CFD Modeling of Flow, Mixing and Reaction

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Outline

1. Introduction
2. Reactive Mixing: Flash Nanoprecipitation
3. Polydisperse Particles: Nanoparticle Flame Synthesis
4. Macroscale Particles in a Fluid: Gas-Liquid Reactors
5. Inertial Particles: Gas-Solid Reactors
6. Conclusions

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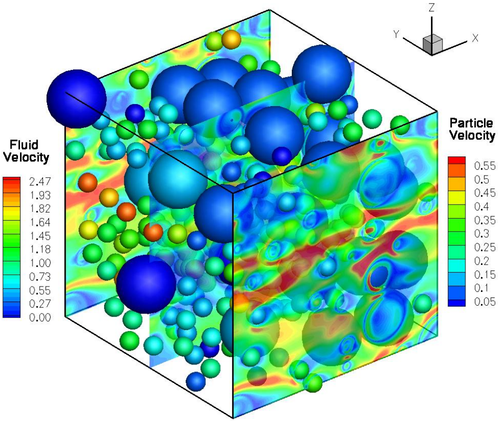
Multiphase Reacting Systems

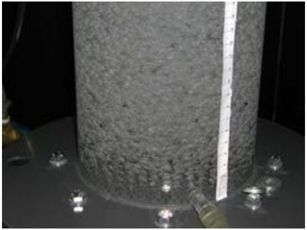
- continuous phase
- disperse phase
- size distribution
- finite particle inertia
- particle collisions
- variable mass loading
- chemical reactions
- heat and mass transfer
- multiphase turbulence



Bidisperse gas-particle flow (DNS of S. Subramaniam)

Multiphase Reacting Systems

Bubble columns



Brown-out

Jet break up

Power stations

Environmental

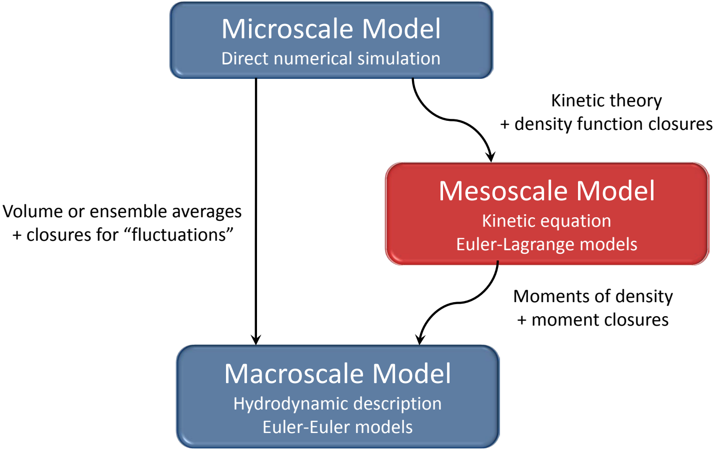
Spray flames

CFD Modeling Challenges in CRE Applications

- Strong coupling between continuous and disperse phases
- Wide range of particle volume fractions (even in same system!)
- Inertial particles with wide range of Stokes numbers
- Collision-dominated to collision-less regimes in same system
- Particle polydispersity (e.g. size, density, shape) is always present
- Chemical reaction in one (or all) phases
- Wide range of chemical and physical time scales

Need a modeling framework that can handle all aspects!

Kinetic-Based Modeling Approach



Mesoscale model incorporates more microscale physics in closures!

Hierarchy of CFD Models

- Liquid-phase reacting flows $\Longrightarrow$ mixing-limited reactions
- Gas-phase reacting flows $\Longrightarrow$ combustion + nanoparticles
- Bubbly flows $\Longrightarrow$ bubble size/chemical distribution
- Gas-particle flows $\Longrightarrow$ size-dependent particle inertia

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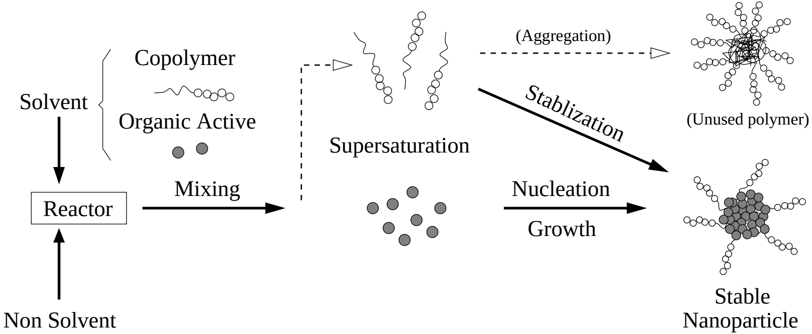
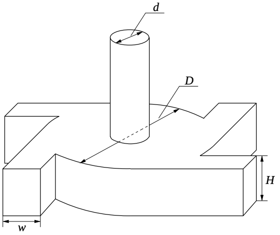
Flash Nanoprecipitation



Product quality depends on fast mixing!

