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Deterministic and Stochastic Optimal Control for Batch Cooling Crystallization

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Abstract

Minimization of operation costs and the enhancement in product quality have been major concerns for all industrial processes. The field under study here is batch crystalllization which is affected heavily by the uncertainities in measurements and other errors. The current work aims to study Deterministic and Stochastic methods for Optimum Control in batch crystallization. All the methods involve maximising an objective function by manipulating the cooling profile. At first, the Deterministic approach uses experimental kinetic parameters, which is then extended to Stochastic optimization to incorporate uncertainities in them. Lastly, a novel approach named Polynomial Chaos Expansions is implemented which has been applied successfully to other domains for Nonlinear Model Predictive Control but was not explored in detail in the field of batch cooled crystalllization. It successfuly includes probability distributions for the parameters into the model to provide a more robust optimization srategy.

This work analyses various optimization approaches for the batch crystallization process that are robust to model error. First, The key challenge in addressing robustness to model error is to propagate the uncertainty in model parameters onto the control or optimization objective.

Keywords: Stochastic Optimal Control, Polynomial Chaos Expansions, Robust Optimization, Batch Crystallization, Predictive Control, Optimum Temperature Profile

1. Introduction

Numerous industries today, such as pharmaceutical, chemical, photographic etc. employ the batch crystallization process for the preparation of crystalline products with high degree of purity. A common goal of each crystallization process is to obtain a narrower Particle size distribution (PSD) of the desired product. The PSD has a strong influence on the downstream processing and, hence, reproducible PSD in each operation is of prime importance. Thus, finding an effective control strategy to obtain the resulting crystals with a desired Crystal Size Distribution becomes significant in order for improving the performance of both the batch crystallization process and the subsequent processes which depend on it.

Crystallization is the (natural or artificial) process where the atoms or molecules are highly organized into a solid structure known as a crystal. Some of the ways which crystals form are through precipitating from a solution, melt or more rarely deposited directly from a gas. In order for crystallization to take place a solution must be "*super-saturated*". **Supersaturation**(ΔC) is a condition in which the solute concentration in the solution is higher than the solubility. It acts as the driving force for the crystallization process and affects the final quantity of product formed. It is mathematically expressed as :

$$\Delta C = C - C_s \tag{1}$$

where C_s is the concentration of the solute in the saturated solution. In the following work, the method in focus is cooling crystallization in which superstauration magnitude is determined by the cooling rate. Thus, determination of an optimal cooling rate or a temperature trajectory becomes the objective of the currrent study.

This work formulates and analyses various control strategies for a cooling crystallization process represented through the population balance equation. **Deterministic Optimal Control** aims at finding the an optimum temperature profile to maximise an objective function selected to achieve a desired volume of the product. Herein, the experimental kinetic parameters are employed to simulate a batch crystalllization process. **Stochastic Optimal Control** undertakes the task of quantifying the uncertainites which creep in due to experimentation. It aims to achive a maximum expected value for the desired product, simultaneously incorporating randomness in the process parameters into the model. Namely, two methods **Ito Process** and a novel approach **Polynomial Chaos Expansions** are employed for this purpose.

Most of the reported works in the this field deal with the determination of optimal temperature or supersaturation trajectory for the batch crystallizer. The concept of programmed cooling in batch crystallizers was first discussed by Mullin and Nyvlt [**?**] in 1971. Later, in 1974, A. G. Jones [**?**] presented a mathematical theory based on moment transformations of population balance equations. He used the continuous maximum principle to predict optimal cooling curves. Rawlings et al. [**?**] discussed issues in crystal size measurement using laser light scattering experiments and optimal control problem formulation. In 1994, Miller and Rawlings [**?**] discussed the uncertain bounds on model parameter estimates for a batch crystallization system. ost importantly optimal temperature prediction for batch crystallization has also been done by Hu et al.hu, Shi et al.[**?**], Paengjuntuek et al.[**?**], and Corriou and Rohani.[**?**], the data and knowledge from which have been used in further work in this project.

Stochastic modeling of particulate processes and parameter estimation using the experimentally measured particle sizes has attracted many researchers. Grosso et al.[**?**] presented a stochastic approach for modeling PSD and comparative assessments of different models. Ma et al.[**?**] presented a worse-case performance analysis of optimal control trajectories by considering features such as the computational effort, parametric uncertainty and control implementation inaccuracies. Monte Carlo simulations have also been used to propogate uncertainities but often present the problem of high computational demand, for which approximations are proposed. Nagy and Braatz (2007) [**?**] have shown that Polynomial Chaos Expansions(PCE) is a computationally efficient alternative to Monte Carlo simulations for propagating uncertainty in dynamic models. PCE is based on orthogonal basis functions thus requiring smaller function evaluations for the calculation of numerical integrations needed for obtaining statistical moments. The computational advantages of PCEs for robust control and optimization has been shown by Nagy and Braatz [**?**], Kim et al.[**?**], Kumar and Budman[**?**].

The focus of the current research activity is to incorporate parametric uncertainties in the mathematical formulations of batch crystallization process for building a robust model. In the deterministic approach, kinetic parameters from the experimental data have been used to model the system. Next, stochastic Ito processes are used to assimilate the errors in the experimental data. Finally, PCE are demostrated as an effective and novel method to achieve the desired objective function value. A case study of an unseeded crystallization process is also included to authenticate the methodology.

