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

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Abstract

Crystallization models that take into account direction-dependent growth rates give rise to multi-dimensional population balances that require a high computational cost. We propose a model reduction technique based on the quadrature method of moments (QMOM) that simplifies a two-dimensional population balance to a one-dimensional advection system. Our method returns the crystal volume distribution and other volume dependent moments of the crystal size distribution, in contrast to many other QMOM based reduction methods that lose all the volume dependent information. The method is applied to the direction-dependent growth of barium sulphate crystals, showing a close agreement with the solution of the full two-dimensional population balance.

Keywords: barium sulphate crystallization, direction-dependent growth, two-dimensional population balances, quadrature method of moments

1.Introduction

Crystallization processes are often modeled using one-dimensional population balances. Models of this kind are able to describe the distribution of crystals in a reactor with respect to one size coordinate, e.g. the diameter of spherical crystals. However, crystalline particles are in general of a far more complex shape which, in addition, may change during a crystal's growth process. The growth rate is often different along certain directions of the crystal, which results e.g. in needle-shaped or plate-like crystals. In order to simulate crystallization processes more accurately, one is interested in models that cover the complicated structure of crystal shape and growth mechanisms. As a motivating example one could look at the reaction crystallization of benzoic acid presented in [1]. There the authors showed that appropriate modeling is possible only when the shape evolution of crystal population is taken into account. In this regard several authors have included more than one length coordinate and direction-dependent growth rates, resulting in multi-dimensional population balances, see e.g. Puel et al. [2], Ma et al. [3] and Gunawan et al. [4].

Multi-dimensional population balances can be solved numerically, but are often computationally expensive. To overcome this problem, several model reduction techniques have been suggested. Some of them are based on the quadrature method of moments (QMOM), a method originally introduced by McGraw [5] to simplify one-dimensional population balances. Extensions to multi-dimensional population balances have been proposed, e.g. the method DQMOM of Marchisio and Fox [6]. However, this higher-dimensional variant of QMOM returns only moments of the crystal size distribution (CSD) that are averaged over all length coordinates. Thus the moments obtained from

DQMOM contain much less information than the CSD. It is, for example, not possible to reconstruct the crystal volume distribution (CVD) from these moments.
Recently Briesen [7] has proposed a model reduction technique resulting in moments of the CSD that are still volume dependent. Therefore, this method is able to keep important information like the CVD, and only the additional shape information is sacrificed. This method approximates the CSD with a Gaussian distribution ansatz. It gives very good results if the ansatz is closely met by the CSD. However, for a more general CSD, e.g. a multi-modal one, Briesen's approach is not well suited.
In this paper we introduce a method to compute the same volume dependent moments as in Briesen's approach, but the Gaussian distribution ansatz is replaced by a QMOM ansatz. Therefore, our method combines the advantage of Briesen's method, to keep the volume dependence, with the flexibility of QMOM which can also cover crystal distributions that are far from a Gaussian one. We apply our method to a model of direction-dependent growth of plate-like barium sulphate crystals.

The model

As an example for a two-dimensional crystal growth process we consider the following model for a direction-dependent growth of barium sulphate crystals in a batch reactor of volume V_{reac}. It is assumed that every crystal has the shape of a prism with hexagonal base. Crystals of this particular form have been observed in our precipitation experiments, see Fig. 1, and are reported in the literature, see Voigt and Sundmacher [8], and Niemann and Sundmacher [9]. The dimensions of the hexagonal base are indicated in Fig. 2. The thickness of the prism is denoted by L_2 and is assumed to be always less than a given value $L_{2,\max} > 0$. All hexagonal bases are assumed to be self-similar, i.e. the relations $L_3 = \beta L_1$ and $L_4 = \gamma L_1$ hold for all crystals with given constants $\beta,\gamma > 0$. We set $\alpha = \beta + \gamma$. Let $f(t,L_1,L_2)$ be the crystal size distribution (CSD). The volume of one crystal is given by $V_{\text{cr}}(L_1,L_2) = \alpha L_1^2 L_2/2$, the total volume of crystals in the reactor is $V_{\text{cr,tot}}(t) = \int_0^\infty \int_0^\infty V_{\text{cr}}(L_1,L_2) f(t,L_1,L_2) dL_1 dL_2$. It is assumed that for all crystals $V_{\text{cr}}(L_1,L_2) \geq V_0 > 0$ holds, where V_0 is a given minimum crystal volume. In our example we set $V_{\text{reac}} = 10\ \text{m}^3$, $V_0 = 10^{-20}\ \text{m}^3$, $L_{2,\max} = 3 \cdot 10^{-6}\ \text{m}$, $\beta = 2$ and $\gamma = 1.5$, which gives $\alpha = 3.5$. The crystal growth in the direction of L_1 and L_2 is given by the growth rates $G_1(c) = dL_1/dt = G(c)$ and $G_2(L_2,c) = dL_2/dt = (1 - L_2/L_{2,\max})G(c)$, where c is the concentration of barium sulphate in solution and G is defined according to Bałdyga et al. [10] as

