Computers and Chemical Engineering 99 (2017) 271–279

Contents lists available at ScienceDirect

Computers and Chemical Engineering

journal homepage: www.elsevier.com/locate/compchemeng

Comparison of objective functions for batch crystallization using a simple process model and Pontryagin's minimum principle

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ARTICLE INFO

Article history:
Received 24 June 2016
Received in revised form 9 January 2017
Accepted 11 January 2017
Available online 16 January 2017

Keywords:
Batch crystallization
Pontryagin's minimum principle
Optimal control theory

ABSTRACT

In this contribution different objective functions based on the moments of the product crystal size distribution are compared using optimal control theory to solve for the optimal batch trajectory for each objective. For a simple crystallization process model with only nucleation and ordinary crystal growth, and neglecting the contribution of the nucleated mass to the nucleation rate and material balance, mostly analytic expressions are obtained for the optimal control vector. Different objective functions lead to different final values for the costates, which lead to different sets of coupled differential and algebraic equations which must be solved to determine the values of constants numerically. The results of nine different objective functions for three crystal systems are presented. The objective functions based on the lower moment of the nucleated crystals lead to late-growth trajectories while the objective functions based on the higher moment of the nucleated crystals lead to early-growth trajectories, consistent with previous findings. The effect of seed loading is also investigated.

1. Introduction

Batch crystallization is an important and widely-used operation in chemical engineering for the separation and purification of solid products. The quality of crystalline products is primarily determined by the crystal polymorph, crystal shape and crystal size distribution. Achieving a desired or optimal product crystal size distribution is important for many applications in industrial crystallization and has been the subject of considerable study. Most of the research on the operation of batch crystallization has focused on the question of how the supersaturation (which may depend on the batch temperature, evaporation rate or rate of anti-solvent addition) should change with time during the batch to maximize or minimize the value of some objective function.

Because the supersaturation changes continuously during the batch, the problem is a continuous optimization problem. The most widely-used method for solving this problem is control vector parameterization (Kraft, 1985; Schlegel et al., 2005), which has been applied to solve a number of complicated problems including optimization with aggregation (O'Ciardha et al., 2012), crystal shape evolution (Acevedo et al., 2015; Liu et al., 2013; Ma et al., 2002), enantiomer separation (Angelov et al., 2008) polymorphic phase transition (Sheikholeslamzadeh and Rohani, 2013), simultaneous cooling and antisolvent addition (Nagy et al., 2008) and imperfect mixing (Ma et al., 2002). Several review articles (Nagy and Braatz, 2012; Nagy et al., 2013) discuss control of batch crystallization more generally.

A number of researchers have applied Pontryagin's minimum principle to determine optimal trajectories for batch crystallization over the years (Jones 1974; Ajinkya and Ray 1974; Morari 1980; Corriou and Rohani 2008). Raisch and coworkers (Vollmer and Raisch 2003; Vollmer and Raisch 2006; Hofmann and Raisch, 2010; Bajcinca et al., 2010) proposed a transformation of the standard method of moments model which permits a nearly analytical solution to the optimal control problem to be determined for a simple crystallization process model including only nucleation and ordinary growth (no size-dependent growth or growth-rate dispersion). The nearly analytical nature of the result permits greater understanding of the nature of the solution than was previously possible. For example, they proved mathematically that when certain relationships hold between parameters in the nucleation and growth kinetic models and the objective function is to minimize the nucleated mass, the growth rate trajectory always passes through a minimum and reaches the maximum constraint near the end of the batch. Further extensions and applications of the method were presented by Bajcinca and coworkers (Bajcinca and Hofmann 2011; Bajcinca 2013).

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http://dx.doi.org/10.1016/j.compchemeng.2017.01.017
0098-1354/

Notation

B	Nucleation rate (#/m^3 s)
C	Concentration (kg/m^3)
C_s	Seed loading ratio (−)
C_s^*	Critical seed loading ratio (−)
C_{sat}	Saturated concentration (kg/m^3)
f	Crystal size distribution function (#/m^3 m)
G	Crystal growth rate (m/s)
k_b	Nucleation parameter (#/m^3 s)
k_g	Growth parameter (m/s)
k_v	Volumetric shape factor (−)
L_p	Product volume mean size (m)
L_s	Seed volume mean size (m)
m_s	Seed mass (kg)
n_0	Number of seed crystals (#/m^3)
S	Relative supersaturation (−)
t_f	Final time (s)
w	Solvent mass (kg)
W_s	Seed mass (kg)
W_{th}	Theoretical crystal yield (kg)
x_0	Seed mean size (m)
μ_i	ith moment of the crystal size distribution (m^i/m^3)
ρ_c	Crystal density (kg/m^3)

