

Nonlinear Model-Based Control of a Semi-Industrial Batch Crystallizer Using a Population Balance Modeling Framework

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Abstract—This paper presents an output feedback nonlinear model-based control approach for optimal operation of industrial batch crystallizers. A full population balance model is utilized as the cornerstone of the control approach. The modeling framework allows us to describe the dynamics of a wide range of industrial batch crystallizers. In addition, it facilitates the use of performance objectives expressed in terms of crystal size distribution. The core component of the control approach is an optimal control problem, which is solved by the direct multiple shooting strategy. To ensure the effectiveness of the optimal operating policies in the presence of model imperfections and process uncertainties, the model predictions are adapted on the basis of online measurements using a moving horizon state estimator. The nonlinear model-based control approach is applied to a semi-industrial crystallizer. The simulation results suggest that the feasibility of real-time control of the crystallizer is largely dependent on the discretization coarseness of the population balance model. The control performance can be greatly deteriorated due to inadequate discretization of the population balance equation. This results from structural model imperfection, which is effectively compensated for by using the online measurements to confer an integrating action to the dynamic optimizer. The real-time feasibility of the output feedback control approach is experimentally corroborated for fed-batch evaporative crystallization of ammonium sulphate. It is observed that the use of the control approach leads to a substantial increase, i.e., up to 15%, in the batch crystal content as the product quality is sustained.

Index Terms—Batch crystallization, direct multiple shooting strategy, dynamic optimization, moving horizon estimation, population balance equation (PBE), real-time control.

I. INTRODUCTION

BATCH crystallization is prevalent in the specialty chemical, food, and pharmaceutical industries to separate and to purify high value-added chemical substances. Crystalliza-

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tion processes are governed by several physico-chemical phenomena such as nucleation, crystal growth, and agglomeration, which arise from the co-presence of a continuous phase and a dispersed phase. These kinetic phenomena result in distributed nature of the physical and chemical properties of the manufactured crystals. One of the key characteristics of crystals is their size distribution, providing the critical link between various product quality attributes and the process operating conditions [23].

Despite their extensive use, optimal operation of batch crystallizers is particularly challenging. The difficulties primarily result from the complexity of process models, uncertainties of the crystallization kinetics, sensor limitations in reliably measuring the process variables, and the inherent process uncertainties that may impair the effectiveness of advanced control strategies [3]. In addition, the optimal operation of batch crystallizers is often hampered by the lack of process actuation.

Numerous strategies have been proposed for advanced control of batch crystallizers. These strategies can be broadly categorized into the model-based control approach [6], [16], [25], [26], [34], [37], [45], [47] and the direct design approach [12], [36], [53]. In the former approach, a process model is used to optimally exploit the degrees of freedom of the process to achieve the desired product properties in accordance with a performance objective. A desirable product typically consists in crystals with a large mean size and a narrow size distribution [42]. In recent years, the advent of process analytical technology has led to the emergence of the direct design approach. This approach aims to control the crystallization within the metastable zone bounded by the solubility curve and the metastable limit. In the direct design approach, a supersaturation profile is determined experimentally with the aid of different process analytical techniques. Subsequently, the setpoint profile is tracked in the phase diagram by means of a supersaturation controller, which relies on *in-situ* measurements of process variables. Although the direct design approach circumvents the need for derivation of first-principles models and accurate determination of crystallization kinetics, it only ensures near-optimal operation of the process.

It is self-evident that the cornerstone of any model-based control approach is its dynamic process model, describing the relation between the relevant inputs and outputs of the system. The population balance equation provides a natural framework for mathematical modeling of the evolution of crystal size distribution (CSD) in crystallization processes [17]. The prime difficulty in the synthesis of feedback model-based controllers arises from the distributed nature of the population balance modeling

framework. Finite dimensional approximations of the population balance equation typically lead to high order sets of ordinary differential equations. This often makes real-time implementation of the model-based controllers computationally prohibitive.

Over the past decade, several studies have been carried out to synthesize low-order feedback controllers for real-time control of batch crystallizers [7], [10], [21], [22], [31], [52]. The model-based controllers mostly utilize a computationally affordable nonlinear moment model for online repetitive optimization of certain characteristics of crystal size distribution, e.g., mean crystal size. Shi *et al.* [48], [49] utilized reduced-order moment models to optimize the shape of crystal size distribution by incorporating appropriate constraints expressed in terms of the moments of CSD into the optimization problem. Nonetheless, the complex dynamics of industrial batch crystallizers may not be adequately described by a moment model when the knowledge of full crystal size distribution is required, e.g., crystallizers equipped with fines removal or product classification systems. In addition, the moment model allows merely the use of performance objectives expressed in terms of the moments of CSD that may lead to conservative operation of the batch crystallizer.

With developments in solution methods of the population balance equation, few attempts have recently been made to devise nonlinear feedback controllers that exploit the full population balance equation. Sheikhzadeh *et al.* [46] proposed a model-based approach for real-time control of an anti-solvent batch crystallizer by formulating single- and multiple-objective optimal control problems. An *in-situ* focused beam reflectance measurement (FBRM) probe was utilized in combination with attenuated total reflection-Fourier transform infrared (ATR-FTIR) spectroscopy to provide the dynamic optimizer with online estimations of the crystallization kinetics. This facilitated the feedback implementation of the control approach on a laboratory-scale crystallizer. The experimental results showed that real-time optimization of the process led to improved quality of the crystalline product. Nagy [33] presented a robust control methodology to achieve the desired shape of CSD. The approach combined the concepts of the direct design approach and the model-based control in a hierarchical framework. The optimizer used solute concentration, temperature, and chord length distribution measurements to repeatedly compute the optimal supersaturation profile. This profile was then applied as the setpoint for the supersaturation controller. The solute concentration and chord length distribution measurements were performed by an attenuated total reflection-ultraviolet (ATR-UV) spectrometer and an FBRM probe, respectively. Real-time implementation of the control approach on a laboratory-scale cooling crystallizer indicated that the target CSD could be achieved even in the case of a sudden change in the nucleation rate. Hermanto *et al.* [15] developed a nonlinear model predictive control strategy for batch polymorphic transformation of L-glutamic acid from the metastable α -form to the stable β -form crystals. The authors expected that solution of the full population balance equation would significantly increase the computation time, prohibiting the application of nonlinear programming. Therefore, a more practical nonlinear model predictive control strategy based on the extended predictive self-adaptive control (EPSAC)

was adopted. The EPSAC approximates the nonlinear process variables by iterative linearization around future trajectories so that they converge to the same nonlinear optimal solution [44]. To implement the proposed NMPC strategy, an unscented Kalman filter was utilized to estimate the unmeasurable states. The robustness of the feedback control approach to parameter perturbations was compared to the established control strategies. The simulation results suggested that the nonlinear model predictive control strategy had good overall robustness for different performance objectives.

This paper intends to address the inherent challenges of real-time control of industrial batch crystallizers by developing an output feedback nonlinear model-based control approach. The primary requirement on the control approach is its applicability to a wide range of industrial batch crystallizers. This necessitates the use of the full population balance modeling framework, which allows us to describe the dynamics of diverse crystallization kinetics of any complexity and to incorporate the effect of different actuating mechanisms. In addition, the modeling framework facilitates the use of performance objectives defined in terms of the CSD instead of classical objectives, which may lead to conservative and economically inefficient operation of the crystallizer. The real-time feasibility of the output feedback control approach is experimentally corroborated by implementation on a semi-industrial crystallizer, in which fed-batch evaporative crystallization of ammonium sulphate takes place. To the best of the authors' knowledge to date, this article is one of the first contributions that investigate real-time output feedback control of industrial batch crystallizers using a full population balance modeling framework.

II. POPULATION BALANCE MODELING FRAMEWORK

The dynamic behavior of a solution crystallization process can be described by the population balance equation, along with conservation balance equations and kinetic relations. The population balance equation (PBE) is a nonlinear partial integro-differential equation that provides a deterministic description of the distributed characteristics of the dispersed phase. The PBE describes the integral effect of crystallization kinetics on the dynamic evolution of crystal size distribution.

