Numerical Solutions of Population-Balance Models in Particulate Systems

Shamsul Qamar Gerald Warnecke

Institute for Analysis and Numerics
Otto-von-Guericke University Magdeburg, Germany

In collaboration with Max-Planck Institute for Dynamical Systems, Magdeburg

Harrachov, August 19–25, 2007

Outline

Outline

Motivation

Aim

To model and simulate nucleation, growth, aggregation and Breakage phenomena in processes engineering by solving population balance equations (PBEs).

Numerical Methods

To solve population balance models we use the high resolution finite volume schemes as well as their combination with the method of characteristics

Outline

Industrial Applications

Applications

- Pharmaceutical
- Chemical industries
- Biomedical science
- Aerosol formation
- Atmospheric physics
- Food industries

Outline

General Population Balance Equation (PBE)

$$\frac{\partial f(t,x)}{\partial t} + \frac{\partial [G(t,x)f(t,x)]}{\partial x} = \mathcal{Q}^{\pm}_{\text{agg}}(t,x) + \mathcal{Q}^{\pm}_{\text{break}}(t,x) + \mathcal{Q}^{+}_{\text{nuc}}(t,x)$$
$$f(0,x) = f_0\,, \quad x \in \mathbb{R}_+ :=]0, +\infty[, t \geq 0$$

1. $f(t,x)$ is the number density function,
2. t denotes the time and x is an internal coordinate
3. $G(t,x)$ is the growth/dissolution rate along x,
4. $\mathcal{Q}^{\pm}_{\alpha}(t,x)$ are the aggregation, breakage and nucleation terms for $\alpha = \{\text{agg}, \text{break}, \text{nuc}\}$.
5. The entities in the population density can be crystals, droplets, molecules, cells, and so on.

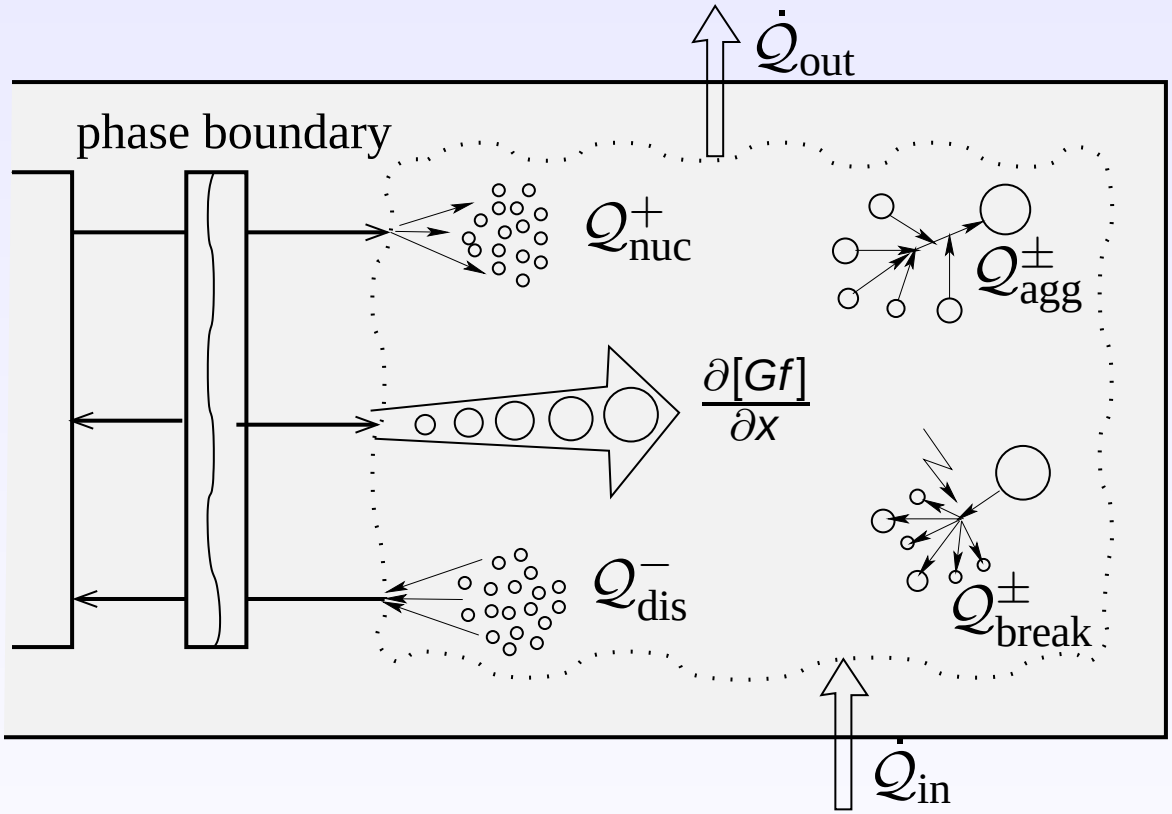


Figure: A schematic representation of different particulate processes

$$\mathcal{Q}^{\pm}_{\text{agg}}(t,x) = \frac{1}{2}\int_0^x \beta(t,x',x-x')\,f(t,x')\,f(t,x'-x)dx' - \int_0^\infty \beta(t,x,x')\,f(t,x)\,f(t,x')dx' .$$

Where: $\beta = \beta(t,x,x')$ is the rate at which the aggregation of two particles with respective volumes x and x' produces a particle of volume $x + x'$ and is a nonnegative symmetric function,

$$0 \leq \beta(t,x,x') = \beta(t,x',x), \quad x' \in]0,x[, \quad (x,x') \in \mathbb{R}^2_+ .$$

$$\mathcal{Q}^{\pm}_{\text{break}}(t,x) = \int_x^{\infty} b(t,x,x')\, S(x')\, f(t,x')dx' - S(x)\, f(t,x)\,.$$

$b := b(t,x,x')$ is the probability density function for the formation of particles of size x from particle of size x'. The selection function $S(x')$ describes the rate at which particles are selected to break.