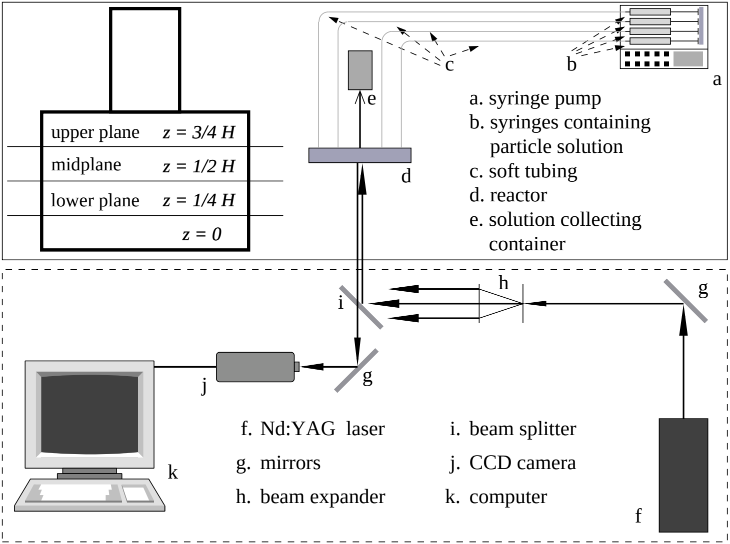
Multi-Inlet Vortex Reactor (MIVR)



$$w = 1.19 \text{ mm}$$
$$H = 1.78 \text{ mm}$$
$$D = 6.26 \text{ mm}$$
$$d = 1.40 \text{ mm}$$

$$\mathrm{Re_j} = \frac{u_\mathrm{j} D_\mathrm{h}}{\nu}$$

Experimental Setup



Experimental Velocity Vector Fields

Laminar

Turbulent

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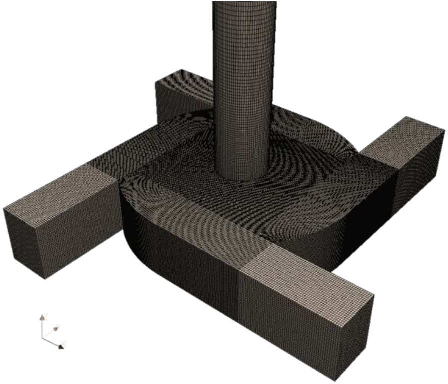
Image taken at mid-plane

CFD Simulation Overview

- Large-eddy simulation (LES) with Probability Density Function methods
- Platform: OpenFOAM



Probability Density Function Methods

Derive a transport equation for PDF of scalars:

closed ← → unclosed

$$\frac{\partial f}{\partial t} + \underbrace{\bar{u} \cdot \nabla f = \nabla \cdot (\Gamma \nabla f)}_{\text{macro/mesomixing}} + \frac{\partial}{\partial \psi}(S(\psi) f) - \underbrace{\frac{\partial}{\partial \psi}\left(D\langle \nabla^2 \phi | \psi \rangle f\right)}_{\text{micromixing}}$$

<ins>Advantages</ins>:

- Clear separation of mixing phenomena
- Micromixing model is defined locally in space (depends on local length and time scales)
- Numerical implementation can easily capture limiting case of no micromixing (*D*=0)

Micromixing model has a critical (unclosed) role!

Multi-Environment Mixing Model

Discrete representation of PDF:

$$f = \sum_{\alpha=1}^{N} p_\alpha \delta\left(\psi - \phi_\alpha\right) = N \text{ environments with concentrations } \phi_\alpha$$

Find transport equations for p_α and ϕ_α from PDF transport equation:

$$\frac{\partial p_\alpha}{\partial t} + \bar{u} \cdot \nabla p_\alpha = \nabla \cdot (\Gamma \nabla p_\alpha)$$

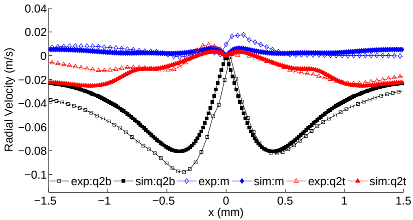
$$\frac{\partial p_\alpha \phi_\alpha}{\partial t} + \bar{u} \cdot \nabla (p_\alpha \phi_\alpha) = \nabla \cdot (\Gamma \nabla p_\alpha \phi_\alpha) + \underbrace{\frac{p_\alpha}{\tau_\phi}\left(\langle \phi \rangle - \phi_\alpha\right)}_{\text{micromixing}} + \underbrace{p_\alpha S(\phi_\alpha)}_{\text{reactions}}$$

