Dimension reduction of bivariate population balances using the quadrature method of moments

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\textbf{A B S T R A C T}

Crystallization models with direction-dependent growth rates give rise to multi-dimensional population balance equations (PBE) that require a high computational cost. We propose a model reduction based on the quadrature method of moments (Q MOM). Using this method a two-dimensional population balance is reduced to a system of one-dimensional advection equations. Despite the dimension reduction the method keeps important volume dependent information of the crystal size distribution (CSD). It returns the crystal volume distribution as well as other volume dependent moments of the two-dimensional CSD. The method is applied to a model problem with direction-dependent growth of barium sulphate crystals, and shows good performance and convergence in these examples. We also compare it on numerical examples to another model reduction using a normal distribution ansatz approach. We can show that our method still gives satisfactory results where the other approach is not suitable.

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1. Introduction

Crystallization processes are often modelled using one-dimensional population balances. Models of this kind are able to describe the distribution of crystals in a reactor with respect to one internal property. However, crystalline particles are in general of a far more complex shape that may, in addition, change during a growth process. Moreover, the speed of crystal growth might be different along the axes of a crystal. In order to simulate crystallization processes more accurately, one is interested in models that are able to cover the complicated structure of crystal shapes and growth mechanisms. As a step in this direction several authors have included more than one internal crystal property and direction-dependent growth speeds, resulting in multi-dimensional population balances. To name a few examples, two-dimensional population balances have been used by Puel, Marchal, and Klein (1997) and Puel, Févotte, and Klein (2003a, 2003b) for the description of hydroquinone (C\textsubscript{6}H\textsubscript{4}(OH)\textsubscript{2}) crystallization, and by Ma, Tafti, and Braatz (2002), Gunawan, Fusman, and Braatz (2004) and Gunawan, Ma, Fujiwara, and Braatz (2002) to model the direction-dependent growth of potassium dihydrogen phosphate (K\textsubscript{2}HPO\textsubscript{4}) crystals. While these authors track the crystal distribution with respect to two characteristic length coordinates, (Gerstlauer, Mitrovic, Motz, & Gilles, 2001) consider a two-dimensional population balance model covering one length coordinate and the inner lattice strain as a second crystal property.

Multi-dimensional population balances are computationally rather costly. To overcome this problem, several model reduction approaches have been suggested. Some of them are based on the quadrature method of moments (Q MOM), a method that approximates the moments of the crystal distribution by finite sums. The method was originally introduced by McGraw (1997) to simplify one-dimensional population balances. Extensions to multi-dimensional population balances have been proposed by Wright, McGraw, and Rosner (2001), Rosner and Pykkonen (2002), Yoon and McGraw (2004), and Marchisio and Fox (2005). However, all these higher-dimensional QMOM approaches are only able to return moments of the crystal distribution that are averaged over all internal coordinates. Therefore, using those methods one can not calculate a crystal volume distribution or other moments that are functions of a crystal volume coordinate.

On the other hand, Briesen (2006) has introduced a model reduction that keeps information on the volume distribution of crystals. Briesen approximates the crystal size distribution (CSD) by a normal distribution ansatz. This method is shown to perform well if the ansatz is closely met by the CSD of the problem. However, it is questionable if Briesen's method can produce accurate results if the underlying ansatz is not a good approximation of the CSD. This
distribution (CSD) is described by a density function. Regarding this aspect, our approach is similar to the method of Briesen. However, our method will be better suited to deal with crystal size distributions that are not approximately normally distributed, such as multi-modal ones. Such distributions frequently occur in crystallization experiments. A number of numerical examples will illustrate this issue.

The method presented here is designed to reduce the dimension of two-dimensional population balances that result from modelling crystals growing with different speed in two characteristic directions, e.g. in length and in width. Therefore we will restrict ourselves to the growth mechanism. Our method could be further extended to include other processes like crystal nucleation.

The article is structured as follows: In Section 2, the crystal growth is modelled using a two-dimensional population balance. A special coordinate transformation that will allow us to calculate crystal volume distributions is introduced in Section 3. A number of model problems is presented in Section 4, the normal distribution method of Briesen is described in Section 5, the population balance equation is transformed into a system of moments of the CSD. The normal distribution method of Briesen is described in Section 6, while in Section 7 we present our QMOM based dimension reduction method. In Section 8 the numerical performance of Briesen’s normal distribution method and our QMOM based method is shown for the given model problems. Numerous plots of the numerical error depending on the number of quadrature points are included.

2. The population balance

The growth process of crystals showing direction-dependent growth can be modelled using a multi-dimensional population balance. We first consider a simple model where every crystal is assumed to be in the shape of a parallelepiped with length $l_1 > 0$ and equal width and depth $l_2 > 0$ so that the volume of a crystal is given by

$$v = l_1^2 l_2.$$ (1)

The crystal shall have the growth rate $G_1(l_1, l_2)$ in its length-direction and the growth rate $G_2(l_1, l_2)$ in the direction of its width and depth where $G_1$ and $G_2$ might differ. As indicated, both growth rates are allowed to be size-dependent, i.e. they can depend on the characteristic quantities $l_1$ and $l_2$ of the crystal. The crystal size distribution (CSD) is described by a density $n(t, l_1, l_2)$ defined in such a way that the number of crystals over $[l_1 \min, l_1 \max] \times [l_2 \min, l_2 \max]$ at time $t \geq 0$ is given by

$$\int_{l_1 \min}^{l_1 \max} \int_{l_2 \min}^{l_2 \max} n(t, l_1, l_2) dl_1 dl_2.$$ (3)

We assume $G_1(l_1, l_2) = G_1(l_1, l_2)$ and $G_2(l_1, l_2) = G_2(l_1, l_2)$ to be a continuously differentiable vector field and consider the two-dimensional crystal growth process modeled by the population balance equation

$$\frac{\partial n}{\partial t} + \frac{\partial G_1(l_1, l_2) n(t, l_1, l_2)}{\partial l_1} + \frac{\partial G_2(l_1, l_2) n(t, l_1, l_2)}{\partial l_2} = 0,$$

$$0 < t < t_{\max}, (l_1, l_2) \in \Omega,$$ (2)

over a domain $\Omega \subset \mathbb{R}^2$. This population balance is a two-dimensional advection equation. With the notations $V n = (\partial n / \partial l_1, \partial n / \partial l_2)$ and $\nabla \cdot G = \partial G_1 / \partial l_1 + \partial G_2 / \partial l_2$ we rewrite Eq. (2) in the compact form

$$\frac{\partial n}{\partial t} + \nabla \cdot (V n + (\nabla \cdot G) n) = 0.$$ (3)

Remark 2.1. In commonly used crystallization models the growth rate $G$ usually depends also on quantities like the concentration of solute in the supersaturated liquid, the temperature etc., see e.g. Mersmann (2001). A problem with concentration dependent growth will be introduced in Section 4. The model considered here is restricted to crystal growth and does not cover processes like crystal nucleation or breakage. We have chosen such a simple model to illustrate the dimension reduction method described in the sequel. Extensions to include crystal nucleation are possible.

For simple growth functions $G_1, G_2$ the solution of problem (2) is best obtained by the method of characteristics. For more complicated growth rates $G_1, G_2$, e.g. in case the growth functions $G_1, G_2$ depend on $n$, analytic expressions for the characteristic curves may not be computable so that the method of characteristics is to be combined with numerical discretization. In such a case one might prefer to use a finite difference or finite volume scheme for the solution of the advection Eq. (2). Alternatively, a model reduction technique might be applied reducing the computational effort by converting the population balance equation into a system of equations for certain moments of $n$. Such moments provide only partial information of the solution $n$, but in many applications this will be sufficient.