Moments: $$\mu_i(t) = \int_0^{\infty} x^i f(t,x)dx\,, \quad i = 0,1,2,\cdots,$$

$\mu_0(T)$ =total number of particles, $\mu_1(t)$ =total volume of particles

Outline

Reformulation of PBE

Multiply the original PBE with x and re-arrange the terms, we get

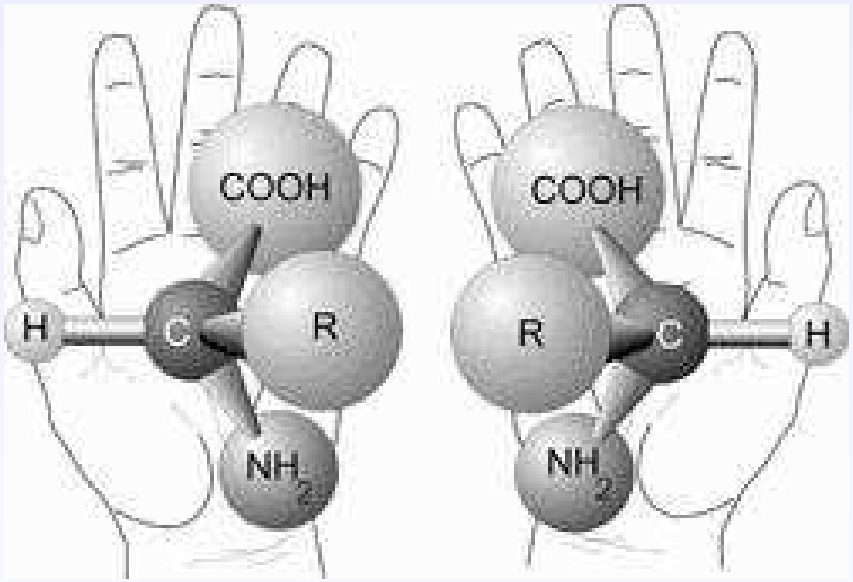
$$\frac{\partial \tilde{f}(t,x)}{\partial t} + \frac{\partial[(G\tilde{f})(t,x)]}{\partial x} - \frac{(G\tilde{f})(t,x)}{x} = -\frac{\partial \mathcal{F}_{\text{agg}}(t,x)}{\partial x} + \frac{\partial \mathcal{F}_{\text{break}}(t,x)}{\partial x} + \tilde{\mathcal{Q}}_{\text{nuc}}\,,$$
$$\tilde{f}(0,x) = \tilde{f}_0\,, \qquad x \in \mathbb{R}_+\,,\ t \geq 0\,,$$

where $\tilde{f}(t,x) := xf(t,x)$, $\tilde{\mathcal{Q}}_{\text{nuc}} = x\,\mathcal{Q}^{+}_{\text{nuc}}$ and

$$\mathcal{F}_{\text{agg}}(t,x) = -\int_0^x \int_{x-u}^{\infty} u\,\beta(t,u,v)\,f(t,u)\,f(t,v)\,dvdu \quad \textit{(Filbet \& Laurencot, 2004)}$$

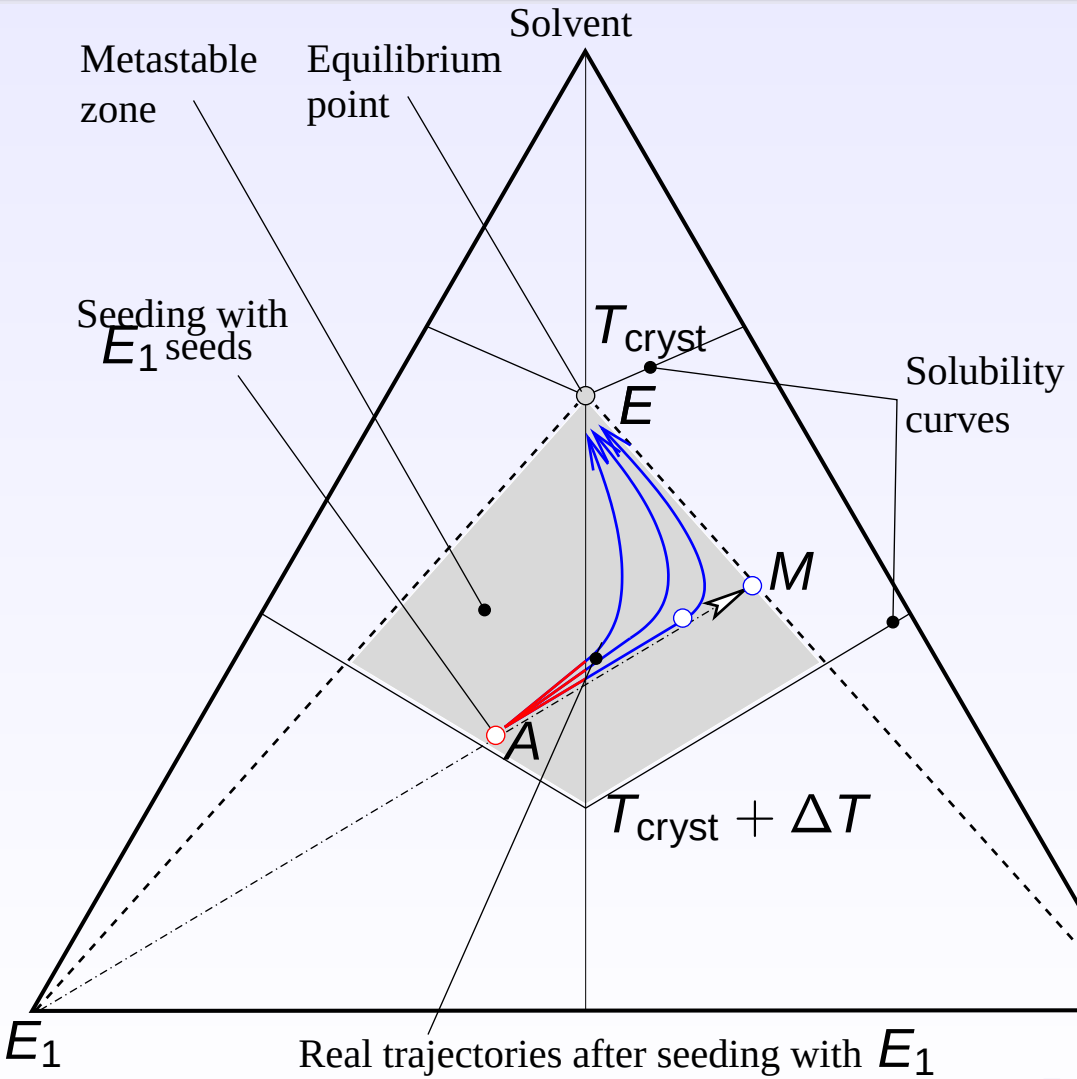
$$\mathcal{F}_{\text{break}}(t,x) = \int_0^x \int_x^{\infty} u\,b(t,u,v)\,S(v)\,f(t,v)\,dvdu\,.$$

