

Batch-to-batch Optimization of Batch Crystallization Processes

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Abstract It is the fact that several process parameters are either unknown or uncertain. Therefore, an optimal control profile calculated with developed process models with respect to such process parameters may not give an optimal performance when implemented to real processes. This study proposes a batch-to-batch optimization strategy for the estimation of uncertain kinetic parameters in a batch crystallization process of potassium sulfate production. The knowledge of a crystal size distribution of the product at the end of batch operation is used in the proposed methodology. The updated kinetic parameters are applied for determining an optimal operating temperature policy for the next batch run.

Keywords batch crystallization, batch-to-batch optimization, optimal control, population balance model, moment model

1 INTRODUCTION

A batch crystallization process is a major process in fine chemical, pharmaceutical, food, and mineral industries, because of its ability to provide high purity separation and its flexibility to offer product specification change as defined. Due to the widespread use of this process, finding an efficient operating strategy for improving both product quality and process efficiency is very significant. Recently, an optimization of batch crystallizers has received considerable attention to improve product quality characterized by a crystal size distribution [1, 2]. Previous studies mostly concentrate on the determination of an optimal operation policy in terms of crystallizer temperature as it has a direct influence on the supersaturation which is the driving potential for crystal formation [3, 4]. However, it has been known that due to uncertain process and kinetic parameters, an optimal control profile calculated with these process models may not give an optimal performance when applied to real processes. As batch processes are repetitive in nature, it would be possible to exploit information of previous batch results to improve the operation of current batch and so on.

Therefore, the aim of this study is to propose a batch-to-batch optimization strategy to estimate uncertain kinetic parameters. Measurement of a crystal size distribution which is obtained with reliability and accuracy through experimental analysis at the end of run is used in such an optimization-based estimation. The updated kinetic parameters are used to modify an optimal operating temperature policy of the crystallizer for subsequent operation. The optimal control problem formulated is solved by a sequential optimization approach. The proposed method is illustrated via a simulation study of a batch cooling crystallization for the synthesis of potassium sulfate.

2 MODEL OF BATCH CRYSTALLIZER

The classical framework for modelling batch

crystallization processes consists of a population balance equation (PBE) of crystals and mass and energy conservation equations as shown in Eqs. (1)–(3), respectively. It is assumed that crystal agglomeration or breakage phenomena are neglected [5].

$$\frac{\partial f(L, t)}{\partial t} + G(t) \frac{\partial f(L, t)}{\partial L} = 0 \quad (1)$$

$$\frac{dc}{dt} = -3\rho_c k_v G(t) \mu_2(t) \quad (2)$$

$$\frac{dT}{dt} = -3 \frac{\Delta H_c}{c_p} \rho_c k_v G(t) \mu_2(t) - \frac{UA}{Mc_p} [T(t) - T_j(t)] \quad (3)$$

where, $f(L, t)$ is the population density of crystals at a characteristic length (L) and time (t), c is the solute concentration and T is the solution temperature.

In the crystallization process, the formation of a crystal can be classified into two steps: the birth of a new particle and its growth to macroscopic size. The driving potential for both rates is the nonequilibrium state of the system measured by a relative supersaturation (S).

$$S = \frac{c - c_s(T)}{c_s(T)} \quad (4)$$

where, c_s is the saturation concentration of the solute.

The nucleation (B) and growth (G) rates of crystals can be described as given in Eqs. (5) and (6).

$$B(t) = k_b e^{-E_b/RT} S^b \mu_3(t) \quad (5)$$

$$G(t) = k_g e^{-E_g/RT} S^g \quad (6)$$

It is noted that the i th moment of the population density is defined in term of the population density function as in Eq. (7).

$$\mu_i = \int_0^\infty f(L, t) L^i dL \quad (7)$$

where, μ_i ($i = 0, \dots, 3$) means the total of number, length, surface area, and volume of crystals, respectively [5]. These moments represent an average of product qualities.