The one dimensional PBE for a seeded fed-batch crystallization process with nucleation and crystal growth as the dominant physico-chemical phenomena can be expressed as [40]

$$\frac{\partial n(L, t)}{\partial t} + \frac{\partial(G(L, t)n(L, t))}{\partial L} = -\frac{Q_p}{V}n(L, t) \quad (1)$$

with the left boundary condition

$$n(L_0, t) = \frac{B_0(t)}{G(t)|_{L_0}} \quad (2)$$

where n is the number density function ($\#/m^3m$); G is the crystal growth rate (m/s); B_0 is the total nucleation rate ($\#/m^3s$); L is the characteristic crystal size (m); t is the time (s); V is the crystallizer volume (m^3); Q_p is the unclassified product removal flow rate (m^3/s). Note that in the seeded batch runs the injection of seeds in an initially supersaturated solution usually results in a burst of nearly zero sized crystals. Therefore, nucleation is introduced as the left boundary

condition of the PBE to describe the formation of crystals of infinitesimal size.

Numerous models have been proposed in the literature to describe nucleation and crystal growth; see, e.g., [42] for a brief discussion on some of the theories and models. In most control applications, the secondary nucleation rate and the crystal growth rate are described by empirical power-law expressions [4]

$$B_0(t) = k_b \mu_3 S^b \quad (3)$$

$$G(t) = k_g S^g \quad (4)$$

where k_b is the nucleation rate constant ($\#/m^4$); b is the nucleation rate exponent; k_g is the crystal growth rate constant (m/s); g is the crystal growth rate exponent; $S = C - C^*$ is the degree of supersaturation; C is the solute concentration ($k_{g_{\text{solute}}}/k_{g_{\text{solution}}}$); C^* is the saturation concentration at the given operating temperature; μ_i is the i th moment of crystal size distribution defined as

$$\mu_i = \int_{L_0}^{\infty} n(L, t) L^i dL \quad i = 0, 1, \dots, 3. \quad (5)$$

Equations (3) and (4) indicate that the knowledge of the state variable of the continuous phase, namely the solute concentration, is required to solve the PBE. Due to the isothermal operation of the evaporative crystallizer under study, the mass and energy balance equations simplify to a single expression for the solute concentration

$$\frac{dC}{dt} = \frac{Q_p(C_{\text{sat}} - C)}{V} + \frac{3K_v G \mu_2 (k_1 + C)}{1 - K_v \mu_3} + \frac{k_2 H_{\text{in}}}{1 - K_v \mu_3} \quad (6)$$

$$C(t_0) = C_0$$

with the constant coefficients

$$k_1 = \frac{H_v C_{\text{sat}}}{H_v - H_L} \left(\frac{\rho_c}{\rho_L} - 1 + \frac{\rho_L H_L - \rho_c H_c}{\rho_L H_v} \right) - \frac{\rho_c}{\rho_L} \quad (7)$$

$$k_2 = \frac{C_{\text{sat}}}{V \rho_L (H_v - H_L)} \quad (8)$$

where K_v is the crystal volumetric shape factor; H_{in} is the heat input to the crystallizer (kW); ρ_L is the density of saturated solution (kg/m^3); ρ_c is the density of crystals (kg/m^3); H_L , H_c , and H_v are the specific enthalpies of solution, crystal, and vapor phase (kJ/kg), respectively.

It follows from the above analysis that the dynamic evolution of CSD in a batch crystallizer can be described by solving the set of (1)–(8). However, the population balance equation is a hyperbolic partial differential equation, whose numerical solution is often a challenging task. The nonlinear, distributed nature of the PBE has motivated extensive research on specialized numerical techniques allowing for accurate and efficient solution of the PBE; see, e.g., [39] and [8]. Mesbah *et al.* [28] have performed a comparative analysis of the most widely used PBE solution methods in terms of the performance requirements essential for online control applications. The study confirms that the high order finite volume methods [13], [14], [24] in combination with a flux limiting function are well capable of capturing the sharp discontinuities and the steep moving fronts, which are

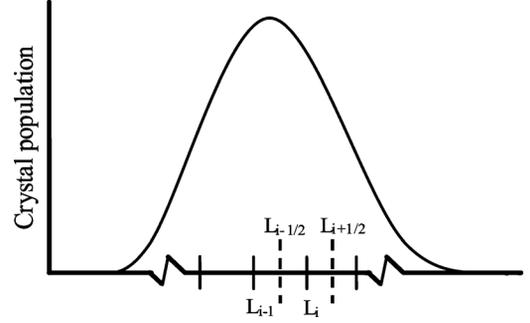


Fig. 1. Cell centered grid mesh.

commonly encountered in simulation of seeded batch crystallizers. In addition, their relatively low computational burden allows us to use a fine grid mesh to further enhance the accuracy of the numerical scheme.

In general, the finite volume methods involve discretization of the spatial variable domain and the use of piecewise functions to approximate the spatial derivatives. By applying these schemes to (1), the hyperbolic partial differential equation reduces to a semi-discrete equation per grid point

$$\frac{\partial n_i}{\partial t} + \frac{1}{\Delta L} \left((Gn)_{L_i^+} - (Gn)_{L_i^-} \right) = -\frac{Q_p}{V} n_i \quad (9)$$

where n_i is the representative value for the number density function in cell i , which is confined to $(L_{i-1/2}, L_{i+1/2})$; see the cell centered grid mesh depicted in Fig. 1. Note that the flux across the inflow boundary, i.e., L_{1-} , need not be approximated since $n_{1-} = B_0/G|_{L_0}$ due to the formation of nuclei of infinitesimal size. To solve (9), crystal fluxes, i.e., Gn , across the cell boundaries should be determined. High order linear or quadratic interpolation schemes such as the piecewise polynomial interpolation formula [38]

$$(Gn)_{L_i^+} = G_{L_i^+} \left(n_i + \frac{1+\kappa}{4} (n_{i+1} - n_i) + \frac{1-\kappa}{4} (n_i - n_{i-1}) \right) \quad (10)$$

$$\kappa \in [-1, 1]$$

can be utilized to approximate the cell-face fluxes. To suppress the oscillatory solutions arisen from the high order interpolation schemes, a flux limiting function should however be exploited to limit the flux across the cell boundaries. The latter function ensures the monotonicity and therefore prevents the occurrence of negative solutions.

In this study, the high order finite volume method with the Van Leer flux limiting function [51] is applied to solve the population balance equation. The dynamics of the seeded batch crystallizer are represented by a set of nonlinear differential algebraic equations of the general form

$$\begin{aligned} \dot{x} &= f(t, x, z, y, u, \theta) & x(t_0) &= x_0 \\ 0 &= g(t, x, z, y, u, \theta) \\ y &= h(t, x, z, y, u, \theta) \end{aligned} \quad (11)$$

where $x(t) = [n(L, t) \quad C(t)]^T$ is the state vector comprised of values of the crystal population in each cell and the solute concentration; $z(t) = [\mu_2(t) \quad \mu_3(t) \quad B_0(t) \quad G(t)]^T$ is the vector

of algebraic variables; $y(t)$ is the vector of measured variables, e.g., crystal size distribution; $u(t) = [H_{in}(t)]$ is the process input; $\theta = [k_b \ b \ k_g \ g]^T$ is the vector of model parameters; f is the vector function of the dynamic state equations; g is the vector function of the algebraic equations; h is the vector function of the measurement equations. Note that the vector x_0 consists of the initial states of the system, namely the seed distribution and the initial solute concentration.

III. NONLINEAR MODEL-BASED CONTROL APPROACH

The formulation of a model-based control approach for optimal operation of batch crystallizers largely depends on the product quality requirements, the properties of the crystallizing system, and the availability of online measurements. The control objectives sought in the operation of batch processes are often conflicting [2]). This necessitates the formulation of an optimal control problem that pushes the process to its most optimal operating regime, while various operational and quality constraints are honored. The optimal control problem to be solved online can be cast as

$$\begin{aligned} \min_{u(t) \in U, t^F} \quad & \Psi(x(t), z(t), u(t), \theta) \\ \text{subject to:} \quad & \text{equation (11)} \\ & \mathcal{H}(x(t), z(t), u(t), \theta) \leq 0 \end{aligned} \quad (12)$$

where Ψ is the performance objective; t^F is the final time; \mathcal{H} is the vector of functions that describe all linear and nonlinear, time-varying or end time algebraic constraints of the system. In its most general form, the performance objective Ψ consists of two parts

$$\begin{aligned} \Psi(x(t), z(t), u(t), \theta) = & \mathcal{M}(x(t^F), z(t^F), \theta) \\ & + \int_t^{t^F} \mathcal{L}(x(\tau), z(\tau), u(\tau), \theta) d\tau \end{aligned} \quad (13)$$

where \mathcal{M} is a terminal cost term and \mathcal{L} is a running cost term. Equation (13) expresses a wide range of performance objectives commonly used in model-based control applications, e.g., set-point tracking, minimization of operation time, etc.