3. A coordinate transformation

As in Briesen (2006), the coordinates $(l_1, l_2)$ are transformed to new coordinates $(v, x)$ according to

$$v = l_1^2 l_2, x = l_2.$$ (4a)

The transformed crystal density is defined by

$$N(t, v, x) = \frac{n(t, l_1, l_2)}{v^2}.$$ (4b)

in the variables $v$ and $x$: At time $t$, the number of crystals over $[v_{\min}, v_{\max}] \times [x_{\min}, x_{\max}]$ is now given by

$$\int_{v_{\min}}^{v_{\max}} \int_{x_{\min}}^{x_{\max}} N(t, v, x) dv dx.$$

Throughout this article we will assume the following conditions to be fulfilled:

Assumption 1. The integrals $\int_{0}^{\infty} \psi N(t, v, x) dv dx$ exist, are finite and differentiable for all $t \geq 0$, $v > 0$ and $i \in \mathbb{R}$. □

Assumption 2. There holds $\lim_{\mu \rightarrow 0} \psi N(t, v, x) = 0$ for all $t \geq 0$, $v > 0$ and $i \in \mathbb{R}$. □

These assumptions are satisfied for all crystal size distributions considered in the model problems we are going to introduce in Section 4. The assumptions are also fulfilled for all processes with a smooth crystal size distribution where the maximum crystal size stays bounded.

The moments of the transformed crystal size distribution $N$ are defined by

$$M_i(t, v) = \int_{0}^{\infty} \psi^n N(t, v, x) dv dx$$

for $i \in \mathbb{Z}$.

In particular, the zeroth moment $M_0(t, v)$ is the crystal volume distribution, meaning that the number of crystals having an individual volume $v \in [v_1, v_2]$ at time $t$ is given by $\int_{v_1}^{v_2} M_0(t, v) dv$. Thus the total volume of crystals can be expressed as

$$V_{cr, tot}(t) = \int_{0}^{\infty} v M_0(t, v) dv.$$ (5)
In the case of some simple growth rates, a model reduction technique leads to a system of partial differential equations for the moments $M_i$. We will illustrate such reduction techniques with some examples in Section 5.

**Remark 3.1.** A model reduction is also possible in terms of the original coordinates $(l_1, l_2)$ leading to the moments $M_i(t, l_1) = \int_0^{l_2} \int_{t_1}^{t_2} n(t, l_1, l_2)dl_2$. However, the results for the moments $M_i$ are not so suited for a comparison with experimental data since the crystal distributions over the length coordinates are difficult to measure in practice. A volume based crystal distribution is much easier to obtain from experiments.

We define
\[ \hat{G}_i(v, x) = G_i(v/x^2, x) = G_i(l_1, l_2) \]  
for $i = 1, 2$ and obtain by chain rule the partial derivatives
\[ \frac{\partial n(t, l_1, l_2)}{\partial t} = \frac{\partial N(t, v, x)}{\partial t}, \quad \frac{\partial n(t, l_1, l_2)}{\partial x} = \frac{\partial N(t, v, x)}{\partial x}, \quad \frac{\partial n(t, l_1, l_2)}{\partial l_2} = 2\sigma N(t, v, x) + x^2 \frac{\partial^2 N(t, v, x)}{\partial x^2}, \]  
and
\[ \frac{\partial \hat{G}_i(l_1, l_2)}{\partial t} = \frac{\partial \hat{G}_i(v, x)}{\partial t}, \quad \frac{\partial \hat{G}_i(l_1, l_2)}{\partial x} = \frac{\partial \hat{G}_i(v, x)}{\partial x}, \quad \frac{\partial \hat{G}_i(l_1, l_2)}{\partial l_2} = 2 \frac{\partial \hat{G}_i(v, x)}{\partial l_2} x. \]  

Thus the population balance (2) transforms to the equation (see Briesen (2006))
\[ \frac{\partial N}{\partial t} + \left( x^2 \frac{\partial \hat{G}_1}{\partial x} + 2x \frac{\partial \hat{G}_2}{\partial x} \right) \frac{\partial N}{\partial x} + \frac{\partial \hat{G}_1}{\partial x} \frac{\partial N}{\partial x} + \frac{\partial \hat{G}_2}{\partial x} \frac{\partial N}{\partial x} = N = 0. \]  

Introducing the vector
\[ \mathbf{\hat{v}} = \left( \frac{\partial \hat{G}_1}{\partial x}, \frac{\partial \hat{G}_2}{\partial x} \right) \]  
and the notation $\hat{\mathbf{v}} = (\hat{\partial}/\hat{\partial}v, \hat{\partial}/\hat{\partial}x)$, Eq. (8) can be written in the form
\[ \frac{\partial \hat{v}}{\partial t} + G \cdot \hat{\mathbf{v}} N + (\hat{\mathbf{v}} \cdot \hat{\mathbf{g}}) N = 0. \]  

In this sense, Eq. (3) is invariant under the coordinate transformation $(l_1, l_2) \rightarrow (v, x)$.

4. Model problems

We will apply the model reduction technique described in this article to the following model problems. The Problems 1–3 are rather academic and not related to the crystallization process of a specific substance. Nevertheless, these problems are useful test cases for our numerical method with a known exact solution. Problem 4 describes the direction-dependent growth of barium sulphate crystals.

**Problem 1 (Unimodal distribution, size-independent growth).** For given $v_0$ and $v_{\text{max}}$ satisfying $0 < v_0 < v_{\text{max}}$ let the domain $\Omega$ be defined by
\[ \Omega = \{(l_1, l_2) \in \mathbb{R}^2 : v_0 < l_1 < l_2 < v_{\text{max}}\}. \]  

We consider the population balance (2) with size-independent growth, i.e. $G_1(l_1, l_2) \equiv c_1$, $G_2(l_1, l_2) \equiv c_2$ with positive constants $c_1$, $c_2$. Initial and boundary conditions are set according to the function.

\[ n(t, l_1, l_2) = e^{-\delta[(l_1-G_1)^2+(l_2-G_2)^2]} \]  
with $\delta > 0$ being the solution.

**Problem 2 (Bimodal distribution, size-independent growth).** Let $\Omega$, $G_1$, and $G_2$ be defined as in Problem 1. Initial and boundary conditions are set according to
\[ n(t, l_1, l_2) = e^{-\delta[(l_1-l_1,0-G_1)^2+(l_2-l_2,0-G_2)^2]} + e^{-\delta[(l_1-G_1)^2+(l_2-l_2,0-G_2)^2]} \]  
with parameters $\delta, l_{1,0}, l_{2,0} > 0$ being the solution.

**Problem 3.** Let $\Omega, G_1$, and $G_2$ be defined as in Problem 1. The initial condition $n(0, l_1, l_2) = n_0(l_1, l_2)$ is set according to
\[ a(v) = \exp(-\alpha(v-v_0)^2), \quad \mu(v) = |\beta|, \quad \sigma(v) = \beta|\beta|. \]  

\[ n_0(v, x) = \begin{cases} a(v) & \text{if } v > 0, \ \sigma(v) > 0, \\ 0 & \text{otherwise.} \end{cases} \]  

where $\alpha > 0$, $\beta > 0$ and $v_0 > 0$ are given parameters. Boundary conditions are set according to the desired solution $n(t, l_1, l_2) = n_0(l_1-G_1, l_2-G_2)$.

We note that Problem 3 has been designed such that initially the crystal size distribution $n$ is a normal distribution with respect to $x$ along the curves $v = \text{const}$. The same kind of problem is considered in Briesen (2006).

