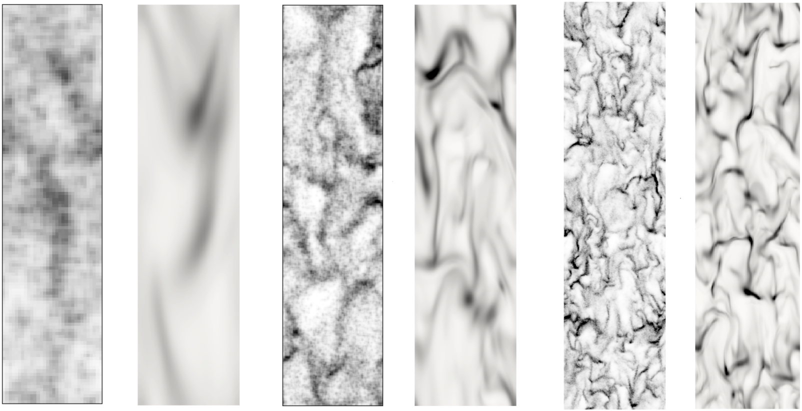
- Average volume fraction: $\alpha_{\mathrm{p}} = 0.01$
- $\rho_{\mathrm{p}}/\rho_{\mathrm{g}} = 1000$, $\mathrm{Re}_{\mathrm{p}} = 0.5$
- terminal velocity: $\mathcal{V} = 0.1$ m/s
- cluster length: $\mathcal{L} = 2.5$ mm
- $L_x/\mathcal{L} = 129$ $(2048 \times 512 \times 512)$

Euler-Lagrange and Euler-Euler simulations performed on same grid, but not with same numerical schemes



Qualitative Comparison of Clustering

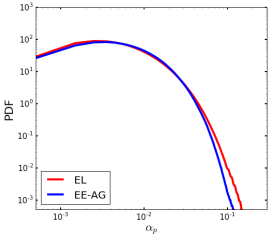
Case 2 Case 4 Case 6



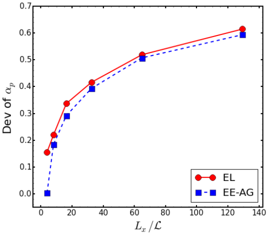
Similar shapes, but EE has slightly longer/wider clusters

Quantitative Comparison

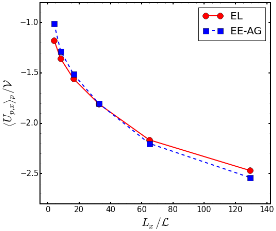
α_p PDF



Inhomogeneity



Drift velocity

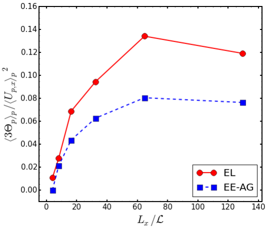


EE has slightly fewer high α_p values (due to numerics?)

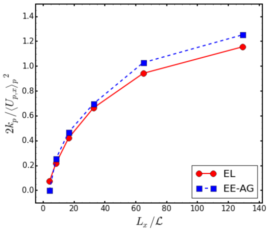
EE has slightly higher drift velocity (due to larger clusters?)

Quantitative Comparison of TKE Statistics

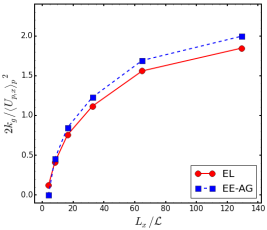
Θ_p



k_p



k_f

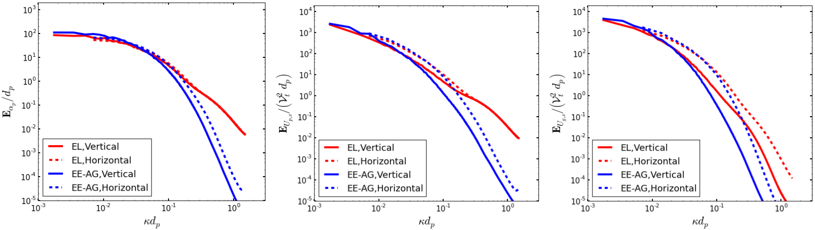


EE has lower uncorrelated TKE (due to EL post-processing?)

EE has higher correlated TKE (due to larger clusters?)

