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Evaluation of controlled cooling for seeded batch crystallization incorporating dissolution

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ABSTRACT

A process control strategy to obtain large product crystals in batch cooling crystallization is investigated through simulation studies. Optimal cooling profiles, which incorporate crystal dissolution by temperature raise, are obtained for various seeding temperatures, with the potassium nitrate/water system as an example. When dissolution of crystals is allowed, sensitivity of the resulting average crystal size to the seeding temperature may be diminished, but the open-loop controlled cooling is still vulnerable to operational perturbations such as shifts in solubility and feed concentration. Feedback control may be a prerequisite to improve the reproducibility of the product crystal size distribution.

Feedback control schemes based on the second moment (μ_2-control) and concentration measurement (C-control) are introduced. The reference tracking control manipulates the crystallizer temperature so that the sensor measurement closely follows the optimal trajectory obtained from the off-line optimization calculation. The early stage control scheme is proposed in which the μ_2 feedback control is applied only at the early stage of the batch, while the rest of the batch is left open-loop. Robustness of the control schemes is evaluated through extensive simulation studies, and it is found that the early stage μ_2 control is capable of providing robust performance despite its simpler implementation.

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 crystal size distribution (CSD) is an important issue in batch crystallization because it has a significant effect on the efficiency of the down stream operations such as filtration and drying.

Traditionally, for batch cooling crystallization, an open-loop temperature profile which yields desired crystal size distribution has been extensively studied. Around 40 years ago, Mullin and Nývlt (1971) derived a temperature profile that gives a constant nucleation and growth rate of the seed. They experimentally validated that such a programmed cooling produces crystals with a larger average size compared with an 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 over those from the previously known operating policies such as natural cooling, linear cooling and constant nucleation rate. Since then, many researchers derived optimal cooling profiles for such objectives as minimization of coefficients of variation, the amount of nucleus-grown crystals, maximization of the seed growth, etc. Ward et al. (2006) summarized the optimal cooling policies for the common objectives.

However, the controlled cooling has been known to exhibit severe robustness problems, when implemented in an open-loop fashion (Bohlin and Rasmuson, 1992; Ma et al., 1999; Nagy and Braatz, 2004). Bohlin and Rasmuson (1992) showed through extensive simulation studies that even the qualitative effects of applying controlled cooling and seeding are highly unpredictable without appropriate kinetics and very accurate control; the controlled cooling may produce even significantly smaller product crystals. Rawlings et al. (1993) pointed out the necessity of feedback control for batch crystallization. Ma et al. (1999) provided a systematic robustness analysis result for batch cooling crystallization of potassium nitrate, and they showed that approximately 50% of the expected benefits of optimal control could be lost due to temperature control implementation uncertainties as small as 0.1 °C, which is very challenging for an industrial crystallizer.

Besides the manipulation of the temperature profile, seeding has been known as one of the most effective manipulated variables for

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controlling the product crystal size distribution, although not much attention has been paid in the optimal control context. Jagadesh et al. (1996, 1999) showed that one can easily obtain mono-sized product crystals even under natural cooling if a sufficient amount of seed is loaded. When a seed is provided above some critical amount, which is defined as a *critical seed loading* (Kubota and Onosawa, 2009), temperature profiles have negligible effects on the product crystal size distribution (Doki et al., 2001). The experimental work for the glycine/water system by Chew et al. (2007a) clearly shows that the cooling profiles, concave, linear, or convex, have only marginal effects on the size distributions of product crystals. Practically, if one wishes to suppress generation of fines, seeding would be the easiest and robust solution without the need to resort to sophisticated temperature control techniques.

The effect of seeding on the product crystal size can be clearly illustrated on the seed chart (Kubota and Onosawa, 2009), in which the volume mean size L_p of the product crystals normalized with the size of seed crystals L_s is plotted as a function of the seed loading ratio C_s with the seed size L_s as a parameter. The volume mean size L_p is calculated as

$$L_p = \frac{\mu_4}{\mu_3}, \tag{1}$$

and the seed loading ratio C_s is defined as the mass ratio of the added seeds to the theoretical crystal yield.

