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MODEL ORDER REDUCTION OF A CONTINUOUS CRYSTALLIZER

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Abstract. Crystallization is one of the most important processes in chemical and pharmaceutical industry, as a large amount of chemical and pharmaceutical agents are produced in crystalline form. The behaviour of industrial crystallizers is often characterised by an intense interaction between fluid flow and particle formation. To improve the operation of such systems requires a deepened understanding of the interaction. This can be obtained from detailed physical process models. Models of adequate complexity comprise up to three external (spatial) coordinates and several internal (property) coordinates. The numerical solution of such models is challenging. To enable model based process design and process control of systems with particle populations in fluid flow, there is a need for reduced models. The reduced models should be of considerably lower system order than the original physical models. Their numerical solution should be much easier and faster. On the other hand, the reduced models should be able to reproduce the system behaviour with sufficient accuracy in the relevant window of operation conditions and in a relevant range of system parameters.

In this contribution, the reduced model is obtained from the original model by projection on a space of problem specific basis functions. The basis functions are computed from snapshot solutions of the original model in a defined range of model parameters. They are chosen such that they are able to reproduce the snapshots accurately in the chosen parameter domain. By Galerkin approximation, a reduction in terms of system order by a factor of about 500 is achieved. However, the reduction in terms of computational time is not so good, when POD alone is applied. The speed-up of the reduced model is only by a factor of 5. Therefore as a second component of the reduction procedure, the best point interpolation method is used. This accelerates the reduced model considerably without a noticeable loss of accuracy. The simulation of the final reduced model is about 100-200times faster than the original model.

1 Introduction

Crystallization is one of the most important processes in chemical and pharmaceutical industry, as a large amount of chemical and pharmaceutical agents are produced in crystalline form. Furthermore, crystallization is an intermediate step used for purification in many chemical production processes.

The behaviour of industrial crystallizers is often characterised by an intense interaction between fluid flow and particle formation. To improve the operation of such systems requires a deepened understanding of the interaction. This can be obtained from detailed physical process models. Models of adequate complexity comprise up to three external (spatial) coordinates and several internal (property) coordinates. The numerical solution of such models is challenging. To date, there is no chance to solve crystallizer models with several internal and external coordinates in real time. Furthermore, the complexity of these models prohibits their direct application to methods of advanced process control. To enable model based process design and process control of systems with particle populations in fluid flow, there is a need for reduced models. The reduced models should be of considerably lower system order than the original physical models. Their numerical solution should be much easier and faster. On the other hand, the reduced models should be able to reproduce the system behaviour with sufficient accuracy in the relevant window of operation conditions and in a relevant range of system parameters.

The model reduction process consists of lumping in the external coordinates as well as in the internal coordinates. For both sets of coordinates reduction techniques are available in the literature. The most widely used class of methods to reduce internal coordinates are generalised moment methods, see e.g. Marchisio et al.⁹, Grosch et al.³ For the external coordinates, reduced-basis approximation^{11,4} or Proper Orthogonal Decomposition (POD)¹² has been applied successfully. The basic idea of this method is to approximate the model solution by a linear combination of time independent basis functions weighted by time dependent coefficients. The basis functions are constructed from numerical simulation results of a detailed reference model. The reduced model consists of ordinary differential equations for the time dependent coefficients.

While model reduction by POD is widely used in complex fluid dynamics^{5,8}, it has hardly been applied to population balance systems, so far. One reason may be that the nonlinear terms in the Navier Stokes equations can be handled rather easily by classical POD methods that result to quadratic polynomial terms in the reduced model. Crystallizer models describing particle formation in fluid flow contain more general nonlinear terms whose efficient treatment requires special reduction techniques. As a new aspect in this work, the best point interpolation method by Nguyen et al.¹⁰ is introduced for the reduction of a crystallizer model. The reduction process will be applied to the model of laboratory crystallizer for the production of urea. The reference model will be presented in the next section. The model reduction strategy is discussed in Section 3. Section 4 compares simulation results of the reduced model and the reference model.



Figure 1: Simplified 2D geometry of the crystallizer (cross-sectional view)

2 Reference model

A tube shaped crystallizer with a rectangular cross sectional area is considered. A schematic view is given in Figure 1.

The inlet of the crystallizer is on the left-hand side, where a supersaturated ureaethanol solution with seeding urea crystals enters the pipe reactor. The seeding crystals grow inside the reactor. Under the chosen operation conditions, nucleation, agglomeration and breakage hardly play a role. At the product outlet on the right-hand side of the reactor, a mixture of liquid and crystals is continuously removed. To model the process a simplified two-dimensional geometry is used that presents an axial cross-section of the reactor. A further simplifying assumption used in the reference model is that the mixture of liquid and particle phase can be seen as a quasi-homogeneous fluid with constant physical properties like density, viscosity and heat conductivity. This is justified, if the particle concentration is sufficiently small.

2.1 Model equations

The crystallizer's fluid phase is described by the incompressible Navier-Stokes equations. The fluid flow is considered to be at steady state, because previous simulations showed that the dynamics of the fluid flow are much faster than those of the energy balance and the population balance. In addition, it is assumed that there is a narrow channel on the left-hand side of the inlet that is not presented in the model space domain. This is in agreement with our experimental setup and allows to treat the inlet velocity as a fully developed laminar flow. The continuity equation and the momentum balance are expressed by the following system of partial differential equations:

$$\nabla \cdot \mathbf{u} = 0 \qquad \text{in } \Omega, \qquad (1)$$
$$\rho(\mathbf{u} \cdot \nabla)\mathbf{u} = -\nabla p + \mu_{\mathrm{E}} \Delta \mathbf{u} \qquad \text{in } \Omega,$$

where $\mathbf{u} = \{u, v\}$ is the vector of flow velocities in $\mathbf{x} = \{x, y\}$ directions, p is the pressure, $\mu_{\rm E}$ and ρ are the fluid viscosity and the fluid density, respectively.