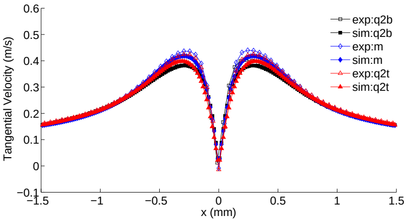
In simplest cases, *N* equals number of inlets

“Minimal” mixing model for liquid-phase reactors

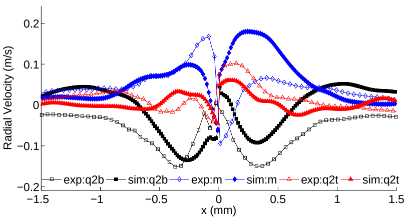
Validation: Mean Velocity Profiles

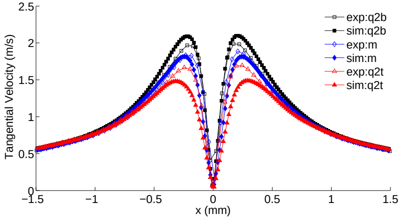
Laminar CFD vs μ-PIV





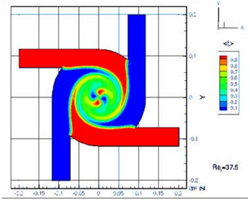
Turbulent CFD vs μ-PIV

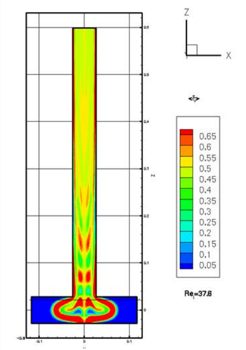




Validation: Passive Scalar Mixing

$\mathrm{Re_j} = 39$





$\mathrm{Re_j} = 240$

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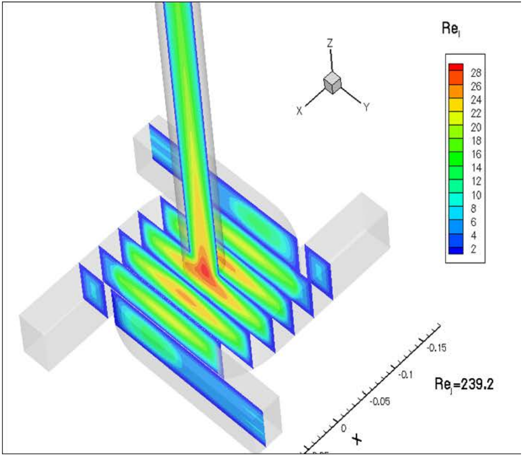
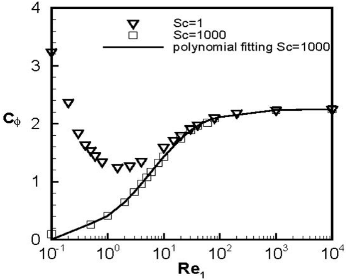
Micromixing Parameter for Reactive Mixing





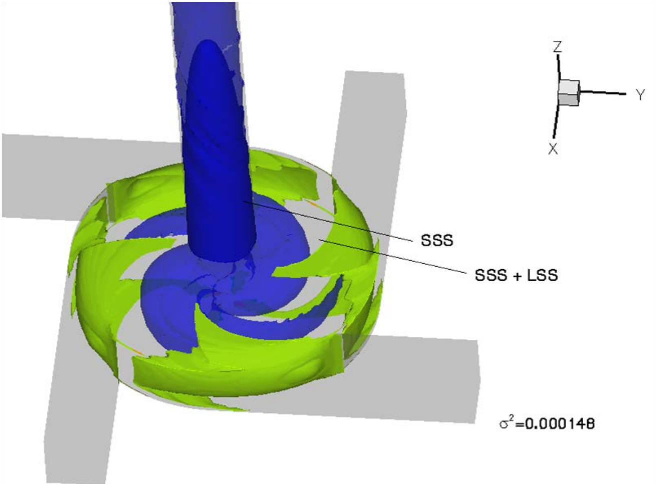
Standard model: $C_\phi = 2$

Turbulent Re is relatively small!

$$\frac{1}{\tau_\phi} = C_\phi \frac{\varepsilon}{k}$$

Mixing Regions in MIVR

$\mathrm{Re}_\mathrm{j} = 240$



SSS = micromixing incomplete

LSS = macromixing incomplete

Summary of CFD Models for Reactive Mixing

- Single-phase reactive mixing is treated with PDF methods
- Large-scale mixing is handled by CFD model for fluid velocity
- Small-scale mixing requires a micromixing model
- Micromixing depends on Schmidt and local Reynolds number

