CFD Models for Polydisperse Solids Based on the Direct Quadrature Method of Moments

Rodney O. Fox
H. L. Stiles Professor
Department of Chemical Engineering
Iowa State University

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Outline

1. Introduction
   - Population Balances
   - Coupling with CFD

2. Population Balances in CFD
   - Population Balance Equation
   - Direct Solvers
   - Quadrature Methods

3. Implementation for Gas-Solid Flow
   - Overview of MFIX
   - Polydisperse Solids Model
   - Application of DQMOM

4. Two Open Problems
Population Balances

- Number density function (NDF)
  
  \[ n(\nu, \alpha; x, t) \]

- Particle surface area
- Particle volume (mass)
- Spatial location
- Time

CFD provides a description of the dependence of \( n(\nu, \alpha) \) on \( x \)

For multiphase flows, the NDF will include the phase velocities (as in kinetic theory)
Population Balances

• Moments of number density function

\[ m_{kl}(x, t) = \int_0^\infty \int_0^\infty v^k a^l n(v, a; x, t) dv da \]

Choice of \( k \) and \( l \) depends on what can be measured

Solving for moments in CFD makes the problem tractable due to smaller number of scalars

Multi-fluid model solves for moments from kinetic theory
Population Balances

• Physical processes leading to size changes
  – Nucleation $J(x,t)$ produces new particles, coupled to local solubility, and properties of continuous phase
  – Growth $G(x,t)$ mass transfer to surface of existing particles, coupled to local properties of continuous phase
  – Restructuring particle surface/volume and fractal dimension changes due to shear and/or physio-chemical processes
  – Aggregation/Agglomeration particle-particle interactions, coupled to local shear rate, fluid/particle properties
  – Breakage system dependent, but usually coupled to local shear rate, fluid/particle properties

CFD provides a description of the local conditions
Population Balances

• What can we compare to in-situ experiments?

  Sub-micron particles ➔ small-angle static light scattering

  \[ I(0) = C_1 \frac{m_2}{m_1} \]
  zero-angle intensity

  \[ \langle R_g \rangle = C_2 \left( \frac{m_2(1+d_f)/d_f}{m_2} \right)^{1/2} \]
  radius of gyration \( 1.8 < d_f < 3 \)

  Larger particles ➔ optical methods

  \[ n(L), \ L = 2\sqrt{A/\pi} \]
  length

  \[ D_{pf} = 2 \ln(P)/\ln(A) \]
  projected fractal dimension

CFD model should predict measurable quantities accurately
Coupling with CFD

• Do particles follow the flow?

Stokes number

\[ St = \frac{\text{particle response time}}{\text{flow response time}} = \frac{\gamma \rho_p d_p^2}{12 \rho_f \nu_f} \]

Particle diameter

Kinematic viscosity

If \( St > 0.14 \), particle velocities must be found from a separate momentum equation in the CFD simulation.
Coupling with CFD

- Do PBE timescales overlap with flow timescales?

- Residence time: \( \tau = \frac{V}{q} \)

- Recirculation time: \( t_c \propto \frac{D_T}{(N_1 D_1)} \) or \( D_T/U_j \)

- Local mixing timescale: \( t_u = \frac{k}{\langle \epsilon \rangle} \)

- Kolmogorov timescale: \( t_\eta = (\nu/\langle \epsilon \rangle)^{1/2} \)

CFD simulations w/o PBE can be used to determine timescales for a particular piece of equipment.
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Population Balance Equation

• Typical NDF Transport Equation (small Stokes)

\[
\frac{\partial n}{\partial t} + \frac{\partial}{\partial x_i} (U_i n) = \text{Advection} \\
\frac{\partial}{\partial x_i} \left( D_T \frac{\partial n}{\partial x_i} \right) \text{ Diffusion} \\
+ J(v) - \frac{\partial}{\partial v} (G(v)n) \text{ Nucleation + Growth} \\
+ \frac{1}{2} \int_0^v \beta(v - s, s)n(v - s)n(s)ds - n(v) \int_0^\infty \beta(v, s)n(s)ds \\
+ \int_v^\infty b(v|s)a(s)n(s)ds - a(v)n(v) \text{ Aggregation Breakage}
\]
Population Balance Equation