The energy balance defines a differential equation for the temperature T that reads

$$\rho c_p \left(\frac{\partial T}{\partial t} + \mathbf{u} \cdot \nabla T \right) = \lambda_{\rm E} \Delta T + \Delta H_{\rm cryst} \cdot h_{\rm gr}, \qquad (2)$$

where c_p and λ_E are the heat capacity and the thermal conductivity of ethanol, and ΔH_{cryst} is the heat of solution.

A component mass balance for the solute gives the following convection-diffusion equation for the solute partial density ρ_c :

$$\frac{\partial \rho_c}{\partial t} + \mathbf{u} \cdot \nabla \rho_c = D_c \Delta \rho_c + h_{\rm gr},\tag{3}$$

where D_c is the diffusion coefficient. The term $h_{\rm gr}$ stands for the mass transfer between fluid phase and particle phase due to crystal growth. It can be written as

$$h_{\rm gr}(T,\rho_c,f) = -3\rho_d k_V G(T,\rho_c) \int_0^\infty L^2 f dL, \qquad (4)$$

where ρ_d is the density of the crystals, G is the growth rate of crystals, and f is the number density function of the crystal population.

The dispersed particle phase is modeled by a population balance for the particle size distribution that accounts for crystal growth and convective transport of crystals in space:

$$\frac{\partial f}{\partial t} + \mathbf{u} \cdot \nabla f + G \frac{\partial f}{\partial L} = D_f \Delta f, \tag{5}$$

here D_f is an artificial diffusion coefficient of particles in the solute which describes a movement of the particles relative to the fluid flow. It is mainly introduced to increase numerical robustness. The particle size distribution f depends on time and space and in addition on the particle size coordinate L.

A power function is used for the growth rate:

$$G = \begin{cases} k_g \sigma(T, \rho_c)^g, & \sigma > 0, \\ 0, & \sigma \le 0, \end{cases}$$
(6)

where σ is the super-saturation defined as

$$\sigma(T, \rho_c) = \frac{\rho_c - \rho_{c,sat}(T)}{\rho_{c,sat}(T)} \,. \tag{7}$$

An empirical expression for the saturation density

$$\rho_{c,sat}(T) = 1.3045(T - 273.15) + 35.3642 \tag{8}$$

based on experimental results⁷ has been chosen.

A simplified description of the particle phase follows if the size distribution function is replaced by its moments μ_k defined as

$$\mu_k = \int_0^\infty L^k f dL \quad \text{for } k = 0, 1, \dots$$
(9)

Inserting the definition (9) into the population balance (5) results in the following closed equation system for the first moments:

$$\dot{\mu}_0 + \mathbf{u} \cdot \nabla \mu_0 = D_f \Delta \mu_0 \dot{\mu}_k + \mathbf{u} \cdot \nabla \mu_k = D_f \Delta \mu_k + k \cdot G \mu_{k-1}, \quad 1 \le k \le 3.$$

$$(10)$$

An obvious physical interpretation can be given for the moments μ_k : μ_0 denotes the total number of crystals at one point in space, the ratio μ_1/μ_0 is a measure for the average crystals characteristic size, and μ_3/μ_0 is the average crystals volume.

The use of moments reduces the rate of mass consumption due to crystal growth $h_{\rm gr}$ to

$$h_{\rm gr}(T,\rho_c,\mu_2) = -3\rho_d k_V G(T,\rho_c)\mu_2.$$
(11)

2.2 Boundary and initial conditions

Boundary conditions in space subject to the geometry in Figure 1 are as follows. The boundary Γ_{wall} has the no slip flow field condition with an isolation of ρ_c and f. The temperature on the upper and lower boundaries is space independent and specified by a parameter T_{wall} . The inlet Γ_{in} of width $w_{\text{in}} = 3.3 \text{ mm}$ has a parabolic velocity profile $\mathbf{u} = \{u_{\text{in}}(1 - (2y/w_{\text{in}})^2), 0\}^T$ for $-w_{\text{in}}/2 \leq y \leq w_{\text{in}}/2$ with the maximal inlet speed u_{in} , equals 1 cm/s. The inlet liquid concentration of the solute at Γ_{in} is given by $\rho_c = \rho_{c,\text{in}}$ with the inflow temperature T_{in} . Further, the feed contains particle seeds of negligible size. That is, we assume $L \to 0$ for all feed particles. This leads to the boundary condition $f(L = 0) = f_0^{in} = 10^{10} \text{ 1/m}^3$. The second boundary condition for the internal coordinate is a regularity condition $f(L \to \infty) = 0$. The wall boundary condition for near the inlet is similar to Γ_{wall} except the homogeneous Neumann condition for the temperature. The outlet presents the outflow boundary condition of the fluid with the zero reference pressure, and convective outflow of both continuous $\mathbf{n} \cdot \nabla \rho_c = 0$ and disperse $\mathbf{n} \cdot \nabla f = 0$ phases with the normal vector \mathbf{n} to the boundary.

The initial conditions are zero concentration and particle size distribution at the initial temperature $T_0 = 18^{\circ}$ C.