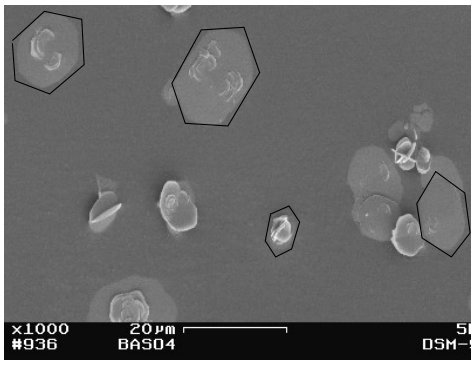


Figure 1: Hexagonal barium sulphate crystals from a precipitation experiment.

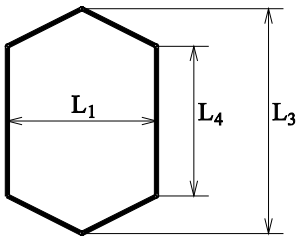


Figure 2: Hexagonal base of the crystals.

$$G(c) = \begin{cases} k_D\left(c - c_{\mathrm{sat}} + c_* - \sqrt{c_*^2 + 2c_*(c - c_{\mathrm{sat}})}\right)^3, & c \geq c_{\mathrm{sat}} \\ 0, & c \leq c_{\mathrm{sat}} \end{cases} \quad (1)$$

with $k_D = 4\cdot10^{-8}$ (m/s)(m^3/mol), $c_{\mathrm{sat}} = 1.05\cdot10^{-2}$ mol/m^3, and $c_* = 0.345$ mol/m^3.

Remark. In Bałdyga et al. [10] direction-dependent growth is not considered, and $G(c)$ is derived there as a uniform growth rate for barium sulphate crystals. Since we have observed plate-like crystals in the experiment, we introduce the growth rates G_1 and G_2 as a simple model that would result in such flat crystals. We would like to stress that our growth rates are not based on empirical measurements; they are rather taken to provide a simple test example for our model reduction technique.

The crystal size distribution $f(t,L_1,L_2)$ satisfies the two-dimensional population balance

$$\frac{\partial f(t,L_1,L_2)}{\partial t} + G_1(c)\frac{\partial f(t,L_1,L_2)}{\partial L_1} + \frac{\partial G_2(L_2,c) f(t,L_1,L_2)}{\partial L_2} = 0 . \quad (2)$$

for all L_1,L_2 with $V_{\mathrm{cr}}(L_1,L_2) > V_0$. As an initial crystal size distribution we consider $f(0,L_1,L_2) = a\, d(L_1,L_2) \exp(-b((L_1 - L_1^*)^2 + (L_2 - L_2^*)^2)$ with $a = 10^{25}$ m^{-2}, $b = 2\cdot10^{12}$ m^{-2}, $V_1 = 5\cdot10^{-19}$ m^3, $L_1^* = (2/\alpha)^{1/2}V_1^{1/3}$, $L_2^* = V_1^{1/3}$, and

$$d(L_1,L_2) = \begin{cases} \frac{1}{2}\cos\left(\frac{\ln V_{\mathrm{cr}}(L_1,L_2) - \ln V_1}{\ln V_1 - \ln V_0}\pi\right) + \frac{1}{2}, & V_0 \leq V_{\mathrm{cr}}(L_1,L_2) \leq V_1 \\ 1, & V_1 \leq V_{\mathrm{cr}}(L_1,L_2) \end{cases} . \quad (3)$$

The factor $d(L_1,L_2)$ is chosen such that $f(0,L_1,L_2)$ is zero at the minimum volume, i.e. for $V_{\mathrm{cr}}(L_1,L_2) = V_0$. We impose zero boundary conditions at the inflow boundary that is located at $V_{\mathrm{cr}}(L_1,L_2) = V_0$. It is assumed that under batch conditions the concentration c decreases with increasing time t since barium sulphate from the solution is needed to grow the crystals:

$$c = c_0 - \left(V_{\mathrm{cr,tot}}(t) - V_{\mathrm{cr,tot}}(0)\right)\rho_{\mathrm{mol}} / V_{\mathrm{reac}} . \quad (4)$$

Here $\rho_{\mathrm{mol}} = 1.928\cdot10^4$ mol/m^3 is the molar density of crystalline barium sulphate and c_0 is the initial concentration. In our example we set $c_0 = 2$ mol/m^3.