Outline

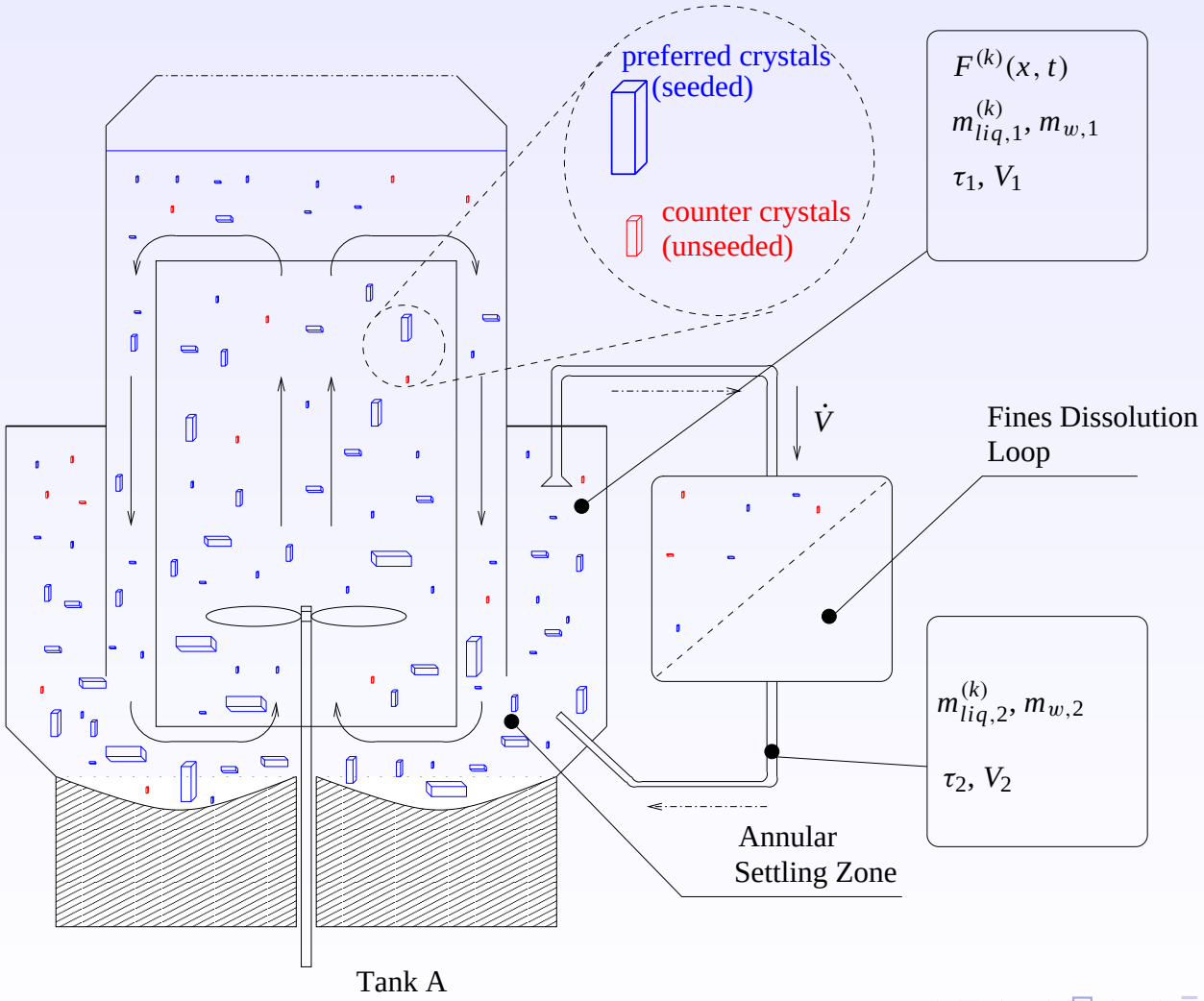


Amino acid enantiomers

Ternary Phase Diagram



Preferential Batch Crystallizer With Fines Dissolution



Model for Preferential Crystallization

Balance for solid phase

$$\frac{\partial f^{(k)}(t,x)}{\partial t} = -G^{(k)}(t)\frac{\partial f^{(k)}(t,x)}{\partial x} - \frac{1}{\tau_1}h(x)f^{(k)}(t,x)\ , \quad k \in [p,c].$$

Mass balance for liquid phase in cyrstallizer

$$\frac{dm^{(k)}(t)}{dt} = \dot{m}_{in}^{(k)}(t) - \dot{m}_{out}^{(k)}(t) - 3\rho k_v G^{(k)}(t)\int_0^\infty x^2 f^{(k)}(t,x)dx.$$

$$f^{(k)}(t,0) = \frac{B^{(k)}(t)}{G^{(k)}(t)}, \qquad w^{(k)}(t) = \frac{m^{(k)}(t)}{m^{(p)}(t) + m^{(c)}(t) + m_W(t)}$$

$$S^{(k)}(t) = \frac{w^{(k)}(t)}{w_{eq}^{(k)}} - 1, \quad G^{(k)}(t) = k_g\,[S^{(k)}(t)]^\alpha,\ \ k_g \geq 0,\ \alpha \geq 1\,.$$

$$B_0^{(p)}(t) = k_b^{(p)} \left(S^{(p)}(t) \right)^{b^{(p)}} \mu_3^{(p)}(t)$$

$$B_0^{(c)}(t) = k_b^{(c)} e^{-\frac{b^{(c)}}{ln\left(S^{(c)}(t)\right)^2}}$$

$$\dot{m}_{out}^{(k)}(t) = w^{(k)}(t)\, \rho_{liq}(T)$$

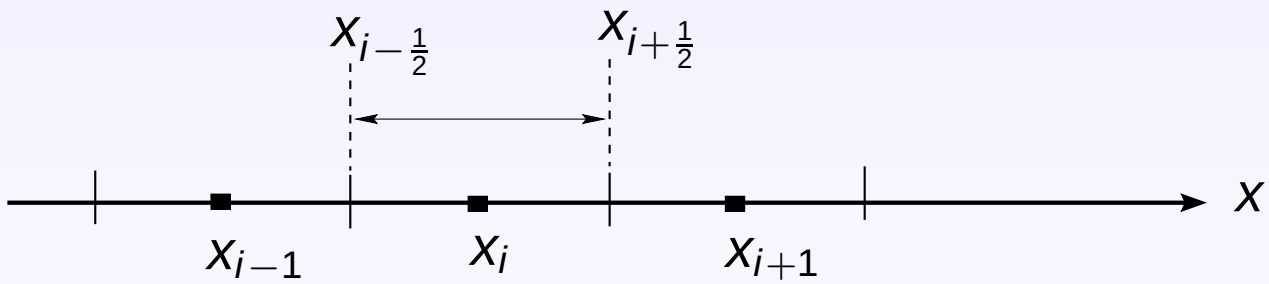
$$\dot{m}_{in}^{(k)}(t) = \dot{m}_{out}^{(k)}(t - \tau_2) + \frac{k_v \rho}{\tau_1} \int_0^\infty x^3 h(x) f^{(k)}(t - \tau_2, x)\, dx\ .$$

Outline

Domain Discretization

Regular/Irregular grid: Let N be a large integer and denote by $(x_{i-\frac{1}{2}})_{i\in\{1,\cdots,N+1\}}$ a mesh of $[x_{\min}, x_{\max}]$. We set

$$x_{1/2} = x_{\min}\,, \quad x_{N+1/2} = x_{\max}\,, \quad x_{i+1/2} = x_{\min} + i\cdot\Delta x_i\,, \quad \forall\, i = 1, 2, \cdots N-1\,.$$



Here $x_i = (x_{i-1/2} + x_{i+1/2})/2\,, \quad \Delta x_i = x_{i+1/2} - x_{i-1/2}\,.$

Geometric grid:

$$x_{1/2} = x_{\min}\,, \quad x_{i+1/2} = x_{\min} + 2^{(i-N)/q}(x_{\max} - x_{\min})\,, \quad \forall\ i = 1, 2, \cdots, N$$

where the parameter q is any positive integer.