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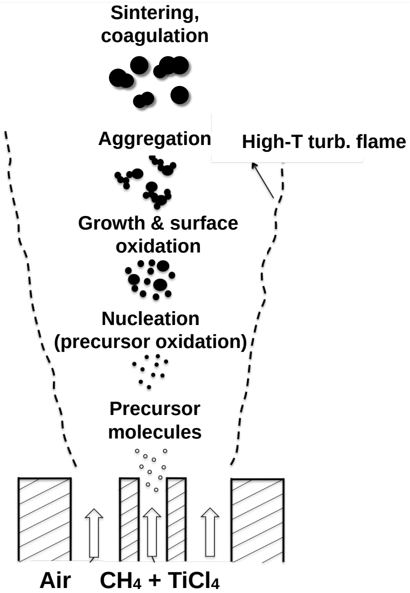
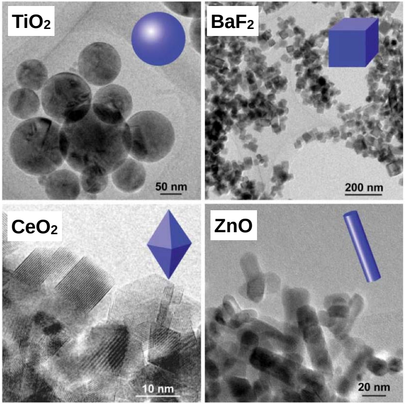
Flame Synthesis of Nanoparticles





(Strobel & Pratsinis 2007)

Population Balance Equation

$$\frac{\partial n(s)}{\partial t} + \underbrace{\nabla \cdot [\mathbf{u}n(s)]}_{\text{advection}} = \underbrace{\nabla \cdot [\Gamma(\Phi, s)\nabla n(s)]}_{\text{diffusion}} + \underbrace{J(\Phi)f(s)}_{\text{nucleation}}$$

$$+ \underbrace{\int_0^s \beta(s - s', s')n(s - s')n(s')\, ds'}_{\text{aggregation - birth}} - \underbrace{\int_0^\infty \beta(s, s')n(s)n(s')\, ds'}_{\text{aggregation - death}}$$

Solved in CFD using Quadrature-Based Moment Methods

Quadrature-Based Moment Methods (QBMM)

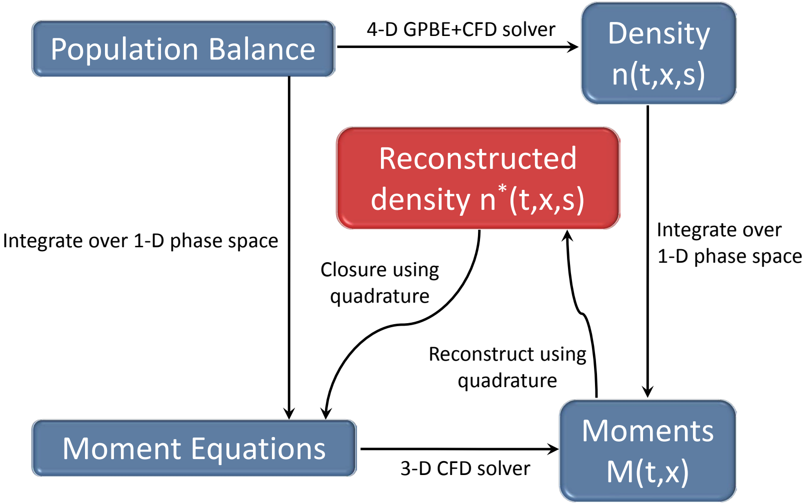
Basic idea: Given a set of moments of number density function (NDF), reconstruct the NDF

Things to consider:

- Which moments should we choose?
- What method should we use for reconstruction?
- How can we extend method to multivariate NDF?
- How should we design the CFD solver for the moments?

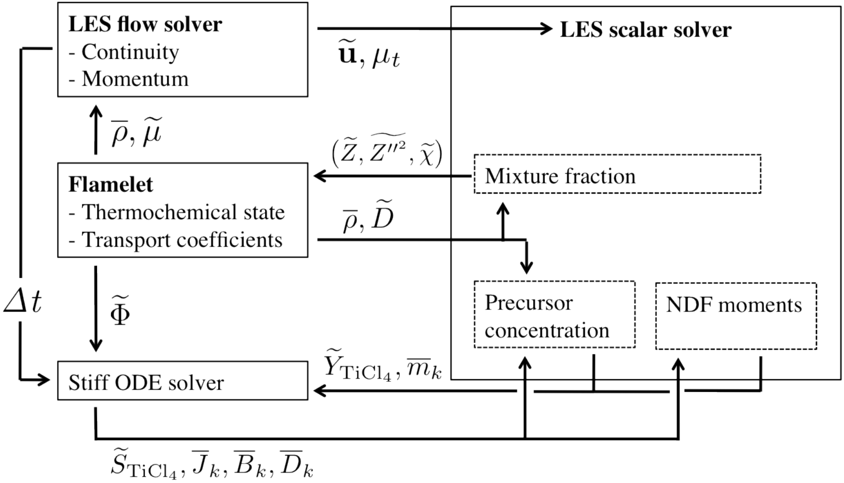
We must demonstrate *a priori* that numerical algorithm is robust!