Fig. 1 shows an example of the seed chart for the potassium nitrate/water system under the three cooling profiles: convex, linear, and concave profiles. For smaller seed loadings, the convex cooling profile provides the largest crystal growth and the concave gives the smallest. As the seed loading ratio becomes larger, the differences due to the cooling profiles become unpronounced and the growth lines asymptotically approach the ideal one; the ideal growth is defined as the crystal growth in which the secondary nucleation can be neglected and only the seed grows. In such a case, the crystal growth can be calculated through the simple mass balance as

$$\frac{L_p}{L_s} = \left(1 + \frac{1}{C_s}\right)^{1/3}. \tag{2}$$

Kubota and Onosawa (2009) showed an empirical expression for the critical seed loading ratio

$$C_s^* = 2.17 \times 10^{-6} L_s^2. \tag{3}$$

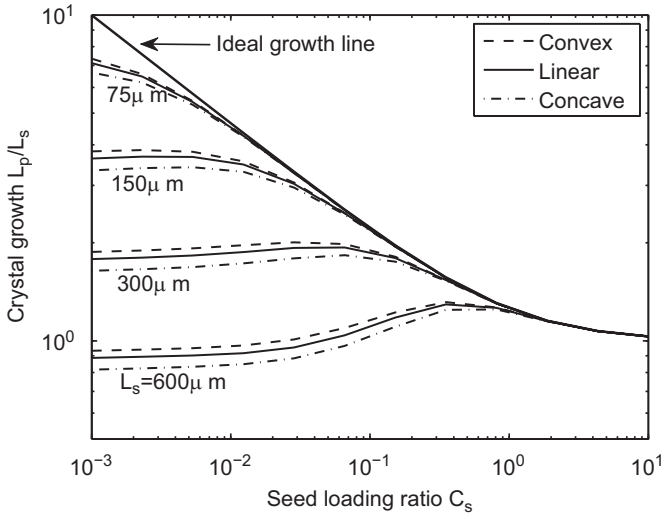

Fig. 1. Seed charts for various temperature profiles: the potassium nitrate/water system.

Since the critical seed loading ratio is determined by the available surface area (Lung-Somarriba et al., 2004), with the second order dependence on the average size of seed crystals as shown in (3), it becomes sometimes difficult to secure enough seed loading for large size seeds. For example, the critical seed loading ratio in the potassium nitrate/water system for $L_s = 300\ \mu m$ is estimated as $C_s^* = 20\%$ according to Fig. 1 and (3), which may be too large; the practical amount of seed is normally between 0.1% and 3% of the final mass of crystals (Choong and Smith, 2004). Too high a seed loading may limit the yield, and consequently lower the productivity, because an undesirably high final slurry concentration has to be avoided.

The use of controlled cooling, that is, sophisticated manipulation of temperature profile during the batch, to reduce secondary nucleation may be justified only when providing the critical amount of seed crystals is difficult in such cases as a desired crystal size is large so that large size seed crystals have to be used. Also, there should be cases where the generation of high quality seed with uniform and reproducible CSD may be difficult. It is known that seed quality has a large effect on the product CSD (Aamir et al., 2010).

In fact, the discussion so far assumes that the crystallizer temperature is a monotonically decreasing function of time, that is, no temperature increase is allowed, so that the dissolution or disappearance of crystals may not be considered. However, experimental works (Moscosa-Santillán et al., 2000; Takiyama et al., 2002; Harner et al., 2009; Bakar et al., 2009) have shown significant improvement in the crystal size distribution if dissolution of crystals is incorporated in the batch operation.

In this paper, optimal temperature profiles are derived through numerical optimization, which incorporate dissolution of crystals by temperature swing during the batch. The potassium nitrate/water system is used as an example (Rawlings et al., 1993; Chung et al., 1999). Since disappearance of crystals has to be considered, the optimization calculation is performed on the basis of explicit solution of a population balance model. Although the moment transformation model of the population balance has been the choice in the previous optimization studies because of its calculation efficiency, it is difficult to apply in this study due to the possibility that disappearance of crystals may occur. Recently, Nagy et al. (2011) provided for the first time an optimal cooling profile based on the population balance model considering dissolution, growth, and nucleation mechanisms. This paper will further make critical evaluation on the robustness of such cooling profiles.

To improve robustness of the controlled cooling, feedback control is introduced. Recent progress in sensor technologies has made available various measurements for more advanced operations of crystallizers (Lewiner et al., 2002; Fujiwara et al., 2002; Doki et al., 2004; Worlitschek and Mazzotti, 2004; Chew et al., 2007b; Nagy et al., 2008; Woo et al., 2009). As a controlled variable, the second moment of the CSD, which may be available through turbidimetry (Moscosa-Santillán et al., 2000; Harner et al., 2009) and laser diffraction measurement (Neuman and Kramer, 2002), is mainly employed in this study. To make the controller implementation as simple as possible, "early stage control" is also proposed, in which the feedback control is applied only at the early stage of the batch. Extensive simulations are performed to evaluate robustness of the feedback controllers.

This paper is organized as follows. In Section 2, a batch cooling crystallizer model is introduced which will be used for optimization and simulation calculations in the following sections. In Section 3, optimal cooling profiles which incorporate dissolution of crystals are obtained by solving nonlinear optimization problems, and robustness issues with their open-loop implementation are elaborated. Furthermore, feedback control schemes based