- Aggregation Kernel

\[ \beta(v, s) = \frac{2K_BT}{3\mu W} \left( v^{1/d_f} + s^{1/d_f} \right) \left( v^{-1/d_f} + s^{-1/d_f} \right) \]

\[ + \gamma \alpha(v, s) v_p \left( v^{1/d_f} + s^{1/d_f} \right)^3 \]

Brownian

Shear-induced

Sub-micron aggregates: Brownian >> Shear-induced

Breakage and restructuring determine fractal dimension \( d_f \)

In granular flow, particle-particle collisions must be added
Population Balance Equation

- Breakage Kernels

\[ a(v) = c \gamma \exp \left( -\frac{B(\gamma)}{\gamma^2 R_p v^{1/d_f}} \right) \] exponential

\[ a(v) = c_1 \gamma^{c_2} \left( R_p v^{1/d_f} \right)^{c_3} \] power law

Breakage due to fluid shear only ==> additional term due to collisions in gas-solid flows

Parameters determined empirically and depend on chemical/physical properties of aggregates
Population Balance Equation

- Daughter Distribution

\[ b(v|s) = \delta (v - fs) + \delta (v - (1 - f)s) \quad \text{binary} \]

Equal sized: \( f = \frac{1}{2} \)
Erosion: \( f << 1 \)

- Number density (#/m^3)

- \( \frac{m(i)}{m(1)} \)

\[ \begin{align*}
\text{Number density} & = \begin{cases} 10^0, & f=0.5 \\
10^5, & f=0.1 \\
10^7, & f=0.01 \\
10^9, & f=0.001 
\end{cases} 
\end{align*} \]
Direct Solvers

- Sectional or Class Methods

Accurate predictions for higher-order moments require finer grid (range: 25-120 bins)
Direct Solvers

- Difficulties encountered when coupled with CFD
  - \( n(\nu; x, t) \) represented by \( N \) scalars \( n_i(x,t) \) where \( 25 < N < 120 \)
  - Depending on kernels, initial conditions, etc., source terms for these scalars can be stiff
  - If particles are large (measured by Stokes number), multiphase models with \( N \) momentum equations required
  - Extension to multi-variate distributions scales like \( N^D \) — accounting for “morphology” changes will be intractable

Need methods that accurately predict experimentally observable moments, but at low computational cost
Quadrature Methods

• Quadrature Method of Moments (QMOM)

\[ n(v; x, t) \approx \sum_{n=1}^{N} w_n \delta(v - v_n) \]

weights \hspace{1cm} abscissas

\( k^{th} \) moment of CSD:

\[ m_k = \sum_{n=1}^{N} w_n v_n^k \]
Quadrature Methods

- Product-Difference algorithm (univariate CSD)

\[ \{m_0, m_1, m_2, m_3, m_4, m_5, m_6, m_7\} \]

\[ \{w_1, w_2, w_3, w_4, v_1, v_2, v_3, v_4\} \]

Inverse problem solved on the fly in CFD simulation
Quadrature Methods

- Transport $2N$ moments in CFD simulation

\[
\frac{\partial m_k}{\partial t} + \frac{\partial}{\partial x_i} (U_i m_k) = \text{Advection}
\]

\[
\frac{\partial}{\partial x_i} \left( D_T \frac{\partial m_k}{\partial x_i} \right) \quad \text{Diffusion}
\]

\[+ J_k + \sum_i kv_i^{k-1} G_i w_i \quad \text{Nucleation + Growth}
\]

\[+ \frac{1}{2} \sum_i \sum_j \left[ (v_i + v_j)^k - v_i^k - v_j^k \right] \beta_{ij} w_i w_j \quad \text{Aggregation}
\]

\[+ \sum_i a_i \left[ b_i^{(k)} - v_i^k \right] w_i \quad \text{Breakage}
\]
Quadrature Methods

- Comparison with direct method

Using $2N = 8$ scalars, QMOM reproduces the grid-independent moments of the direct method
Quadrature Methods