**Problem 4 (Direction-dependent growth of barium sulphate crystals).** We consider the growth of barium sulphate crystals under batch conditions in a supersaturated solution. To simplify the problem, additional processes like crystal nucleation are excluded. The following assumptions are made.

- Every crystal has the shape of a prism with hexagonal base. Crystals of this particular form have been observed in precipitation experiments, see Voigt and Sundmacher (2007), Niemann and Sundmacher (2007) or Voigt, Heineken, Flockner, and Sundmacher (2008). Fig. 1 (left) shows crystals of hexagonal shape that we have observed in precipitation experiments. The dimensions of the hexagonal base of the prism are indicated in Fig. 1 (right). The thickness of the prism is denoted by $L_{\text{max}}$.
- All hexagonal basal planes of the crystals are self-similar, i.e. the relations $L_3 = \beta L_1$, and $L_4 = y L_1$ hold for all crystals with given constants $\beta > 0$ and $\gamma > 0$. We set $\alpha = \beta + \gamma$.
- The thickness $L_2$ of the crystals is assumed to be always less than a given value $L_{\text{max}} > 0$. All crystals have a volume greater than a minimum value $v_{\text{max}} > 0$.
- The growth of $L_1$ is given by the growth rate $\frac{dl_1}{dt} = \hat{G}_1(c) := G(c)$, and the growth of $L_2$ by the growth rate $\frac{dl_2}{dt} = \hat{G}_2(c, L_2) := G(c)(1 - L_2/L_{\text{max}})$ where $c$ is the concentration of barium sulphate in solution and $G$ is defined, according to Baldyga, Podgórski, and Pohorecki (1995), as
\[ G(c) = k_0 \left( c - c_{\text{sat}} + c_\alpha - \sqrt{c_\beta^2 + 2c_\beta(c-c_{\text{sat}})} \right), \quad c \geq c_{\text{sat}} \]  
with $k_0 = 4 \times 10^{-8} \text{m/s} / (\text{m}^3 / \text{mol})$, $c_{\text{sat}} = 1.05 \times 10^{-2} \text{mol} / \text{m}^3$, and $c_\alpha = 0.345 \text{mol} / \text{m}^3$. The CSD $n(t, l_1, l_2)$ is a number density in $L_1$ and $L_2$ and satisfies the population balance
\[ \frac{\partial n}{\partial t} + \frac{\partial \hat{G}_1 n}{\partial l_1} + \frac{\partial \hat{G}_2 n}{\partial l_2} = 0. \]  

\[ \text{(12)} \]
An initial CSD $\hat{n}(0, L_1, L_2)$ is given.

- The concentration $c$ decreases since barium sulphate from the solution is needed to grow the crystals:

$$c(t) = c(0) - \frac{\rho_{\text{mol}}}{V_{\text{react}}} (V_{\text{cr, tot}}(t) - V_{\text{cr, tot}}(0))$$

$$= c(0) - \frac{\rho_{\text{mol}}}{V_{\text{react}}} \int_0^\infty \int_0^{L_1} \int_0^{L_2} V_{\text{cr}}(L_1, L_2) (\hat{n}(t, L_1, L_2) - \hat{n}(0, L_1, L_2)) dL_1 dL_2.$$

Here, $V_{\text{react}}$ is the reactor volume, $\rho_{\text{mol}} = 1.928 \times 10^4 \text{ mol/m}^3$ is the molar density of crystalline barium sulphate, and $V_{\text{cr}}(L_1, L_2) = \alpha 1^2 L_2^2 / 2$ is the volume of a crystal. An initial concentration $c(0) > c_{\text{sat}}$ is given. $c(t) > c_{\text{sat}}$ shall hold for all $t \geq 0$.

As an example we set $L_{\text{max}} = 3 \times 10^{-6} \text{ m}$, $c(0) = 2 \text{ mol/m}^3$, $V_{\text{react}} = 10 \text{ m}^3$, $\beta = 2$, $\gamma = 1.5$, which gives $\alpha = 3.5$.

We introduce the notations

$$l_1 = L_1, \quad l_2 = \frac{\alpha}{2} L_1, \quad G_1 = \left(1 - \frac{l_1}{L_{\text{max}}} \right) G(c), \quad G_2 = \frac{\alpha}{2} G(c),$$

$$n(t, l_1, l_2) = \frac{\sqrt{2}}{\alpha} \hat{n}(t, L_1, L_2),$$

with the coordinates $l_1$ for the length and $l_2$ for the geometry of the hexagonal base so that $G_1$ and $G_2$ are of the form $G_1 = G(\nu, x, c)$ and $G_2 = G_2(c)$ respectively. Then Eq. (2) holds and the volume of a crystal satisfies $V_{\text{cr}}(l_1, l_2) = l_1^2 l_2$ (cf. (1)). Moreover, $n$ is a number density in the variables $l_1$ and $l_2$ and Eq. (13) becomes

$$c(t) = c(0) - \frac{\rho_{\text{mol}}}{V_{\text{react}}} \int_0^\infty v(M_0(t, v) - M_0(0, v)) dv. \quad (14)$$

We consider the problem on the domain

$\Omega = \{(l_1, l_2) \in \mathbb{R}^2 : v_0 < V_{\text{cr}}(l_1, l_2) < v_{\text{max}}\}$

with $v_0 = 10^{-20} \text{ m}^3$ and $v_{\text{max}} = 2 \times 10^{-16} \text{ m}^3$. The initial condition is given by

$$\hat{n}(0, l_1, l_2) = d(l_1, l_2) a e^{-b(l_1-l_1^2) ^2 + (l_2-l_2^2)^2} \quad (15)$$

with $a = 10^{25} \text{ m}^{-2}$, $b = 2 \times 10^{12} \text{ m}^{-2}$, $\nu^* = 5 \times 10^{-19} \text{ m}^3$, $L_1^* = \sqrt{2/\alpha} \sqrt{\nu^*}$, $L_2^* = \sqrt{\nu^*}$, and

$$d(l_1, l_2) = \begin{cases} 1, & 0 \leq V_{\text{cr}}(l_1, l_2) \leq v^* \\ 1, & \nu^* \leq V_{\text{cr}}(l_1, l_2) \leq L_2^* \end{cases}$$

Note that $d$ can be seen as a function of $V_{\text{cr}}$ or even of $\ln V_{\text{cr}}$. The initial condition Problem 4 is a Gaussian distribution multiplied with the function $d$. We have chosen the function $d$ such that the initial condition is zero at the boundary $\nu = v_0$ and for $L_2 \geq L_{\text{max}}$. We impose zero boundary conditions at the inflow boundary that is given by $V_{\text{cr}}(l_1, l_2) = v_0$.

$$\text{Remark 4.1. The growth rate } G(c) \text{ has been taken from Baladyga, Podgór ska, and Pohorecki (1995) where direction dependent growth is not considered. } G(c) \text{ is derived there as a uniform growth rate for barium sulphate crystals. Since we have observed thin plate-like crystals in the experiment, we introduce the growth rates } \tilde{G}_1 \text{ and } \tilde{G}_2 \text{ as a simple model for obtaining such flat crystals. The growth rate } \tilde{G}_2 \text{ is constructed such that it does not allow the length } L_2 \text{ to exceed the maximum value } L_{\text{max}}. \text{ These growth rates are not based on empirical measurement; they are just taken to provide a simple test example for our model reduction technique.}\)$$

It follows from the definitions of $d$, $\tilde{n}(t, l_1, l_2)$ and $N(t, \nu, x)$ (cf. (4b)) and from the Eqs. (12) and (15) that $\tilde{n}(t, L_1, L_2)$ vanishes for $L_2 \geq L_{\text{max}}$ and all $t \geq 0$. This is equivalent to

$$N(t, \nu, x) = 0 \quad \text{for} \quad t \geq 0 \quad \text{and} \quad x \leq \sqrt{\nu/L_{\text{max}}} = x_{\text{min}}(\nu). \quad (16)$$

5. The system of moments

A PDE system for the moments $M_i$ exists for some simple growth rates, especially for size-independent growth rates as in the Problems 1–3 and for the linear size-dependent growth in Problem 4.