2. Mathematical Background

Analysis of a particulate system seeks to synthesize the behavior of the population of particles and its environment from the behavior of single particles in their local environments. The population is described by the density of a suitable extensive variable, usually the **number of particles**, but sometimes by other variables such as the mass or volume of particles. The usual transport equations expressing conservation laws for material systems apply to the behavior of single particles. Particulate processes are characterized by properties such as particle shape, size, surface area, mass, and product purity.
A population balance formulation describes the process of crystal size distribution with time most effectively. Thus, modeling of a batch crystallizer involves the use of population balances to model the crystal size prediction and the mass balance on the system can be modeled as a simple differential equation having concentration as the state variable. The population balance can be expressed as eq :

$$\frac{\partial n(r,t)}{\partial t} + \frac{\partial G(r,t)n(r,t)}{\partial r} = B \tag{2}$$

where **n** is the number density distribution, **t** is the time, **r** represents the characteristic dimension for size measurements, **G** is the crystal growth rate, and **B** is the nucleation rate. Both growth and nucleation processes describe crystallization kinetics, and their expression may vary, depending on the system under consideration.

In this work, the system under consideration is potassium sulfate, which has been studied earlier by Hu et al. [**?**], Shi et al. [**?**], and Paengjuntuek et al.[**?**].

Nucleation kinetics$^{(5-7)}$ are defined by :

$$B(t) = k_b \exp\left(-E_b/RT\right)\left(\frac{C - C_s(T)}{C_s(T)}\right)^b \mu_3 \tag{3}$$

Growth Kinetics$^{(5-7)}$ are given by:

$$G(t) = k_g \exp\left(-E_g/RT\right)\left(\frac{C - C_s(T)}{C_s(T)}\right)^g \tag{4}$$

where k_b and k_g are constants of the system, E_b and E_g are activation energies, and b and g are exponents of nucleation and growth, respectively. $C_s(T)$ is the saturation concentration at a given temperature. The following equations are used to evaluate the saturation and metastable concentrations corresponding to the solution temperature T (expressed in units of °C)[**?**].

$$C_s(T) = 6.29 \times 10^{-2} + 2.46 \times 10^{-3}T - 7.14 \times 10^{-6}T^2 \tag{5}$$

$$C_m(T) = 7.76 \times 10^{-2} + 2.46 \times 10^{-3}T - 8.1 \times 10^{-6}T^2 \tag{6}$$

The mass balance, in terms of concentration of the solute in the solution, is expressed as :

$$\frac{dC}{dt} = -3\rho k_v G(t)\mu_2(t) \tag{7}$$

where ρ is the density of the crystals, k_v the volumetric shape factor, and μ_2 is the second moment of particle size distribution (PSD). Since $n(r,t)$ represents the population density of the crystals, the i-th moment of the particle size distribution(PSD) is given by :

$$\mu_i = \int_0^\infty r^i n(r,t)dr \tag{8}$$

The above equations along with the Population Balance Equation(PBE) represent a complete model of a seeded batch crystallizer. Population balance equations are multidimensional, which poses a problem with their implementation in complex control functions, hence use of a model order reduction becomes imperative.

For simplification, we reduce the population balance equations into **Moment balance equations** which has been estabilished as an efficient method by Yenkie et al.[**?**]. This is done by multiplying the equation (2) with r^i on both sides to generate the expression given by equation (8). Converting the model into Ordinary differential equations proves to be advantageous, since it is difficult and time-consuming to formulate an optimization problem involving PBEs. Thus, the moment method leads to a reduced-order model given by Equations (15-24).
Separate moment equations are used for the seed and nuclei classes of crystals, and they are defined as :

$$\mu_i^n = \int_0^{r_g} r^i n(r,t)dr \tag{9}$$

$$\mu_i^s = \int_{r_g}^\infty r^i n(r,t)dr \tag{10}$$

n in the superscript represents the nucleated crystal whereas **s** stands for the seeded crystal, $\boldsymbol{r_g}$ gives the critical radius separating the two. The moment equations for nucleated and seeded crystals become as follows[**?**] :

1. Nucleated crystals[**? ?**]

$$\frac{d\mu_0^n}{dt} = B(t) \tag{11}$$

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

2. Seeded crystals[**? ?**]

$$\frac{d\mu_0^s}{dt} = G(t) \tag{13}$$

$$\frac{d\mu_i^s}{dt} = iG(t)u_{i-1}^n(t) \quad i = 1, 2, 3 \tag{14}$$

The total moment is obtained as the summation $\mu_i^t = \mu_i^n + \mu_i^s$. The complete set of differential equations are [**?**] :

$$\frac{dy_1}{dt} = -3\rho k_v G(t)(y_4 + y8) \tag{15}$$

$$\frac{dy_2}{dt} = 0 \tag{16}$$

$$\frac{dy_3}{dt} = G(t)y_2 \tag{17}$$

$$\frac{dy_4}{dt} = 2G(t)y_3 \tag{18}$$

$$\frac{dy_5}{dt} = 3G(t)y_4 \tag{19}$$

$$\frac{dy_6}{dt} = B(t) \tag{20}$$

$$\frac{dy_7}{dt} = G(t)y_6 \tag{21}$$

$$\frac{dy_8}{dt} = 2G(t)y_7 \tag{22}$$

$$\frac{dy_9}{dt} = 3G(t)y_8 \tag{23}$$

$$\tag{24}$$

Here the state variables y_i are given by :

$$y_i = \begin{bmatrix} C & \mu_0^s & \mu_1^s & \mu_2^s & \mu_3^s & \mu_0^n & \mu_1^n & \mu_2^n & \mu_3^n \end{bmatrix}$$

3. Deterministic Optimal Control

3.1. Case Study : Seeded batch crystallization

In this section, we derive an optimal control strategy for a batch crystallization process in which seed crystals have been introduced to aid the formation of the product.

4. References

[1] ...