- Multi-variate extension is straightforward

\[
n(v, a; x, t) \approx \sum_{n=1}^{N} w_n \delta(v - v_n) \delta(a - a_n)
\]

\((k,l)\text{th} \text{ moment of CSD:}\)

\[
m_{kl} = \sum_{n=1}^{N} w_n v_n^k a_n^l
\]

But inverse problem cannot be solved on the fly!
Quadrature Methods

- Direct Quadrature Method of Moments (DQMOM)

\[
\frac{\partial w_n}{\partial t} + \frac{\partial}{\partial x_i} (U_i w_n) = \frac{\partial}{\partial x_i} \left( D_T \frac{\partial w_n}{\partial x_i} \right) + \alpha_n \quad \text{Weights}
\]

\[
\frac{\partial w_n v_n}{\partial t} + \frac{\partial}{\partial x_i} (U_i w_n v_n) = \frac{\partial}{\partial x_i} \left( D_T \frac{\partial w_n v_n}{\partial x_i} \right) + \alpha_{1n} \quad \text{Volume}
\]

\[
\frac{\partial w_n a_n}{\partial t} + \frac{\partial}{\partial x_i} (U_i w_n a_n) = \frac{\partial}{\partial x_i} \left( D_T \frac{\partial w_n a_n}{\partial x_i} \right) + \alpha_{2n} \quad \text{Area}
\]

Source terms found from linear system on the fly

\[
\sum_{n=1}^{N} (1-k) \phi_n^k \alpha_n + \sum_{n=1}^{N} k \phi_n^{k-1} (c_v \alpha_{1n} + c_a \alpha_{2n}) = R_k
\]

\[
\phi_n = c_v v_n + c_a a_n
\]
Polydisperse Gas-Solid Flow

- DQMOM with size and momentum of solid phase

\[
\begin{align*}
\frac{\partial w_\alpha}{\partial t} + \nabla \cdot (U_\alpha w_\alpha) &= a_\alpha \\
\frac{\partial \rho w_\alpha v_\alpha}{\partial t} + \nabla \cdot (U_\alpha \rho w_\alpha v_\alpha) &= \rho b_\alpha \\
\frac{\partial \rho w_\alpha v_\alpha U_\alpha}{\partial t} + \nabla \cdot (\rho w_\alpha v_\alpha U_\alpha U_\alpha) &= \rho c_\alpha
\end{align*}
\]

Source terms for mass and momentum can be found from kinetic theory for gas-solid flows

Reduces to two-fluid model when \( \alpha = 1 \)
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Overview of MFIX

Gas-solid multi-fluid model

- Momentum equations
- Mass & energy equations
- Chemical species equations
- Population balance equations

- Kinetic theory
- Mass & heat transfer models
- Detailed chemistry
- Aggregation, breakage and growth

ISAT
DQMOM
MFI X Governing Equations (1)

- **Mass balances**
  \[
  \frac{\partial}{\partial t} (\varepsilon_g \rho_g) + \nabla \cdot (\varepsilon_g \rho_g \mathbf{u}_g) = - \sum_{\alpha=1}^{N} \sum_{n=1}^{N_s} M_{gan} \\
  \frac{\partial}{\partial t} (\varepsilon_{s\alpha} \rho_{s\alpha}) + \nabla \cdot (\varepsilon_{s\alpha} \rho_{s\alpha} \mathbf{u}_{s\alpha}) = \sum_{n=1}^{N_s} M_{gan}
  \]

- **Momentum balances**
  \[
  \frac{\partial}{\partial t} (\varepsilon_g \rho_g \mathbf{u}_g) + \nabla \cdot (\varepsilon_g \rho_g \mathbf{u}_g \mathbf{u}_g) = \nabla \cdot \mathbf{\sigma}_g + \sum_{\alpha=1}^{N} f_{g\alpha} + \varepsilon_g \rho_g \mathbf{g} \\
  \frac{\partial}{\partial t} (\varepsilon_{s\alpha} \rho_{s\alpha} \mathbf{u}_{s\alpha}) + \nabla \cdot (\varepsilon_{s\alpha} \rho_{s\alpha} \mathbf{u}_{s\alpha} \mathbf{u}_{s\alpha}) = \nabla \cdot \mathbf{\sigma}_{s\alpha} - f_{g\alpha} + \sum_{\beta=1, \beta \neq \alpha}^{N} f_{\beta\alpha} + \varepsilon_{s\alpha} \rho_{s\alpha} \mathbf{g}
  \]