In this study, the seeded batch crystallizer of potassium sulfate studied by Shi *et al.* [6] is considered. The model parameters are shown in Table 1. To avoid uncontrolled nucleation, the crystallization process has to be operated in the metastable zone bounded by the saturation and the metastable limit, that is $c_s \leq c \leq c_m$. The following equations are used to calculate the saturation and metastable concentrations which are the function of the solution temperature, T .

$$c_s(T) = 6.29 \times 10^{-2} + 2.46 \times 10^{-3}T - 7.14 \times 10^{-6}T^2 \quad (8)$$

$$c_m(T) = 7.76 \times 10^{-2} + 2.46 \times 10^{-3}T - 8.10 \times 10^{-6}T^2 \quad (9)$$

The set of above-mentioned model equations (called “a population model”), which are used to represent a real crystallization process, is solved using a solution methodology proposed by Hu *et al.* [3] with the initial boundary condition and the initial distribution of the seeded crystals which is assumed to be a parabolic distribution as follows,

$$f(0, t) = \frac{B(t)}{G(t)} \quad (10)$$

$$f(L, 0) = \begin{cases} 0.0032(300 - L)(L - 250) & \text{for } 250\mu\text{m} \leq L \leq 300\mu\text{m} \\ 0 & \text{for } L < 250\mu\text{m} \text{ and } L > 300\mu\text{m} \end{cases} \quad (11)$$

3 BATCH-TO-BATCH OPTIMIZATION APPROACH

As batch processes are repetitive in nature, it would be possible to use information from the previous batch to improve the operation of the current batch. Here a batch-to-batch optimization approach is proposed to estimate kinetic parameters, that is k_b and k_g , that are important for the prediction of crystal formation. The identification relies on the measurement of a crystal size distribution (CSD) obtained at the end

of batch run. Fig. 1 shows the schematic of an integrated batch-to-batch optimization and optimal control methodology proposed in this study. As can be seen from the figure, once the CSD of product at the end of operation is measured, the i th moment of the population density of the process ($\mu_{i,\text{process}}$) is calculated using Eq. (7) and compared with that obtained from a moment model ($\mu_{i,\text{model}}$). New updated value of the parameters is determined by solving a parameter optimization problem with an objective to minimize such an error. It is noted here that although the population model mentioned in the previous section can be directly solved, its implementation in an optimization problem is very time-consuming. Recently, the method of moments, which leads to “the moment model” of crystallizers [Eqs. (12), (13)], is developed, allowing the model of crystallization processes to be solved quickly and efficiently [5]. Therefore, the moment model is applied as a crystallization model to a batch-to-batch optimization and optimal control strategy.

$$\frac{d\mu_0^n}{dt} = B(t),$$

$$\frac{d\mu_i^n}{dt} = iG(t)\mu_{i-1}^n(t), \quad i = 1, 2, 3 \quad (12)$$

$$\mu_0^s = \text{constant},$$

$$\frac{d\mu_i^s}{dt} = iG(t)\mu_{i-1}^s(t), \quad i = 1, 2, 3 \quad (13)$$

It is noted that the overall i th moments are defined as $\mu_i = \mu_i^n + \mu_i^s$.

Optimal control problem

After updating the kinetic parameters via the batch-to-batch optimization strategy, these parameters are used for computing an optimal operating temperature policy for the control of the crystallizer at a new batch run. In batch crystallization processes, the fine crystals usually affect product quality and may cause difficulties in downstream processing equipment (*i.e.*,