To circumvent performance degradation of the optimal operating policies in the presence of model imperfections and process uncertainties, the nonlinear process model should be continuously updated on the basis of new measurements obtained at each sampling time instant. Hence, the control framework depicted in Fig. 2 is adopted for real-time dynamic optimization of industrial batch crystallizers. In this output feedback control approach, the optimal control problem is solved online in a receding horizon mode [20].

The output feedback structure of the control approach necessitates recursive initialization of the dynamic optimizer at each sampling time instant. This is performed by a nonlinear observer that utilizes the process model, along with online measurements, to construct the state profiles. The state estimation in combination with the feedback structure of the control approach compensates for model imperfections and process uncertainties to a large extent. In addition, the nonlinear state estimator allows us to estimate process variables, which may not be measured online due to various technological or economical limitations. A performance evaluation of several nonlinear observers for output

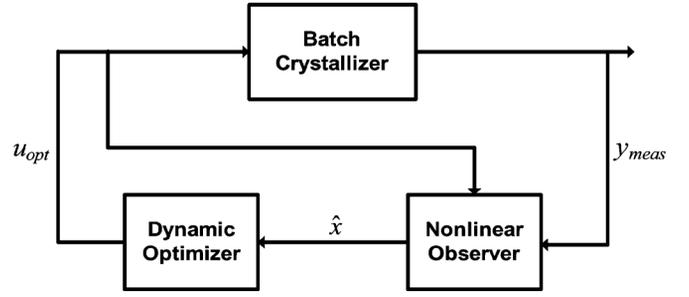


Fig. 2. Output feedback nonlinear model-based control approach.

feedback control of seeded batch crystallization processes can be found in [29].

In this study, a moving horizon estimator (MHE) is exploited for state estimation. The MHE utilizes an optimization-based algorithm, in which the state variables are estimated by solving a minimization problem over a moving and usually fixed-sized time frame [43]. What distinguishes the MHE from the classical estimation techniques, e.g., extended Kalman filtering, is its ability to incorporate physical state constraints into the estimation problem and to use the nonlinear process model without any approximation [41]. The moving horizon estimator can be formulated as

$$\begin{aligned} \min_{x(t-T_{Est})} \quad & \int_{t-T_{Est}}^t \|y_{meas} - y\|_W^2 dt \\ \text{subject to:} \quad & \text{equation (11)} \\ & \mathcal{O}(\hat{x}(t), z(t), u(t), \theta) \leq 0 \end{aligned} \quad (14)$$

where T_{Est} is the estimation horizon; y_{meas} is the vector of measurements; W is the measurement noise matrix; \mathcal{O} is the vector of functions that describe the physical state constraints. Equation (14) suggests that the MHE uses the measurements over time horizon $[t - T_{Est} \ t]$ to reconstruct the state profiles.

The real-time feasibility of the control approach relies on efficient solution of the optimal control and estimation problems. Recently, Mesbah *et al.* [30] have examined the feasibility of different direct optimization strategies for real-time control of the semi-industrial crystallizer considered in this study. It has been shown that the optimization strategies hardly differ in terms of optimal operation of the crystallizer. However, the direct multiple shooting strategy [1], [9] exhibits the highest computational efficiency. The principal idea of this optimization strategy is illustrated in Fig. 3. As can be seen, the state trajectories are discretized over a predetermined number of intervals, which do not necessarily coincide with the discretization points of decision variables. A shooting method is then performed between the successive discretization points. This allows us to convert an infinite dimensional optimization problem into a finite dimensional nonlinear programming (NLP) problem that can be handled by conventional solvers. The model equations and the performance objective are solved independently on the different intervals during each optimization iteration. The solution is obtained on the basis of the current guess of the decision variables and the initial values w_i of the states at the multiple shooting nodes τ_i , which are treated as additional optimization variables. The continuity of the final state trajectories at the

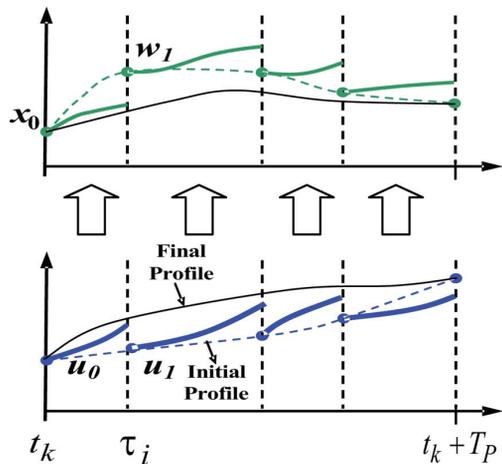


Fig. 3. Illustration of the direct multiple shooting optimization strategy.

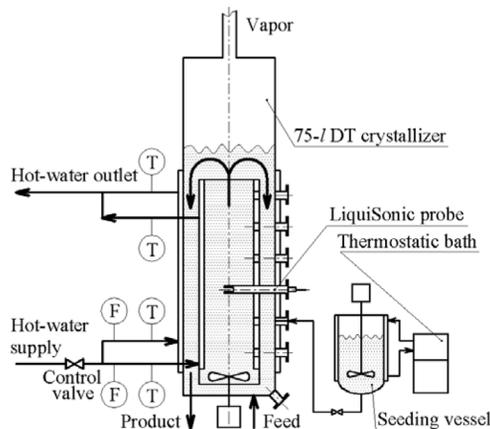


Fig. 4. 75-liter draft tube crystallizer.

end of the optimization is enforced by adding consistency constraints to the NLP problem.

The primary merit of the direct multiple shooting strategy lies in independent solution of the initial value problems on different intervals with a prespecified numerical accuracy. This makes the strategy well suited for parallel computations. The relatively large number of decision variables necessitates the use of tailored NLP algorithms that exploit the special structure of the problem, e.g., sparsity, to yield faster convergence. In this work, the optimal control problem and the moving horizon estimator are implemented in the MATLAB toolbox OptCon [35], [50], which utilizes the large-scale nonlinear optimization solver HQP [11]. The sets of differential algebraic equations on different intervals are solved by the well-known DASPK solver [5].

IV. CASE STUDY: A SEMI-INDUSTRIAL FED-BATCH EVAPORATIVE CRYSTALLIZER

A. Process Description

The nonlinear model-based control approach is employed for optimal operation of a 75-liter draft tube crystallizer, in which seeded fed-batch evaporative crystallization of ammonium sulphate takes place. As shown in Fig. 4, the semi-industrial crystallizer can be considered as a single well-mixed compartment

TABLE I
MODEL PARAMETERS AND PHYSICAL PROPERTIES OF THE AMMONIUM SULPHATE-WATER CRYSTALLIZING SYSTEM

Parameter	Value
b	1.0
C_{sat} , kg _{solute} /kg _{solution}	0.46
g	1.0
H_C , kJ/kg	60.75
H_L , kJ/kg	69.86
H_v , kJ/kg	2.59×10^3
k_b , #/m ³ s	3.84×10^{10}
k_g , m/s	4.91×10^{-5}
K_v	0.43
Q_p , m ³ /s	1.73×10^{-6}
V , m ³	7.50×10^{-2}
ρ_c , kg/m ³	1767.35
ρ_L , kg/m ³	1248.93

with one inlet and two outlet streams. The evaporative crystallization is carried out isothermally at 50 °C. The fed-batch operation is exercised to compensate for losses in the crystallization volume due to the evaporation of solvent, i.e., water, and the slurry sampling. The crystallizer is continuously fed throughout the batch run with a crystal-free feed stream containing saturated ammonium sulphate solution. The outlet flows from the crystallizer include an unclassified product removal stream as well as a vapor stream, which is free from crystal and solute. The small product flow is withdrawn from the crystallizer at regular time intervals every 100 s and diluted with the saturated feed solution for 20 s. The sampling allows us to measure the crystal size distribution online using a laser diffraction instrument (HELOS-Vario, Sympatec, Germany).