\( g \): Gas phase  \\
\( s\alpha \): Solid phases \( \alpha=1, N \)  \\
\( \mathbf{\sigma}_g \): Stress tensor  \\
\( f_{g\alpha} \): Body force  \\
\( f_{\beta\alpha} \): Interaction with gas and other solid phases
MFIX Governing Equations (II)

- Thermal energy balances

\[
\varepsilon_g \rho_g C_{pg} \left( \frac{\partial T_g}{\partial t} + u_g \cdot \nabla T_g \right) = -\nabla \cdot q_g - \sum_{\alpha=1}^{N} H_{g\alpha} - \Delta H_{rg} + H_{wall}(T_{wall} - T_g)
\]

\[
\varepsilon_{s\alpha} \rho_{s\alpha} C_{psa} \left( \frac{\partial T_{s\alpha}}{\partial t} + u_{s\alpha} \cdot \nabla T_{s\alpha} \right) = -\nabla \cdot q_{s\alpha} + H_{g\alpha} - \Delta H_{rsa}
\]

- Chemical species balances

\[
\frac{\partial}{\partial t} (\varepsilon_g \rho_g X_{gn}) + \nabla \cdot (\varepsilon_g \rho_g X_{gn} u_g) = R_{gn} - \sum_{\alpha=1}^{N} M_{gan}
\]

\[
\frac{\partial}{\partial t} (\varepsilon_{s\alpha} \rho_{s\alpha} X_{san}) + \nabla \cdot (\varepsilon_{s\alpha} \rho_{s\alpha} X_{san} u_{s\alpha}) = R_{san} + M_{gan}
\]

**g**: Gas phase  
**s\alpha**: Solid phases \( \alpha = 1, N \)

Heat lost to walls  
Conductive heat flux  
Heat transfer between phases  
Heat of reaction  
Reactions  
Mass transfer
Polydisperse Solids Model

- Population balance equation for solid phase

\[ \frac{\partial n(L, u_s; x, t)}{\partial t} + \nabla \cdot \left[ u_s n(L, u_s; x, t) \right] + \nabla_{u_s} \cdot \left[ F_n(L, u_s; x, t) \right] = S(L, u_s; x, t) \]

Force acting to accelerate particles

Joint size & velocity distribution function

Aggregation, breakage and chemical reaction
Direct Quadrature Method of Moments

\[ \frac{\partial n(L; u_s; x, t)}{\partial t} + \nabla \cdot \left[ u_s n(L, u_s; x, t) \right] + \nabla u_s \cdot \left[ F n(L, u_s; x, t) \right] = S(L, u_s; x, t) \]

Integrate out solid velocity

\[ \frac{\partial n(L; x, t)}{\partial t} + \nabla \cdot \left[ (u_s | L) n(L; x, t) \right] = S(L; x, t) \]

Use distribution function

\[ \frac{\partial}{\partial t} \left[ \sum_{\alpha=1}^{N} \omega_\alpha \delta(L - L_\alpha) \right] + \nabla \cdot \left[ u_{s\alpha} \sum_{\alpha=1}^{N} \omega_\alpha \delta(L - L_\alpha) \right] = S(L; x, t) \]

Simplify equation

\[ \sum_{\alpha=1}^{N} \left[ \delta(L - L_\alpha) a_\alpha - \delta'(L - L_\alpha)(b_\alpha - L_\alpha a_\alpha) \right] = S(L; x, t) \]

Moment transform

\[ \sum_{\alpha=1}^{N} \left[ a_\alpha L_\alpha^k + k L_\alpha^{k-1} (b_\alpha - L_\alpha a_\alpha) \right] = S_k(x, t) \]