CFD with Moment Methods

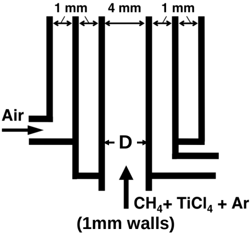


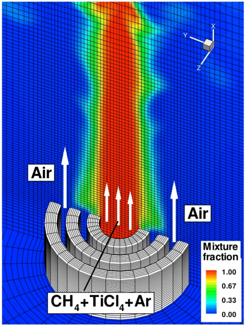
Close moment equations by reconstructing density function

LES Flow and Scalar Solver



Flow Configuration





Sample Results

Temperature

$TiCl_4$

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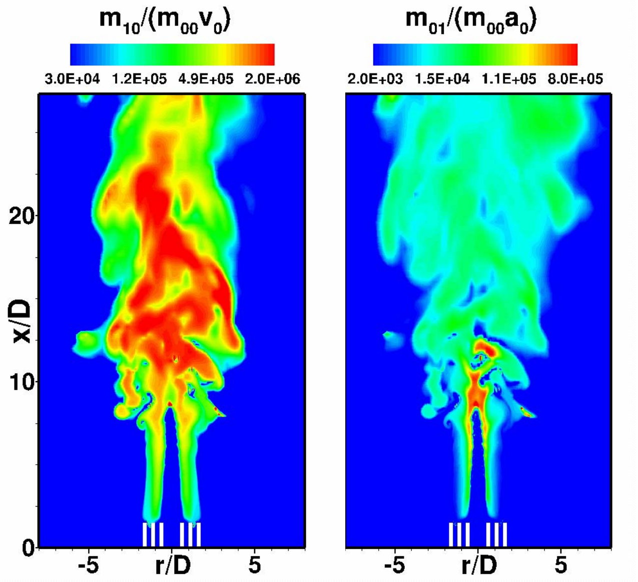
Average Particle Volume and Surface Area



Summary of CFD Models with Population Balances

- PBE arise in many CRE applications
- It is often necessary to use a multivariate PBE
- CFD models based on moment methods are computationally tractable
- Quadrature-Based Moment Methods offer distinct advantages for CFD

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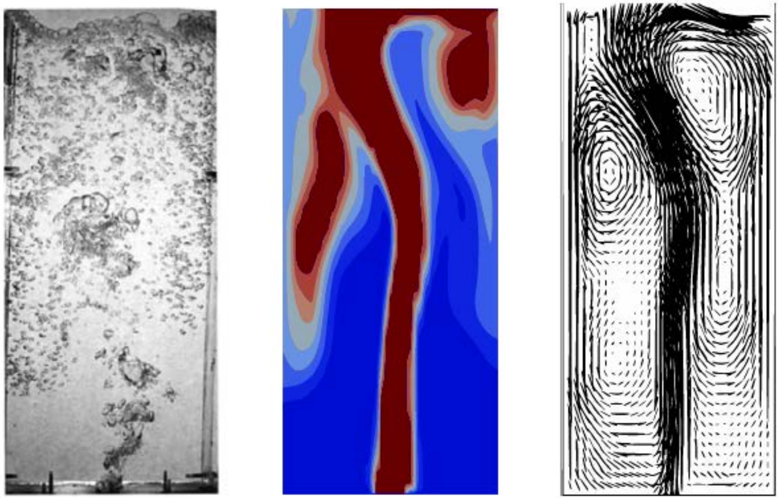
Polydisperse Bubbly Flow



CFD Model for Bubbly Flow

Model must account for

- Liquid-phase continuity and momentum
- Gas-phase continuity and momentum (i.e. moments of NDF)
- Bubble size distribution (with size-dependent velocity)
- Coalescence, breakage, mass transfer, ...