Seeding is performed to ensure the reproducibility of batch runs. Ground seeds are prepared by milling and sieving of the commercial product of ammonium sulphate (DSM, The Netherlands) to collect 0.6 kg of the 90–125 μm sieve fraction. Prior to insertion into the crystallizer, the seed crystals are aged in a seeding vessel for one hour in a saturated solution of ammonium sulphate at 50 °C [19]. An in-line concentration measuring probe (LiquiSonic, SensoTech, Germany) is utilized to detect the predetermined supersaturation, at which the seeds are introduced into the crystallizer.

The dynamics of the process at hand are described by the set of differential algebraic equations given in (11). It follows from the process description that the CSD measurements comprise the output vector, i.e., y . On the other hand, the heat input to the crystallizer is the only mechanism to generate supersaturation and therefore is the process input, i.e., u . The seeding allows us to attain a crystal growth dominant operation envelop, in which the crystallization kinetics are most sensitive to the heat input [18]. The model parameters and physical properties of the ammonium sulphate-water crystallizing system are listed in Table I. The adequacy of the model in describing the dynamics of the process under study is demonstrated in [27].

Often, a tradeoff between nucleation and crystal growth is sought in the control of batch crystallizers. For crystallizing systems with a relatively large metastable zone, a crystal growth dominant operation is typically preferred. In such operation, the crystal growth rate largely determines the product characteristics and the batch productivity. However, excessively high

crystal growth rates may adversely affect the product quality due to increased impurity uptake, increased liquid inclusions as well as undesirable attrition and/or agglomeration. The optimal control problem of the semi-industrial batch crystallizer under study is formulated as

$$\begin{aligned} \min_{H_{in}(t)} & \frac{\int_0^{t_b} (100 \frac{G(t) - G_{max}}{G_{max}})^2 dt}{\int_0^{t_b} dt} \\ \text{subject to:} & \text{equation (11)} \\ & H_{min} \leq H_{in}(t) \leq H_{max} \end{aligned} \quad (15)$$

where H_{in} is the parameterized heat input profile; t_b is the fixed batch time; G_{max} is the desired crystal growth rate to circumvent the detrimental effects of excessive crystal growth rates on the product quality, e.g., irregular crystal shapes, impurity uptakes, etc. In (15), the heat input is the manipulated variable exploited to optimize the crystal growth rate profile and consequently control the supersaturation. The heat input's lower bound $H_{min} = 9$ kW is to ensure a high survival efficiency of seeds in the beginning of the batch run, whereas the upper bound of the heat input $H_{max} = 13$ kW is due to heat transfer limitations of the process. Clearly, (15) seeks a tradeoff between the process productivity, i.e., batch crystal content, and the product quality by attaining a desired crystal growth rate throughout the batch run.

B. Simulation Results

The control approach is applied to a plant simulator, which simulates the process using a full population balance (PB) model. The closed-loop control performance is examined in the presence of structural and parametric model imperfections as well as process uncertainties. The following scenarios are considered.

- State feedback control, where the observer is not incorporated in the control approach. Therefore, the online measurements are directly fed to the dynamic optimizer. This scenario is intended to examine the real-time feasibility of dynamic optimization of the population balance model.
- Output feedback control, in which the moving horizon estimator is used in conjunction with the dynamic optimizer. This is to compensate for the detrimental effect of model imperfections and process uncertainties on the optimal operating policies.

In both scenarios, the evolution of crystal size distribution and solute concentration is measured throughout the batch run.

1) *State Feedback Control*: In this scenario, the effect of the complexity of a population balance model in terms of the number of state variables on optimal operation of the semi-industrial batch crystallizer is investigated. The number of state variables of the PB model is determined by the coarseness of the finite volume grid mesh adopted to discretize the population balance equation. Fig. 5 demonstrates the optimal profiles of the state feedback control approach obtained on the basis of PB models with different numbers of state variables; the optimizer's model is identical to that of the plant simulator. The optimizer manipulates the heat input to the crystallizer such that the crystal growth rate is kept at its desired value, i.e., $G_{max} = 2.5 \times 10^{-8}$ m/s. G_{max} is a conservatively chosen

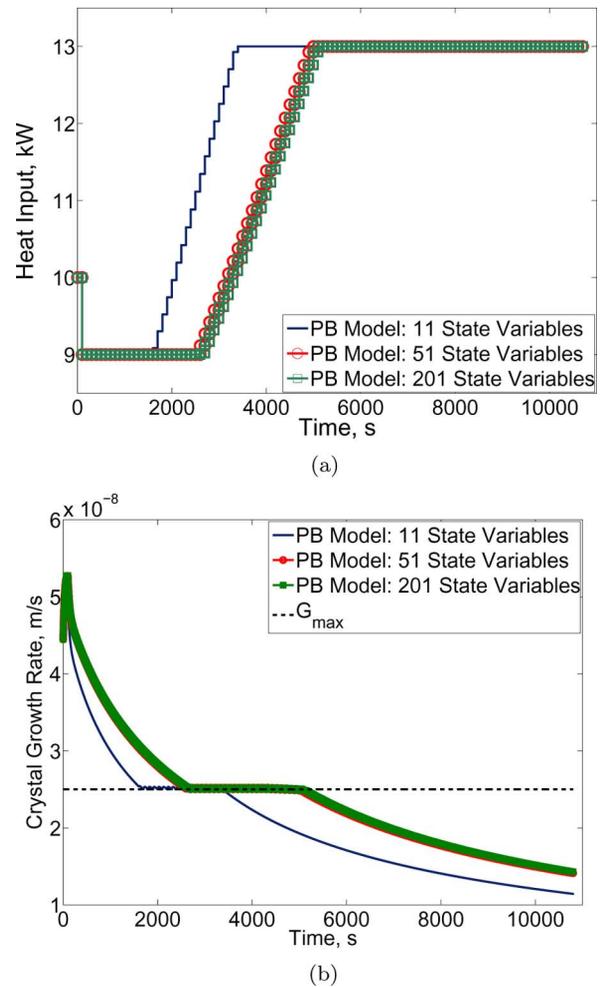


Fig. 5. Simulation results of the state feedback control approach in relation to the discretization coarseness of the population balance model. (a) Heat input profiles. (b) Crystal growth rate profiles.

crystal growth rate to avoid the formation of irregularly shaped crystals and to limit the undesirable effects of high supersaturation. The heat input profiles are depicted in Fig. 5(a). As can be seen, the optimizer readily reduces the heat input to the crystallizer when the batch run is initiated at the heat input of 10 kW. This is to suppress the excessively high crystal growth rates in the beginning of the batch run. However, the desired crystal growth rate cannot be achieved in the initial phase of the batch run due to the heat input's lower bound, i.e., 9 kW. As the crystals grow larger, the supersaturation in the crystallizer depletes and consequently the crystal growth rate reaches its desired value. Thereafter, G_{max} is optimally tracked till the heat input hits its upper bound of 13 kW. In the final phase of the batch run, the crystal growth rate starts decaying again. This is due to the heat input's upper bound, making the tracking of G_{max} no longer possible.

Fig. 5 indicates that when the control approach exploits a PB model with 11 state variables, namely the crystal population in 10 crystal cells and the solute concentration, the reference trajectory tracking is attained over a shorter time frame. This can be attributed to the large numerical diffusion in predictions of the PB model with 10 crystal cells. On the other hand, Fig. 5