Ax = b
Modifications to MFI X

• Relation between volume fractions and weights:

$$\varepsilon_{sa} = k_v L_{\alpha}^3 \omega_{\alpha}$$

$k_v$: volumetric shape factor

• Transport equations for volume fractions and lengths:

$$\frac{\partial(\varepsilon_{sa} \rho_{sa})}{\partial t} + \nabla \cdot (\varepsilon_{sa} \rho_{sa} \mathbf{u}_{sa}) = 3k_v \rho_{sa} L_{\alpha}^2 b_{\alpha} - 2k_v \rho_{sa} L_{\alpha}^3 a_{\alpha}$$

$$\frac{\partial(\varepsilon_{sa} L_{\alpha} \rho_{sa})}{\partial t} + \nabla \cdot (\varepsilon_{sa} L_{\alpha} \rho_{sa} \mathbf{u}_{sa}) = 4k_v \rho_{sa} L_{\alpha}^3 b_{\alpha} - 3k_v \rho_{sa} L_{\alpha}^4 a_{\alpha}$$
DQMOM Source Terms

Matrix $A$ relates moments to weights and lengths

Source term $x$ is obtained by forcing moments to be exact
Aggregation and Breakage

Birth due to aggregation $\dot{b}_k(x,t)$

Death due to aggregation $\dot{d}_k(x,t)$

Death due to breakage $\dot{d}_k(x,t)$

Birth due to breakage $\dot{b}_k(x,t)$

$$\overline{S}_k(x,t) = \overline{B}_k^a(x,t) - \overline{D}_k^a(x,t) + \overline{B}_k^b(x,t) - \overline{D}_k^b(x,t)$$

$$\overline{B}_k^a(x,t) = \frac{1}{2} \int_0^{+\infty} n(\lambda;x,t) \int_0^{+\infty} \beta(u,\lambda)(u^3 + \lambda^3)^{k/3} n(u;x,t) du d\lambda$$

$$\overline{D}_k^a(x,t) = \int_0^{+\infty} L n(L;x,t) \int_0^{+\infty} \beta(L,\lambda)n(\lambda;x,t)d\lambda dL$$

$$\overline{B}_k^b(x,t) = \int_0^{+\infty} L \int_0^{+\infty} a(\lambda)b(L|\lambda)n(\lambda;x,t)d\lambda dL$$

$$\overline{D}_k^b(x,t) = \int_0^{+\infty} L a(L)n(L;x,t) dL$$

Apply DQMOM

$$\overline{S}_k(x,t) = \frac{1}{2} \sum_{i=1}^{N} \sum_{j=1}^{N} \omega_i \omega_j (L_i^3 + L_j^3)^{k/3} \beta_{ij} - \sum_{i=1}^{N} \sum_{j=1}^{N} \omega_i \omega_j L_i^k \beta_{ij} + \sum_{i=1}^{N} \omega_i a_i^{(i)} - \sum_{i=1}^{N} \omega_i L_i^k a_i$$
Aggregation and Breakage Kernels

• Aggregation and breakage kernels are obtained from kinetic theory

Number of collisions:

\[ N_{ij} = \pi \omega_i \omega_j \sigma_{ij}^3 g_{ij} \left[ \frac{4}{\sigma_{ij}} \left( \frac{\theta_s}{\pi} \frac{m_i + m_j}{2m_i m_j} \right)^{\frac{1}{2}} - \frac{2}{3} (\nabla \cdot \mathbf{u}_s) \right] \]

Aggregation kernel:

\[ \beta_{ij} = \frac{N_{ij}}{\omega_i \omega_j} \psi_a \]

Breakage kernel:

\[ a_i = \sum \frac{N_{ij}}{\omega_i} \psi_b \]

Efficiencies (\( \psi_a \) and \( \psi_b \)) depend on temperature, particle size, etc.
PSD Effect on Fluidization

- No aggregation and breakage
- Breakage dominant, average size decreases, FB expands
- Aggregation dominant, average size increases, FB defluidizes
Volume-Average Mean Diameter

Case 1
\[ \beta = 0; \ a = 0 \]

Case 2
\[ \beta = 1 \times 10^{-5} \text{ m}^3 / \text{s}; \ a = 0.1 \text{ s}^{-1} \]

