Robust Optimal Temperature Operation for Seeded Batch Crystallization Processes

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Abstract— **Robust optimal temperature trajectories for seeded batch crystallization processes under size-dependent crystallization kinetics are proposed, which minimize the amount of fine crystals. First, a deterministic optimal temperature-swing trajectory is calculated by Particle Swarm Optimization (PSO). Furthermore, robust trajectories are proposed by considering parametric perturbations in the process model and initial conditions. Two robust versions of PSO, named AMPSO and WMPSO, are introduced to cope with the uncertain optimization problem. The result of Monte Carlo simulation demonstrates that the optimal trajectories are capable of producing fine-minimized crystals with high robustness against initial operating condition changes.**

I. INTRODUCTION

Batch and semi-batch operation is important and popular mode of operation to produce high quality and value-added product in the food, chemical, fine chemical, and pharmaceutical industries. Batch and semi-batch processes are preferred because of their better flexibility and smaller capital cost. Although the economics have not been the dominant factor in batch and semi-batch production and there have been only a few attempts in the industry to optimize their operations through mathematical modeling and optimization techniques, competition is clearly making the cost factor an important issue and the potential gain of applying these techniques could be significant [1, 2].

Crystallization is a common but important unit operation to produce solid phase products through separation and purification. In batch crystallization, crystal size distribution of final products is desired to be optimized, since it always significantly influences the quality of crystalline product and the efficiency of downstream process operations such as filtration, washing and drying.

Traditionally, the optimization of seeded batch crystallization processes has been performed for several surrogate objective functions based on some properties of crystal size distribution, although it is difficult to relate the downstream processing cost directly to the size distribution. According to the previous work of Ward [3], the optimization objective of the crystal size distribution at the end of batch was summarized into four common categories: (1) minimize the "amount" of nucleus-grown crystals; (2) maximize the "average size" of the total crystals; (3) minimize the "variation in size" of the total crystals; (4) grow the seed crystals as "large" as possible.

Besides the single objective problem, Sarkar et al. [4] investigated various constrained multi-objective optimization problems which are related to the quality of final crystal size distribution by applying an adapted GA (Genetic Algorithm) code. Another work by Nagy [5] solved a single nucleation kinetic uncertainty optimization problem by utilizing designed shapes of both monomodal and bimodal crystal size distribution as the optimization objective for batch crystallization. However, it has been found that some optimized temperature profiles exhibit severe robustness problems when there are changes in operating conditions or crystallization kinetics [6, 7].

Traditional discussion on temperature operation is focused on monotonically decreasing crystallizer temperature, which means no temperature increase is allowed during batch process. An experimental study conducted by Barkar et al. [8] demonstrated that temperature rise operation is capable of improving final crystal size distribution. In terms of simulation works, Seki et al. [9] proposed feedback control schemes based on the second moment measurement incorporating temperature rise operation. Moreover, under size-dependent crystallization kinetics, the temperature swing operation is known to be particularly effective for improving the size distribution [10, 11].

In this work, optimal temperature operation to minimize fine part of final crystal size distribution for seeded batch crystallization processes is proposed and investigated through simulation studies. Deterministic and robust optimal temperature trajectories, which incorporate both crystal growth and dissolution by temperature swings, are obtained through a potassium nitrate/water system as an example. By considering it as an uncertain optimization design problem, PSO (Particle Swarm Optimization) and its robust versions have been adapted to find the optimal trajectories. The Monte Carlo simulation result demonstrates that the PSO and its robust variants are capable of generating reliable and robust optimal solutions to minimize fines for the batch crystallization model under both deterministic and uncertain environments.

The remaining of this paper is organized as follows: in Section 2 we introduce the mathematical model for seeded batch cooling crystallization. In Section 3 a brief overview of uncertain optimization problem, PSO and the proposed robust variants is presented, while results of numerical optimization and analysis of optimized profiles are given in Section 4. Finally, in Section 5 we give some concluding remarks of this work.