Quantitative comparison of energy spectra

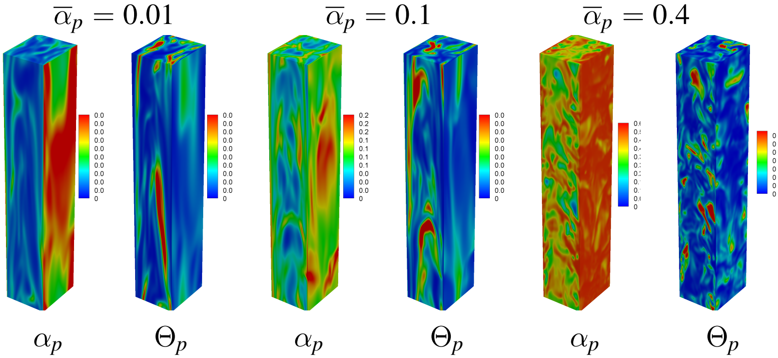
α_p $\qquad$ U_p $\qquad$ U_f



Good agreement at small wavenumbers ($\kappa d_\mathrm{p} < 0.1$)

EE has less energy than EL at large wavenumbers
(due to EL filter for coupling, numerics?)

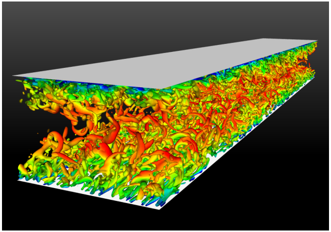
Particle-Laden Channel Flow



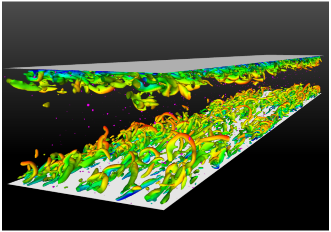
Transition from clusters to bubbles

Effect of Increasing Particle Mass Loading

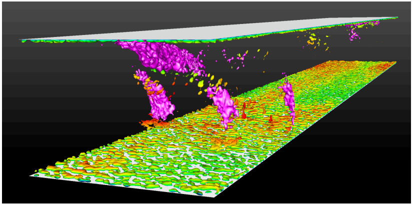
0.2



1.2



20



Courtesy of Dr. J Capecelatro

Bubbling Fluidized Bed Reactor

α_p h_2 $U_{p,y}$ Θ_p $\sigma_{p,xy}$

Outline

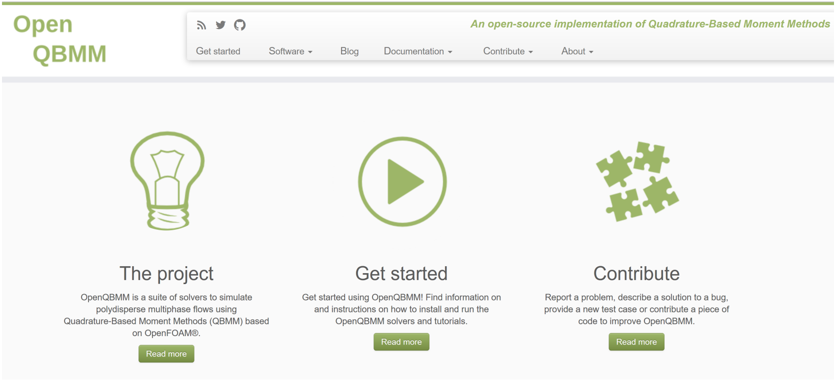
Conclusions

- Computational fluid dynamics can be applied to a wide range of technologically relevant flow reactors
- Multi-scale modeling is required to treat the disparate length and time scales present in most applications
- Development of computational models based on fundamental physics and chemistry is a key step
- Computational methods targeted at accurately solving the computational models are equally important

Successful CFD modeling requires a team effort
with complementary skill sets

OpenQBMM Project (www.openqbmm.org)

Polydisperse flow simulation tools in OpenFOAM



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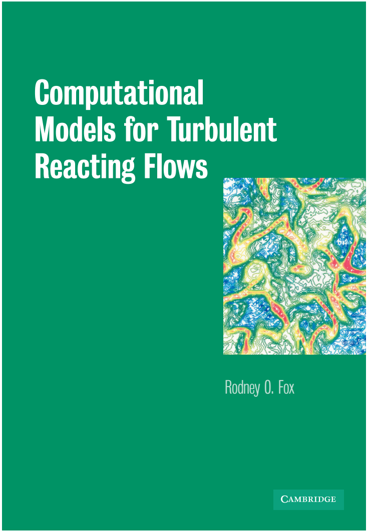
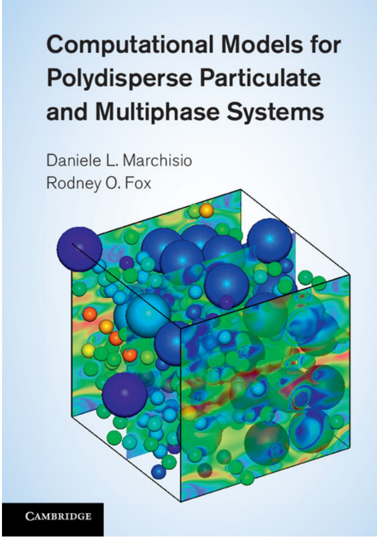
Further Reading





Thanks for your attention!

Questions?