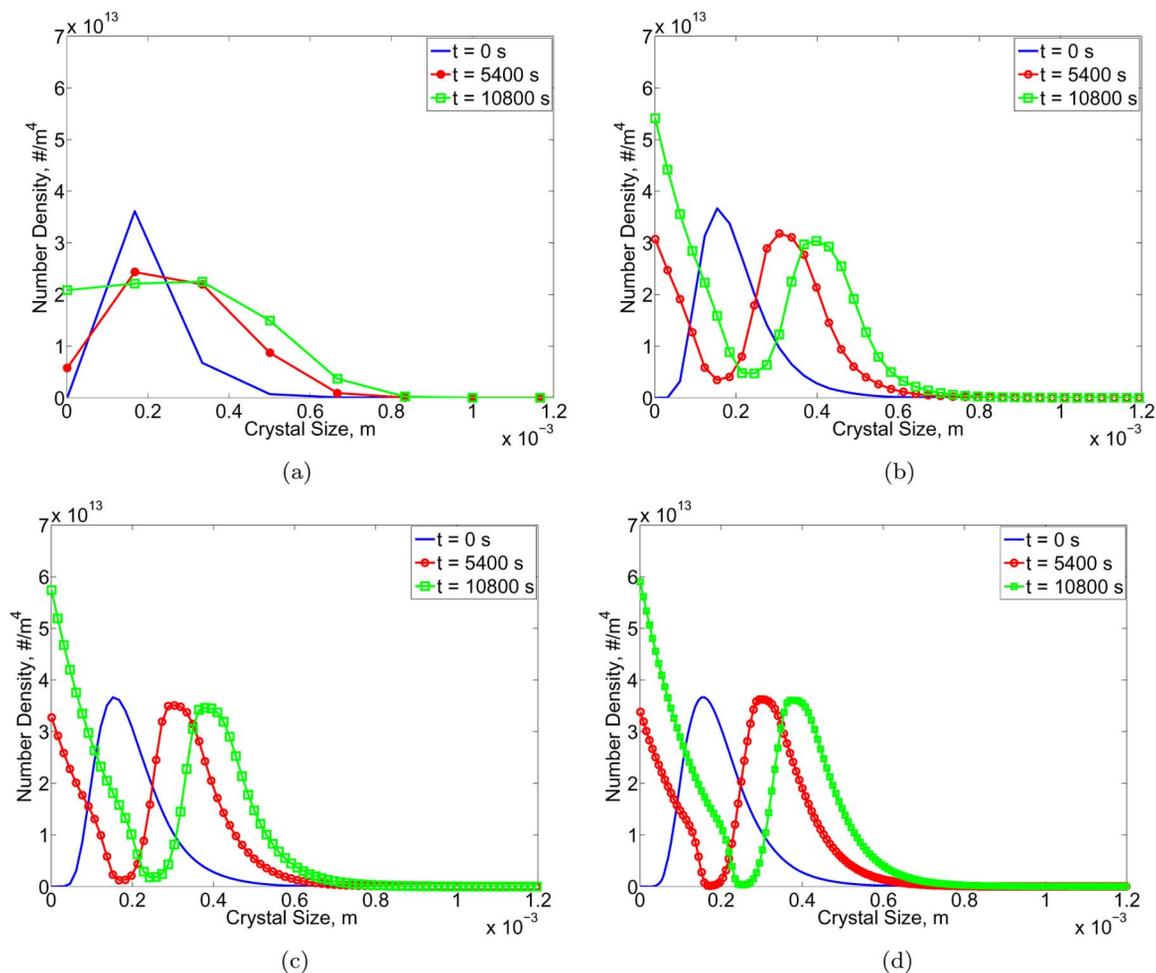


Fig. 6. Crystal size distribution at different time instants during the batch run. (a) Population balance model with 11 state variables. (b) Population balance model with 51 state variables. (c) Population balance model with 101 state variables. (d) Population balance model with 201 state variables.

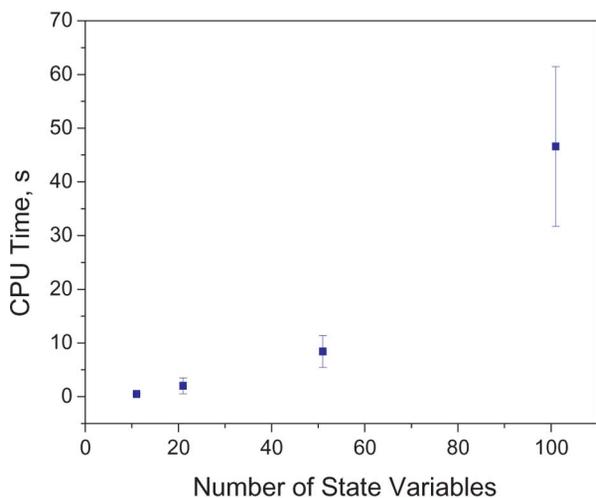


Fig. 7. Average computation times of one optimization step in relation to the number of state variables of the population balance model. The vertical lines indicate the standard deviations of computation times around the average values.

suggests that the PB models with 51 and 201 state variables allow the controller to fulfill its objective adequately. Fig. 6 shows the evolution of crystal size distribution in the course of a batch run of 10 800 s. It is observed that a coarse grid mesh

TABLE II
AVERAGE COMPUTATION TIMES OF ONE OPTIMIZATION STEP IN THE STATE FEEDBACK CONTROL APPROACH

Number of state variables	CPU time ^a , s
11	0.48 ± 0.06
21	2.02 ± 0.75
51	8.42 ± 1.48
101	46.59 ± 15.43
201	1204.37 ± 205.79

^aThe reported CPU times correspond to the Microsoft Windows XP (Professional) operating system running on a Genuine Intel(R) T2050 @1.60GHz processor with 1 GB RAM.

results in non-smooth crystal size distributions. A sufficiently large number of crystal cells leads to adequate representation of the distributions at the expense of a higher optimization burden.

Fig. 7 depicts the average computation times required for one optimization step in relation to the number of state variables of the PB model. The computational burden of the state feedback control approach seems to increase exponentially with the number of state variables. Table II lists the computation times of one optimization step in the control approach when different PB models are used. It is observed that the control approach based on the PB model with 201 state variables cannot be used for real-time control of the crystallizer under study. The average

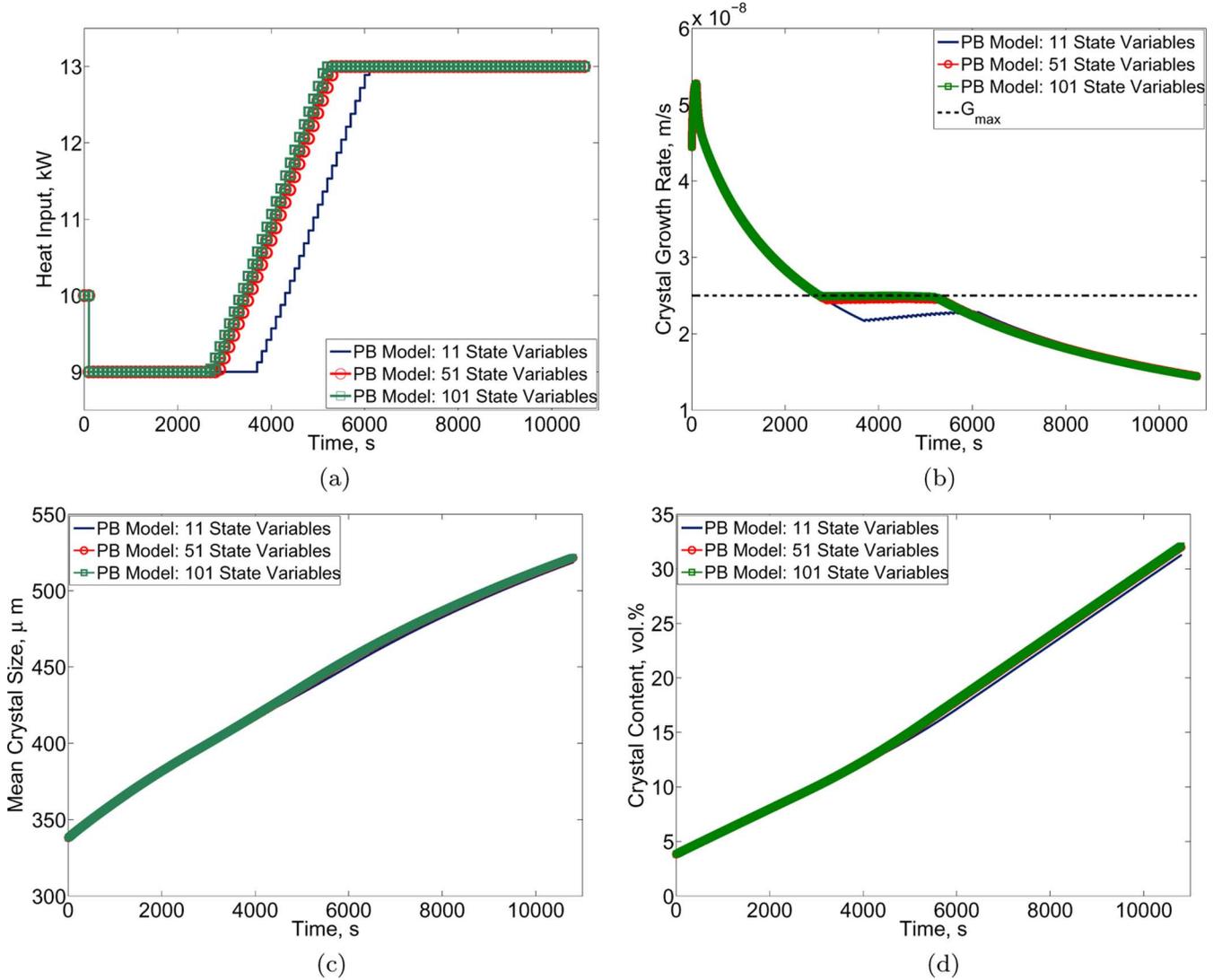


Fig. 8. Simulation results of the state feedback control approach in the presence of structural plant-model mismatch. (a) Heat input profiles. (b) Crystal growth rate profiles. (c) Mean crystal size. (d) Batch crystal content.

