STATE ESTIMATION IN BATCH CRYSTALLIZATION USING REDUCED POPULATION MODELS

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Abstract: This contribution deals with the design of an observer for state estimation of a batch crystallizer, which is described by a detailed population balance model. Therefore, the rigorous model containing (partial) integro-differential equations is first reduced by applying an integral approximation technique to a model of moments that consists of only a few ordinary differential equations. This reduced model serves then as the basis for the design of a Luenberger type observer. The performance of the observer is finally demonstrated by using the rigorous population balance model for the simulation of the crystallizer plant.

Keywords: state estimation, observer, population balance, model of moments, model reduction, integral approximation

1. INTRODUCTION

In a lot of crystalline product manufacturing applications the product quality is determined by the crystal size distribution. The main difficulty in batch production is thus to accomplish uniform and reproduceable particle size distributions. A suitable model based technique for monitoring each batch with respect to a reference batch is the state estimation by an observer (Ray 1981). Detailed descriptions of particulate processes are usually based on population balances, which generally leads to a complex mathematical model structure. An observer design based on such a population balance model is thus very difficult, if not impossible. Therefore, a model reduction technique will be applied in this contribution, which reduces the model formulation to a system of only a few ordinary differential equations. With the help of this reduced model it will then be possible to apply standard design techniques for a nonlinear observer (Schaffner and Zeitz 1995).
The paper is organized as follows: After a compendious description of the population balance model for the considered batch crystallizer, the applied model reduction technique, which is based on integral approximation, will be illustrated. An observer will then be designed based on the derived reduced model. Simulation results will finally illustrate the performance of the observer.

2. MODELING OF BATCH CRYSTALLIZERS

The batch cooling crystallizer considered in this contribution is a two phase system, which consists of a continuous liquid phase L and a dispersed solid phase S, see Fig. 1. The continuous liquid phase, which contains a binary mixture of *dissolved crystals* and *solvent*, is modeled by material balances. The population of individual crystals within the dispersed solid phase is described us-

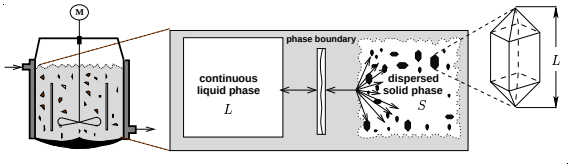

Fig. 1. Decomposition of a batch crystallizer.

ing the population balance approach (Ramkrishna 2000). In order to take the cooled operation of the batch crystallizer on different temperature levels into account, the model is completed by energy balances for the overall crystallizer content and for the coolant inside the cooling jacket.
In the following, the structure of the mathematical model for the considered crystallizer will be shortly described. For more details about the modeling of this type of crystallization processes, the reader is referred to (Gerstlauer *et al.* 2002).

2.1 Modeling of the dispersed solid phase

The application of the population balance approach in order to model the dispersed solid phase S leads to the following population balance equation for the number density function F depending on time t and the characteristic particle length L:

$$\frac{\partial F}{\partial t} = -\frac{\partial (G\,F)}{\partial L} + \dot{F}_{nu}^{+} + \dot{F}_{at}^{\pm} \qquad (1)$$

$$F(L, t=0) = F_{seed}(L)\,; \quad F(L=0, t) = 0\,.$$

The first term on the right hand side of Eq. (1) accounts for crystal growth with the growth rate G. The source due to nucleation is denoted by $\dot{F}_{nu}^{+}$. The term $\dot{F}_{at}^{\pm}$ summarizes all sinks and sources due to attrition of crystals at the stirrer. The initially added seed crystals are denoted by F_{seed}. All considered phenomena are described using detailed kinetic relations. The number of primary nuclei is calculated following Mersmann (1996), considering homogeneous and heterogeneous nucleation. The growth rate

$$\frac{G(\Delta c_{L,A}, L)}{2k_d(L)} = \frac{\Delta c_{L,A}}{c_S} + \frac{k_d(L)}{2k_r c_S} - \sqrt{\left(\frac{k_d(L)}{2k_r c_S}\right)^2 + \frac{k_d(L)}{k_r c_S}\frac{\Delta c_{L,A}}{c_S}} \qquad (2)$$

accounting for integration and diffusion limited crystal growth (Mersmann *et al.* 1992, Gahn and Mersmann 1999) depends on the supersaturation $\Delta c_{L,A}$ and on the size dependent mass transfer coefficient