5.1. The system of moments for size-independent growth

Since in the Problems 1–3 the growth rates $G_1$ and $G_2$ do not depend on $l_1$ and $l_2$, hence $\tilde{G}_1 = G_1$ and $\tilde{G}_2 = G_2$ defined in (7) and (9a) are independent on $\nu$ and $x$. Due to (9a), $\tilde{G}_1$ depends on $\nu$ and $x$ too.

Multiplication of (8) with $x^i$ and integration with respect to $x$ from 0 to $\infty$ leads to

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\[ \frac{\partial}{\partial t} \int_0^\infty x_i^j N \, dx_i + G_1 \frac{\partial}{\partial v} \int_0^\infty x_i^{j+2} N \, dx_i + 2vG_2 \frac{\partial}{\partial v} \int_0^\infty x_i^{j-1} N \, dx_i = 0. \]

Integrating the term involving \( \partial N/\partial x_i \) by parts we get

\[ G_2 \int_0^\infty x_i^{j-1} \frac{\partial N}{\partial x_i} \, dx_i = -ig_2 \int_0^\infty x_i^{j-1} N \, dx_i. \]

Note that no boundary terms are present due to the Assumptions 1 and 2. We finally obtain the system of moments

\[ \frac{\partial M_i}{\partial t} + G_1 \frac{\partial M_{i+2}}{\partial v} + 2vG_2 \frac{\partial M_{i+1}}{\partial v} + (2 - i)G_2 M_{i-1} = 0. \] \hspace{1cm} (17)

The partial differential Eq. (17) hold for all \( i \in \mathbb{Z} \). However, choosing \( i \) from any finite subset \( S \) of \( \mathbb{Z} \), the resulting system does not

\[ \{ \hat{M}_0 = a, \hat{M}_1 = a\mu, \hat{M}_2 = a(\mu^2 + 3\mu\sigma^2), \hat{M}_3 = a(\mu^4 + 6\mu^2\sigma^2 + 3\sigma^4), \hat{M}_4 = a(\mu^5 + 10\mu^3\sigma^2 + 15\mu\sigma^4) \}. \]

5.2. The system of moments for Problem 4

In Problem 4 the growth rate \( G_1 \) depends linearly on \( t \). If we multiply the population balance (8) with \( x_i^j \) and integrate with respect to \( x_i \) from 0 to \( \infty \), we get the system of moments

\[ \frac{\partial M_i}{\partial t} + \sqrt{2}vG(c) \frac{\partial M_{i-1}}{\partial v} - vG(c) \frac{\partial M_i}{\partial v} + G(c) \frac{\partial M_{i+1}}{\partial v} + \frac{\partial^2}{\partial v^2} G(c) M_i - (2 - i) \sqrt{2} \frac{G}{c^2} M_i = 0. \] \hspace{1cm} (18)

for \( i \in \mathbb{Z} \). As in the case of size-independent growth in Section 5.1, the system of moments is not closed. In Section 7, an approach to approximate its solution is introduced.

6. Closure of the moment equations for size-independent growth using a normal distribution ansatz

In order to close the moment system (17), Briesen (2006) uses the normal distribution ansatz

\[ N(t, v, \xi) \approx N_{ND}(t, v, \xi) = \frac{a(t, v)}{\sigma(t, v) \sqrt{2\pi}} e^{-\frac{[\xi - \mu(t,v)]^2}{2\sigma(t,v)^2}} \]. \hspace{1cm} (19)

The function \( N(t, v, \xi) \) is extended to negative values of \( \xi \):

\[ N_{\xi}(t, v, \xi) = \begin{cases} N(t, v, \xi), & \xi > 0, \\ 0, & \xi \leq 0. \end{cases} \]

We denote by \( \hat{M}_i(t, v) \) the moments of the ansatz function \( N_{ND} \), i.e. \( \hat{M}_i(t, v) := \int_{-\infty}^\infty x_i^j N_{ND}(t, v, \xi) \, d\xi \) for \( i = 0, 1, 2, \ldots \). We define \( k! \) for \( k = -1, 0, 1, \ldots \) in the following way:

\[ k! = \begin{cases} \prod_{t=0}^{k/2} (2t+1), & k \text{ odd}, k > 0, \\ 1, & k = -1 \text{ or } k = 0. \end{cases} \]

The moments \( \hat{M}_i \) can be expressed analytically in \( a, \mu \) and \( \sigma \) as

\[ \hat{M}_i(t, v) = a(t, v) \sum_{k=0}^{i} \binom{i}{k} (k-1)! \mu(t, v)^{i-k} \sigma(t, v)^k, \quad i = 0, 1, 2, \ldots \]

For \( i = 0, 1, 2, 3, 4, 5 \) the moments \( \hat{M}_i \) are given by

\[ \hat{M}_0 = a(\mu^2 + \sigma^2)^2, \quad \hat{M}_1 = a(\mu^2 + \sigma^2), \quad \hat{M}_2 = a(\mu^4 + 6\mu^2\sigma^2 + 3\sigma^4), \quad \hat{M}_3 = a(\mu^5 + 10\mu^3\sigma^2 + 15\mu\sigma^4). \] \hspace{1cm} (20)

We consider the Eq. (17) for \( i = 1, 2, 3 \) and replace the moments \( M_i \) by the moments \( \hat{M}_i \) of the normal distribution ansatz \( N_{ND} \). Using (20) one gets a system of three equations of advection-reaction type, namely

\[ \frac{\partial}{\partial t} a + \sqrt{2}vG(c) \frac{\partial a}{\partial v} - vG(c) \frac{\partial a}{\partial v} + G(c) \frac{\partial a}{\partial v} + \frac{\partial^2}{\partial v^2} G(c) a - (2 - i) \sqrt{2} \frac{G}{c^2} a = 0 \]

for the three unknown functions \( a, \mu \) and \( \sigma \) of \( (t, v) \). With \( U = (a, \mu, \sigma)^T \) this system can be written in matrix form as

\[ A(U)v + B(U, v) \frac{\partial U}{\partial v} + C(U) = 0 \] \hspace{1cm} (22)

where the entries of \( A \), \( B \) and \( C \) are easily computed from (21).