2.3 Simulation results

The finite volume CFD tool openFOAM⁶ is used to simulate the described crystallizer model. A complete list of model parameters is given in Krasnyk et al.⁷

The numerical simulation shows that the system reaches a steady-state after about 2000 seconds of the simulation time. Figure 2 illustrates the development of the average crystal size μ_1/μ_0 in time by some snapshots. One can see that initially crystals with nonzero length exist only close to the inlet on the left-hand side. The seeding crystals fed to the system move with the fluid flow. At the end of the simulation, the largest crystals are found near the walls at the outlet because the crystals in this area have the highest residence time in the reactor.



Figure 2: Development in time of the average crystal size μ_1/μ_0 with $u_{\rm in} = 1$ cm/s (bright shading corresponds to high values)

3 Model reduction by Proper Orthogonal Decomposition

As a first step of the model reduction procedure, the temperature T, the concentration c, and the moments $\mu_{0..3}$ are approximated by the following expressions:

$$T \approx \sum_{i=1}^{N_{\rm b}^{\rho_{\rm c}}} \varphi_i^T(t) \psi_i^T(\mathbf{x}) + T_{\rm in} \psi^{T_{\rm in}}(\mathbf{x}) + T_{\rm wall} \psi^{T_{\rm wall}}(\mathbf{x}),$$

$$\rho_c \approx \sum_{i=1}^{N_{\rm b}^{\rho_c}} \varphi_i^{\rho_c}(t) \psi_i^{\rho_c}(\mathbf{x}) + \rho_{c,\rm in} \psi^{\rho_{c,\rm in}}(\mathbf{x}),$$

$$\mu_k \approx \sum_{i=1}^{N_{\rm b}^{\mu_k}} \varphi_i^{\mu_k}(t) \psi_i^{\mu_k}(\mathbf{x}) + \mu_{k,in} \psi^{\mu_{k,in}}(\mathbf{x}), \quad k = 0, \dots, 3$$
(12)

The first part of the approximations in (12) consists of time-dependent coefficients φ and space-dependent modes ψ that represent orthonormal bases in space. The remaining terms on the right-hand side of (12) are introduced in order to make the bases ψ independent of the inlet and ambient conditions, as was suggested in Graham et al.² In this work, control functions $\psi^{T_{wall}}$ and $\psi^{T_{in}}$ are zero everywhere in Ω except unity on the corresponding boundaries Γ_{wall} and Γ_{in} , respectively. The control functions $\psi^{\rho_{c,in}}$ and $\psi^{\mu_{k,in}}$ can be chosen as steady-state solutions of the reference model normalized with corresponding parameter values at Γ_{in} .

In the case of a dynamic flow field, one could approximate the flow velocities u and v in a similar way. However, the quasi-stationarity assumptions for the flow make this unnecessary and lead to a trivial formulation of \mathbf{u} with an empty basis set and one control function that corresponds to a fluid flow steady-state solution. For the sake of simplicity, further in text \mathbf{u} denotes such a steady-state solution of the fluid flow.

To simplify notation a bit, some abbreviations will be introduced:

$$\Phi^{T} := \left(\varphi_{1}^{T}, \dots, \varphi_{N_{b}^{T}}^{T}, T_{in}, T_{wall}\right), \qquad \Psi^{T} := \left(\psi_{1}^{T}, \dots, \psi_{N_{b}^{T}}^{T}, \psi^{T_{in}}, \psi^{T_{wall}}\right),
\Phi^{\rho_{c}} := \left(\varphi_{1}^{\rho_{c}}, \dots, \varphi_{N_{b}^{\rho_{c}}}^{\rho_{c}}, \rho_{c,in}\right), \qquad \Psi^{\rho_{c}} := \left(\psi_{1}^{\rho_{c}}, \dots, \psi_{N_{b}^{\rho_{c}}}^{\rho_{c}}, \psi^{\rho_{c,in}}\right),$$

$$\Phi^{\mu_{k}} := \left(\varphi_{1}^{\mu_{k}}, \dots, \varphi_{N_{b}^{\mu_{k}}}^{\mu_{k}}, \mu_{k,in}\right), \qquad \Psi^{\mu_{k}} := \left(\psi_{1}^{\mu_{k}}, \dots, \psi_{N_{b}^{\mu_{k}}}^{\mu_{k}}, \psi^{\mu_{k,in}}\right),$$
(13)

where Φ^l are vectors of $N_{\rm b}^l$ time-dependent coefficients and $N_{\rm c}^l$ model or control parameters $(N_{\rm c}^T = 2, N_{\rm c}^{\rho_c} = N_{\rm c}^{\mu_k} = 1)$, and Ψ^l are corresponding basis and control functions for a variable $l = \{T, \rho_c, \mu_0, \ldots, \mu_3\}$.

Using this notation, the variables in (12) can be presented in the reduced form as

$$T \approx \sum_{i=1}^{N_{\rm b}^T + N_{\rm c}^T} \Phi_i^T \cdot \Psi_i^T, \quad \rho_c \approx \sum_{i=1}^{N_{\rm b}^{\rho_c} + N_{\rm c}^{\rho_c}} \Phi_i^{\rho_c} \cdot \Psi_i^{\rho_c}, \quad \mu_k \approx \sum_{i=1}^{N_{\rm b}^{\mu_k} + N_{\rm c}^{\mu_k}} \Phi_i^{\mu_k} \cdot \Psi_i^{\mu_k}.$$
(14)