$$k_d(L) = \frac{D_{AB}}{L}\left[0.8\left(\frac{\bar{\varepsilon} L^4}{\nu_L^3}\right)^{\frac{1}{5}}\left(\frac{\nu_L}{D_{AB}}\right)^{\frac{1}{3}} + 2\right], \qquad (3)$$

where D_{AB} is the diffusion coefficient and $\bar{\varepsilon}$ and ν_L denote the specific energy input and the kinematic viscosity. For the calculation of the attrition rate β and the number N_{frag} and size distribution f_{frag} of abraded fragments a very detailed model is used, which takes the stirrer geometry and the hardness of the crystalline material into account (Gahn and Mersmann 1999). In the population balance (1), this leads to a sink

$$\dot{F}_{at}^{-}(L) = \beta(L) F(L) \qquad (4)$$

due to the crystals that collided with the stirrer, to a source term $\dot{F}_{at,1}^{+}$ accounting for the resulting large crystal with a length $L' - \Delta L(L')$ somewhat smaller than the size L' of the original crystal and to a source term $\dot{F}_{at,2}^{+}$ for all produced fragments:

$$\dot{F}_{at,1}^{+} = \int_L^\infty \delta\left(L - (L' - \Delta L(L'))\right) \dot{F}_{at}^{-}(L')\, dL' \qquad (5)$$

$$\dot{F}_{at,2}^{+} = \int_L^\infty N_{frag}(L') f_{frag}(L, L') \dot{F}_{at}^{-}(L')\, dL'\,. \qquad (6)$$

2.2 Modeling of the continuous liquid phase

The continuous liquid phase L inside the crystallizer consists of two components: the solute and the solvent, i.e. component A and B, respectively. The fundamental balance equations for the liquid phase are thus a balance for the total number of moles n_L

$$\frac{dn_L}{dt} = (a+1)\cdot(-\dot{n}_{nu} - \dot{n}_{gr})\,, \qquad (7)$$

and a component mole balance for the number of moles $n_{L,A}$ of dissolved crystals (component A)

$$\frac{dn_{L,A}}{dt} = -\dot{n}_{nu} - \dot{n}_{gr}\,. \qquad (8)$$

Initial conditions for the balance equations (7) and (8) are given by

$$n_L(t=0) = n_{L,0} \text{ and } n_{L,A}(t=0) = n_{L,A,0}\,.$$

The variable a in Eq. (7) denotes the number of solvent molecules that form a crystal hydrate together with a single molecule of solute. The total molar fluxes $\dot{n}_{nu}$ and $\dot{n}_{gr}$ on the right hand sides of Eqs. (7) and (8) describe the exchange of material between the continuous liquid phase and the dispersed solid phase due to primary nucleation and crystal growth. With the nucleation rate B_{nu} and the growth rate G these molar fluxes are given by

$$\begin{aligned} \dot{n}_{nu} &= k_V \cdot c_S \cdot L_{crit}^3 \cdot B_{nu} \\ \dot{n}_{gr} &= k_V \cdot c_S \int_0^\infty L^3 \frac{\partial (G\,F)}{\partial L}\, dL\,. \end{aligned} \qquad (9)$$

The symbols k_V and c_S in Eqs. 9 denote the volume shape factor of the crystals and their molar density. The minimum size, which nuclei must have to be stable, is characterized by the critical crystal length L_{crit}.

2.3 Energy balances

Besides the population balance (1) and the material balances (7) and (8) energy balances accounting for the overall crystallizer temperature T_{cr} and for the temperatute T_j of the coolant inside the cooling jacket are required to complete the crystallizer model.
With the heat capacity $C_{P,cr}$ of the crystallizer content, the change of the crystallizer temperature with time is determined by

$$C_{P,cr}\frac{dT_{cr}}{dt} = -\Delta h^*_{cr}\,(\dot{n}_{nu}+\dot{n}_{gr}) + J_{cool} + W_{st} \qquad (10)$$

depending on the heat due to crystallization Δh^*_{cr}, the energy exchange J_{cool} with the cooling jacket and the energy dissipation W_{st} by the stirrer. In a similar way, the change of the temperature T_j inside the cooling jacket is given with the heat capacity $C_{P,j}$ of the coolant as

$$C_{P,j}\frac{dT_j}{dt} = c^*_{P,j,in}\dot{n}_{cool}\,(T_{j,in}-T_j) - J_{cool}. \qquad (11)$$

In this equation, the symbol $c^*_{P,j,in}$ denotes the molar heat capacity of the inflowing coolant $\dot{n}_{cool}$ and $T_{j,in}$ is the temperature of the inflowing coolant, which is the only manipulating variable to operate the process. Initial conditions for the energy balances (10) and (11) are given by

$$T_{cr}(t=0) = T_{cr,0} \text{ and } T_j(t=0) = T_{j,0}\,.$$

This concludes the description of the rigorous crystallizer model, which is made up of a partial integro-differential equation (1) for $F(L,t)$ and of four ordinary integro-differential equations (7), (8), (10) and (11) for $n_L(t)$, $n_{L,A}(t)$, $T_{cr}(t)$ and $T_j(t)$. This rigorous model will be the starting point for the following model reduction, and it will be used to validate the subsequently designed observer.