The advection-reaction system (22) is solved by a second order splitting scheme, which is described in detail in Supplementary Material, Section A. The scheme consists of a second order upwind scheme for the advection part (cf. Toro (1998)) and a second order Runge-Kutta method for the source term. Initial and boundary conditions needed in the numerical scheme are computed from the initial and boundary moments \( M_0, M_1 \) and \( M_2 \): If \( u(t, v_*) = (a(t, v_*), \mu(t, v_*), \sigma(t, v_*))^T \) is needed at a point \( (t, v_*) \) of the initial or boundary conditions, it is calculated by

\[ a(t, v_*) = M_0(t, v_*), \quad \mu(t, v_*) = M_1(t, v_*)/M_0(t, v_*), \quad \sigma(t, v_*) = \sqrt{M_0(t, v_*)M_2(t, v_*) - M_1^2(t, v_*)/M_0(t, v_*)}. \] \hspace{1cm} (23)

The moments \( M_0(t, v_*), \quad i = 0, 1, 2, \) are derived from known initial and boundary values of the crystal size distribution \( n \) by numerical quadrature. The expressions in (23) are only defined if \( M_0(t, v_*) > 0 \) and \( M_0(t, v_*)M_2(t, v_*) > M_1^2(t, v_*)/M_0(t, v_*) \) holds. The first inequality is violated if the function \( n \) is zero initially or along a boundary; it can be avoided by adding a negligible positive perturbation to \( n \). The second inequality was always fulfilled in the numerical computations considered in this article. If the solution \( U = (a, \mu, \sigma)^T \) has been computed with the numerical scheme, the moments \( M_i \) can be derived by (20).
It can be expected that, as long as the function $N$ is well approximated by the normal distribution ansatz (19), the moments $\bar{M}_i$ are also good approximations of the moments $M_i$. In Briesen (2006) such an example is numerically investigated, and Problem 3 defined in Section 4 is similar to Briesen’s example. However, if the normal distribution ansatz is a rather poor approximation to the function $N$, it turns out that this closure method does not give acceptable results. This will be illustrated with the numerical examples in Section 8. For problems far from normal distribution, as e.g. Problem 2 in Section 4, we alternatively propose a moment closure using the quadrature method of moments. This approach will be described in the next section.

7. Closure of the moment equations using the quadrature method of moments (QMOM)

The quadrature method of moments was introduced by McGraw (1997). In this method the ODE system of moments is closed using Gaussian quadrature. An algorithm for the inversion of moments needs to be carried out in every time step of the numerical solver. A modification of this method that avoids the moment inversion during the computation was given by Motz (2003). Here the system of moments is replaced by an ODE system for the abscissae and weights of the Gaussian quadrature rule. The idea of directly tracking abscissae and weights instead of the moments is also the basis of the direct quadrature method of moments (DQMOM) of Marchisio and Fox (2005). The quadrature method of moments was originally given for one-dimensional population balances, describing abscissae and weights of the Gaussian quadrature rule. The idea of directly tracking abscissae and weights instead of the moments is also the basis of the direct quadrature method of moments (DQMOM) of Marchisio and Fox (2005). The quadrature method of moments was originally given for one-dimensional population balances, describing crystals that are only characterized by a single length coordinate $l$. There exist a number of higher-dimensional extensions of this method, the first one given by Wright et al. (2001). Other such extensions were presented by Rosner and Pykkonen (2002), Yoon and McGraw (2004), and Marchisio and Fox (2005). However, all these higher-dimensional QMOM approaches deal with moments that are averaged over all crystal size coordinates and only depend on time. In contrast to these approaches, our method is able to compute moments of the CSD that still depend on the crystal volume. Therefore, it is still possible to evaluate a crystal volume distribution using our method. Regarding this aspect, our approach is

while for Problem 4 with moment system (18) we have $I_1 = \{-1, 0, 2\}$ and $I_2 = \{-1, 0\}$ with

$$a_{-1} = 2\sqrt{2}G_2, \quad a_2 = G_1, \quad b_{-1} = -G_2, \quad d_{-1} = 2G_2$$

$$a_{-1} = \sqrt{2\pi}G(c), \quad a_0 = -\sqrt{\pi}G(c), \quad a_2 = G(c)$$

and

$$b_{-1} = 0, \quad b_0 = 0, \quad d_{-1} = \sqrt{2\pi}G(c), \quad d_0 = -\frac{G(c)}{\sqrt{\pi}}.$$ 

The quadrature method of moments is based on Gaussian quadrature rules. For a given number of quadrature points $n_{QP} \in \mathbb{N}$ the abscissae $x_k(t, v)$ and weights $w_k(t, v)$ of a Gaussian quadrature rule satisfy

$$M_i(t, v) = \sum_{k=1}^{n_{QP}} w_k(t, v)x_k(t, v), \quad i = 0, \ldots, 2n_{QP} - 1.$$ 

with the $x_k^{(P1)}(v)$ from (16). The system (25) can be solved using the PD algorithm of Gordon (1968), see also McGraw (1997). This algorithm is included in Supplementary Material, Section B. The Gaussian quadrature rule approximates an integral of the form

$$\int_0^\infty f(x)N(t, v, x)dx$$ 

by the finite sum

$$\sum_{k=1}^{n_{QP}} f(x_k(t, v))w_k(t, v).$$ 

In particular, the moments $M_i(t, v)$ for arbitrary $i \in \mathbb{Z}$ are approximated by $\sum_{k=1}^{n_{QP}} x_k(t, v)w_k(t, v)$. By (25), the moments $M_i(t, v)$ for $i = 0, 1, \ldots, 2n_{QP} - 1$ are evaluated exactly by the quadrature rule. For all other moments the quadrature is in general not exact.

In the quadrature method of moments, the moments $\bar{M}_i$ in system (24) are formally replaced by the corresponding Gaussian quadrature expressions. In addition, system (24) is only considered for $i = 0, 1, \ldots, 2n_{QP} - 1$. The resulting system is

$$\sum_{k=1}^{n_{QP}} \tilde{x}_k \frac{\partial \tilde{w}_k}{\partial t} + \left( \sum_{j \in I_1} a_{j,k} \frac{\partial \tilde{w}_k}{\partial v} + \sum_{j \in I_2} b_{j,k} \tilde{w}_k \right) \tilde{w}_k = 0$$ 

for $i = 0, 1, \ldots, 2n_{QP} - 1$. Here we denote the abscissa by $\tilde{x}_k$ and the weights by $\tilde{w}_k$ in order to distinguish them from the abscissae and weights defined in (25) since, in general, they are not equal. The reason is that the moments $\bar{M}_i$ in (24) are replaced by quadrature expressions that are only approximations. We introduce the $2n_{QP} \times n_{QP}$ matrices

$$P = (p_{ik}), \quad Q = (q_{ik})$$

and the $n_{QP}$-dimensional row vectors

$$r = (r_1, \ldots, r_{n_{QP}}), \quad s = (s_1, \ldots, s_{n_{QP}}),$$

whose elements are given by

$$p_{ik} = \tilde{x}_k \frac{\partial \tilde{w}_k}{\partial t} + \left( \sum_{j \in I_1} a_{j,k} \frac{\partial \tilde{w}_k}{\partial v} + \sum_{j \in I_2} b_{j,k} \tilde{w}_k \right) \tilde{w}_k,$$

for $i = 0, 1, \ldots, 2n_{QP} - 1$. In the approach of Motz mentioned above.