2.Transformation to a volume based distribution

In this section we are going to derive an ODE system for volume based moments of the crystal size distribution f. Following Briesen [7], we transform the coordinates (L_1,L_2) to new coordinates (v,κ), where v is the volume of a crystal and κ^2 is the area of its hexagonal base. The transformation is given by $v = \alpha L_1^2 L_2/2$ and $\kappa = (\alpha/2)^{1/2}L_1$. This results in the transformed CSD $F(t,v,\kappa) = (2/\alpha)^{1/2}\kappa^{-2} f(t,L_1,L_2)$. The population balance, Eq. (2), transforms to

$$\frac{\partial F(t,v,\kappa)}{\partial t}+\frac{\partial G_v(v,\kappa,c)F(t,v,\kappa)}{\partial v}+G_\kappa(c)\frac{\partial F(t,v,\kappa)}{\partial \kappa}=0 \tag{5}$$

with $G_v(v,\kappa,c) = (\kappa^2 - v/L_{2,\max} + (2\alpha)^{1/2}v\kappa^{-1})G(c)$ and $G_\kappa(c) = (\alpha/2)^{1/2}G(c)$. We introduce the moments $M_i(t,v)=\int_0^\infty \kappa^i F(t,v,\kappa)d\kappa$ for $i \in \mathbb{Z}$ and assume that all moments required in the method below are finite. Note that M_0 is the crystal volume distribution. Eq. (4) is equivalent to

$$c=c_0-\frac{\rho_{\mathrm{mol}}}{V_{\mathrm{reac}}}\int_{V_0}^\infty v\big(M_0(t,v)-M_0(0,v)\big)dv\,. \tag{6}$$

If we multiply Eq. (5) with κ^i and integrate, we obtain the following PDE system for the moments M_i:

$$\begin{aligned}&\frac{\partial M_i}{\partial t}+\sqrt{2\alpha}vG(c)\frac{\partial M_{i-1}}{\partial v}-\frac{vG(c)}{L_{2,\max}}\frac{\partial M_i}{\partial v}+G(c)\frac{\partial M_{i+2}}{\partial v}\\&+(2-i)\sqrt{\alpha/2}G(c)M_{i-1}-\frac{G(c)}{L_{2,\max}}M_i=0,\qquad i\in I\end{aligned} \tag{7}$$

where I is chosen to be a set of $2n$ integers. This is a system in only one spatial variable v, while the population balance (5) contains two spatial variables v and κ. However, the system of moments is not closed. This means that from whatever index set $I \subset \mathbb{Z}$ the indices i are taken, the system will always contain more moments than equations. In the next section we apply the quadrature method of moments (QMOM) in order to close the system (7).

3.Closure of the system of moments using QMOM

In QMOM, the moment $M_i(t,v)$ in Eq. (7) is replaced by the sum $\sum_{j=1}^n \kappa_j(t,v)^i w_j(t,v)$. The functions $\kappa_j(t,v)$ and $w_j(t,v)$ are the abscissae and weights of a Gaussian quadrature rule. This leads to the PDE system

$$\begin{aligned}&\sum_{j=1}^n i\kappa_j^{i-1}w_j\left[\frac{\partial \kappa_j}{\partial t}+A(\kappa_j,v,c)\frac{\partial \kappa_j}{\partial v}+B(c)\right]\\&+\sum_{j=1}^n \kappa_j^i\left[\frac{\partial w_j}{\partial t}+C(\kappa_j,w_j,v,c)\frac{\partial \kappa_j}{\partial v}+A(\kappa_j,v,c)\frac{\partial w_j}{\partial v}+D(\kappa_j,w_j,c)\right]=0,\qquad i\in I\end{aligned} \tag{8}$$