Let $\Omega_i = \left[x_{i-1/2}, x_{i+1/2}\right]$ for $i \geq 0$. We approximate the initial data $f_0(x)$ in each grid cell by

$$f_i = \frac{1}{\Delta x_i} \int_{\Omega_i} f_0(x)dx\,.$$

Outline

Method 1: Combination of MOC and FVS

Let us substitute the growth rate $G(t,x)$ by

$$\frac{dx}{dt} := \dot{x}(t) = G(t,x)\,.$$

Then we have to solve:

$$\frac{d\tilde{f}_i}{dt} = -\frac{1}{\Delta x_i(t)}\left[(\mathcal{F}_{\text{agg}})_{i+\frac{1}{2}} - (\mathcal{F}_{\text{agg}})_{i-\frac{1}{2}}\right] + \frac{1}{\Delta x_i(t)}\left[(\mathcal{F}_{\text{break}})_{i+\frac{1}{2}} - (\mathcal{F}_{\text{break}})_{i-\frac{1}{2}}\right]$$
$$+ \frac{G_{i+\frac{1}{2}}\tilde{f}_i}{x_i(t)} - \left(G_{i+\frac{1}{2}} - G_{i-\frac{1}{2}}\right)\frac{\tilde{f}_i}{\Delta x_i(t)} + \tilde{\mathcal{Q}}_i$$

$$\frac{dx_{i+\frac{1}{2}}}{dt} = G_{i+\frac{1}{2}}\,, \qquad \forall\, i = 1,2,\cdots,N \quad \text{with i.c.} \quad \tilde{f}(0,x_i) = \tilde{f}_0(x_i)$$

where

$$(\mathcal{F}_{\text{agg}})_{i+1/2} = \sum_{k=0}^{i} \Delta x_k(t)\tilde{f}_k \left\{ \sum_{j=\alpha_{i,k}}^{N} \int_{\Lambda_j^h} \frac{\beta(x', x_k)}{x'} dx' \tilde{f}_j + \int_{x_{i+1/2}-x_k}^{\alpha_{i,k}-1/2} \frac{\beta(x', x_k)}{x'} dx' \tilde{f}_{\alpha_{i,k}-1} \right\},$$

$$(\mathcal{F}_{\text{break}})_{i+1/2} = \sum_{k=0}^{i} \int_{\Omega_k} x^* \left(\sum_{j=i+1}^{N} \tilde{f}_j \int_{\Omega_j} b(x^*, x') \frac{S(x')}{x'} dx' \right) dx^* + \mathcal{O}(\Delta x^3).$$

Here, the integer $\alpha_{i,k}$ corresponds to the index of the cell such that $x_{i+1/2}(t) - x_k(t) \in \Omega_{\alpha_{i,k}-1}(t)$.

A standard ODE-solver can be used to solve the above ODEs.

Outline

Method 2: Semidiscrete HR-schemes

Integration of PBE over the control volume $\Omega_i = \left[x_{i-\frac{1}{2}}, x_{i+\frac{1}{2}}\right]$ implies

$$\int_{\Omega_i} \frac{\partial \tilde{f}(t,x)}{\partial t}\, dx + \int_{\Omega_i} \frac{\partial [G(t,x)\tilde{f}(t,x)]}{\partial x}\, dx - \int_{\Omega_i} \frac{G(t,x)\tilde{f}(t,x)}{x}\, dx$$

$$= -\int_{\Omega_i} \frac{\partial \mathcal{F}_{\text{agg}}(t,x)}{\partial x}\, dx + \int_{\Omega_i} \frac{\partial \mathcal{F}_{\text{break}}(t,x)}{\partial x}\, dx + \int_{\Omega_i} \tilde{\mathcal{Q}}(t,x)\, dx\,.$$

Let $\tilde{f}_i = \tilde{f}_i(t)$ and $\tilde{\mathcal{Q}}_i = \tilde{\mathcal{Q}}_i(t)$ be the averaged values, then we have

$$\frac{\partial \tilde{f}_i}{\partial t} = -\frac{1}{\Delta x}\left[\mathcal{F}_{i+\frac{1}{2}} - \mathcal{F}_{i-\frac{1}{2}}\right] - \frac{1}{\Delta x}\left[(\mathcal{F}_{\text{agg}})_{i+\frac{1}{2}} - (\mathcal{F}_{\text{agg}})_{i-\frac{1}{2}}\right]$$

$$+ \left[(\mathcal{F}_{\text{break}})_{i+\frac{1}{2}} - (\mathcal{F}_{\text{break}})_{i-\frac{1}{2}}\right] + \frac{G_{i+\frac{1}{2}}\,\tilde{f}_i}{x_i} + \tilde{\mathcal{Q}}_i\,,$$

where $\mathcal{F}_{i+\frac{1}{2}} = (G\tilde{f})_{i+\frac{1}{2}}$ and $(\mathcal{F}_{\text{agg}})$ & $(\mathcal{F}_{\text{break}})$ are as given in Method 1.

The flux $\mathcal{F}_{i+\frac{1}{2}}$ at the right cell interface is given as (*Koren, 1993*):

$$\mathcal{F}_{i+\frac{1}{2}} = \left(\mathcal{F}_i + \frac{1}{2} \Phi \left(r_{i+\frac{1}{2}} \right) \left(\mathcal{F}_i - \mathcal{F}_{i-1} \right) \right)$$

and Φ is defined as:

$$\Phi(r_{i+\frac{1}{2}}) = \max \left(0, \min \left(2r_{i+\frac{1}{2}}, \min \left(\frac{1}{3} + \frac{2}{3} r_{i+\frac{1}{2}}, 2 \right) \right) \right) .$$

The argument $r_{i+\frac{1}{2}}$ of the function Φ is given as

$$r_{i+\frac{1}{2}} = \frac{\mathcal{F}_{i+1} - \mathcal{F}_i + \varepsilon}{\mathcal{F}_i - \mathcal{F}_{i-1} + \varepsilon} .$$

Analogously, one can formulate the flux $\mathcal{F}_{i-\frac{1}{2}}$. Here, $\varepsilon = 10^{-10}$.

There are several other limiting functions, namely, minmod, superbee and MC limiters, etc. Each of them leeds to a different HR-scheme (*LeVeque 2002, Koren 1993*).