3.1 Preliminary equations of the reduced model

The reduced model will be a set of equations for the coefficients φ . To obtain these equations, the approximation (12) is inserted into the equations of the reference model. Galerkin approximation is applied, i.e. the projection of the residuals on the space spanned by the basis functions φ is requested to be zero. In this work, the inlet temperature T_{in} and the wall temperature T_{wall} are assumed as varying in time, while the partial density $\rho_{c,\text{in}}$ and the moments $\mu_{k,in}$ at the inlet are assumed as constant, i.e. $\dot{\rho}_{c,in} = \dot{\mu}_{k,in} = 0$. As a result of the reduction process, one obtains the following low-order ODE model:

$$\rho c_{p} \dot{\varphi}_{i}^{T} = -\rho c_{p} A_{ij}^{T} \Phi_{j}^{T} + \lambda_{E} B_{ij}^{T} \Phi_{j}^{T} + \Delta H_{cryst} \cdot (\psi_{i}^{T}, h_{gr})_{\Omega}, \quad 1 \leq i \leq N_{b}^{l}, \\
-(\psi_{i}^{T}, \psi^{T_{in}})_{\Omega} \cdot \rho c_{p} \dot{T}_{in} - (\psi_{i}^{T}, \psi^{T_{wall}})_{\Omega} \cdot \rho c_{p} \dot{T}_{wall} \quad 1 \leq j \leq N_{b}^{l} + N_{c}^{l}, \\
\dot{\varphi}_{i}^{\rho_{c}} = -A_{ij}^{\rho_{c}} \Phi_{j}^{\rho_{c}} + D_{c} B_{ij}^{\rho_{c}} \Phi_{j}^{\rho_{c}} + (\psi_{i}^{\rho_{c}}, h_{gr})_{\Omega}, \\
\dot{\varphi}_{i}^{\mu_{0}} = -A_{ij}^{\mu_{0}} \Phi_{j}^{\mu_{0}} + D_{f} B_{ij}^{\mu_{0}} \Phi_{j}^{\mu_{0}}, \\
\dot{\varphi}_{i}^{\mu_{k}} = -A_{ij}^{\mu_{k}} \Phi_{j}^{\mu_{k}} + D_{f} B_{ij}^{\mu_{k}} \Phi_{j}^{\mu_{k}} + k \cdot (\psi_{i}^{\mu_{k}}, G\mu_{k-1})_{\Omega}, \quad 1 \leq k \leq 3,$$
(15)

where $(g_1, g_2)_{\Omega}$ denotes an inner product in the space domain Ω :

$$(g_1, g_2)_{\Omega} = \int_{\Omega} g_1 g_2 \, d\mathbf{x} \tag{16}$$

The coefficient matrices in (15) are

$$\begin{aligned}
A_{ij}^{l} &= (\psi_{i}^{l}, \mathbf{u} \cdot \nabla \Psi_{j}^{l})_{\Omega}, & 1 \leq i \leq N_{\mathrm{b}}^{l}, \\
B_{ij}^{l} &= (\psi_{k}^{l}, \Delta \Psi_{j}^{l})_{\Omega}, & 1 \leq j \leq N_{\mathrm{b}}^{l} + N_{\mathrm{c}}^{l}.
\end{aligned} \tag{17}$$

The first terms on the right-hand-side of each equation correspond to the convective parts of the original equations, and the second parts correspond to diffusion. The third terms result from nonlinearities due to crystal growth. Therefore, the reduced model (15) is nonlinear as the original reference model.

One can see from the first equations in (15) that a jump in T_{in} and T_{wall} causes a jump in φ_i^T which may seem surprising at first glance, because the temperature T of course does not jump in such a case. However, a closer look reveals that the jump of φ_i^T exactly compensates the jump of T_{in} and T_{wall} in (12), and the temperature T is still continuous.

While the structure of the reduced model is given by (15), two important questions are still open. The first question is how to choose suitable basis functions Φ that lead to a low order model with a good approximation quality. The second question concerns the efficient treatment of the inner products in the reduced model, which is crucial in order to obtain a considerable reduction in terms of computation time. Both questions will be discussed in the next two sections.

3.2 Construction of basis functions

The basis functions ψ are obtained from piecewise continuous reference simulations of the detailed model, so-called snapshots, which are taken at different time points and for different values of free model parameters or control parameters. Let ξ denote the set of the time variable t and the control parameters, so in this case $\xi = \{t, T_{in}, T_{wall}\} \in \mathcal{M}$ for some box domain \mathcal{M} . The snapshots used to construct basis functions for the temperature T, the partial density ρ_c and the moments μ_k are written as $z^T(\xi, \mathbf{x}), z^{\rho_c}(\xi, \mathbf{x})$ and $z^{\mu_k}(\xi, \mathbf{x})$, respectively. As mentioned at the beginning of Section 3, "shifted" reference data are used in order to obtain homogeneous boundary conditions for the basis functions, i.e.

$$z^{T}(\xi, \mathbf{x}) = T(\xi, \mathbf{x}) - T_{in}\psi^{T_{in}}(\mathbf{x}) - T_{wall}\psi^{T_{wall}}(\mathbf{x}),$$

$$z^{\rho_{c}}(\xi, \mathbf{x}) = \rho_{c}(\xi, \mathbf{x}) - \rho_{c,in}\psi^{\rho_{c,in}}(\mathbf{x}),$$

$$z^{\mu_{k}}(\xi, \mathbf{x}) = \mu_{k}(\xi, \mathbf{x}) - \mu_{k,in}\psi^{\mu_{k,in}}(\mathbf{x}).$$
(18)

For simplicity, the super-scripts T, ρ_c , and μ_k will be skipped in the further discussion.