TABLE III
RELATIVE ERRORS OF THE MOMENTS OF THE PRODUCT CRYSTAL SIZE DISTRIBUTION

Number of state variables of the PB model	Relative Error				
	μ_0	μ_1	μ_2	μ_3	μ_4
11	0.039	0.043	0.130	0.318	0.629
21	0.022	0.013	0.042	0.125	0.242
51	0.009	0.008	0.015	0.047	0.088
101	0.004	0.003	0.008	0.023	0.041

computation time of one optimization step is approximately 120 s, which exceeds the measurement sampling time interval of 100 s.

The ability of the state feedback control approach to cope with structural plant-model mismatch due to a coarse finite volume grid mesh is examined. The control approach is applied to a plant simulator, consisting of the PB model with 201 state variables. It is assumed that the plant simulator provides a true representation of the process dynamics. On the other hand, the control approach uses a PB model with less state variables, which leads to structural model imperfection with

respect to the plant simulator. Fig. 8 shows the simulation results of the state feedback control approach in the presence of structural plant-model mismatch. As can be seen in Fig. 8(b), the structural plant-model mismatch results in a poor reference trajectory tracking when the control approach is based on the PB model with 11 state variables. The deterioration of the control performance is due to inadequate representation of the evolution of crystal size distribution, which arises from the coarse grid mesh. Table III lists the relative errors of the moments of the product CSD simulated by the different PB models with respect to predictions of the plant simulator

$$\text{Relative Error} = \left(\frac{\tilde{\mu}_i(t) - \mu_i(t)}{\tilde{\mu}_i(t)} \right) \quad (16)$$

where $\tilde{\mu}_i(t)$ denotes the moments of CSD simulated by the plant simulator. It is evident that the errors of the PB model with 11 state variables are significantly larger than those of the other PB models, particularly in the higher moments. The errors diminish as more crystal cells are used for spatial discretization of the

population balance equation. As shown in Fig. 8(b), the performance objective, namely the reference trajectory tracking, can be effectively realized when PB models with 51 and 101 state variables are adopted in the control approach. Fig. 8(c) and (d) suggest that more effective reference trajectory tracking will in turn lead to slightly larger crystals and increased batch crystal content.

To enhance the effectiveness of the control approach in the presence of structural plant-model mismatch, the optimal control problem is modified as

$$\min_{H_{in}(t)} \frac{\int_0^{t_b} \left(100 \frac{(G(t) - G_{max}) + (G_{meas} - \hat{G}(t))}{G_{max}} \right)^2 dt}{\int_0^{t_b} dt}$$

subject to: equation (11)

$$H_{min} \leq H_{in}(t) \leq H_{max} \quad (17)$$

where G_{meas} denotes the measured value of the crystal growth rate that is inferred from the solute concentration measurements; $\hat{G}(t)$ is the estimated crystal growth rate. The correction term $G_{meas} - \hat{G}(t)$ is introduced in the performance objective to suppress the large offset in the reference trajectory tracking. The correction term aims to diminish the large discrepancy between the predicted crystal growth rate and its measured value. This term confers an integrating action to the optimizer that decreases the effects of plant-model mismatch according to the generally used method in linear model predictive control [32]. The simulation results of the control approach with the modified performance objective are depicted in Fig. 9. It is observed that the correction term allows us to eliminate the large deviation of the crystal growth rate from its reference trajectory. This illustrates the significance of solute concentration measurements, whose availability is essential for effective application of the control approach.

2) *Output Feedback Control*: Industrial batch crystallizers are often prone to various process uncertainties, mainly arisen from measurement deficiencies and uncertain initial conditions due to improper seeding. In addition, the great degree of uncertainty typically associated with the parameters of crystallization kinetic expressions may not allow the process model to provide an adequate description of the system dynamics. To compensate for the adverse impact of process uncertainties and model imperfections on optimal process operation, the dynamic optimizer is applied in an output feedback fashion. The PB model with 51 state variables is exploited to devise the output feedback control approach. A moving horizon estimator is designed to facilitate receding horizon implementation of the dynamic optimizer. The performance of the control approach in the presence of model imperfections and process uncertainties is examined when applied to a plant simulator that utilizes the PB model with 201 state variables. A correction term is added to the performance objective to diminish the effects of structural plant-model mismatch; see (17).

A comparison of the simulation results of the state feedback and the output feedback control approaches is demonstrated in Fig. 10. The specifications of model imperfections and process uncertainties are given in Table IV. Fig. 10(b) suggests that the parametric plant-model mismatch and process uncertainties

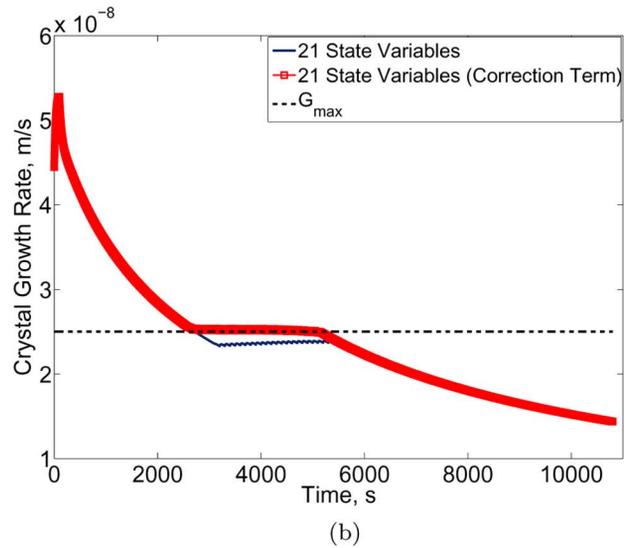
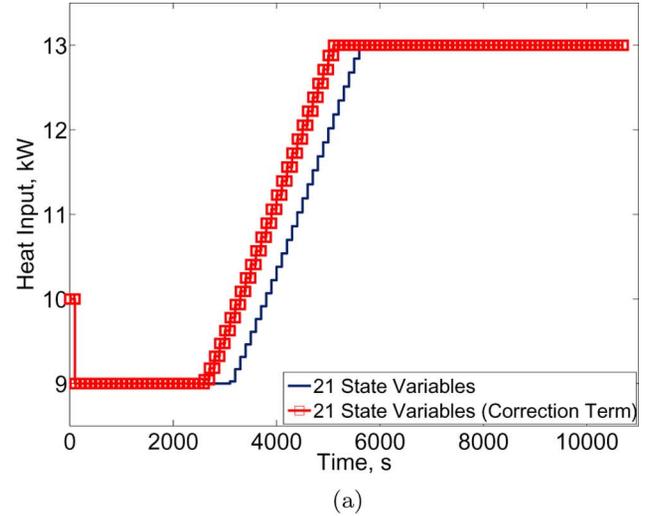


Fig. 9. Simulation results of the state feedback control approach with the modified performance objective in the presence of structural plant-model mismatch. (a) Heat input profiles. (b) Crystal growth rate profiles.

slightly deteriorate the performance of the state feedback control approach in terms of the reference trajectory tracking. On the other hand, the output feedback implementation of the dynamic optimizer allows us to follow the reference crystal growth rate trajectory more effectively.