Example 1: All Processes

The initial data:

$$f(0,x) = \begin{cases} 100 & \text{for } 0.4 \leq x \leq 0.6\,, \\ 0.01 & \text{elsewhere}\,. \end{cases}$$

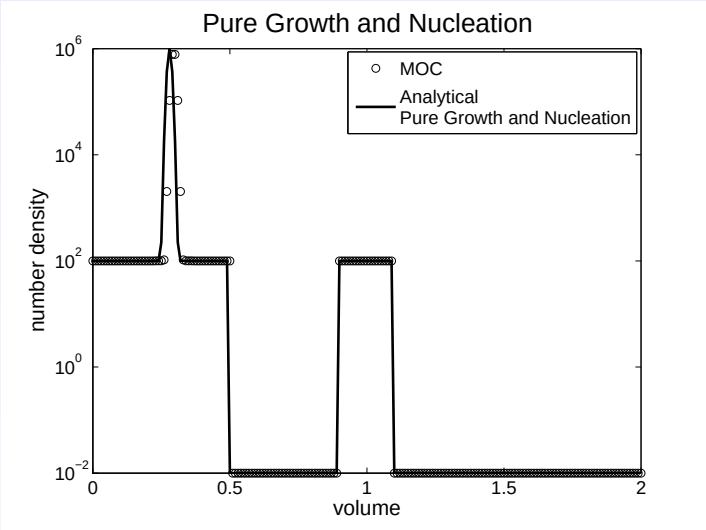
$$\text{B.C.:} \quad f(t,0) = 100 + 10^6 \exp(-10^4 (t - 0.215)^2)\,.$$

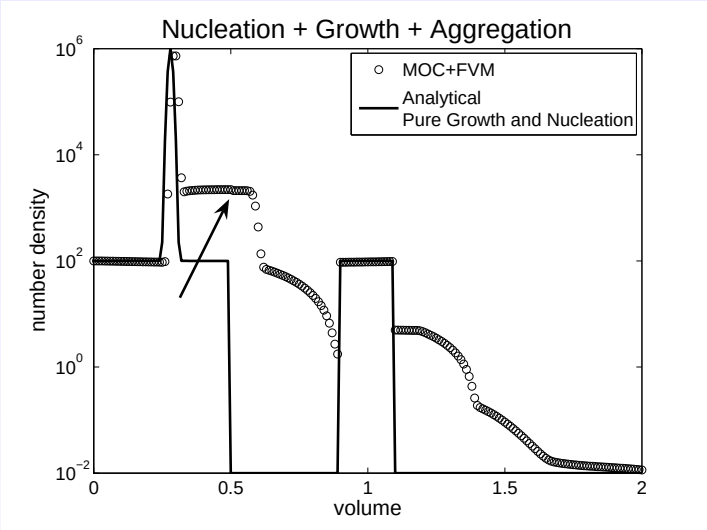
$G = 1.0$, $\beta = 1.5 \cdot 10^{-5}$, $b(t,x,x') = \frac{2}{x'}$ and $S(x) = x^2$. The exact solution in growth and nucleation case is:

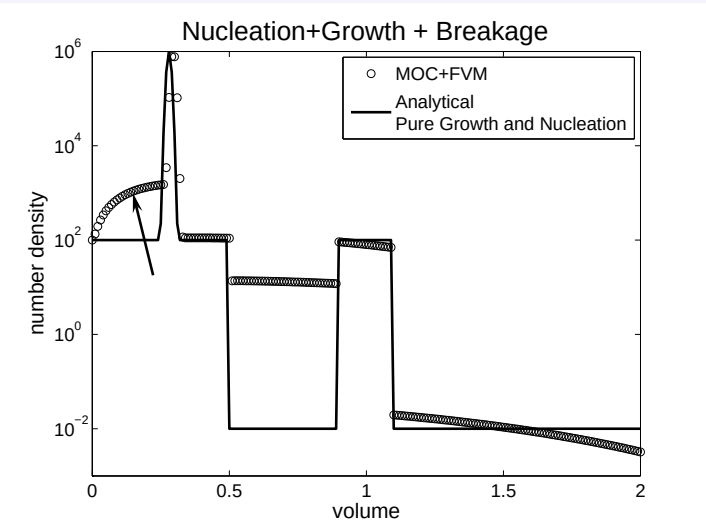
$$f(t,x) = \begin{cases} 10^2 + 10^6 \exp(-10^4((G't - x) - 0.215)^2) & \text{for } 0 \leq x \leq Gt\,, \\ 10^2 & \text{for } 0.4 \leq x - Gt \leq 0.6\,, \\ 0.01 & \text{elsewhere}\,. \end{cases}$$

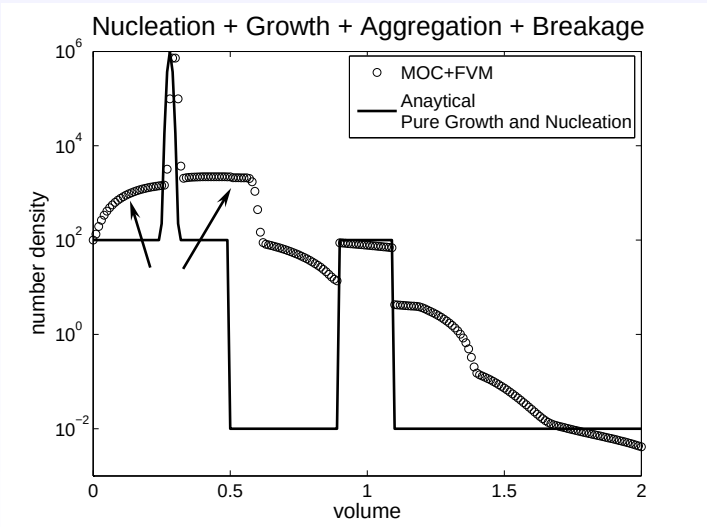
$t_{\max} = 0.5$ and $N = 200$.