Table 1 Parameter values for the seeded batch crystallizer of potassium sulfate

b	$k_b/s^{-1} \cdot \mu\text{m}^{-3}$	$E_b/R/K$	$U/kJ \cdot m^{-2} \cdot h^{-1} \cdot K^{-1}$	$\Delta H_o/kJ \cdot kg^{-1}$	M/kg	k_v	g	$k_g/\mu\text{m} \cdot s^{-1}$	A/m^2	$E_g/R/K$	$c_p/kJ \cdot kg^{-1} \cdot K^{-1}$	$\rho_o/g \cdot \mu\text{m}^{-3}$	t_f/min
1.45	285.0	7517.0	1800.0	44.5	27.0	1.5	1.5	1.44×10^8	0.25	4859.0	3.8	2.66×10^{-12}	30.0

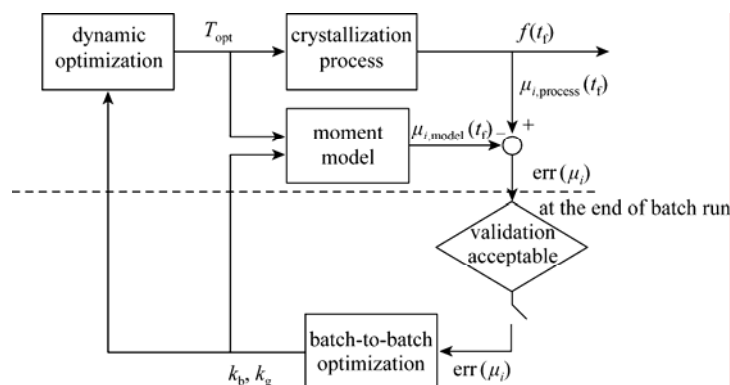


Figure 1 An integrated batch-to-batch optimization and optimal control strategy

filtration, drying). For this reason, the aim of an optimal control problem is to determine an optimal temperature profile minimizing the total volume of fine crystals, $\mu_3^n(t_f)$, whereas the total volume of seeded crystals, $\mu_3^s(t_f)$, has to satisfy the product quality requirement. In addition, the solution concentration has to be maintained in the metastable zone. The formulation of the optimal control problem can be mathematically explained as follows,

$$\min_{T(t)} \mu_3^n(t_f) \quad (14)$$

subject to

the model of crystallizer [Eqs. (2), (12) and (13)]

$$c_s \leq c \leq c_m$$

$$303 \leq T \leq 323$$

$$\mu_3^s(t_f) \geq 8.3301 \times 10^9$$

$$t_f = 30 \text{ min}$$

The resulting optimal control problem is solved via a sequential approach in which a dynamic optimization problem is converted to a nonlinear programming (NLP) problem by parameterizing the control variables (i.e. solution temperature) as a piecewise constant function [7]. With the initial guess of the temperature value in each time interval, the crystallizer model equations are integrated using the Ode15s routine in Matlab and the objective function is then determined. The resulting optimization is solved using the Fmincon routine to compute a new set of the control variable. The sequence continues until the optimal value of the temperature in each time interval is found satisfying a specified accuracy.

4 SIMULATION RESULTS

Simulation study is first investigated under nominal case where all parameters in the model of the crystallizer as given in Table 1 are known exactly. Fig. 2 (a) shows the optimal temperature profile obtained from the calculation. It can also see that under such the optimal operating temperature policy, the solution concentration can be maintained within the metastable zone keeping away from the uncontrolled nucleation of crystals. The total volume of fine and seeded crystals are 5.9010×10^8 and 8.4229×10^9 , respectively. The corresponding CSD at the end of batch run is shown in Fig. 2 (b). From the figure, the distribution can be characterized by the classes of nucleated and seeded crystals. The average product qualities in terms of the i th moments computed from the CSD at final batch time and those obtained from the moment model are provided in Table 2. The results show a good agreement; small relative errors (less than 5%) are observed. This confirms that the moment model can be reasonably used to describe crystallization processes.