The systems of moments (17) and (18) can be written in the general form

$$\frac{\partial M_i}{\partial t} + \sum_{j \in I_1} a_{j,k} \frac{\partial M_{i+j}}{\partial v} + \sum_{j \in I_2} b_{j,k} M_{i+j} = 0.$$

where $I_1, I_2 \subseteq \mathbb{Z}$ are certain index sets. In particular, in the case of size-independent growth with moment system (17) we have $I_1 = \{-1, 2\}$ and $I_2 = \{-1\}$ with

$$a_{-1} = 2\sqrt{2}G_2, \quad a_2 = G_1, \quad b_{-1} = -G_2, \quad d_{-1} = 2G_2$$

for $i = 0, 1, \ldots, 2n_{QP} - 1, k = 1, \ldots, n_{QP}$. The systems of moments (17) and (18) can be written in the general form

$$\frac{\partial M_i}{\partial t} + \sum_{j \in I_1} a_{j,k} \frac{\partial M_{i+j}}{\partial v} + \sum_{j \in I_2} b_{j,k} M_{i+j} = 0.$$
Then the system (28) can be written in matrix form as 
\[(P|Q)(\mathbf{r}|s)^T = 0.\] Thus, the matrix \((P|Q)\) is regular provided the \(\hat{s}_k\) as well as the \(\hat{w}_k\) are pairwise different. So the system (28) is almost everywhere equivalent to the system \((\mathbf{r}|s) = 0\) which we are going to solve now.

\begin{align*}
\frac{\partial \hat{s}_k}{\partial t} + \left( \sum_{j \in I_1} a_{kj}^2 \hat{s}_j \right) \frac{\partial \hat{s}_k}{\partial v} + \sum_{j \in I_2} b_{kj} \hat{s}_{j+1} &= 0, \\
\frac{\partial \hat{w}_k}{\partial t} + \left( \sum_{j \in I_1} j a_{kj}^2 \hat{s}_j \right) \frac{\partial \hat{w}_k}{\partial v} + \left( \sum_{j \in I_1} a_{kj}^2 \right) \frac{\partial \hat{w}_k}{\partial v} + \sum_{j \in I_2} d_{kj} \hat{w}_j &= 0
\end{align*}
(29a)
\( (29b) \)

for \(k = 1, \ldots, n_{QP}\). It is a system of \(2n_{QP}\) equations of advection-reaction type in the \(2n_{QP}\) unknown functions \(\hat{s}_k(t, v)\) and \(\hat{w}_k(t, v)\) for \(k = 1, \ldots, n_{QP}\). If we set
\[
\mathbf{u} = (\hat{s}_1, \ldots, \hat{s}_{n_{QP}}, \hat{w}_1, \ldots, \hat{w}_{n_{QP}})^T,
\]
\[
\mathbf{B} = \begin{pmatrix} B_1 & 0 \\ B_2 & B_1 \end{pmatrix}, \quad B_1 = \text{diag} \left( \sum_{j \in I_1} a_{kj}^2 \right), \quad B_2 = \text{diag} \left( \sum_{j \in I_1} a_{kj}^2 \right)
\]
and
\[
\mathbf{c} = (g_1, \ldots, g_{n_{QP}}, h_1, \ldots, h_{n_{QP}})^T, \quad g_k = \sum_{j \in I_2} b_{kj}, \quad h_k = \sum_{j \in I_2} d_{kj}
\]
we can write system (29a) in matrix form as
\[
\frac{\partial \mathbf{u}}{\partial t} + \mathbf{B} \mathbf{u} \cdot \mathbf{v} \frac{\partial \mathbf{u}}{\partial v} + \mathbf{c} \mathbf{u} = 0.
\]
(29c)

The PDE system (29a) is equipped with initial and boundary conditions. With
\[
\Gamma = \{(0) \times [v_0, v_{\text{max}}]\} \cup \{(0, \infty) \times \{0\}\}
\]
the initial and boundary conditions are given by
\[
\hat{s}_k(t_0, v_i) = s_k(t_0, v_i), \quad \hat{w}_k(t_0, v_i) = w_k(t_0, v_i)
\]
(30)
for \((t_0, v_i) \in \Gamma\) and \(k = 1, \ldots, n_{QP}\) where the initial and boundary values \(s_k(t_0, v_i)\) and \(w_k(t_0, v_i)\) are defined by
\[
M_j(t_0, v_i) = \sum_{k=1}^{n_{QP}} s_k(t_0, v_i) \hat{s}_k(t_0, v_i), \quad i = 0, \ldots, 2n_{QP} - 1,
\]
see (25). They can be calculated from the moments
\[
M_0(t_0, v_i), \ldots, M_{2n_{QP}-1}(t_0, v_i)
\]
using the PD algorithm (Gordon, 1968; McGraw, 1997) – see Section B in Supplementary Material of this article. The moments \(M_j(t_0, v_i)\) are derived from the known initial and boundary values of the crystal size distribution \(n(t, l_1, l_2)\) by numerical quadrature.

If we assume that for the Problems 1–4, the initial and boundary conditions for \(\hat{s}_k\) are given by smooth functions and that the solution \(\hat{s}_k(t, v)\) of (29c) is smooth for \((t, v) \in [0, t_{\text{max}}] \times [v_0, v_{\text{max}}] =: \Omega_v\), then advection is directed always towards increasing \(v\). This result will be proved in Section D in Supplementary Material. It follows that the boundary conditions (30) are set correctly since at \(v = v_0\) there is an inflow boundary and at \(v = v_{\text{max}}\) there is an outflow boundary.

Remark 7.1. In some of the numerical applications studied in Section 8 the PD algorithm returns a zero or negative abscissa \(s_k(t_*, v_*) \leq 0\) due to numerical error. Then the advection matrix \(B\) is not defined since it contains negative powers of \(s_k\). We have reported the occurrence of such problems in Section 8.1.

Remark 7.2. The system (29c) is solved numerically using the same second order splitting scheme as for (22), see Section A in Supplementary Material of this article. The solution of system (29a) is used to approximate the moments \(M_j(t, v)\) defined in (5) by the expression
\[
\hat{M}_j(t, v) = \sum_{k=1}^{n_{QP}} s_k(t, v) \hat{s}_k(t, v), \quad i \in \mathbb{Z}.
\]
(31)

Remark 7.3. If Problem 4 is treated, the coefficients \(a_i, b_i, d_i\) in (29a) depend on the concentration \(c\) and the \(g_i\) in (42) do depend on \(c\) too. Since the exact concentration is not known, we work with the estimate
\[
\tilde{c}(t) = c(0) - \frac{\partial \text{mol}}{V_{\text{react}}} \int_{v_0}^{v_{\text{max}}} v(\hat{M}_0(t, v) - M_0(0, v)) \, dv
\]
which we get by replacing \(M_0(t, v)\) with \(\hat{M}_0(t, v)\) in (14). The integral in (32) is evaluated by quadrature.

In Problem 4, the CSD and therefore all moments \(M_i\) are zero at the inflow boundary. With zero moments it is not possible to evaluate the boundary condition of system (29a). We overcome this problem by adding a negligibly small positive perturbation to the CSD at the inflow boundary:
\[
\hat{n}(t, l_1, l_2) = 10^{-8} \left(1 - e^{-t(l_1 - 2l_2)^2}\right)
\]
where the constants \(a\) and \(b\) are the same as in (15).
Table 1
Parameters used in the simulation of Problems 1–4.

<table>
<thead>
<tr>
<th>Problem</th>
<th>problem parameters</th>
<th>method parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$G_1 = G_2 = 1$; $t_{\text{max}} = 1$; $v_0 = 0.1$; $v_{\text{max}} = 1$; $\delta = 2.5$</td>
<td>$\Delta v = 0.005, 0.007, 0.01, 0.014, 0.02$; $n_{QP} = 2, \ldots, 8$</td>
</tr>
<tr>
<td>2</td>
<td>$G_1 = G_2 = 1$; $t_{\text{max}} = 1$; $v_0 = 0.1$; $v_{\text{max}} = 1$; $\delta = 2.5$; $t_1^* = t_2^* = 1$</td>
<td>$\Delta v = 0.005, 0.007, 0.01, 0.014, 0.02$; $n_{QP} = 2, \ldots, 8$</td>
</tr>
<tr>
<td>3</td>
<td>$G_1 = G_2 = 1$; $t_{\text{max}} = 0.4$; $v_0 = 0.1$; $v_{\text{max}} = 1$; $\alpha = 50$; $\beta = 0.1$; $\nu = 0.1$</td>
<td>$\Delta v = 0.005, 0.007, 0.01, 0.014, 0.02$; $n_{QP} = 2, \ldots, 8$</td>
</tr>
<tr>
<td>4</td>
<td>$t_{\text{max}} = 400$ s</td>
<td>$\Delta v = 2.5 \times 10^{-12}$ m$^3$; $n_{QP} = 2, \ldots, 8$</td>
</tr>
</tbody>
</table>

Fig. 2. Problem 1, $\delta = 2$: error $\text{err}(\hat{M}_0)$ (left) and $\text{err}(\hat{M}_1)$ (right) over mesh size $\Delta v$ for the moment closure methods ND and Q2, Q3, Q7. The results for methods Q4–Q6 are omitted to enhance visibility, they are very close to Q7.