The idea of the reduction method is to find such basis functions that a relatively small finite subset of $N_{\rm b}$ basis functions $\{\psi_i\}_1^{Nb}$ will minimize a weighted total error E, defined as a norm of a difference between the reference data and their projection on the space spanned by the basis functions:

$$\min_{\substack{(\psi_i,\psi_j)=\delta_{ij}\\1\le i,j\le N_{\rm b}}} E(\{\psi_i\}) := \frac{1}{V} \int_{\mathcal{M}} ||z - \sum_{i=1}^{N_{\rm b}} (z,\psi_i)_{\Omega} \psi_i||_{\Omega}^2 \,\mathrm{d}\xi,\tag{19}$$

where δ_{ij} is the Kronecker delta, $V := \int_{\mathcal{M}} d\xi$ is a volume of the \mathcal{M} -domain, and the induced norm is $||.||_{\Omega} = (.,.)_{\Omega}^{1/2}$.

One can show⁸ that under the orthonormality constraint, the above optimization problem leads to the following integral equation for the orthonormal solution functions φ_k

$$\frac{1}{V} \int_{\mathcal{M}} C(.,\xi') \varphi_k(\xi') \mathrm{d}\xi' = \lambda_k \varphi_k(.), \quad k = 1, \dots,$$
(20)

where the L^2 -kernel $C(\xi, \xi')$ is defined as

$$C(\xi,\xi') := (z(\xi,.), z(\xi',.))_{\Omega}.$$
(21)

Solving of the eigenvalue problem results in a set of orthonormal eigenfunctions $\{\varphi_k(\xi)\}_1^{N_b}$ with corresponding real positive eigenvalues λ_k . The solution of the optimization problem (19) will be a set of N_b basis functions that are computed as

$$\psi_k(\mathbf{x}) = \frac{1}{\sqrt{\lambda_k}} \frac{1}{V} \int_{\mathcal{M}} z(\xi, \mathbf{x}) \varphi_k(\xi) \mathrm{d}\xi, \quad k = 1, \dots, N_\mathrm{b},$$
(22)

for the dominant $N_{\rm b}$ eigenvalues λ_k and corresponding eigenfunctions φ_k .

For the finite number of snapshots and the corresponding integral approximation in \mathcal{M} , equation (20) transforms to an eigenvalue problem.

3.3 Interpolation of nonlinear terms

Nonlinear terms in the original model result in computationally expensive terms in the reduced model. These are the inner products or integrals over the space domain containing the nonlinearities, or to be precise the expressions $(\psi_i^T, h_{\rm gr})_{\Omega}, (\psi_i^{\rho_c}, h_{\rm gr})_{\Omega}$, and $(\psi_i^{\mu_k}, G\mu_{k-1})_{\Omega}$ in (15). An analytical solution of the integrals is not possible. A solution by numerical quadrature is not satisfactory, as it increases the computation time of the reduced model considerably. There is a need for an efficient approximation method that generates low computational costs during the runtime of the reduced model. Such an approximation may be the best point interpolation by Nguyen et al.¹⁰, which is used here. The method is explained for a general function first and then it is applied to the reduced model.

3.3.1 General method

Let $g(\xi; \mathbf{x})$ by a nonlinear function on a spatial domain $\mathbf{x} \in \Omega$ defined for any parameter $\xi \in \mathcal{M}$. Assume that snapshots of this function are available at N_s discrete values of the parameter vector $\xi_i, i = 1, \ldots, N_s$. Now the objective is to find some approximation based on the available snapshots that reproduces $g(\xi; \mathbf{x})$ quite well but requires evaluation of the nonlinearity only at a few points in space.

As a first step, an orthonormal basis $\{\psi_1^g, \ldots, \psi_{N_b}^g\}$ in space is constructed for g, using the same method as described in the previous section. The result is a reasonable approximation of g for the parameter values ξ_i in the ensemble of snapshots:

$$g(\xi_i; \mathbf{x}) \approx \sum_{n=1}^{N_{\rm b}} \varphi_n(\xi_i) \, \psi_n^g(\mathbf{x}), \quad i = 1, \dots, N_s.$$
(23)

The question is how to obtain a good guess of g for some parameter value ξ that is not in the snapshots. For high-dimensional parameter vectors ξ , this is a non-trivial interpolation problem. A possible solution procedure suggested in Nguyen et al.¹⁰ is as follows. One approximates g at the parameter value ξ of interest as

$$g(\xi; \mathbf{x}) \approx g_{N_{\rm b}}(\xi; \mathbf{x}) = \sum_{n=1}^{N_{\rm b}} \beta_n(\xi) \psi_n^g(x).$$
(24)

Equations for $\beta_n(\xi)$ are obtained by demanding that g_{N_b} matches the original g exactly at N_b interpolation points in space $\{\mathbf{x}_1 \in \Omega, \ldots, \mathbf{x}_{N_b} \in \Omega\}$:

$$\sum_{n=1}^{N_{\rm b}} \psi_n^g(\mathbf{x}_m) \,\beta_n(\xi) \stackrel{!}{=} g(\xi; \mathbf{x}_m), \quad m = 1, \dots, N_{\rm b}$$
(25)

