Dimension reduction of two-dimensional population balances based on the quadrature method of moments

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Motivation

- Crystal production for chemical and pharmaceutical products
- Quality criteria - Control of crystal size distribution and crystal shape distribution
- Process design and process control via modeling and simulation
- Employment of population balance equations (PBE) for crystallization modeling
- Shape-dependent crystal growth leads to multi-dimensional PBE
- Solution of multi-dimensional PBE → complex and numerically advanced problem
- Model reduction via moment methods → application of quadrature method of moments (QMOM)
- Loss of information due to QMOM application → volume-dependent information usually not available

Aim of the Research

- Application of advanced QMOM method
- Simplification of two-dimensional PBE to a → one-dimensional advection system
- Restoration of volume-dependent information
- Calculation of crystal volume distribution and volume-dependent properties → now possible

Experimental data on shape-dependent growth of crystals

- Bulk precipitation [1] or emulsion-based precipitation [2,3] of → Barium sulfate crystals

$$BaCl_2 + K_2SO_4 \longrightarrow BaSO_4 \downarrow + 2KCl$$

- crystal shape observation via microscopy (i.e. TEM, REM)

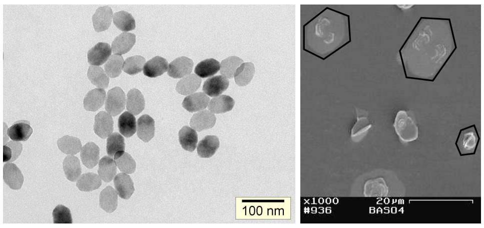

Figure 1: Left: TEM picture of Barium Sulfate crystals from an emulsion-based precipitation process; Right: REM picture of Barium Sulfate crystals from a bulk-phase precipitation.

- Assumption of self-similar hexagonal shape
- Experimental shape observation
  - variable width L_1
  - corresponding $L_3 = 2L_1$ and $L_4 = 1.5L_1$
  - variable thickness L_2

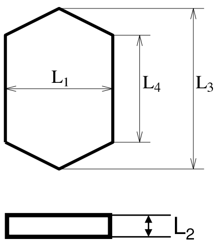

Figure 2: Idealized Barium Sulfate crystal with hexagonal shape.

- different growth rates for width and thickness
  - Growth rate for L_1: $G_1(c) = dL_1/dt = G(c)$
  - Experimental observation of plate thickness L_2
  - Growth rate for L_2: $G_2(L_2, c) = dL_2/dt = (1 - L_2/L_{2,max})G(c)$ with $L_{2,max} = 3\mu m$ (from experiments)
- Baldyga Ansatz [4] for $G(c)$: $G(c) = k_D(c - c_{sat} + c_* - \sqrt{c_*^2 + 2c_*(c - c_{sat})}; c \geq c_{sat}$
- Total volume of a single crystal: $V_{cr}(L_1, L_2) = 3.5L_1^2 L_2/2$
- Total volume of all crystals by integration: $V_{cr,tot}$
- Coupling with concentration balance for Barium Sulfate

Model Equations

- Population balance equation for a shape-dependent crystal size distribution:

$$\frac{\partial f(t,L_1,L_2)}{\partial t} + \underbrace{G_1(c)\frac{\partial f(t,L_1,L_2)}{\partial L_1}}_{\text{Growth term for } L_1} + \underbrace{\frac{\partial G_2(L_2,c) f(t,L_1,L_2)}{\partial L_2}}_{\text{Growth term for } L_2} = 0$$

Initial size distribution:

$$f(0,L_1,L_2) = ad(L_1,L_2)e^{-b(L_1-L_2^*)^2+(L_2-L_2^*)^2}$$

- a, b, L_1^*, L_2^* based on experimental data
- Coupling with balance equation for oversaturated solution: $c = c_0 - V_{cr,tot}(t) - V_{cr,tot}(t=0))\sigma_{mol}/V_{reac}$
- Transformation into volume-based distribution with crystal volume v and the hexagonal base area $\varkappa^2$ $F(t, v, \varkappa) = (2/\alpha)^{1/2}\varkappa^{-2} f(t, L_1, L_2)$
- Derivation of the moment equations (i=0,2,4,6,8,...): $\frac{dM_i}{dt} + \sqrt{2\alpha}vG(c)\frac{dM_{i-1}}{dv} - (vG(c)/L_{2,max})\frac{dM_i}{dv} + G(c)\frac{dM_{i+2}}{dv} + (2-i)\sqrt{\alpha/2}G(c)M_{i-1} - (G(c)/L_{2,max})M_i = 0$
- More moments than equations! → Solve a closure problem!

Closure of moment system

- Replacement of $M_i(t, v)$ by sum of Gaussian quadrature with abscissae K_j and weights W_j: $M_i(t,v) = \sum_{j=1}^{n} K_j(t,v)^i W_j(t,v)$
- New PDE system for K_j and W_j → Solution by closure!
- Matrix regularisation → decoupled subsystem of n independent equations
- Coefficients can be solved using all initial and boundary conditions for M_i
  Numerics with Gordon PD-algorithm
- Solution of advection system of equations for K_j and W_j with numerics:
  → second-order upwind scheme for advection term
  → second-order Runge-Kutta for source term

Numerical Results

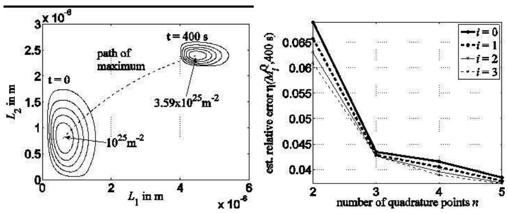

Figure 3: Left: Contour plot of the crystal size distribution with a dynamical path from t=0 to t=400s; Right: Estimated relative error of the moments M_i at t=400s.

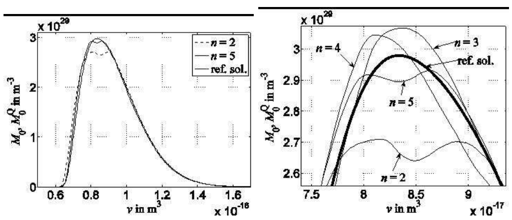

Figure 4: Left: Contour plot of the crystal size distribution with a dynamical path from t=0 to t=400s; Right: Estimated relative error of the moments M_i at t=400s.

Conclusion

- QMOM-based dimension reduction for two-dimensional PBE with → direction-dependent growth
- Numerical solution of advection system of equations
- Advantage - Calculation of volume-dependent properties
- Approach for shape-dependent crystallization modeling
- Extension of additional phenomena like nucleation possible

Literature

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