for the unknown functions $\kappa_j(t,v)$ and $w_j(t,v)$, where the functions A, B, C and D are given according to $A(\kappa_j,v,c) = ((2\alpha)^{1/2}v/\kappa_j - v/L_{2,\max} + \kappa_j^2)G(c)$, $B(c) = -(\alpha/2)^{1/2}G(c)$, $C(\kappa_j,w_j,v,c) = (-(2\alpha)^{1/2}v/\kappa_j^2 + 2\kappa_j)G(c)w_j$, $D(\kappa_j,w_j,c) = ((2\alpha)^{1/2}/\kappa_j - L_{2,\max}^{-1})G(c)w_j$, and c is given in Eq. (6). Introducing the matrices $\mathbf{P} = (P_{ij}) = (i\kappa_j^{i-1}w_j)$, $\mathbf{Q} = (Q_{ij}) = (\kappa_j^i)$ and the line vectors $\mathbf{r} = (r_j) = (\partial\kappa_j/\partial t + A\partial\kappa_j/\partial v + B)$, $\mathbf{s} = (s_j) = (\partial w_j/\partial t + C\partial\kappa_j/\partial v + A\partial w_j/\partial v + D)$ for $i = 1,\ldots,2n$ and $j = 1,\ldots,n$, system (8) can be written in matrix form as $(\mathbf{P}\ \mathbf{Q})(\mathbf{r}\ \mathbf{s})^T = \mathbf{0}$. The matrix $(\mathbf{P}\ \mathbf{Q})$ is almost everywhere regular, i.e. it is singular only for $(\kappa_1,\ldots,\kappa_n) \in$

K_s, where K_s is a set of measure zero in $\mathbb{R}^n$. Thus, system (8) is almost everywhere equivalent to the system ($\mathbf{r}$ $\mathbf{s}$) = $\mathbf{0}$ which we are going to solve. Note that this system is independent on the choice of I. The subsystem $\mathbf{r} = \mathbf{0}$ decouples into n independent advection equations. The system is equipped with initial values $\kappa_j(0,v) > 0$, $w_j(0,v) > 0$ and boundary values $\kappa_j(t,V_0) > 0$, $w_j(t,V_0) > 0$ for $j = 1,\ldots,n$. It follows that $\kappa_j(t,v) > 0$, $w_j(t,v) > 0$ and $A(\kappa_j,v,c) > 0$ hold for all $t \geq 0$, $v \geq V_0$, $j = 1,\ldots,n$. The latter inequality means that advection is directed towards increasing v. Thus the boundary condition is imposed correctly, i.e. at the inflow boundary of the problem. The initial and boundary values $\kappa_j(t^*,v^*)$, $w_j(t^*,v^*)$ for $(t^*,v^*) \in \Gamma := \left(\{0\} \times [V_0,\infty)\right) \cup \left([0,\infty) \times \{V_0\}\right)$ are derived from the initial and boundary moments $M_i(t^*,v^*)$ using the PD-algorithm of Gordon [11], see e.g. McGraw [5]. If this algorithm produces a solution $\kappa_j(t^*,v^*) > 0$, $w_j(t^*,v^*) > 0$, then $M_i(t^*,v^*) = \sum_{j=1}^{n} \kappa_j(t^*,v^*)^i \, w_j(t^*,v^*)$ holds for $i = 0,\ldots,2n-1$.

The initial and boundary moments $M_i(t^*,v^*)$ are calculated from the known initial and boundary values of f by quadrature. The advection system is numerically solved using a splitting scheme that consists of a second order upwind scheme [12] for the advection part and a second order Runge-Kutta method for the source term. The numerical method returns a solution $\kappa_j(t_k,v_l)$, $v_j(t_k,v_l)$ on a time-volume grid. The moments M_i are then approximated at the grid by $M_i^{\mathrm{Q}}(t_k,v_l) = \sum_{j=1}^{n} \kappa_j(t_k,v_l)^i \, v_j(t_k,v_l)$, where the superscript Q stands for QMOM. Off the grid, $M_i^{\mathrm{Q}}(t,v)$ is linearly interpolated. The concentration c is approximated at the time grid by

$$c^{\mathrm{Q}}(t_k) = c_0 - (\rho_{\mathrm{mol}} / V_{\mathrm{reac}}) \int_{V_0}^{\infty} v \left(M_0^{\mathrm{Q}}(t_k,v) - M_0(0,v) \right) dv .$$

Here the integral is evaluated by quadrature.

The above mentioned PD-algorithm is not applicable if $M_0(t^*,v^*) = 0$ holds for any $(t^*,v^*) \in \Gamma$. Therefore, it is numerically problematic if the CSD f is zero in the initial or boundary condition. We overcome this problem by adding a negligibly small positive perturbation to the initial and boundary conditions on f which are given in Section 2.