3. MODEL REDUCTION

With the model reduction technique described in this section, the infinite dimensional population balance (1) can be reduced to a set of six ordinary differential equations for the lower order moments

$$\mu_k(t) = \int_0^\infty L^k\,F(L,t)\,dL \quad k=0,\ldots,5 \qquad (12)$$

of the crystal size distribution $F(L,t)$. From a process engineering point of view, this reduction is not really a restriction, since the knowledge of the lower order moments is sufficient for most practical applications, but the great advantage of the resulting reduced model of moments is its model structure, which finally consists of only ten ordinary differential equations.
Such a moment based model reduction has already been applied by Hulburt and Katz (1964), but as mentioned by many authors, the analytical derivation of the moment equations

$$\frac{d\mu_k}{dt} = \int_0^\infty L^k\left(-\frac{\partial\,(GF)}{\partial L} + \dot{F}^+_{nu} + \dot{F}^\pm_{at}\right)dL\,, \qquad (13)$$

which can be derived by differentiating Eq. (12) with respect to time t, leads in general to an unclosed set of ordinary differential equations. In contrast to the analytical derivation (Hulburt and Katz 1964), in this contribution a numerical integral approximation, which is based on Gaussian quadrature rule (McGraw 1997), will be applied to evaluate the integral on the right hand side of Eq. (13). To illustrate this technique, the approximation of the k$^{\text{th}}$ moment yields for example

$$\mu_k(t) = \int_0^\infty L^k\,F(L,t)\,dL \simeq \sum_{i=1}^{n} L_i^k(t)\,w_i(t), \qquad (14)$$

where L_i amd w_i are so called abscissas and weights (Lanczos 1956). Since the sum on the right hand side of Eq. (14) results in the exact value of the integral for $k \le 2n-1$, the time dependent abscissas $L_i(t)$ and weights $w_i(t)$ can be calculated from

$$\begin{aligned} w_1 + w_2 + \ldots + w_n &= \mu_0 \\ L_1w_1 + L_2w_2 + \ldots + L_nw_n &= \mu_1 \\ L_1^2w_1 + L_2^2w_2 + \ldots + L_n^2w_n &= \mu_2 \\ \vdots \qquad\qquad\qquad & \quad \vdots \\ L_1^{2n-1}w_1 + \ldots + L_n^{2n-1}w_n &= \mu_{2n-1}\,. \end{aligned} \qquad (15)$$

For the solution of this *problem of weighted moments* (Lanczos 1956), several methods have been reported, see e.g. (Sack and Donovan 1972). All these methods are based on the fact that the abscissas L_i for the Gaussian quadrature rule can be determined as the zeros of orthogonal polynomials, which can be computed as the eigenvalues $[L_i\boldsymbol{I}-\boldsymbol{J}]=0$ of a tridiagonal matrix

$$\boldsymbol{J} = \begin{pmatrix} \beta_0 & \alpha_0 & 0 \\ \alpha_0 & \beta_1 & \alpha_1 \\ 0 & \alpha_1 & \beta_2 \end{pmatrix} \qquad (16)$$

of size $n\times n$ (3×3 for $k=0,\ldots,5$), where α_i and β_i depend on the moments μ_k. The weights w_i can finally be obtained from the first components of the corresponding eigenvectors v_i

$$w_i = \mu_0 v_i(1)^2 \,. \tag{17}$$

Using the abscissas L_i and the weights w_i, the integrals on the right hand side of Eq. (13) and in Eqs. (9) can thus be approximated by applying

$$\int_0^\infty L^k \, \Phi(\underset{n}{L}) \, F(L,t) \, dL \approx \sum_{i=1} L_i^k(t) \, \Phi(L_i(t)) \, w_i(t) \tag{18}$$