In case of a mismatch in kinetic parameters, the 15% decrease of k_b and the 15% increase of k_g are introduced in the moment model. It is found that when applying the calculated temperature profile from solving the optimal control problem based on incor-

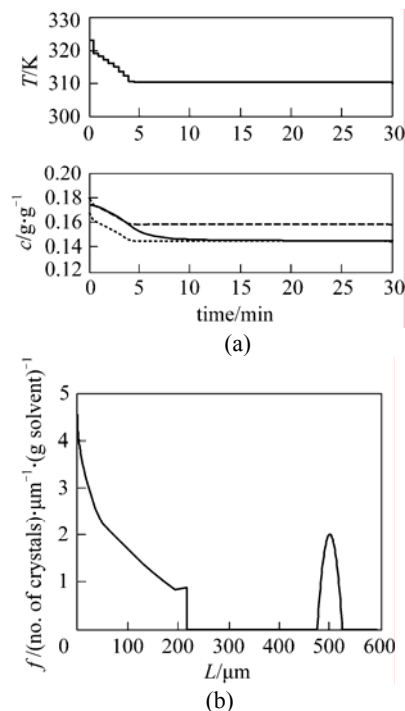


Figure 2 (a) The optimal temperature and concentration profiles (c_s : dotted; c : solid; c_m : dash) and (b) the CSD at final batch time in the nominal case

Table 2 Final-time product properties of crystals in the nominal case

i th moments	$\mu_0(t_f)$	$\mu_1(t_f)$	$\mu_2(t_f)$	$\mu_3(t_f)$
population model (actual process)	456	6.4506×10^4	2.0692×10^7	9.0130×10^9
moment model	482	6.5908×10^4	2.0134×10^7	8.7701×10^9

rect kinetic parameters to the crystallizer (Batch I), the final crystal product obtained is out of specification; the average total volume of seeded crystals (μ_3^s) is 8.3196×10^9 which is less than the desired specification ($\mu_{3,spec}^s \geq 8.3301 \times 10^9$). In addition, the result indicates that at the initial time period, the solution concentration stays outside the metastable zone, leading to the uncontrolled crystal nucleation. To avoid this problem for the next batch run (Batch II), a batch-to-batch optimization approach is considered. Information of the CSD from the previous run is used to update the kinetic parameters following the method mentioned in Section 3. It can be seen from Table 3 that the updated kinetic parameters approach the actual values (Table 1). With these updated values, the optimal temperature profile is re-computed and implemented to control the crystallizer at the subsequent batch. The results indicate that at the final time, the total volume of seeded crystals obtained ($\mu_3^s =$

Table 3 The value of kinetic parameters used in the moment model

Case	$k_b/s^{-1} \cdot \mu m^{-3}$	$k_g/\mu m \cdot s^{-1}$
Batch I with mismatch kinetic parameters	242.25	1.656×10^8
Batch II with updated kinetic parameters	283.15	1.440×10^8

8.4205×10^9) satisfies the requirement while the total volume of fine crystals is minimized ($\mu_3^n = 5.8954 \times 10^8$). From these results, it is clearly indicated that the product quality and process efficiency can be improved by using the batch-to-batch optimization strategy integrated with the optimal control. It is noted that in this study, the temperature of the crystallizer is assumed to be perfectly controlled following the desired optimal temperature profile. However, if advanced model-based control methodologies, that is generic model control [7] and model predictive control [8], are applied to on-line control the crystallizer temperature, the updated kinetic parameters obtained from the batch-to-batch optimization approach can also be used to design such the controllers, resulting in an improved control performance.

5 CONCLUSIONS

A batch-to-batch optimization strategy for a kinetic parameter estimation is developed and implemented to a seeded batch crystallization of potassium sulfate production in this study. Based on the knowledge in a crystal size distribution measured from a previous batch operation, the kinetic parameters involving the nucleation and growth of crystals are updated and then used to determine an optimal operating temperature policy for a next batch. From simulation results, it is clearly shown that the product quality and process efficiency can be improved by using the proposed batch-to-batch optimization integrated with the optimal control strategy.