An important consideration in the formulation of any optimization problem is the selection of objective function, and this is particularly true for batch crystallization, where subtle differences in the choice of objective function can have a significant effect on the outcome (Chung et al., 1999; Ma et al., 2002; Ward et al., 2006; Hsu and Ward, 2013). In some instances, the best choice for the objective function will be clear because of regulatory requirements or other factors, but in other instances the engineer must choose the objective function using his or her own best judgement. Although it is not the only possibility, most researchers formulate objective functions based on the moments of the crystal size distribution.

Chung et al. (1999) and Ma et al. (2002) were among the first to call attention to the effect of the objective function on the results of the optimization in batch crystallization processes. They considered several different objective functions, including some that were based on lower moments of the crystal size distribution and others that were based on higher moments of the crystal size distribution and found significantly different trajectories for different objective functions. Ward et al. (2006) surveyed objective functions considered by different authors and argued that objective functions based on lower moments of the nucleated mass lead to so-called early growth trajectories, while those based on higher moments of the nucleated crystals or any moment of the seed-grown crystals lead to late-growth trajectories. Later, Hsu and Ward (2013) compared the results of using different objective functions and argued that minimizing the nucleated mass was the most appropriate objective function in most circumstances, but in that work the optimization problem was solved using control vector parameterization and therefore the results were entirely numerical.

Besides the effect of the supersaturation trajectory, researchers have also investigated the effect of seed loading (Jagadesh et al., 1999; Kubota et al., 2001; Doki et al., 2002; Hojjati and Rohani, 2005; Tseng and Ward, 2014). Generally increasing the seed mass and decreasing the seed size is found to improve performance, and in some cases nucleation can be suppressed almost entirely by suitable seeding.

In the present contribution, the method of Hofmann and Raisch (2010) is applied to determine nearly-analytical expressions for the optimal supersaturation trajectory in seeded batch crystallization for nine different objective functions. Because the results are mostly analytical, they can be more readily understood and compared. Because the choice of objective function has a profound effect on the results of the optimization, it is necessary for the engineer to understand the effect of the choice of objective function on the resulting supersaturation trajectory and product crystal size distribution. Without this understanding, the engineer may choose an objective function that leads to an undesirable result.

The remainder of this article is organized as follows: In the next section, some mathematical theory related to batch crystallization processes and optimization is reviewed, including a brief summary of the method of Hofmann and Raisch and modifications necessary to solve the problem for different objective functions. In the third section, results are presented. Comparison is made between the optimal trajectories for different objective functions for three different crystallization systems, and the effect of seed loading is also investigated. Finally, conclusions are presented in the final section.

2. Theory

In this section a model for a batch crystallization process is introduced, the method of Hofmann and Raisch is briefly reviewed and modifications necessary to consider different objective functions are presented. Results are illustrated using three complete batch crystallization models taken from the literature: a model for the crystallization of potassium nitrate from water by Miller and Rawlings (1994) (also used by Chung et al. (1999)), a model for the crystallization of succinic acid from water by Qiu and Rasmuson (1991) and a model for the crystallization of pentaerythritol from water by Bernardo and Giulietti (2010).

2.1. Batch crystallization model

In this work a standard moment model that describes a batch crystallization system is considered:

$$\frac{d\mu_0}{dt} = B \tag{1}$$

$$\frac{d\mu_i}{dt} = iG\mu_{i-1} i = 1, 2, \ldots \tag{2}$$

Where G is size independent crystal growth rate (m/s) and B is nucleation rate (#/m^{3}s). The definition of the moments is:

$$\mu_i = \int_0^\infty L^i f(L) dLi = 0, 1, 2\ldots \tag{3}$$

The driving force for crystal nucleation and growth is the supersaturation. The relative supersaturation is given by:

$$S = \frac{C - C_{\text{sat}}}{C_{\text{sat}}} \tag{4}$$

where C and C_{sat} are the solute concentration and saturation solute concentration in units of kg/kg solvent. The following empirical expressions can be employed for the nucleation and growth rates:

$$G = k_g S^g \tag{5}$$

$$B = k_b S^b \mu_3{}^j \tag{6}$$

where μ_3 is the third moment of crystal size distribution. An expression for a mass balance on the solute is

$$\frac{dC}{dt} = -3G\rho_c k_v \mu_2 \tag{7}$$