Describe bubble phase using **Generalized Population Balance Equation**

Generalized Population Balance Equation

- GPBE has 4-D phase space: bubble velocity $\mathbf{v}$ and bubble size s

$$\frac{\partial n}{\partial t} + \mathbf{v} \cdot \frac{\partial n}{\partial \mathbf{x}} + \frac{\partial}{\partial \mathbf{v}} \cdot [\mathbf{A}(t, \mathbf{x}, \mathbf{v}, s)n] + \frac{\partial}{\partial s}[G(t, \mathbf{x}, \mathbf{v}, s)n] = \mathbb{C}$$

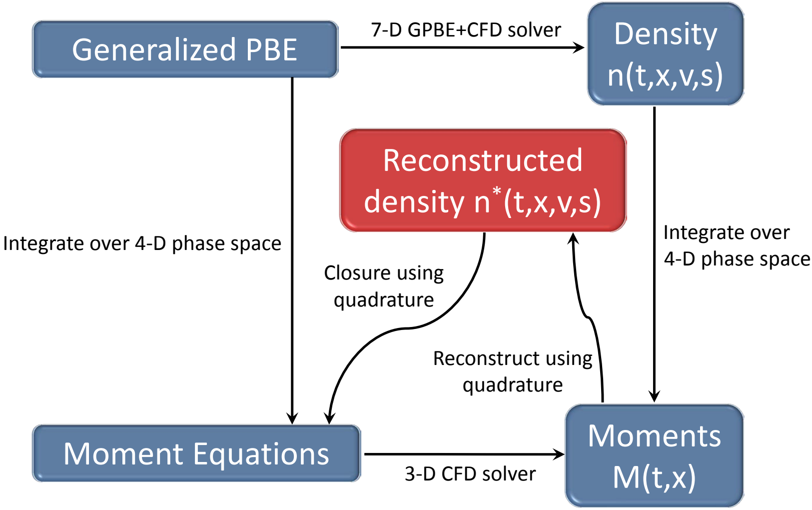
  with known acceleration $\mathbf{A}$, growth G and coalescence $\mathbb{C}$ functions

- In principle, a 4-D reconstruction of $n(\mathbf{v}, s)$ is required

- However, bubbles have small inertia relative to liquid

- Use a monokinetic NDF approximation

CFD with Generalized Population Balance Equation



Close moment equations by reconstructing density function

Monokinetic NDF

Bubbles have small Stokes number $\Longrightarrow$ velocity is function of size

$$n(v, s) = n(s)\delta(v - u(s))$$

- $u(s) = u_0 + u_1 s + u_2 s^2 + u_3 s^3$ found from velocity-size moments

$$M_{i,1} = \int s^i u(s) n(v, s) \, \mathrm{d}v \, \mathrm{d}s = u_0 M_{i,0} + u_1 M_{i+1,0} + u_2 M_{i+2,0} + u_3 M_{i+3,0} \quad \text{for } i = 0, 1, 2, 3$$

- Linear system ($\Longrightarrow$ EQMOM for $n(s)$ with 3 nodes):

$$\begin{bmatrix} M_{0,0} & M_{1,0} & M_{2,0} & M_{3,0} \\ M_{1,0} & M_{2,0} & M_{3,0} & M_{4,0} \\ M_{2,0} & M_{3,0} & M_{4,0} & M_{5,0} \\ M_{3,0} & M_{4,0} & M_{5,0} & M_{6,0} \end{bmatrix} \begin{bmatrix} u_0 \\ u_1 \\ u_2 \\ u_3 \end{bmatrix} = \begin{bmatrix} M_{0,1} \\ M_{1,1} \\ M_{2,1} \\ M_{3,1} \end{bmatrix}$$

Solve for a total of 19 multivariate moments for 3-D velocity

Sample Results

Uniform injection

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Point injection

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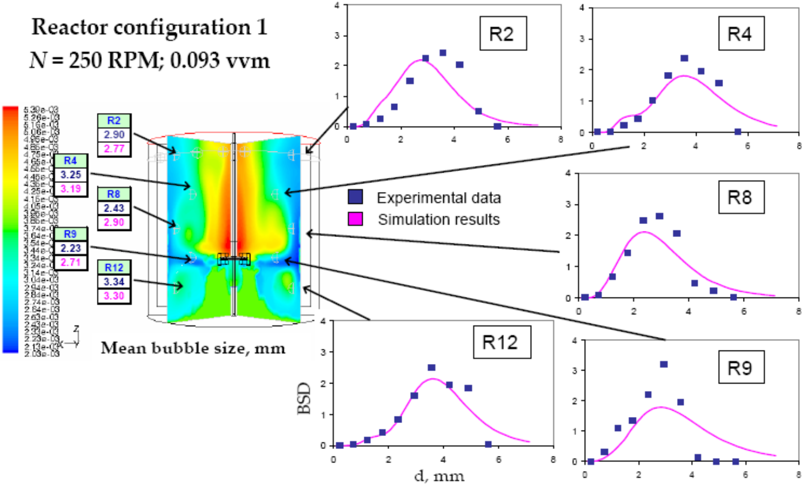
Application to Stirred Vessels