Fig. 3. Problem 1, $\delta = 5$: error $\text{err}(\hat{M}_0)$ (left) and $\text{err}(\hat{M}_1)$ (right) over mesh size $\Delta v$ for the moment closure methods ND and Q2, Q6. The results for methods Q3–Q5 are omitted to enhance visibility, they are very close to Q6.

8.1. Numerical problems

The numerical methods described in this article were not applicable if one of the following problems occurred:

- In the advection scheme, i.e. in Algorithm 1, Step 1 given in Supplementary Material, the matrix $C_{1/2}$ used therein has non-real eigenvalues, or not a full-rank eigenspace, see Remark A.1 in Supplementary Material. In this case, the numerical scheme is not applicable.
- The PD algorithm breaks down due to division by zero.
- The PD algorithm produces a non-positive abscissa $\hat{m}_i \leq 0$. See Remark 7.1.

We report in Table 2 the model problems and parameter values where we have observed such difficulties.

**Result.** If the ND method was used for Problem 2, the advection scheme was in some cases not applicable. If QMOM was applied, the PD algorithm broke down and negative abscissae occurred in some cases for $n_{QP} \geq 6$, and always for $n_{QP} = 8$. In general it can be observed that algorithmic problems increase with the number of quadrature points.

8.2. Comparison of closure methods for size-independent growth

In this section we compare the accuracy of the methods ND and QMOM using as examples the Problems 1–3 given in Section 4. The problem and method parameters are chosen as in Table 1. The accuracy of the numerical solution is measured by computing a relative error of the moments $M_i$ that is defined in the following way:

$$
\text{err}(\hat{M}_i) = \frac{\int_0^{t_{\text{max}}} \int_{v_0}^{v_{\text{max}}} |\hat{M}_i(t, v) - M_i(t, v)| \, dv \, dt}{\int_0^{t_{\text{max}}} \int_{v_0}^{v_{\text{max}}} |\hat{M}_i(t, v)| \, dv \, dt}, \quad i = 0, 1.
$$

Table 2
Numerical difficulties with Problems 1–4 for all $\Delta v$ (except for the case explicitly stated).

<table>
<thead>
<tr>
<th>Problem</th>
<th>$\delta$</th>
<th>ND</th>
<th>Q6</th>
<th>Q7</th>
<th>Q8</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>2</td>
<td>0</td>
<td></td>
<td></td>
<td>PDbreak</td>
</tr>
<tr>
<td>2</td>
<td>5</td>
<td></td>
<td></td>
<td></td>
<td>PDbreak</td>
</tr>
<tr>
<td>2</td>
<td>2</td>
<td>ADVbreak</td>
<td></td>
<td></td>
<td>PDbreak</td>
</tr>
<tr>
<td>2</td>
<td>5</td>
<td>ADVbreak for $\Delta v = 0.02$</td>
<td></td>
<td></td>
<td>PDbreak</td>
</tr>
<tr>
<td>3</td>
<td></td>
<td>absc $\leq 0$</td>
<td>PDbreak</td>
<td>PDbreak</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td></td>
<td>absc $\leq 0$</td>
<td>PDbreak</td>
<td>PDbreak</td>
<td></td>
</tr>
</tbody>
</table>

Legend: ADVbreak – advection scheme breakdown; PDbreak – PD algorithm breakdown, absc $\leq 0$ – abscissa $\leq 0$. 
In Figs. 2–6 we plot the error err(\(\tilde{M}_0\)) and err(\(\tilde{M}_1\)) over \(\Delta v\). Note that no result could be displayed if one of the numerical errors reported in Table 2 occurred. Fig. 7 shows for Problem 2 (with \(\delta = 5\)) the exact and numerically obtained moment \(M_0\) at time \(t = 0.3\).

**Results.**

**Problem 1:** The results for Problem 1 are shown in Figs. 2 and 3.

The initial condition of Problem 1 is a normal distribution with respect to the coordinates \(l_1\) and \(l_2\). However, it is **not** a normal distribution with respect to \(x\) along the lines \(v = \text{const}\). Therefore, the normal distribution ansatz of Briesen’s method ND is not exactly fulfilled, even not at time \(t = 0\). The method ND shows acceptable results for \(\delta = 5\) (see Fig. 3), but it is not convergent for \(\delta = 2\) (see Fig. 2). However, also for \(\delta = 5\) the methods Q3–Q6 give better results than ND (Fig. 3). The method Q2 has a rather irregular convergence behaviour. For some mesh sizes \(\Delta v\), Q2 approximates the moment \(M_0\) unexpectedly well, i.e. better than the QMOM methods with higher \(n_{QP}\) (Fig. 2, left). This seems to be an error cancellation
effect. The QMOM methods for \( n_{QP} \geq 3 \) converge smoothly as the mesh size \( \Delta v \) is decreased, but the theoretical second order convergence of the upwind scheme is not achieved. For \( \delta = 2 \), Q7 is the most accurate method (Fig. 2) and should be used if one is interested in highly accurate results. In the case of \( \delta = 5 \), the methods Q3 – Q6 have nearly the same accuracy (Fig. 3). Therefore it seems to be sufficient to use Q3 in this case.

Problem 2: The results for Problem 1 are shown in Figs. 4 and 5. The solution of this problem has a double peak, and the normal distribution ansatz of the ND method is a poor approximation in this case. As expected, the ND method is not well suited to solve this problem. For \( \delta = 2 \), the ND method leads to a system which cannot be solved by the upwind scheme. Therefore, results for the ND method are not shown in Fig. 4. For \( \delta = 5 \), the system of the ND method can be used with the advection scheme except for \( \Delta v = 0.02 \), but the accuracy is very poor, see Fig. 5. The method Q2 is divergent as \( \Delta v \) gets small (see Figs. 4 and 5, left), but the QMOM methods with higher \( n_{QP} \) converge. The order of convergence in \( \Delta v \) increases with increasing \( n_{QP} \), but second order convergence is again not reached. As expected, Q7 is the most accurate method. As an example, we show the moments \( M_0 \) and \( \bar{M}_0 \) at time \( t = 0.3 \) for Problem 2 with \( \delta = 5 \) in Fig. 7.

Problem 3: See Fig. 6 for results with Problem 3. The solution of this problem is initially a normal distribution with respect to \( x \) along the lines \( v = \text{const} \). This problem is well suited for the ND method and for the QMOM methods with \( n_{QP} \leq 5 \). For higher \( n_{QP} \) some algorithmic problems occurred, see Section 8.1. ND and Q2–Q5 are second order convergent with respect to the mesh size \( \Delta v \). For fixed \( \Delta v \), ND and Q2–Q5 have nearly the same accuracy.