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NOMENCLATURE

A	total heat transfer surface area, m^2
B	nucleation rate, $(\text{no. of crystal}) \cdot \text{s}^{-1} \cdot (\text{g solvent})^{-1}$
b	nucleation rate exponent
c	solution concentration, $(\text{g solute}) \cdot (\text{g solvent})^{-1}$

c_m	metastable concentration, $(\text{g solute}) \cdot (\text{g solvent})^{-1}$
c_p	heat capacity of the solution, $\text{kJ} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$
c_s	saturation concentration, $(\text{g solute}) \cdot (\text{g solvent})^{-1}$
E	activation energy
f	population density of crystals, $(\text{no. of crystals}) \cdot \mu\text{m}^{-1} \cdot (\text{g solvent})^{-1}$
G	growth rate, $\mu\text{m} \cdot \text{s}^{-1}$
g	growth rate exponent
ΔH_c	heat of crystallization, $\text{kJ} \cdot \text{kg}^{-1}$
k_b	birth rate coefficient, $\text{s}^{-1} \cdot \mu\text{m}^{-3}$
k_g	growth rate coefficient, $\mu\text{m} \cdot \text{s}^{-1}$
k_v	volumetric shape factor
L	characteristic crystal length, μm
M	mass of solvent in the crystallizer, kg
R	gas constant
T	reactor temperature, K
T_j	cooling jacket temperature, K
t	time, min
U	overall heat transfer coefficient, $\text{kJ} \cdot \text{m}^{-2} \cdot \text{h}^{-1} \cdot \text{K}^{-1}$
μ	moment of the CSD
μ_0	zeroth moment of the CSD, $(\text{no. of crystals}) \cdot (\text{g solvent})^{-1}$
μ_1	first moment of the CSD, $\mu\text{m} \cdot (\text{g solvent})^{-1}$
μ_2	second moment of the CSD, $\mu\text{m}^2 \cdot (\text{g solvent})^{-1}$
μ_3	third moment of the CSD, $\mu\text{m}^3 \cdot (\text{g solvent})^{-1}$
ρ_c	density of crystals, $\text{g} \cdot \mu\text{m}^{-3}$

Superscripts

n	nucleation
s	seeded crystal

REFERENCES

- 1 Fujiwara, M., Nagy, Z. K., Chew, J. W., Braatz, R. D., "First-principles and direct design approaches for the control of pharmaceutical crystallization", *J. Process Contr.*, **15**, 493–504 (2005).
- 2 Larsen, P.A., Patience, D.B., Rawlings, J.B., "Industrial crystallization process control", *Control System Magazine, IEEE*, **26** (4), 70–80 (2006).
- 3 Hu, Q., Rohani, S., Wang, D.X., Jutan, A., "Optimal control of a batch cooling seeded crystallizer", *Powder Technology*, **156**, 170–176 (2005).
- 4 Miller, S. M., Rawlings, J. B., "Model identification and control strategies for batch cooling crystallizers", *AIChE J.*, **40**, 1312–1327 (1994).
- 5 Matthews, H. B., "Model identification and control of batch crystallization for an industrial chemical system", Ph.D. thesis, University of Wisconsin at Madison, U.S.A. (1997).
- 6 Shi, D., El-Farra, N. H., Li, M., Mhaskar, P., Christofides, P. D., "Predictive control of particle size distribution in particulate processes", *Chem. Eng. Sci.*, **61**, 268–281 (2006).
- 7 Arpornwichanop, A., Kittisupakorn, P., Mujtaba, I. M., "On-line dynamic optimization and control strategy for improving the performance of batch reactors", *Chem. Eng. Process.*, **44** (1), 101–114 (2005).
- 8 Arpornwichanop, A., Kittisupakorn, P., "Dual mode NMPC for regulating the concentration of exothermic reactor under parametric uncertainties", *J. Chem. Eng. Jap.*, **37**, 698–710 (2004).