Summary of CFD Models with Low-Stokes Particles

- Generalized PBE includes the velocity of the disperse phase
- Monokinetic NDF approximation valid for small Stokes
- Quadrature-Based Moment Methods applied to reconstruct the NDF
- CFD solver modified to treat size-dependent velocity of disperse phase

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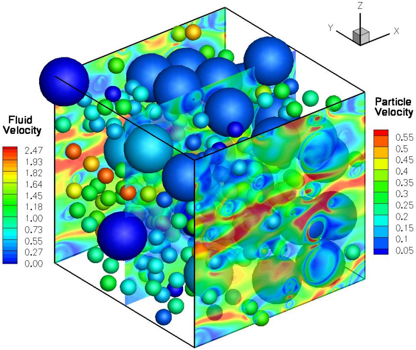
Gas-Solid Systems

- solid density $\gg$ gas density
- particle diameter $\gg 1\ \mu$m
- particle size distribution
- finite particle inertia (St $\gg$ 1)
- inelastic collisions



Kinetic Theory of Granular Flow coupled to gas-phase continuity and momentum balances

Particle Trajectory Crossing

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Dilute inertial particle jets with high Stokes number

CFD Model for Monodisperse Gas-Solid Flow

Particle-phase Kinetic Equation

$$\frac{\partial n}{\partial t} + \mathbf{v} \cdot \frac{\partial n}{\partial \mathbf{x}} + \frac{\partial}{\partial \mathbf{v}} \cdot (\mathbf{A} n) = \mathbb{C}$$

- $n(t, \mathbf{x}, \mathbf{v})$: velocity NDF
- $\mathbf{v}$: particle velocity
- $\mathbf{A}$: particle acceleration (drag, gravity, lift, ...)
- $\mathbb{C}$: rate of change of n due to particle-particle collisions

Fluid-phase equations

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

$$\frac{\partial}{\partial t}(\rho_g \alpha_g \mathbf{U}_g) + \nabla \cdot (\rho_g \alpha_g \mathbf{U}_g \mathbf{U}_g) = \nabla \cdot \alpha_g \boldsymbol{\tau}_g + \beta_g + \rho_g \alpha_g \mathbf{g}$$

- $\alpha_g = 1 - \alpha_p$: gas volume fraction
- β_g: mean particle drag

Equations coupled through moments of velocity NDF

Lagrangian vs. Eulerian Simulations

$$\frac{\partial n}{\partial t} + \mathbf{v} \cdot \frac{\partial n}{\partial \mathbf{x}} + \frac{\partial}{\partial \mathbf{v}} \cdot (\mathbf{A} n) = \mathbb{C}$$

Lagrangian method

For large ensemble, particle positions and velocities are tracked

$$\frac{\mathrm{d}\mathbf{x}^{(\alpha)}}{\mathrm{d}t} = \mathbf{v}^{(\alpha)}$$

$$\frac{\mathrm{d}\mathbf{v}^{(\alpha)}}{\mathrm{d}t} = \mathbf{A}^{(\alpha)} + \mathcal{C}^{(\alpha)}$$

Limited by statistical "noise" and coupling errors

Eulerian method

Velocity moments are tracked

$$M^0 = \alpha_\mathrm{p} = \int n \, \mathrm{d}\mathbf{v}$$

$$M_i^1 = \alpha_\mathrm{p} U_{\mathrm{p}i} = \int v_i \, n \, \mathrm{d}\mathbf{v}$$

$$\vdots$$

$$M_{i,j,\ldots}^n = \int (v_i v_j \cdots) \, n \, \mathrm{d}\mathbf{v}$$

Moments closed with QBMM

Complexity of Solutions

Collisions No collisions

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Multiphase turbulence generated by momentum coupling

KTGF Model for Collisional Gas-Solid Flow

Particle phase

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

$$\frac{\partial}{\partial t}(\rho_p \alpha_p \mathbf{U}_p) + \nabla \cdot \rho_p \alpha_p (\mathbf{U}_p \mathbf{U}_p + \boldsymbol{\tau}_p) = \rho_p \alpha_p \beta_p + \rho_p \alpha_p \mathbf{g}$$

$$\frac{\partial}{\partial t}(\rho_p \alpha_p \Theta_p) + \nabla \cdot \rho_p \alpha_p (\mathbf{U}_p \Theta_p + \mathbf{q}_p) = -\rho_p \alpha_p \boldsymbol{\tau}_p : \nabla \mathbf{U}_p - \gamma_\Theta$$

Fluid phase

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

$$\frac{\partial}{\partial t}(\rho_g \alpha_g \mathbf{U}_g) + \nabla \cdot \rho_g \alpha_g (\mathbf{U}_g \mathbf{U}_g + \boldsymbol{\tau}_g) = -\rho_p \alpha_p \beta_p + \rho_g \alpha_g \mathbf{g}$$

- $\alpha_p + \alpha_g = 1$
- $\beta_p = \frac{1}{\tau_D}(\mathbf{U}_g - \mathbf{U}_p)$
- τ_D: drag time scale
- γ_Θ: granular energy dissipation

Equivalent to Navier-Stokes Eq. $\Longrightarrow$ Need a turbulence model!