with $\Phi(L)$ being any sufficiently smooth (kinetic) expression.
The application of this model reduction technique leads thus to a reduced model of moments that contains the material balances (7) and (8), the energy balances (10) and (11) and six ordinary differential equations for the first six moments $\mu_0, \ldots, \mu_5$ instead of the population balance (1). The resulting model can thus be formulated as

$$\begin{aligned} \dot{\boldsymbol{x}} &= \boldsymbol{f}(\boldsymbol{x}) + \boldsymbol{g}(\boldsymbol{u}) \,; \;\; \boldsymbol{x}(t=0) = \boldsymbol{x}_0 \\ \boldsymbol{y} &= \boldsymbol{h}(\boldsymbol{x}) \end{aligned} \tag{19}$$

with the state vector,

$$\boldsymbol{x} = \left(n_L \;\; n_{L,A} \;\; T_{cr} \;\; T_j \;\; \mu_0 \;\; \mu_1 \;\; \mu_2 \;\; \mu_3 \;\; \mu_4 \;\; \mu_5 \right)^T, \tag{20}$$

the output vector $\boldsymbol{y}$ and the control vector $\boldsymbol{u}$, which in case of this batch crystallizer only contains the temperature $T_{j,in}$ of the inflowing coolant. This form allows the derivation of a standard Luenberger type observer design (see e.g. in (Schaffner and Zeitz 1995)), which will be subject of the next section.

4. OBSERVER DESIGN

For state estimation, either for process monitoring tasks or with the objective of process control, the entire state of the batch crystallizer in terms of the number of moles n_L and $n_{L,A}$, the temperatures T_{cr} and T_j and the moments $\mu_0 - \mu_5$ of the crystal size distribution has to be reconstructed from available measurements. Nowadays, very efficient sensors are available for temperature measurement, as well as for the online determination of supersaturation, see e.g. the crystallizer setups described in (Miller 1993, Neumann 2001). Besides these sensors, also techniques are available for determining particle size distributions, e.g. from light scattering (MALVERN) or from chord length (LASENTEC) measurements. However, a drawback of these methods is that they work quite well, if additional information about the shape of the crystal size distribution is available, e.g. to calculate the size distribution from a chord length distribution (Ruf *et al.* 2000). But from these measurements values for certain moments can be derived, as e.g. μ_1 from the LASENTEC FBRM (Ruf *et al.* 2000) or μ_2 from the application of the MALVERN sensor (Miller 1993). Therefore, the following approach for an observer design will be based on the availability of either the first moment μ_1 or the second moment μ_2.
In case of the here considered batch crystallizer, the measured supersaturation, which is equivalent with the knowledge of the mole fraction $x_{L,A} = n_{L,A}/nL$, leads directly to the determination of the model states n_L and $n_{L,A}$, since the overall content of material keeps constant throughout the batch time. Moreover, the knowledge of $x_{L,A}$ allows the calculation of the third moment μ_3 using

$$\mu_3 = \frac{1}{k_V c_S} \left[n_{seed} + \frac{1}{1-(1+a)x_{L,A}} \left(n_{L,A,0} - x_{L,A} n_{L,0} \right) \right], \tag{21}$$

with n_{seed} being the number of moles of the seed crystals F_{seed}.
Thus, an output vector $\boldsymbol{y}$ can be defined containing six state variables, which are measured or directly related to measurements

$$\boldsymbol{y} = \left(n_L \;\; n_{L,A} \;\; T_{cr} \;\; T_j \;\; \mu_* \;\; \mu_3 \right)^T \tag{22}$$

with μ_* defining either μ_1 or μ_2. Based on this output vector $\boldsymbol{y}$, a Luenberger type observer (Schaffner and Zeitz 1995) of the form

$$\begin{aligned} \dot{\hat{\boldsymbol{x}}} &= \boldsymbol{f}(\hat{\boldsymbol{x}}) + \boldsymbol{g}(\boldsymbol{u}) + \boldsymbol{L}(\boldsymbol{y} - \hat{\boldsymbol{y}}); \;\; \hat{\boldsymbol{x}}(t=0) = \hat{\boldsymbol{x}}_0 \\ \hat{\boldsymbol{y}} &= \boldsymbol{h}(\hat{\boldsymbol{x}}) \end{aligned} \tag{23}$$