Case 3
\[ \beta = 1 \times 10^{-5} \text{ m}^3 / \text{s}; \ a = 1 \text{ s}^{-1} \]

Symbols:
- 2 filled symbols
- 3 empty symbols
- 4 lines
Volume-Average Normalized Moments

\[ m_k(x,t) = \int_0^{+\infty} n(L;x,t) L^k dL \approx \sum_{\alpha=1}^{N} \omega_{\alpha} L_{\alpha}^k \]

\( N = 2 \) filled symbols
\( N = 3 \) empty symbols
\( N = 4 \) lines
Extension to Energy/Species Balances

- Thermal energy balance

\[ ε_{sα} \rho_{sα} C_{psα} \left( \frac{∂T_{sα}}{∂t} + u_{sα} \cdot \nabla T_{sα} \right) = -∇ \cdot q_{sα} + H_{gα} - ΔH_{rsa} \]

\[ + k_v ρ_s L^3 C_{ps} c_{T,α} - k_v ρ_s L^3 C_{ps} T_{sα} a_α \]

Changes due to aggregation and breakage

**Multi-variate DQMOM**
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Two Open Problems

1. How to extend DQMOM to systems with unknown fluxes at boundaries in phase space?

Model problem: pure evaporation

\[ \frac{\partial n}{\partial t} = \frac{\partial n}{\partial v} \quad \Rightarrow \quad \frac{dm_k}{dt} = -\delta_{k,0} n(0, t) - km_{k-1} \]

How can we estimate it?

Estimate flux in DQMOM variables, test with exact solutions:

Define vectors:
\[ x_{\alpha} = \frac{w_{\alpha} v_{\alpha}}{m_{0}} \]
\[ \dot{x} = \frac{dx}{dt} \]

Define "cross product":
\[ c = \dot{x} \times x \]

Linear constraint:
\[ \sum c_{\alpha} = 0 \]
Simple case with monotone flux ($N = 2$):
Harder case with multimode flux ($N = 2$):
Harder case with $N = 3$: 

- **Flux**: Comparison between exact flux and flux with DQMOM.
- **Moments**: Plot of moments for different cases with and without DQMOM.
- **Abscissas & Weights**: Graphs showing the evolution of abscissas and weights over time for different cases.
Two Open Problems

2. What is “best” choice of moments for multivariate DQMOM?

Model problem: homogeneous aggregation

\[ \frac{dw_n}{dt} = a_n \]

\[ \frac{dw_n v_n}{dt} = b_n \]

\[ \frac{dw_n a_n}{dt} = c_n \]

Moments (mass \( k=1, l=0 \); area \( k=0, l=1 \) )
• Choice of moments affects the condition of matrix

All choices yield nearly same weights and abscissas. Choose moments with lowest condition number?
Another example: Williams’ Spray Equation

\[ \partial_t f + \mathbf{u} \cdot \partial_x f + \partial_v (R_v f) + \partial_u \cdot (F f) = \Gamma \]

\[ f(v, u; x, t) = \text{volume, velocity number density function} \]

\[ R_v = \text{evaporation rate} \]

\[ F = \text{drag force} \]

\[ \Gamma = Q^- + Q^+ = \text{coalescence operator} \]

\[ Q^- = - \int \int B(|u - u^*|, v, v^*) f(v, u) f(v^*, u^*) \, dv^* \, du^* \]

\[ Q^+ = \frac{1}{2} \int \int B(|u^\diamond - u^*|, v^\diamond, v^*) f(v^\diamond, u^\diamond) f(v^*, u^*) J \, dv^* \, du^* \]
Coefficients depend on choice of $5N$ moments:

$$\langle v^k u_1^l u_2^m u_3^p \rangle$$

Condition number of $A$ depends on choice of $k, l, m, p$

In general, $A$ matrix will become singular if $1 < l + m + p$

Choose $l, m, p = (0,1)$ and vary $k$ to yield $5N$ distinct moments

Number: $(k, l, m, p) = 0$

Mass: $k = 1, (l, m, p) = 0$

X-Mom: $k = 1, l = 1$  Y-Mom: $k = 1, m = 1$  Z-Mom: $k = 1, p = 1$

Is there a general method for choosing moments?