8.3. Direction-dependent growth of barium sulphate crystals

The quadrature method of moments has been applied to Problem 4 that models the direction-dependent growth of barium sulphate crystals. For this problem an exact solution is not known. Therefore we compare the results of our QMOM based closure method with a reference solution of the CSD \( \hat{n} \) that has been calculated numerically using a discretization of the two-dimensional population balance (12) with a high resolution finite volume scheme. This scheme is described in Supplementary Material, Section. The scheme has been taken from Gunawan et al. (2002). From the reference solution \( \hat{n} \) we compute the moments \( M_i \). In the QMOM simulations we have used the parameters given in Table 1.

Remark 8.1. In the QMOM computations of Problem 4 the lengths \( l_1 \) and \( l_2 \) as well as the crystal size distribution \( n \) need to be scaled in order to prevent the occurrence of extremely high or low numeric values which would severely damage accuracy. With constants \( \bar{l} \) and \( \bar{n} \) we introduce the scaled variables \( \bar{l} \) and \( \bar{n} \) which should be of moderate order of magnitude. Thus Eq. (2) transforms to

\[
\frac{\partial \bar{n}(t, \bar{l}_1, \bar{l}_2)}{\partial t} + \frac{1}{\bar{l}_1} \frac{\partial G_1(\bar{l}_1, \bar{l}_2) \bar{n}(t, \bar{l}_1, \bar{l}_2)}{\partial \bar{l}_1} + \frac{1}{\bar{l}_2} \frac{\partial G_2(\bar{l}_1, \bar{l}_2) \bar{n}(t, \bar{l}_1, \bar{l}_2)}{\partial \bar{l}_2} = 0.
\]

which is then solved with the quadrature method of moments as described in Section 7. Of course, domain, initial condition, boundary condition, moments, etc. have also to be scaled in the obvious way. In the example of Problem 4 we have used \( l = 10^{-6} \) m and \( \bar{n} = 10^{25} \) m\(^{-2}\) for scaling.
In Figs. 9 and 10 we compare the moments expected, the estimated error with a good agreement with the solution obtained from FV2D. As especially at the peak of the solution, while Q3–Q5 are in quite good agreement with the concentration obtained from FV2D and approximation $\tilde{M}_i$ obtained with Q2, Q5. Results for Q3, Q4 are omitted to enhance visibility. Right figure shows zoom into left figure, with Q3, Q4 included.

In Fig. 8 we present the initial CSD, see (15), and the CSD at time $t = t_{\text{max}} = 400$ s obtained with the finite volume scheme. In Figs. 9 and 10 we compare the moments $M_i(t_{\text{max}}, v)$, $i = 0, 1$ obtained from the reference solution with the moments $\tilde{M}_i(t_{\text{max}}, v)$ calculated from the QMOM based moment closure method. As $\Delta L = 5 \times 10^{-9}$ m has been chosen so small that the graphs for FV2D would not visibly change if $\Delta L$ were decreased any further. The relative error of the moments $\tilde{M}_i$ can only be estimated by comparison with the moments $M_i$ of the reference solution. We define the estimated relative error

$$\eta(\tilde{M}_i, M_i) = \frac{\int_{v_{\text{min}}}^{v_{\text{max}}} |\tilde{M}_i(t, v) - M_i(t, v)| \, dv}{\int_{v_{\text{min}}}^{v_{\text{max}}} M_i(t, v) \, dv}.$$ 

Fig. 11 (right) shows the estimated relative error $\eta(\tilde{M}_i, t_{\text{max}})$ for $i = 0, 1, 2, 3$ and $n_{\text{QP}} = 2, 3, 4, 5$. The transformation (4b) of the CSD was defined such that $N(t, v, \bar{N}) = x^{s-2} \tilde{N}(t, l_1, l_2)$, where $s \in \mathbb{Z}$ is a parameter. Note that $s = 0$ gives the original transformation. We now construct a dimension reduction method along the lines of Section 7 which is based on $\bar{N}$ instead of $N$. The moments of $\bar{N}$ are $\bar{M}_i(t, v) = \int_0^\infty \bar{N}(t, v, x) \, dx = M_i(t, v)$, and we replace the QMOM ansatz (25) by $\bar{M}_i(t, v) = \sum_{k=0}^{n_{\text{QP}}} \hat{q}_i(t, v) \bar{w}_k(t, v)$. The system (29a) changes to

$$\frac{\partial \hat{w}_k}{\partial t} + \sum_{j \in I_1} q_{i,j} \hat{w}_k - \sum_{j \in I_2} b_j \hat{w}_{k+1} = 0, \quad \frac{\partial \bar{w}_k}{\partial t} + \sum_{j \in I_1} q_{i,j} \bar{w}_k + \sum_{j \in I_2} a_j \bar{w}_k = 0,$$

where $a_j$ and $b_j$ are parameters. Note that

$$\eta(\bar{M}, 400 \text{ s})$$

for $i = 0, 1, 2, 3$ and Q2–Q5.
where the coefficients \(a_i, b_i,\) and \(d_i\) are defined as in Section 7. Initial and boundary conditions are calculated from the moments \(M_0, \ldots, M_{2\text{drug}−1}\). From the numerical solution of this system we construct the moments \(\bar{M}_i\) according to \(\bar{M}_i = \sum_{n=0}^{\infty} \bar{Z}_n(t,v)^i \bar{w}(t,v)\). The numerical error is defined as in (33). For Problem 1, \(\delta = 5\) and for Problem 3, Fig. 12 shows how the numerical error depends on the parameter \(s \in \mathbb{Z}\).

**Result.** The investigated accuracy of \(\bar{M}_0\) depends strongly on the parameter \(s\). There is little accuracy if \(s\) is either very large or very small. The “optimum” value of \(s\), which gives the highest accuracy, depends on the problem and on the number of quadrature points \(n_{\text{QP}}\). Since the initial and boundary conditions of system (34) are derived from the moments \(M_0, \ldots, M_{2\text{drug}−1}\), the moment \(M_0\) is initially and at the inflow boundary exact if \(s \in \mathbb{Z}\) is in the interval \([1−2n_{\text{QP}}, 0]\). It is remarkable that for Problem 1, \(\delta = 5\), the optimal \(s\) is outside this interval for Q3–Q6. This means that here a choice of \(s\) that introduces initial and boundary error of \(M_0\) leads to better results for \(M_0\) than a choice of \(s\) without initial and boundary error of \(\bar{M}_0\). This unexpected result is due to an error cancellation effect. For Problem 3, the optimal parameter was \(s = -2\) for all methods. In this case \(\bar{M}_0\) has indeed no initial and boundary error. The optimum value of \(s\) depends strongly on the particular problem and the numerical methods used. Based only on the few numerical examples we have studied, we are not able to give general rules regarding the optimal choice of \(s\). Of course, many other transformations of the CSD would also be possible. The intention of this section is only to indicate by a simple example that the accuracy might be improved by a transformation of the CSD. A detailed error analysis would be required in order to decide which kind of transformation could be a suited to reduce the numerical error.

### 9. Conclusion

We have proposed a QMOM based dimension reduction method for the numerical simulation of direction-dependent crystallization processes. The method reduces a two-dimensional population balance to a small system of advection equations. The method is able to retain the volume-dependence of the crystal distribution. It was shown to perform well for a number of model problems. Good convergence was observed also for problems that are not well-suited for the normal distribution method of Brück, especially for problems with bimodal crystal size distribution. The method could be extended to cover additional processes like crystal nucleation. It would also be a valuable extension to consider the flow of crystals in a reactor that is not ideally mixed. For this purpose, the Eq. (29c) need to be coupled with a CFD code.

### Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.compchemeng.2010.06.012.

### References