Results of Method 1: MOC+FVM

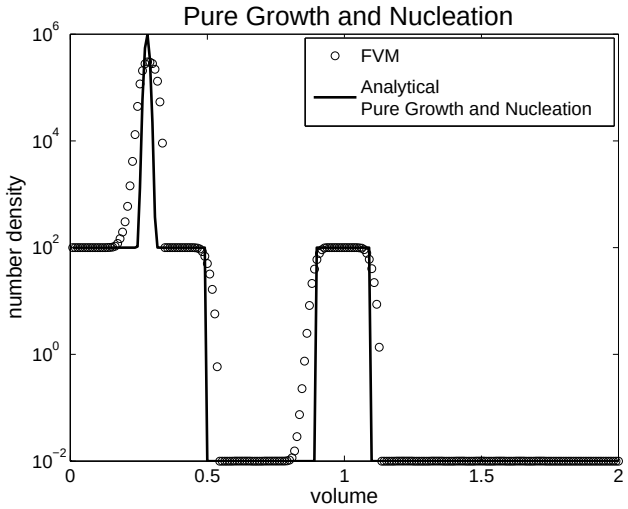


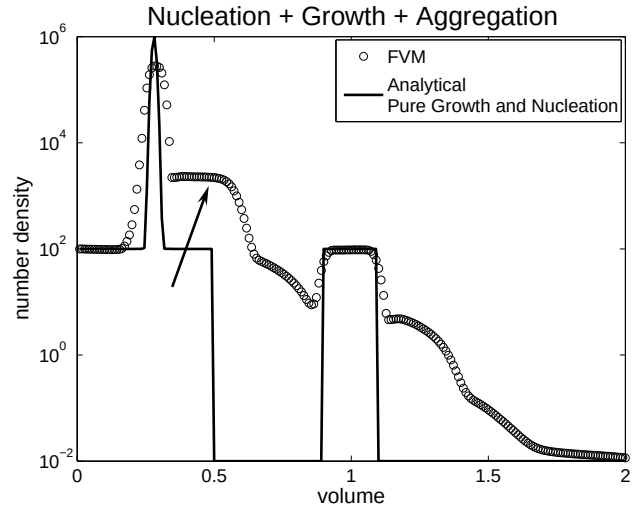


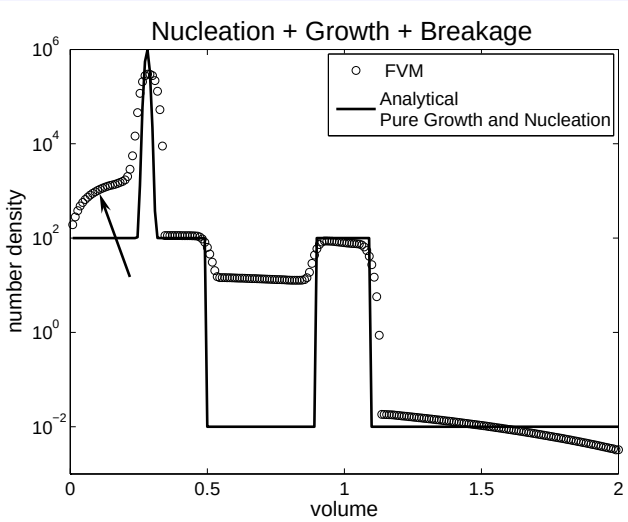


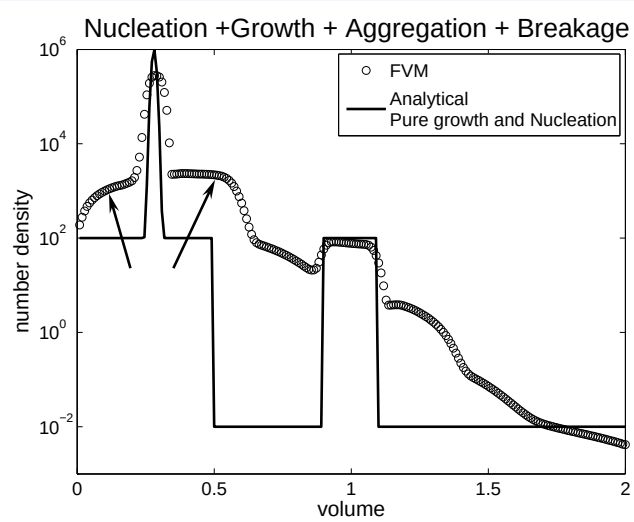


Results of Method 2: FVM







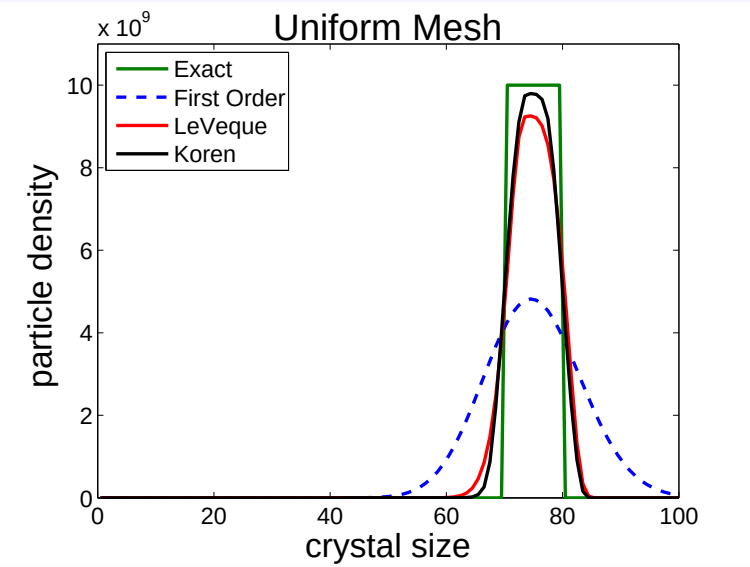


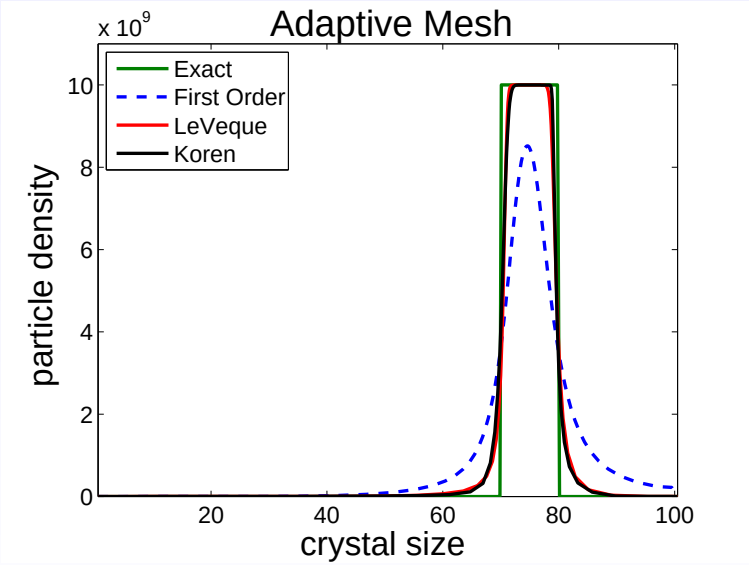
Example 2: Pure Growth

The initial data are:

$$f(0,x) = \begin{cases} 1 \times 10^{10} & \text{if } 10 < x < 20\,, \\ 0 & \text{elsewhere}\,. \end{cases}$$

with $G = 1.0$, $N = 100$, $t = 60$.