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II. Mathematical Model of Seeded Batch Crystallization

A population balance model for crystallization processes with the characteristic length L can be described as the following partial differential equation:

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

subject to the initial and boundary conditions:

$$F(L,0) = F_0 \tag{2}$$

$$F(0,t) = \frac{B(t)}{G(0,t)}, \tag{3}$$

where $F(L,t)$ is the population density function, $G(L,t)$ represents size-dependent growth and dissolution rate of crystals. F_0 represents initial population density of seed crystals charged at the beginning of batch. $B(t)$ represents the nucleation rate. Secondary mechanism nucleation is assumed and the nuclei are considered arbitrarily small. The i-th moment μ_i of the population density function F is given by:

$$\mu_i(t) = \int_0^\infty L^i F(L,t)dL. \tag{4}$$

Material balance for the solute concentration $C(t)$ and the relative supersaturation $S(t)$ are defined as:

$$C(t) = C(0) - \rho_c k_v \big(\mu_3(t) - \mu_3(0)\big), \tag{5}$$

$$S(t) = \frac{C(t) - C_{sat}(T)}{C_{sat}(T)}, \tag{6}$$

where ρ_c is the crystal density, k_v is the volumetric shape factor, and μ_3 is the third moment. $C_{sat}(T)$ represents the saturation concentration at temperature T and T is a function of operating time t.

Since crystal dissolution is incorporated, the nucleation rate and growth rate are described as:

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

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

A potassium nitrate/water system is investigated as a case study [13], and details of parameters are shown in Table I. The parameters concerning the dissolution and size dependency are hypothetical. Note that the parameters concerning crystal growth and dissolution are determined as $\alpha_g > 0$, $\beta_g > 0$ and $\alpha_d > 0$, $\beta_d < 0$ to ensure larger crystals grow faster and smaller crystals dissolve faster.

Generally, (1) can be solved by the method of moments. However, the population balance equation is directly solved in this study, since it is difficult to incorporate crystal disappearance due to dissolution into the method of moments. To solve the population balance numerically, the MATLAB code provided by Ward et al. [12], which is based on the space time conservation element and solution element (CE/SE) method, is modified to account for crystal disappearance due to dissolution.

TABLE I. PARAMETER VALUES OF POTASSIUM NITRATE/WATER CRYSTALLIZATION MODEL

Parameter	Value
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
Dissolution parameter k_b(m/s)	1.16×10^{-4}
Density ρ_c(kg/m³)	2.11×10^3
Volumetric shape factor k_v	1
Growth parameter α_g	1×10^3
Growth parameter β_g	0.5
Dissolution parameter α_d	1×10^3
Dissolution parameter β_d	-1
Solubility $C^*(T)$(g/g-solve)	$0.129 - 0.00588T + 0.000172T^2$

III. Uncertain Optimization Problem and PSO Algorithm

A. Uncertain Optimization Problem

So far, most studies on engineering optimal design problems have been mainly focused on locating the global optima using deterministic models. However, in many real-world engineering optimization problems, uncertainties are often present. If the solution is very sensitive to small perturbation either in the design variables or operating conditions, it often leads to undesirable final performance. It is considered necessary to take uncertainties into account during optimization.

A constrained optimization problem is formed as follow:

$$\text{Maximize:} \quad f(x) \tag{9}$$

$$\text{Subject to:} \quad x_{low} \leq x \leq x_{up} \tag{10}$$

where $f(x)$ is a fitness function, x_{low} and x_{up} are the lower and upper bounds of design variable vector x. In general, it is possible to classify uncertainties in an optimization problem into three main categories: (1) intrinsic uncertainties from fitness function; (2) uncertainties in design variables; (3) uncertainties from fluctuations in operating conditions [17].

In seeded batch crystallization processes, the initial conditions always change from batch to batch and have significant effects on the final quality of product crystals, hence we may regard this uncertain problem of batch crystallization as Category (3), which can be formed as:

$$\text{Maximize:} \quad F(x) = f(x, s + \delta) \tag{11}$$

where $s = [s_1, s_2, \cdots, s_n]$ is the nominal value vector of the operating parameters such as initial solute concentration, and δ denotes a random vector representing the uncertainties.

B. Particle Swarm Optimization

To solve many complex engineering design problems, PSO has been widely studied as an evolutionary computational algorithm [17]. In the standard PSO introduced by Kennedy and Eberhart [16], many individuals move around in a D-dimensional research space, each representing a possible solution to a numerical problem, and each individual memorizes its own position and velocity $X = [X_1, X_2, \cdots, X_D]$ and $V = [V_1, V_2, \cdots, V_D]$, as well as the spot $pbest$ where the

individual has acquired the best fitness value. Furthermore, all of them share the best-fitness spot information $gbest$ of the whole swarm.