Multiphase Turbulence Model

Phase-average quantities: $\langle A \rangle_\mathrm{p} = \frac{\langle \alpha_\mathrm{p} A \rangle}{\langle \alpha_\mathrm{p} \rangle}$, $\langle A \rangle_\mathrm{g} = \frac{\langle \alpha_\mathrm{g} A \rangle}{\langle \alpha_\mathrm{g} \rangle}$

- Particle-phase mean variables: $\langle \alpha_\mathrm{p} \rangle$, $\langle \mathbf{U}_\mathrm{p} \rangle_\mathrm{p}$, $\langle \Theta_\mathrm{p} \rangle_\mathrm{p}$
- Gas-phase mean variables: $\langle \alpha_\mathrm{g} \rangle$, $\langle \mathbf{U}_\mathrm{g} \rangle_\mathrm{g}$
- Particle-phase turbulence variables: k_p, ε_p
- Gas-phase turbulence variables: k_g, ε_g

Derive turbulence model directly from KTGF model

Multiphase Turbulence Model: Gas Phase

Turbulent kinetic energy

$$\frac{\partial \rho_g \alpha_g k_g}{\partial t} + \nabla \cdot \rho_g \alpha_g k_g \mathbf{u}_g = \nabla \cdot \left(\mu_g + \frac{\mu_{gt}}{\sigma_{gk}} \right) \nabla k_g + \rho_g \alpha_g \Pi_g - \rho_g \alpha_g \varepsilon_g$$

$$+ \frac{2\rho_p \alpha_p}{\tau_D} (k_p^{1/2} - k_g^{1/2}) k_g^{1/2} + \rho_p \alpha_p \frac{C_g}{\tau_D} (\mathbf{u}_p - \mathbf{u}_g)^2$$

Phase coupling   TKE production

Turbulent dissipation

$$\frac{\partial \rho_g \alpha_g \varepsilon_g}{\partial t} + \nabla \cdot \rho_g \alpha_g \varepsilon_g \mathbf{u}_g = \nabla \cdot \left(\mu_g + \frac{\mu_{gt}}{\sigma_{g\varepsilon}} \right) \nabla \varepsilon_g + \frac{\varepsilon_g}{k_g} (C_1 \rho_g \alpha_g \Pi_g - C_2 \rho_g \alpha_g \varepsilon_g)$$

$$+ C_3 \frac{2\rho_p \alpha_p}{\tau_D} (\varepsilon_p^{1/2} - \varepsilon_g^{1/2}) \varepsilon_g^{1/2} + C_4 \frac{\varepsilon_p}{k_p} \rho_p \alpha_p \frac{C_g}{\tau_D} (\mathbf{u}_p - \mathbf{u}_g)^2$$

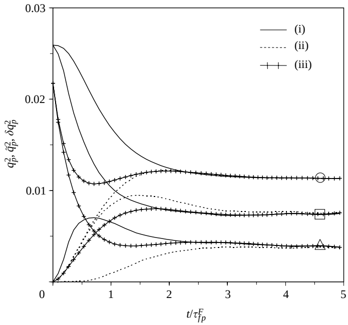
Phase coupling   TKE production

Each term has clear physical significance

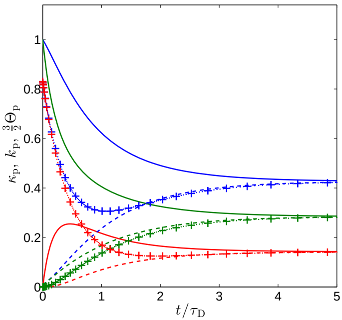
Validation of Phase Coupling with DNS

Dynamics of energy partition between total energy, TKE and $\langle \Theta_p \rangle$

DNS (Fevrier et al. 2005)



Turbulence model



Phase coupling closure has correct physics!

Summary of CFD Models for Fluid-Particle Systems

- Generalized PBE describes the disperse phase
- Particle trajectory crossing leads to full NDF for particle velocity
- Quadrature-Based Moment Methods used to reconstruct velocity NDF
- Momentum coupling leads to multiphase turbulence
- Rigorous turbulence model can be developed from KTGF model

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Final Remarks

- CFD modeling capabilities have grown enormously in last 20+ years
- Kinetic-based modeling approach uses mesoscale models
- Multiphase reacting systems lead to a Generalized PBE
- Quadrature-based moment methods lead to tractable CFD models
- Predictive multiphase turbulence models are still an open problem

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