How to choose the interpolation points \mathbf{x}_m is still open. A first requirement is that the set of interpolation points is admissible¹⁰. This means that equation (25) can be solved uniquely for all $\beta_n(\xi)$ or in other words that the left-hand side matrix of (25) is invertible. Further, it is plausible that the quality of the approximation (24) depends on the position of the interpolation points. Therefore, the \mathbf{x}_m should be chosen such that the quadratic error of $g_{N_{\rm b}}$ integrated over the space and the parameter domain is minimized:

$$\min_{x_1,\dots,x_{N_{\mathrm{b}}}} J\left(\mathbf{x}_1,\dots,\mathbf{x}_{N_{\mathrm{b}}}\right) \tag{26}$$

with

$$J(\mathbf{x}_1, \dots, \mathbf{x}_{N_{\mathrm{b}}}) = \int_{\mathcal{M}} \int_{\Omega} \left(g(\xi; \mathbf{x}) - \sum_{n=1}^{N_{\mathrm{b}}} \beta_n(\xi) \psi_n^g(\mathbf{x}) \right)^2 \mathrm{d}\mathbf{x} \mathrm{d}\xi.$$
(27)

In the form (27) the optimization problem for $\{\mathbf{x}_1 \in \Omega, \ldots, \mathbf{x}_{N_b} \in \Omega\}$ is very hard to solve, and simplifications are needed. The first simplification is to replace the integration in the parameter domain by a quadrature rule with N_s function evaluations at the parameter values ξ_i of the snapshots and corresponding weights w_i :

$$J(\mathbf{x}_1, \dots, \mathbf{x}_{N_{\mathrm{b}}}) \approx \sum_{i=1}^{N_{\mathrm{s}}} \int_{\Omega} \left(g(\xi_i; \mathbf{x}) - \sum_{n=1}^{N_{\mathrm{b}}} \beta_n(\xi_i) \psi_n^g(\mathbf{x}) \right)^2 \mathrm{d}\mathbf{x} \cdot w_i.$$
(28)

The second simplification is to substitute $g(\xi_i; \mathbf{x})$ with the approximation (23). Using the orthonormality of the basis functions, one can express the space integral in (28) as

$$\int_{\Omega} \left(g(\xi_i; \mathbf{x}) - \sum_{n=1}^{N_{\rm b}} \beta_n(\xi_i) \psi_n^g(\mathbf{x}) \right)^2 \mathrm{d}\mathbf{x} \approx \int_{\Omega} \left(\sum_{n=1}^{N_{\rm b}} \left(\varphi_n(\xi_i) - \beta_n(\xi_i) \right) \psi_n^g(\mathbf{x}) \right)^2 \mathrm{d}\mathbf{x}$$
$$= \sum_{n=1}^{N_{\rm b}} \left(\varphi_n(\xi_i) - \beta_n(\xi_i) \right)^2. \tag{29}$$

So the final optimization problem for obtaining the interpolation points reads

$$\min_{x_1,\dots,x_{N_{\rm b}}} \sum_{i=1}^{N_{\rm s}} \sum_{n=1}^{N_{\rm b}} \left(\varphi_n(\xi_i) - \beta_n(\xi_i)\right)^2 \cdot w_i, \quad 1 \le m \le N_{\rm b}.$$
(30)

subject to the constraints (25). Note that the optimization problem has to be solved only once to get the position of the interpolation points. It is *not* part of the evaluation of the reduced model.



Figure 3: Optimal position of the interpolation points for the approximate computation of the growth rate G

3.3.2 Application to the reduced crystallizer model

The best point interpolation method is now applied to the nonlinear source terms in (15). The nonlinearities are caused by the growth rate $G(T, \rho_c)$. As in the reduced model T and ρ_c are expressed by basis functions Ψ and weighting coefficients Φ , $G(T, \rho_c)$ is rewritten as

$$G(T,\rho_c) = G\left(\sum_i \Phi_i^T \cdot \Psi_i^T(\mathbf{x}), \sum_j \Phi_j^{\rho_c} \cdot \Psi_j^{\rho_c}(\mathbf{x})\right) = G\left(\Phi_i^T, \Phi_j^{\rho_c}; \mathbf{x}\right) =: g\left(\xi; \mathbf{x}\right).$$
(31)

So in the notation of the previous section, Φ_i^T and $\Phi_j^{\rho_c}$ belong to the parameter vector ξ . The series approximation of G reads

$$G(T, \rho_c) \approx \sum_{n=1}^{N_b^G} \beta_n^G(\Phi_i^T, \Phi_j^{\rho_c}) \cdot \psi_n^G(\mathbf{x}), \qquad (32)$$

with basis functions ψ_n^G obtained as described above. The coefficients β_n^G follow from

$$\sum_{n=1}^{N_{\rm b}^G} \psi_n^G(\mathbf{x}_m) \,\beta_n^G(\xi) \stackrel{!}{=} G(T(\mathbf{x}_m), \rho_c(\mathbf{x}_m)), \quad m = 1, \dots, N_{\rm b}^G, \tag{33}$$

where the optimal location of the interpolation points \mathbf{x}_m is computed offline before solving the reduced model. The position of the interpolation points is visualized in Figure 3. $N_b^G = 21$ points are found to be sufficient for an accurate approximation of the growth rate. The position of the interpolation points corresponds to one of several local minima of the objective function (27). The initial guess for numerical optimization is the first family of Padua points¹. Although the model domain and the initial guess of point positions are symmetrical with respect to y = 0, the optimizer converges to an unsymmetrical solution. This is because symmetrical interpolation points would give the same information and hence are redundant.