4.Numerical results

In this section we compare the moments M_i^{Q} obtained with the QMOM based closure method with the moments M_i that are calculated from a reference solution of system (2), (4). Since an exact solution of this system is not known, the reference solution is computed numerically using a second order finite volume method [13], see also [4], for the two-dimensional advection problem (2). Fig. 3 shows contour plots of $f(0,L_1,L_2)$ and $f(400\mathrm{s},L_1,L_2)$ obtained from the reference solution. The problems are computed on such a fine grid that any further refinement would lead only to negligible changes in the moments. The relative error of M_i^{Q} is estimated by the expression

$$\eta(M_i^{\mathrm{Q}},t) = \int_{V_0}^{\infty} | M_i^{\mathrm{Q}}(t,v) - M_i(t,v) | dv \, / \int_{V_0}^{\infty} M_i(t,v) dv .$$

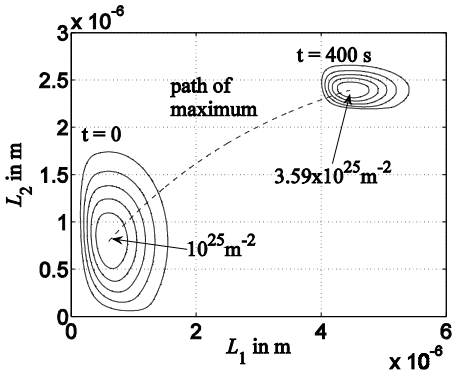


Figure 3: Contour plot of the crystal size distribution f at times $t = 0$ and $t = 400$ s.

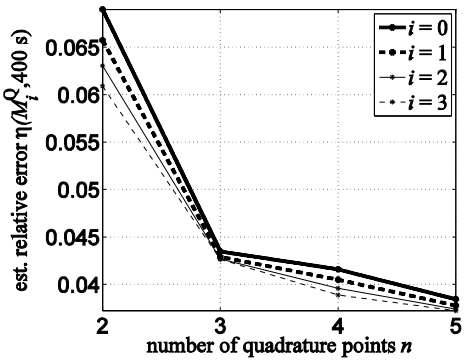
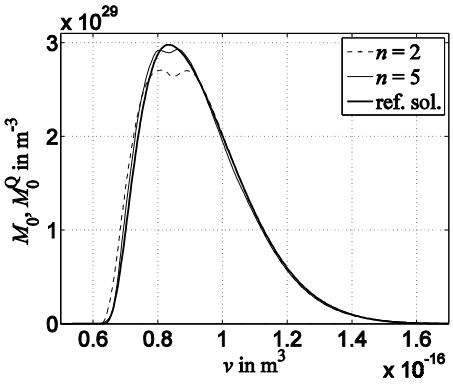


Figure 4: Estimated relative error $\eta(M_i^{\mathrm{Q}},400\ \mathrm{s})$.



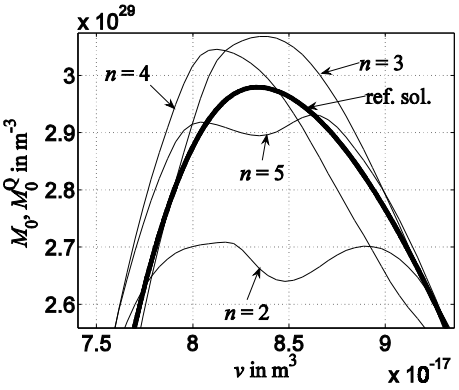


Figure 5: M_0 and M_0^{Q} at time $t = 400$ s. Right plot shows zoom into left plot.

In Fig. 4 we show the estimated error for $t = 400$ s, $i = 0, 1, 2, 3$ and $n = 2, 3, 4, 5$. As expected, it turns out that the estimated error can be decreased by increasing the number n of quadrature points in QMOM. However, for $n \geq 6$ the numerical method solving the advection problem in QMOM was unstable.
In Fig. 5 the moments M_0 and M_0^{Q} are shown for time $t = 400$ s and $n = 2, 3, 4, 5$. In the left plot the cases $n = 3, 4$ are omitted to enhance visibility. The moments M_0^{Q} are shown to lie close to M_0.

5.Conclusion

We have proposed a QMOM based dimension reduction method for the numerical simulation of direction-dependent crystal growth. Our method reduces a two-dimensional population balance to a small system of advection equations. The method returns volume-dependent moments of the CSD which might be an advantage over reduction methods that exclude all information on volume dependence of the CSD. Our dimension reduction was shown to give satisfactory results when it was applied to a problem that models the growth of hexagonal barium sulphate crystals. The method could be extended to cover additional processes like crystal nucleation.

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