For mesh adaptation we have used a moving mesh technique of T. Tang et al. (2003)

Preferential Crystallization

Isothermal Case

$Temperature = 33C^o.$

Non-isothermal Case

$$T(t)[C^o] = -1.24074e^{-7}t^3 + 4.50926e^{-5}t^2 - 0.00405556t + 33\,.$$

Example 3: Preferential Crystallization

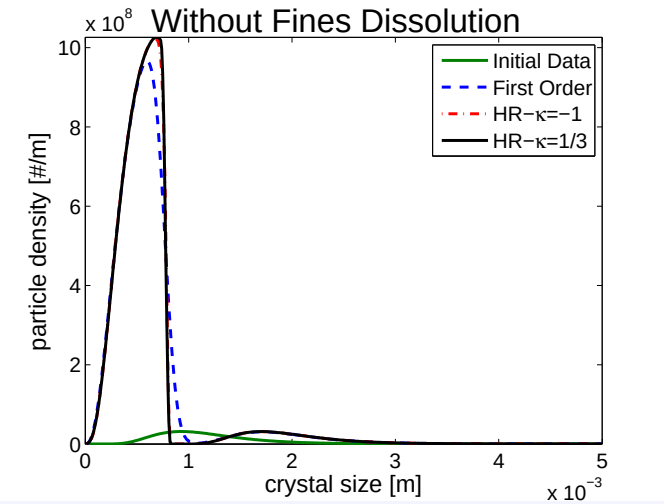
The initial data:

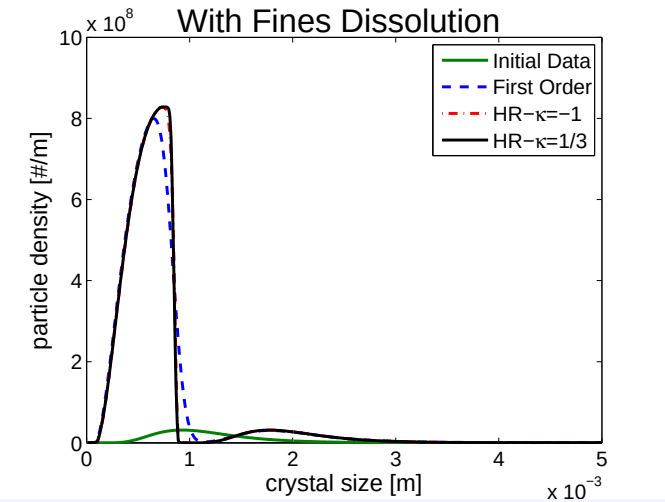
$$f^{(p)}(0,x) = \frac{1}{\sqrt{2\pi}\sigma I_a} \cdot \frac{1}{x} \cdot \exp\left[-\frac{1}{2} \cdot \left(\frac{ln(x)-\mu}{\sigma}\right)^2\right],$$

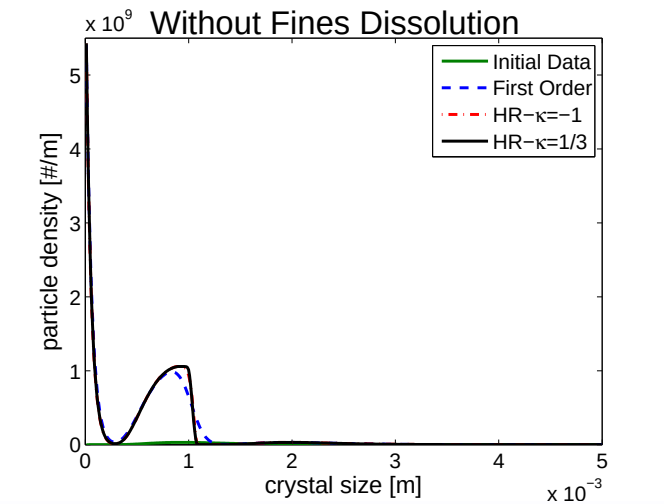
$$f^{(c)}(0,x) = 0\,, \qquad \textit{with} \qquad I_a = \frac{k_V \cdot \rho_S}{M_S} \mu_3^{(p)}(0)\,.$$

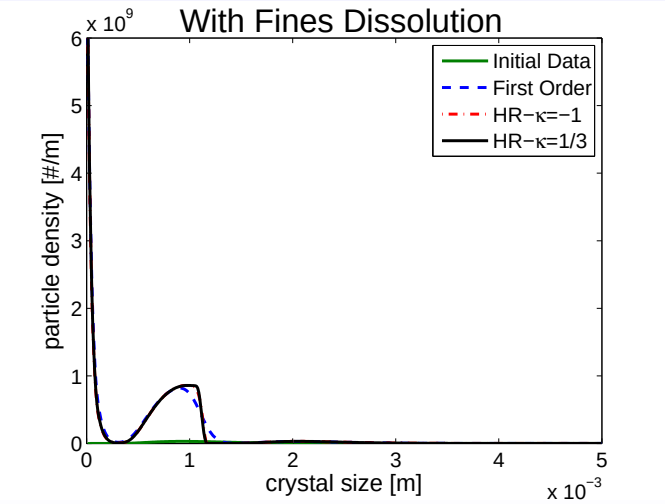
Here $M_S = 2.5 \cdot 10^{-3} kg$ is the mass of initial seeds. The maximum crystal size is $x_{\max} = 0.005\,m$ with $N = 500$ for $t = 600$ min.