The approximation of G can now be used for an efficient evaluation of the inner products

in the reduced model. One obtains

$$\begin{aligned} \left(\psi_i^T, \sigma_{gr}\right)_{\Omega} &= \left(\psi_i^T, -3\rho_d k_V G \mu_2\right)_{\Omega} \\ &\approx -3\rho_d k_V \left(\psi_i^T, \sum_n \beta_n^G \psi_n^G \cdot \sum_j \Phi_j^{\mu_2} \Psi_j^{\mu_2}\right)_{\Omega} \\ &= -3\rho_d k_V \sum_n \sum_j \beta_n^G \Phi_j^{\mu_2} \cdot \left(\psi_i^T, \psi_n^G \cdot \Psi_j^{\mu_2}\right)_{\Omega}, \\ \left(\psi_i^{\rho_c}, \sigma_{gr}\right)_{\Omega} &\approx -3\rho_d k_V \sum_n \sum_j \beta_n^G \Phi_j^{\mu_2} \cdot \left(\psi_i^{\rho_c}, \psi_n^G \cdot \Psi_j^{\mu_2}\right)_{\Omega}, \\ \left(\psi_i^{\mu_k}, G \mu_{k-1}\right)_{\Omega} &\approx \sum_n \sum_j \beta_n^G \Phi_j^{\mu_{k-1}} \cdot \left(\psi_i^{\mu_k}, \psi_n^G \cdot \Psi_j^{\mu_{k-1}}\right)_{\Omega} \end{aligned}$$

with indices $1 \le n \le N_{\rm b}^G$, $1 \le i \le N_{\rm b}^l$ for the corresponding ψ^l , and $1 \le j \le N_{\rm b}^{\mu_k} + N_{\rm c}^{\mu_k}$ for the corresponding Ψ^{μ_k} .

The big advantage of the best point interpolation is that the inner products on the righthand side of (34) depend only on the basis functions. Therefore they can be evaluated during the generation of the reduced model. At run time of the reduced model, no more numerical quadrature is required. Further, only $N_{\rm b}^G$ evaluations of the nonlinear function G are needed to get β_n^G .

3.4 Final equations of the reduced model

The best point interpolation procedure, applied to the nonlinear inner products of the model equations (15), results in the final form of the reduced model

$$\rho c_{p} \dot{\varphi}_{i}^{T} = -\rho c_{p} A_{ij}^{T} \Phi_{j}^{T} + \lambda_{E} B_{ij}^{T} \Phi_{j}^{T} - 3\Delta H_{cryst} \rho_{d} k_{V} \cdot C_{inj}^{T} \beta_{n}^{G} \Phi_{j}^{\mu_{2}} \\
-(\psi_{i}^{T}, \psi^{T_{in}})_{\Omega} \cdot \rho c_{p} \dot{T}_{in} - (\psi_{i}^{T}, \psi^{T_{wall}})_{\Omega} \cdot \rho c_{p} \dot{T}_{wall}, \qquad 1 \leq i \leq N_{b}^{l}, \\
\dot{\varphi}_{i}^{\rho_{c}} = -A_{ij}^{\rho_{c}} \Phi_{j}^{\rho_{c}} + D_{c} B_{ij}^{\rho_{c}} \Phi_{j}^{\rho_{c}} - 3\rho_{d} k_{V} \cdot C_{inj}^{\rho_{c}} \beta_{n}^{G} \Phi_{j}^{\mu_{2}}, \qquad 1 \leq j \leq N_{b}^{l} + N_{c}^{l}, \\
\dot{\varphi}_{i}^{\mu_{0}} = -A_{ij}^{\mu_{0}} \Phi_{j}^{\mu_{0}} + D_{f} B_{ij}^{\rho_{c}} \Phi_{j}^{\mu_{0}}, \qquad 1 \leq n \leq N_{b}^{G}, \\
\dot{\varphi}_{i}^{\mu_{k}} = -A_{ij}^{\mu_{k}} \Phi_{j}^{\mu_{k}} + D_{f} B_{ij}^{\rho_{c}} \Phi_{j}^{\mu_{k}} + k \cdot C_{inj}^{\mu_{k}} \beta_{n}^{G} \Phi_{j}^{\mu_{k-1}}, \qquad 1 \leq k \leq 3. \\
\end{cases}$$
(34)

where the coefficient matrices

$$C_{inj}^{T} = \left(\psi_{i}^{T}, \psi_{n}^{G} \cdot \Psi_{j}^{\mu_{2}}\right)_{\Omega}$$

$$C_{inj}^{\rho_{c}} = \left(\psi_{i}^{\rho_{c}}, \psi_{n}^{G} \cdot \Psi_{j}^{\mu_{2}}\right)_{\Omega}$$

$$C_{inj}^{\mu_{k}} = \left(\psi_{i}^{\mu_{k}}, \psi_{n}^{G} \cdot \Psi_{j}^{\mu_{k-1}}\right)_{\Omega}, \quad 1 \le k \le 3$$

$$(35)$$

are computed in offline. The reduced model is completed by $N_{\rm b}^G$ linear equations (33) for the coefficients β_n^G .

The final reduced model is nonlinear, because the β_n^G depend nonlinearly on φ_i^T and $\varphi_i^{\rho_c}$. The chosen notation makes it a bit hard to see the influence of the control parameters T_{in} and T_{wall} , which are hidden in the vector Φ^T . The control parameters enter the linear as well as the nonlinear terms of the model equations (34). Also time derivatives of the control parameters appear; the origin of this is the use of shifted reference data for the

construction of basis functions for the temperature. If desired, one can make the model equations independent from the time derivatives of the control parameters by choosing the control functions $\psi^{T_{\text{in}}}$ and $\psi^{T_{\text{wall}}}$ such that they are orthogonal to the basis functions ψ_i^T . Such a choice was made in this work.