Fig. 10(a) shows that the optimal heat input profile obtained under the output feedback control scenario exhibits a sudden increase at time 300 s. This results from the rather long estimation horizon, i.e., 500 s, required to build up information to initialize the moving horizon estimator. Hence, the estimator is unable to readily take action against the detrimental effects of uncertain initial conditions. Nonetheless, once the moving horizon estimator is initialized at time 500 s, the recursive state estimation allows us to adequately compensate for model imperfections and process uncertainties. In addition to increased process productivity, the effective crystal growth rate control will result in slightly larger crystals; see Fig. 10(c) and (d).

The computation time profiles of dynamic optimization and state estimation along a batch run of 10 800 s are depicted in Fig. 11. It is observed that the computational burden of the

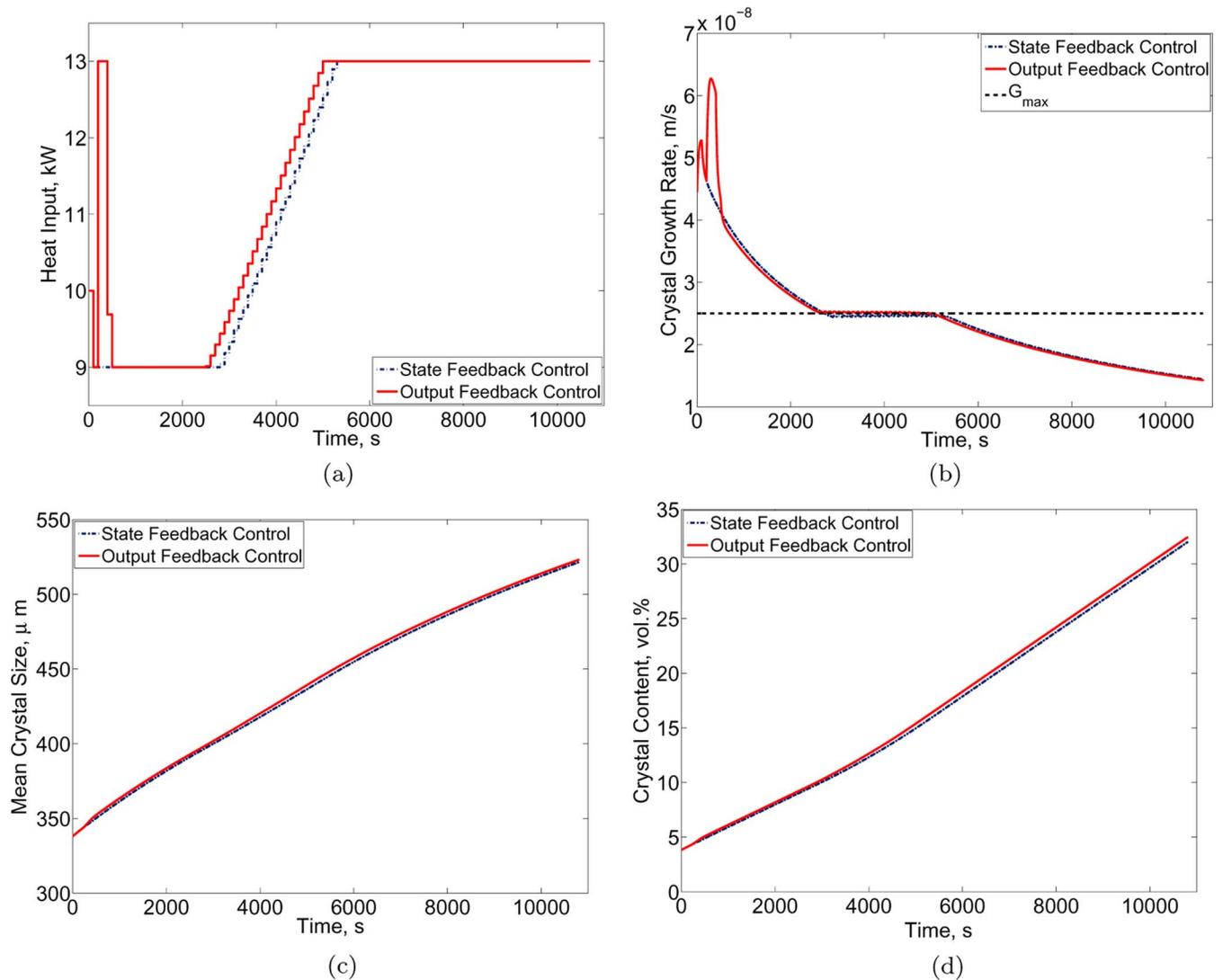


Fig. 10. Comparison of simulation results of the state feedback control and the output feedback control approaches in the presence of model imperfections and process uncertainties. The control approach exploits the population balance model with 51 state variables, whereas the plant simulator consists of the population balance model with 201 state variables. (a) Heat input profiles. (b) Crystal growth rate profiles. (c) Mean crystal size. (d) Batch crystal content.

output feedback control approach is considerably less than the measurement sampling time interval, i.e., 100 s, at all times during a batch run. Therefore, the control approach based on the full population balance model with 51 state variables can be applied for real-time control of the semi-industrial batch crystallizer under study.

C. Experimental Results

The real-time feasibility of the output feedback control approach is experimentally demonstrated for fed-batch evaporative crystallization of ammonium sulphate. The crystallization takes place in the 75-liter draft tube crystallizer, which is equipped with a distributed control system (DCS CENTUM CS3000, Yokogawa, Japan). The hierarchical control framework depicted in Fig. 12 is adopted to apply the optimal operating policies to the semi-industrial crystallizer. The optimal input sequences are implemented using conventional proportional integral derivative (PID) controllers. The PID controllers are embedded in the DCS, which forms

the lowest control layer. The timed signal exchange among different components of the control approach is facilitated by an OPC [object linking and embedding (OLE) for process control] communication interface (IPCOS, The Netherlands). Synchronization and sequencing of tasks are carried out by means of a timer and a scheduler, respectively.

Variations of the crystal growth rate in relation to different heat input profiles are examined. The crystal growth rate largely affects the product quality of the seeded batch runs, whereas the heat input serves as the only driving force for supersaturation generation in the evaporative crystallizer under study. Fig. 13 depicts the heat input and the crystal growth rate profiles of two seeded batch runs. The experimental settings are given in Table V.

In Exp. 1, the heat input to the crystallizer is kept constant at 9 kW. Consequently, the crystal growth rate gradually decays, being unable to follow the reference trajectory, i.e., G_{max} . To attain the desired crystal growth rate throughout the batch run, the output feedback control approach is applied in Exp. 2. Fig. 13

TABLE IV
SPECIFICATIONS OF MODEL IMPERFECTIONS AND PROCESS UNCERTAINTIES IN THE OUTPUT FEEDBACK CONTROL SCENARIO

Parametric plant-model mismatch:	+15.0% error in kinetic parameters
Systematic measurement error:	+2.0% error in the solute concentration +5.0% error in the crystal size distributions
Uncertain initial conditions:	+2.0% error in the initial solute concentration +5.0% error in the mean of the initial crystal size distribution

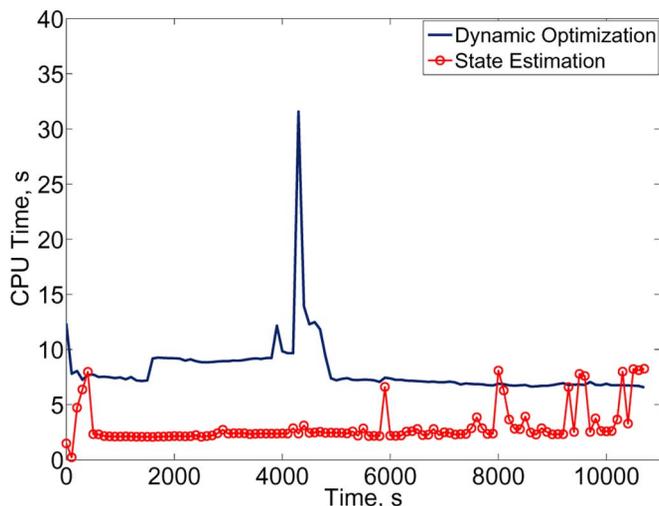


Fig. 11. CPU time profiles of dynamic optimization and state estimation tasks in the output feedback control approach based on the population balance model with 51 state variables.

