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Robust optimal temperature swing operations for size control of seeded batch cooling crystallization

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HIGHLIGHTS

- Temperature swing profiles for batch crystallization are optimized.
- Model and operating condition perturbations are considered in the optimization.
- Robust performance is evaluated through Monte Carlo simulation.
- Measures for improvement are suggested based on the *k*-means clustering analysis.

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ABSTRACT

Robust optimal temperature swing operations for a seeded batch cooling crystallization process with size-dependent kinetics were derived for minimizing the amount of fines, and their robust performances were evaluated through simulation studies. The nominal optimal temperature swing profile, which does not incorporate any model errors or operating condition perturbations in the optimization calculation, was found to have a robustness problem. Complete dissolution of seed crystals may occur because of the temperature swing operation, although the temperature swing, if appropriately applied, can significantly reduce the amount of fine crystals. The robust optimal trajectory, which was obtained from a robust particle swarm optimization calculation by considering model errors, outperformed the nominal trajectory model, avoiding complete dissolution in all 50 000 cases in a Monte Carlo experiment. Furthermore, *k*-means clustering analysis was applied to the simulation data to reveal that the performance was still sensitive to initial perturbations in the operating conditions, i.e., the initial super-saturation and seed size.

1. Introduction

Batch crystallization is used extensively in the chemical and pharmaceutical industries for separation and purification, to produce small-volume, high-value-added specialty chemicals. Control of the crystal size distribution is an important issue because it has a significant effect on the efficiency of down-stream operations such as filtration and drying.

Open-loop temperature profiles that yield desired crystal size distributions have been extensively studied. Mullin and Nývlt (1971) derived a profile that gives seeded systems with constant nucleation and growth rates. They experimentally confirmed that the derived programmed cooling produces crystals with a larger average size compared with those obtained by uncontrolled natural cooling. Jones (1974) derived a size-optimal-controlled cooling profile, which aims at maximizing the terminal size of the seed crystals based on the moment transformation model of the population balance. He experimentally showed that the size-optimal cooling policy results in an improved terminal mean crystal size compared with those obtained using previous operating policies such as natural cooling, linear cooling, and a constant nucleation rate. Since then, many researchers have derived optimal cooling profiles for such objectives as minimization of coefficients of variation, the amount of nucleus-grown crystals, and maximization of seed growth. Ward et al. (2006) summarized the optimal cooling policies for these common objectives.

Although most of those studies assumed monotonic decreasing temperature trajectories for ease of operation, some experimental studies (Moscosa-Santillán et al., 2000; Takiyama and Sindo, 2002; Harner et al., 2009; Bakar et al., 2009) have achieved significant improvements in the crystal size distribution by incorporating crystal dissolution into the batch operation. It has also been shown that the influence of temperature swings is further enhanced if the crystallization kinetics is size dependent (Shoji et al., 2011; Nagy et al., 2011; Jiang et al., 2014a). Generally, smaller crystals dissolve

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faster, but larger crystals grow faster; this implies that temperature swing operations are advantageous for reducing the amounts of crystal fines.

However, controlled cooling, defined here as a more sophisticated temperature trajectory compared with simple cooling such as linear and natural cooling implemented in an open-loop fashion, has severe robustness problems (Bohlin and Rasmuson, 1992; Ma et al., 1999; Nagy and Braatz, 2004, 2012; Seki et al., 2012). Bohlin and Rasmuson (1992) showed through extensive simulation studies that the performance of controlled cooling is very sensitive to model errors and perturbations, especially to perturbations in the initial super-saturation. The effects of using controlled cooling and seeding are highly unpredictable without appropriate kinetics and very accurate control; controlled cooling may even produce significantly smaller product crystals. Ma et al. (1999) provided a systematic robustness analysis for batch cooling crystallization of potassium nitrate, and they showed that approximately 50% of the expected benefits of optimal control could be lost as a result of temperature control implementation uncertainties as small as 0.1 °C; this is very challenging for an industrial crystallizer. A recent review of optimization and robustness issues in crystallization can be found in Nagy and Braatz (2012).

In this study, we derived robust optimal temperature swing trajectories for minimizing the amount of fines through numerical simulation studies. Robust optimal temperature trajectories have already been derived for some sets of uncertain parameters in the crystallization model (Ma et al., 1999; Nagy and Braatz, 2004), but we consider perturbations in the initial super-saturation for the robust optimization problem, which has a significant effect on the product size distribution, as discussed by Bohlin and Rasmuson (1992) (changes in solubility may typically occur in practice because of the presence of inorganic salts or other contaminants in the feedstock, Nagy et al., 2008). First, the nominal optimal temperature trajectories were calculated, using the particle swarm optimization (PSO) method (Kennedy and Eberhart, 1995), to show the advantage of temperature swing operations. A robust optimization problem, which took model errors and operating condition perturbations into account, was then introduced to find robust optimal temperature trajectories. Finally, a Monte Carlo simulation was performed to evaluate the robust performances of the obtained optimal temperature trajectories.