4 Simulation results of the reduced model

Reference simulations with the detailed model deliver a set of normalized basis functions for the model variables T, ρ_c , μ_0, \ldots, μ_3 . The snapshots are solutions of the detailed model on a non-equidistant time grid for $t = 0, 20, \ldots, 2000s$ and $2100, 2200, \ldots, 5000s$ with wall temperature values $T_{\text{wall}} = 18, 19, \ldots, 28$ °C. The following numbers of basis functions are found to give a good approximation of the snapshots: $N_b^T = 5$, $N_b^{\rho_c} = 40$, $N_b^{\mu_{0..3}} = 20$, and $N_b^G = 21$. Consequently, the size of the reduced order model is 125 ordinary differential equations plus 21 linear algebraic equations compared to 73728 differential equations of the discretized reference model in OpenFOAM.

In a first step, the behaviour of the reduced model is studied for constant time independent wall temperatures. Figure 4 shows a dynamic simulation of the reduced model (34). The diagrams contain results for μ_1 , when different constant values are used for the wall temperature T_{wall} . The solid lines are the first five coefficients φ^{μ_1} and the markers are projected points of the reference solutions on the basis functions ψ^{μ_1} . One can see that at all times the reduced model agrees well with the original model and that at the end of simulation the reduced model settles at the correct steady state. Figure 5 gives more detailed information on the approximation error of the reduced model. It shows time averaged relative errors for different wall temperatures in the interval between 18 and 28 $^{\circ}$ C. The dashed line marks the error of the original reduced model (15), the solid line is the error of the final reduced model with best point interpolation (34). One can see that the reduction error is bounded in time and less then 4% for all variables. The approximation is also very good for values of the wall temperature that are not contained in the snapshots of the reference model. This documents that the reduced model is able to interpolate solutions of the reference model with respect to the wall temperature T_{wall} . Finally, there is hardly a difference in accuracy between the reduced model with numerical quadrature (15) and the reduced model with best point interpolation (34), while the differences in computation time are quite dramatic: the best point interpolation reduced model is at least ten times faster than the original reduced model. In conclusion, the best point interpolation is a valuable improvement of the model reduction procedure.

In the next step, the performance of the reduced model is studied for a time varying wall temperature T_{wall} . This is an important scenario for control applications: The wall temperature can be used to modify the crystal growth and to achieve a desired particle size distribution at the reactor outlet. Therefore, the reduced model should be able to cope well with dynamic changes of T_{wall} . To test the reduced model a time-dependent profile for T_{wall} has been used. The profile is a step change of T_{wall} from 28°C to 18°C at t = 1500s and back to 28°C at t = 3500s. Such a wall temperature pulse causes an appearance of



Figure 4: First five coefficients $\varphi_i^{\mu_1}$ during a dynamic simulation; solid lines denote solutions of the reduced model (34), markers indicate projections of the reference solution on the basis functions



Figure 5: Relative error plots for different values of T_{wall} ; solid lines: reduced model with best point interpolation (34); dashed lines: reduced model without best point interpolation (15)

larger crystals at the outlet for a limited time. Figure 6 presents simulation results of the reduced model (34). To visualize the situation at the outlet, an averaging functional s(.) on the outlet boundary Γ_{out} is introduced:

$$s(.) = \frac{1}{1 \text{ cm}} \int_{\Gamma_{\text{out}}} .\mathrm{d}y.$$
(36)

As before, the solid lines stand for the reduced model, while markers are the reference simulations. From Figure 6 and from the corresponding error plot in Figure 7 one can recognize a good predictive quality of the reduced model for time varying wall temperatures. The agreement with the reference simulation is certainly satisfactory enough to qualify the reduced model for application to controller design. The results confirm the findings of the first step that the reduced model responds well to dynamic changes of control parameters. Results of both simulation scenarios show that the maximal error is bounded in time and time-average relative errors do not exceed 5%.

5 Conclusions

A reduced order model for a crystallizer has been developed by using Proper Orthogonal Decomposition (POD) and best point interpolation. The reduced model is obtained from the original model by projection on a space of problem specific basis functions. The basis functions are computed from snapshot solutions of the original model in a defined range of wall temperatures. They are chosen such that they are able to reproduce the snapshots accurately in this wall temperature range. By Galerkin approximation, a reduction in terms of system order by a factor of about 500 is achieved. However, the reduction in terms of computational time is not so good, when POD alone is applied. The speed-up of the reduced model is only by a factor of 5. Therefore as a second component of the reduction procedure, the best point interpolation method by Nguyen et al.¹⁰ is used. This accelerates the reduced model considerably without a noticeable loss of accuracy. The simulation of the final reduced model is about 100–200 times faster than the original model. These properties qualify the reduced model for applications in the area of process design and process control. Especially for controller design a less accurate model may be sufficient. This would allow the use of a smaller number of basis functions and an even stronger reduction in system order and computation time.

In this work, POD was applied only to the external (spatial) coordinates of the crystallizer, while the internal (property) coordinate was treated by the method of moments. But actually a discrimination between internal and external coordinates is not necessary for model reduction. Therefore, the next step will be to apply the developed reduction methodology simultaneously to internal and external coordinates.

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Figure 6: Dynamic response of the crystallizer to a pulse of the wall temperature T_{wall} ; T_{wall} is shown as a dashed line in the upper left diagram; solid lines denote simulation results of the reduced model (34), markers are simulation results of the reference model; s(.) is an averaging functional on the outlet boundary defined in (36)



Figure 7: Relative error of the reduced model for the pulse input test shown in Figure 6; solid lines: reduced model with best point interpolation (34); dashed lines: reduced model without best point interpolation (15)

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