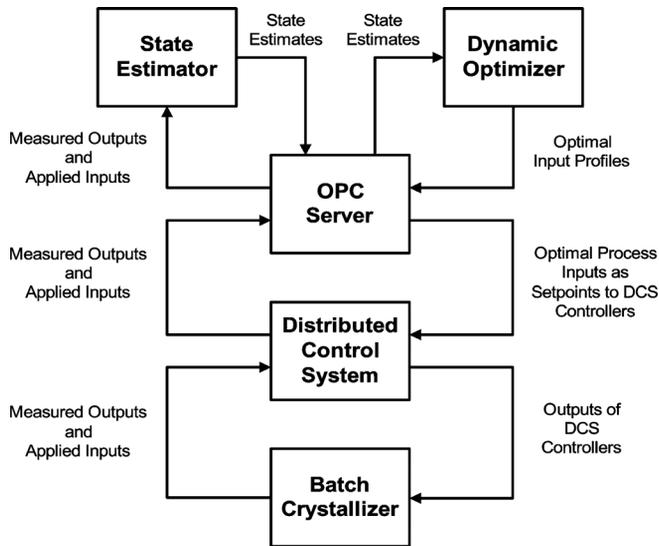
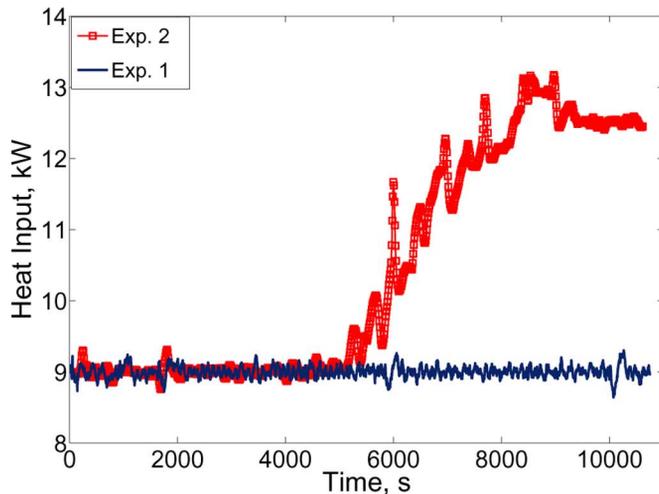


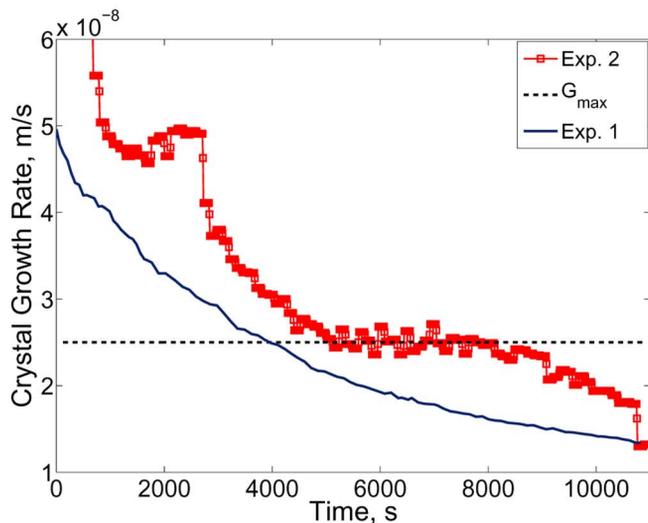
Fig. 12. Hierarchical output feedback control framework.

shows that once the crystal growth rate crosses the reference trajectory the heat input is raised by the control approach to facilitate tighter tracking of the desired crystal growth rate. However, the reference trajectory can no longer be closely followed when the heat input reaches its upper bound. The crystal growth rate steadily decreases thereafter, while the heat input remains at 13 kW.

Fig. 13(b) suggests that the optimal process operation cannot be realized at all times during the batch run. The reference trajectory is violated in the beginning and towards the end of the



(a)



(b)

Fig. 13. Experimental results of the output feedback control approach based on the full population balance model with 51 state variables. (a) Heat input profiles. (b) Crystal growth rate profiles.

batch run due to actuation limitations. In the optimal control problem, the heat input's lower bound is defined as 9 kW to avoid possible dissolution of seeds. On the other hand, the heat transfer limitations of the process restrict the heat input to an upper bound of 13 kW. Mesbah *et al.* [31] have demonstrated that inserting the seeds into the crystallizer at lower heat inputs, i.e., 4.5 kW, will allow a more effective reference trajectory tracking in the beginning of the batch run as the seeds remain intact. Note that the differences in initial crystal growth rates of the two batch runs result from different supersaturation levels at the seeding point; see Table V.

TABLE V
DESCRIPTION OF THE SEEDED FED-BATCH EVAPORATIVE CRYSTALLIZATION EXPERIMENTS

Experiment	Operating conditions				Seeding conditions	
	Temperature, °C	Pressure, mbar	Impeller frequency, rpm	Heat input, kW	Seed aging time, min	Initial supersaturation
Exp. 1	50	100	450	9.0	57	0.01627
Exp. 2	50	100	450	Figure 13(a)	76	0.00953

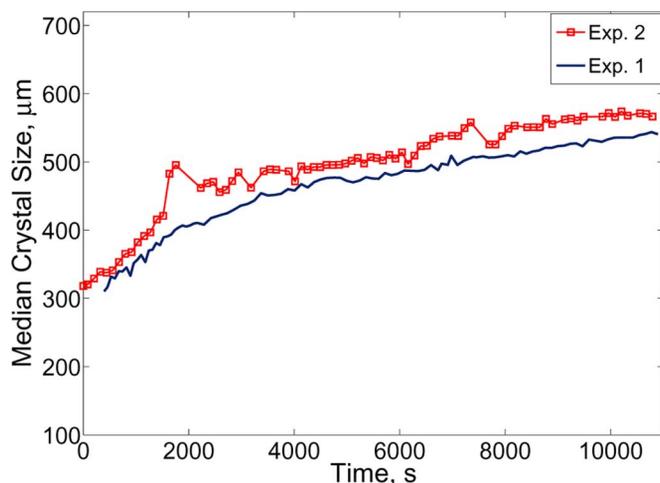


Fig. 14. Evolution of median crystal size throughout the batch experiments.

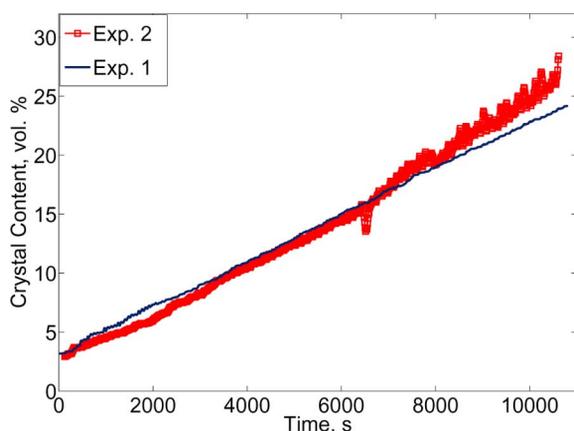


Fig. 15. Evolution of crystal volume content throughout the batch experiments.

The evolution of median crystal size and crystal volume content throughout the batch experiments is depicted in Figs. 14 and 15, respectively. It is observed that the reference crystal growth rate trajectory tracking results in larger crystals. In addition, the application of the control approach leads to a substantial increase, i.e., up to 15%, in the process productivity as compared to the batch run with constant heat input. The increased crystal content at the end of the controlled batch run results from the higher crystal growth rates.

V. CONCLUSION

An output feedback nonlinear model-based control approach has been presented for optimal operation of industrial batch crystallizers. The distinct contribution of this article is the use of a full population balance model as the cornerstone of the control approach. Successful application of the control approach for

real-time control of a semi-industrial batch crystallizer has been corroborated by simulation case studies as well as experimental implementation. The simulation results indicate that the discretization coarseness of the population balance model largely affects the real-time feasibility of the control approach. To compensate for the detrimental effect of structural model imperfections arisen from inadequate discretization, the optimal control problem has been slightly modified to confer an integrating action to the dynamic optimizer. It has been shown that the moving horizon state estimator allows us to adequately fulfill the control objective in the presence of parametric model imperfections and process uncertainties. The experimental results suggest that the use of the output feedback control approach leads to increased process productivity, i.e., batch crystal content, as the product quality is sustained. The generic structure of the presented control approach facilitates its application to a wide range of industrial batch crystallizers.

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