Example 3: Preferential Crystallization

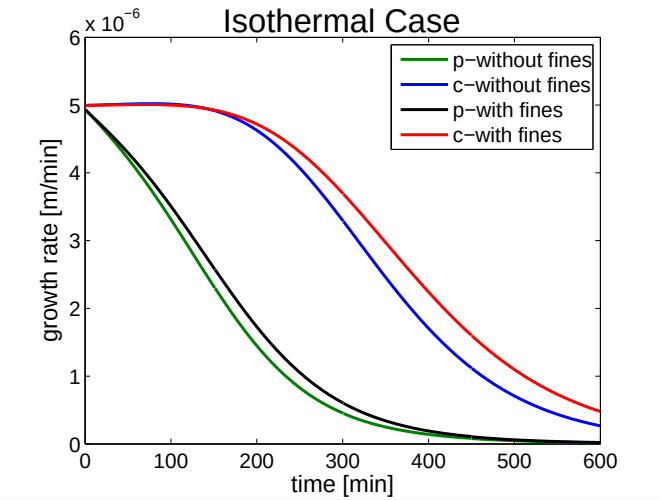


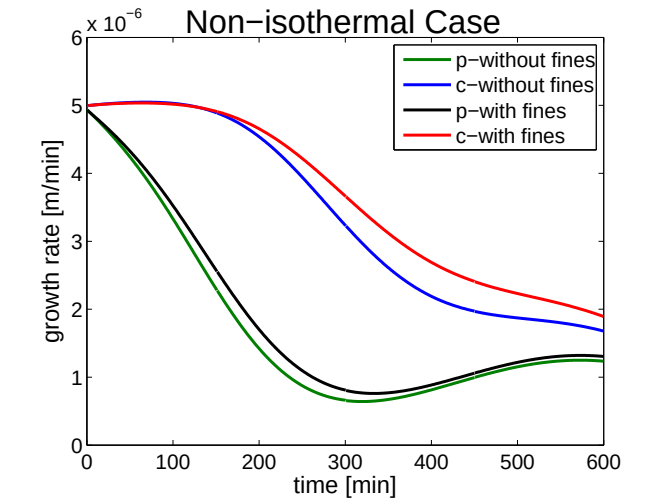


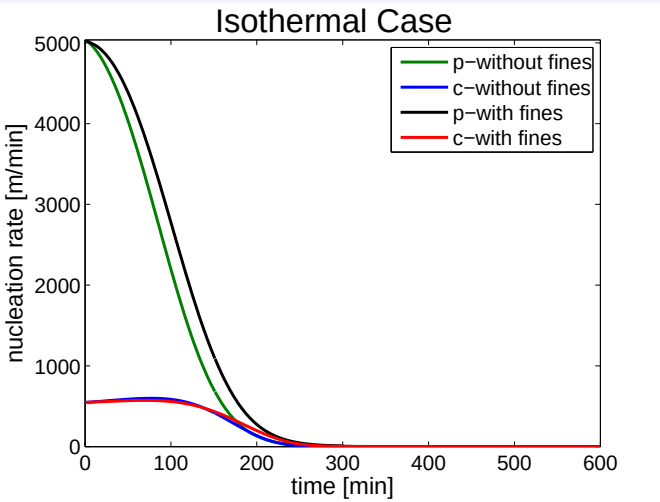


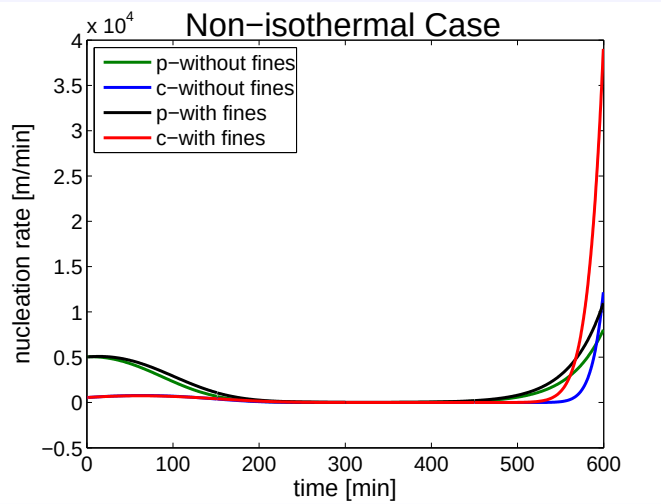


Example 3: Preferential Crystallization









Example 3: Mass Preservation in the Schemes

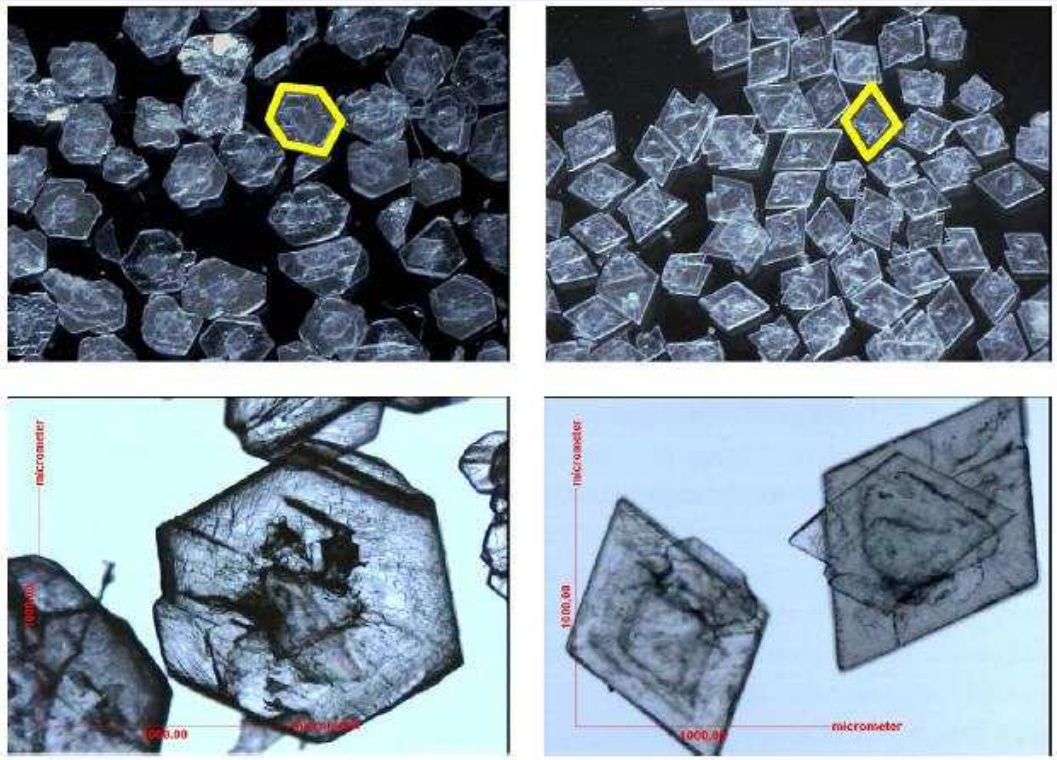
Table: Percentage errors in mass preservation without fines dissolution.

Method	*Isothermal*		*Non-isothermal*		*CPU time (s) (isothermal)*	
	N=500	*N=1000*	*N=500*	*N=1000*	*N=500*	*N=1000*
First order	3.737	3.775	4.460	4.669	1.5	3.1
HR-$\kappa = -1$	3.811	3.813	4.733	4.736	2.2	4.4
HR-$\kappa = 1/3$	3.813	3.814	4.736	4.737	2.3	4.6
MOC	2.604	1.844	3.792	2.917	0.34	0.41

Table: Percentage errors in mass preservation with fines dissolution.

Method	*Isothermal*		*Non-isothermal*		*CPU time (s) (isothermal)*	
	N=500	*N=1000*	*N=500*	*N=1000*	*N=500*	*N=1000*
First order	2.801	2.838	2.841	2.904	2.3	5.5
HR-$\kappa = -1$	2.873	2.875	2.962	2.965	3.1	7.5
HR-$\kappa = 1/3$	2.875	2.876	2.965	2.967	3.5	7.7
MOC	1.823	1.30	2.055	1.086	0.39	0.71

Current Project



Results of isothermal (30 °C) seeded growth experiments with mandelic acid in water. Left:without counter enantiomer; Right: with counter-enantiomer (Lorenz et al., 2006).

For Further Reading


S. Qamar, M. Elsner, I. Angelov, G. Warnecke and A. Seidel-Morgenstern
A comparative study of high resolution schemes for solving population balances in crystallization.
Compt. & Chem. Eng., Vol. 30, 1119-1131, 2006.


S. Qamar, and G. Warnecke
Solving population balance equations for two-component aggregation by a finite volume scheme.
Chem. Sci. Eng., 62, 679-693, 2006.


S. Qamar, and G. Warnecke
Numerical solution of population balance equations for nucleation growth and aggregation processes.
Compt. & Chem. Eng. (in press), 2007.

Thanks for your Attention