can be established. Here, $\hat{\boldsymbol{x}}$ represents the vector containing the estimated states, $\boldsymbol{f}(\hat{\boldsymbol{x}}) + \boldsymbol{g}(\boldsymbol{u})$ is a copy of the right hand sides of the reduced model, $\boldsymbol{L_{obs}}(\boldsymbol{y} - \hat{\boldsymbol{y}})$ the correction term, and $\hat{\boldsymbol{y}}(\hat{\boldsymbol{x}})$ represents the estimator output vector. The initial values $\hat{\boldsymbol{x}}_0$ for the observer states are given by the initial states $\boldsymbol{x}_0$ of the reduced model of moments, which can be calculated from the initial conditions of the original rigorous population balance model.
For convergence of the estimated states $\hat{\boldsymbol{x}}$ against the states of the plant, the difference $\boldsymbol{y} - \hat{\boldsymbol{y}}$ has to converge to zero. In order to obtain this, a matrix $\boldsymbol{L_{obs}}$ has to be designed. Since the reduced model of moments has due to the involved eigenvalue problem a very complex nonlinear structure, it is not possible to determine $\boldsymbol{L_{obs}}$ by an analytical design method. But from physical considerations the structure of the matrix $\boldsymbol{L_{obs}}$ can be identified for the here considered batch crystallizer as

$$\boldsymbol{L_{obs}} = \begin{pmatrix} X_1^1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & X_2^2 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & X_3^3 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & X_4^4 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & X_5^5 & X_5^6 & X_5^7 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & X_6^8 & X_6^9 & X_6^{10} \end{pmatrix}^T . \tag{24}$$

All the entries $X_{i,j}$ represent adequate gain values, depending on the availability of μ_1 or μ_2. These gain values can be adjusted as constant values following again physical considerations. With this gain matrix $\boldsymbol{L_{obs}}$, the lower order moments $\mu_0 - \mu_2$ are adjusted with the measured μ_1 or μ_2, the higher order moments $\mu_3 - \mu_5$ by μ_3. Due to the ratio between the moments, this leads to typical gain values of e.g. $X_6^8 = 1 \cdot 10^{-2}$ and $X_6^9 = 1.84 \cdot 10^{-6}$.

As another consequence of the complex nonlinear system used for the observer design, it is not possible to investigate stability properties analytically. Therefore, simulation studies for different disturbed and undisturbed operation modes have to be carried out, in order to verify the applicability of the designed observer.

5. SIMULATION RESULTS

In this section, simulation results will be presented, in order to demonstrate the performance of the observer designed on the basis of the reduced model of moments. Therefore, the crystallizer setup by Miller (1993) will be considered, for which the detailed population balance model has already been validated (Gerstlauer *et al.* 2002). In this setup, a commonly used linear cooling regime for $T_{j,in}(t)$ is applied, as depicted in Fig. 2. All the following investigations are performed by

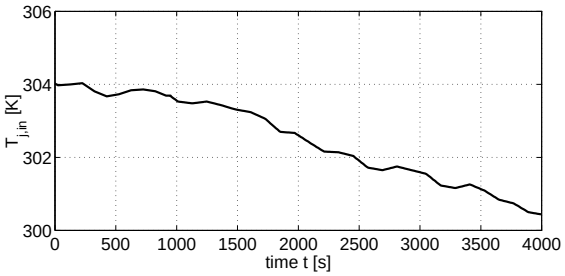

Fig. 2. The applied, approximately linear cooling rate in the crystallizer setup by Miller (1993).

using the rigorous population balance model for the simulation of the batch plant. The population balance is therefore discretized by a Method of Lines approach (Schiesser 1991) using 1000 grid points. For all simulations $10g$ of normally distributed seed crystals (mean: $500\mu m$; deviation $50\mu m$) are used. Both the plant model as well as the observer are solved using a standard ODE solver in MatLab [1] .

The robustness and performance of the observer based on the reduced model of moments and described in Eqs. (23) and (24) will in the following be analyzed by adding disturbances to the programmed cooling profile in Fig. 2. Therefore, the considered batch process will be simulated under rather heavily disturbed cooling profiles as can be seen in Fig. 3. For the further investigation

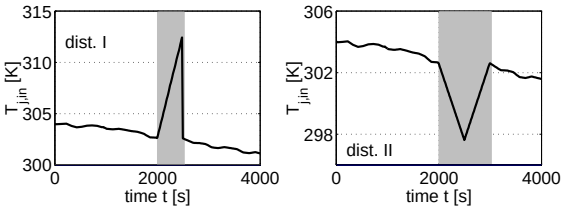

Fig. 3. Applied disturbed cooling profiles.

of the observer behavior, two state estimators will be used, one for the availability of μ_1 and another for the measurement of μ_2. In the following comparisons between the estimated observer states with the states of the rigorous plant model, only those states will be discussed, for which no measurements are available.