This paper is organized as follows. In Section 2, a batch cooling crystallizer model and its operating scenario are introduced; these are used for the optimization and simulation calculations in the following sections. In Section 3, a nominal optimization problem, which does not consider any model perturbations, is formulated to find a cooling profile that minimizes the amount of fine crystals. In Section 4, a robust optimization problem is considered, which incorporates model and operating condition perturbations. Section 5 compares the robust performance of the nominal and robust optimal trajectories obtained in the previous sections through a Monte Carlo experiment. Furthermore, cluster analysis is applied to the data obtained in the Monte Carlo experiment to identify the effects of model perturbations on the robust performance. Finally, conclusions and future work are given in Section 6.

2. Mathematical model and operating scenario

2.1. Mathematical model

A population balance model for a batch crystallizer with one characteristic length L is described by (Rawlings et al., 1993)

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

subject to the initial condition:

$$f(L,0)=f_0(L), \tag{2}$$

and the boundary condition:

$$f(0,t)=\frac{B(S,\mu_3(t))}{G(0,S)}, \tag{3}$$

where $f(L,t)$ is the population density function, $G(L,S)$ is the size-dependent linear growth rate which is also a function of the relative super-saturation S, $f_0(L)$ is the initial population density of the seed crystals, and $B(S,\mu_3(t))$ is the rate of nucleation which is a function of S and the third moment $\mu_3(t)$ of the size distribution. The relative super-saturation S and the ith moment of the distribution are defined respectively by

$$S=\frac{C(t)-C^*(T(t))}{C^*(T(t))}, \tag{4}$$

$$\mu_i(t)=\int_0^\infty L^i f(L,t)\,dL, \tag{5}$$

where $C(t)$ and $T(t)$ are the solute concentration and the crystallizer temperature at time t respectively, and $C^*(T(t))$ is the saturation concentration at temperature $T(t)$. The solute concentration $C(t)$ is determined by the material balance and is described by

$$C(t)=C(0)-\rho_c k_v\{\mu_3(t)-\mu_3(0)\}, \tag{6}$$

where ρ_c is the density of the crystals, k_v is the volumetric shape factor.

The kinetics of crystal birth, growth, and dissolution are given by

$$B(S,\mu_3(t))=k_b S^b \mu_3(t) \tag{7}$$

$$G(L,S)=\begin{cases} k_g S^g(1+\alpha_g L)^{\beta_g} & S\geq 0 \\ k_d S(1+\alpha_d L)^{\beta_d} & S<0 \end{cases}. \tag{8}$$

The model parameters, which are those for a potassium nitrate/water system (Chung et al., 1999), are listed in Table 1. It should be noted that the dissolution kinetics as well as the size dependence of the growth and dissolution kinetics are fictitious. The dissolution rate parameter is set so that the dissolution rate is of the same order of magnitude as the growth rate, and a first-order dependence on the degree of under-saturation is assumed (Noyes and Whitney, 1897; Lu et al., 1993; Gu et al., 2002). The size dependence parameters are determined so that the assumed size dependence is not far from the reported values for other substances (Mydlarz and Jones, 1993; Shoji et al., 2011; Jiang et al., 2014a).

2.2. Operating scenario

The assumed scenario for crystallizer operation is as follows. Initially, the crystallizer is loaded with a feed solution whose saturation temperature is T_{feed}. At the beginning of the batch, the crystallizer is cooled to a temperature below the saturation temperature and then

Table 1
Crystallizer model parameters (Chung et al., 1999).

Nucleation parameter k_b (#/s/g-water)	4.64×10^5
Nucleation parameter b	1.78
Growth parameter k_g (m/s)	1.1612×10^{-4}
Growth parameter g	1.32
Growth parameter α_g (1/m)	1000
Growth parameter β_g	0.5
Dissolution parameter k_d (m/s)	1.16×10^{-4}
Dissolution parameter α_d (1/m)	1000
Dissolution parameter β_d	−1.0
Density ρ_c (kg/m^3)	2.11×10^3
Volumetric shape factor k_v	1
Solubility $C^*(T(t))$ (g/g-water)	$0.129+0.00588T(t)+0.000172T(t)^2$

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