In the first case, an observer based on the availability of μ_1 is considered. As can be seen in Fig. 4, this observer achieves very good results for an undisturbed operation of the batch crystallizer as well as for a disturbed cooling profile ('dist I' in Fig. 3). Fig. 4 shows clearly the significant

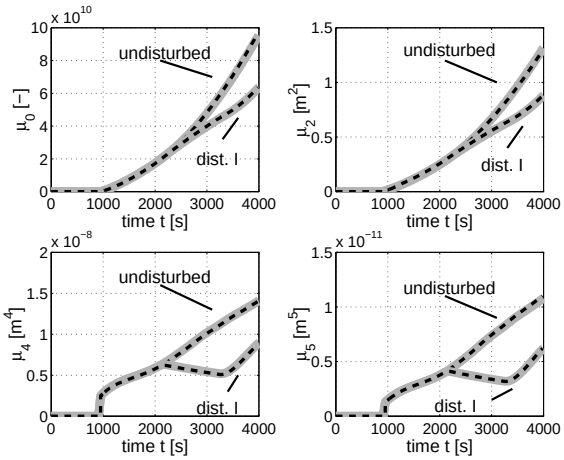

Fig. 4. Comparison of estimated moments μ_0, μ_2, μ_4 and μ_5 (dashed black lines) with the moment values calculated from the rigorous plant model (thick grey lines) for an observer based on the availability of μ_1.

consequences of the applied disturbance. The estimated states (dashed black lines) follow the plant states (thick grey lines) very well, even after the disturbance occured.

The observer considered in the second case, which is based on the availability of μ_2 has some more difficulties to follow the rigorous plant model, as can be seen in Fig. 5. As a consequence of disturbance II the estimated moments μ_0 and μ_1 start slightly drifting away from the plant states at about $2500s$, but due to the correction term $\boldsymbol{L_{obs}}(\boldsymbol{y} - \hat{\boldsymbol{y}})$ in Eq. (23) both μ_0 and μ_1 converge again and reach the plant states again after $3000s$.

[1] MatLab 5.3, The MathWorks Inc., 3 Apple Hill Drive, Natick, MA 01760-2098, USA

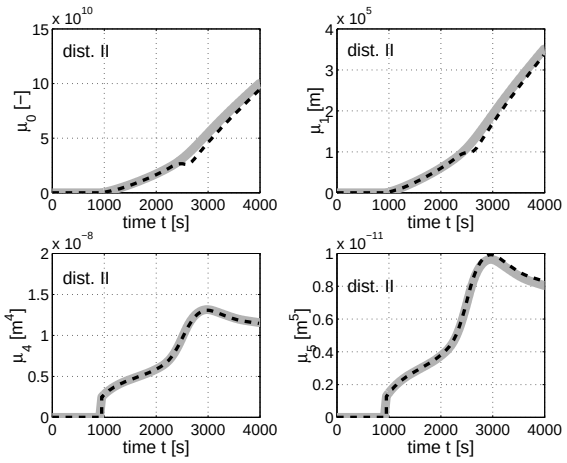

Fig. 5. Comparison of estimated moments μ_0, μ_1, μ_4 and μ_5 (dashed black lines) with the moment values calculated from the rigorous plant model (thick grey lines) for an observer based on the availability of μ_2.

The higher moments μ_4 and μ_5 are in perfect accordance during the whole process.
As depicted in Figs. 4 and 5 observers can be designed that achieve quite good results for both cases, the availability of μ_1 or μ_2.

6. SUMMARY AND CONCLUSIONS

In this contribution, an observer is designed for the state estimation of a batch crystallizer. The rigorous population balance model that describes this crystallization plant consists of (partial) integro-differential equations (Gerstlauer *et al.* 2002). After applying a model reduction technique, which is based on integral approximation using Gaussian quadrature rule (McGraw 1997), this infinite dimensional model can be reduced to a finite dimensional model that consists of only ten ordinary differential equations. On the basis of this reduced model of moments standard Luenberger type observers (Schaffner and Zeitz 1995) can be constructed, based on the availability of measurements of the first or the second moment of the crystal size distribution. As a consequence of the complex nonlinear design model, these observers are designed and tuned on the basis of process knowledge and physical considerations.
In the finally presented results, the rigorous population balance model is used to simulate the undisturbed and disturbed operation of the considered batch crystallizer. For all performed simulation studies, both observers behave very well, which demonstrates the applicability of these observers either for process monitoring or for the objective of process control.

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