At k-th iteration, the velocity of each individual is updated according to their best encountered position and the swarm best-fitness spot in the following way:

$$V_{k+1} = \omega V_k + c_1 r_1(pbest_k - X_k) + c_2 r_2(gbest - X_k) \quad (12)$$

where c_1 and c_2 are the acceleration constants which usually are set as 2, r_1 and r_2 are independent random numbers from 0 to 1. ω is the inertia weight showing the impact of previous velocities on the next move. A suitable value ω is important to balance the global and local exploration abilities. Therefore, a general and reliable strategy used here suggests linearly decreasing ω during the iteration. Furthermore, if the velocity exceeds a prefixed limit, another restriction V_{max} is used to keep the individuals inside the search space.

Meantime, the position is updated according to the new individual velocity by generation and expressed by the equation:

$$X_{k+1} = X_k + V_{k+1} \quad (13)$$

C. PSO for Uncertain Optimization

To solve the uncertain optimization problem for our batch crystallization process, a multiple-evaluation method has been proposed and combined with evolutionary computational approaches. Basic idea of this approach is replacing fitness value by the average or worst case value among m new intermediate model evaluations as perceived fitness which reflects several random vector uncertainties $\delta = [\delta_1, \delta_2, \cdots, \delta_m]$ in the original model [14, 17].

According to AVS (Average Value Scheme), the perceived fitness is calculated by the average fitness of all m perturbed models, i.e. $average\{f(x, s + \delta_1), f(x, s + \delta_2), \cdots, f(x, s + \delta_m)\}$. On the other hand, for WCS (Worst Case Scheme), the perceived fitness is chosen as the worst among m perturbed evaluations, i.e. $worst\{f(x, s + \delta_1), f(x, s + \delta_2), \cdots, f(x, s + \delta_m)\}$. It is worth noting that the worst case would represent the minimum for a maximization problem or maximum on a minimization problem.

It is possible to adapt multiple-evaluation method into PSO algorithm. In this work, we incorporated both AVS and WCS into a standard PSO code and proposed two robust versions of multiple-evaluating PSO, AMPSO and WMPSO, to cope with the uncertain optimization problem in seeded batch crystallization. The procedure is shown as follows:

Step 1: Initialize each swarm individual by random position X and velocity V.

Step 2: Evaluate the perceived fitness values of all particles through m multiple-evaluations according to AVS or WCS then initialize $pbest$ of each individual and the $gbest$.

Step 3: Update the velocity V and position X of each individual according to (12) and (13).

Step 4: Update the perceived fitness values of all particles according to AVS or WCS.

Step 5: Update $pbest$ of each particle. Determine the current best individual which performs the best perceived fitness value then update the current $gbest$.

Step 6: If the stopping criterion is met, then output $gbest$ and its objective fitness value; otherwise go back to Step 3.

IV. OPTIMIZATION PROBLEM SETTING FOR SEEDED BATCH CRYSTALLIZATION

A. Deterministic Optimization Problem

In order to repress the generation of fine crystals at the end of batch, the optimization objective is considered to minimize the fine part, where crystal size is less than 500μm, of the final third moment crystal size distribution. The deterministic optimization problem can be expressed as:

Minimize: $\mu_{3_{L \leq 500\mu m}}(T)$ (14)

Subject to: $T_{min} \leq T(t_n) \leq T_{max}, \ n = 1, 2, \ldots, N$ (15)

$$T_0^{min} \leq T(t_0) \leq T_{max} \quad (16)$$

$$R_{min} \leq dT(t)/dt \leq R_{max} \quad (17)$$

$$T(t_{cool}) = T(t_{end}) = T_{fin} \quad (18)$$

where T_{min} and T_{max} are the lower and upper temperature limits, and T_0^{min} is the lower limit of seeding temperature. From t_{cool} to the end of batch t_{end}, crystallizer temperature is kept constant as T_{fin} to ensure that the system is not supersaturated at the end of the batch. R_{min} and R_{max} are lower and upper rates of temperature change. To simplify the infinite dimensional nonlinear problem to a finite dimensional problem, N temperature changing points are determined as the design variables, while the entire profile is defined as $T = [T(t_0), T(t_1), \cdots, T(t_{N-1}), T(t_{cool}), T(t_{end})]$.

The use of sieved seed crystals is assumed and its size distribution is assumed to be parabolic with the mean size L_s and width w as shown in Fig.1 and it is used as the initial condition F_0. Details of parameter settings in this problem are listed in Table II.
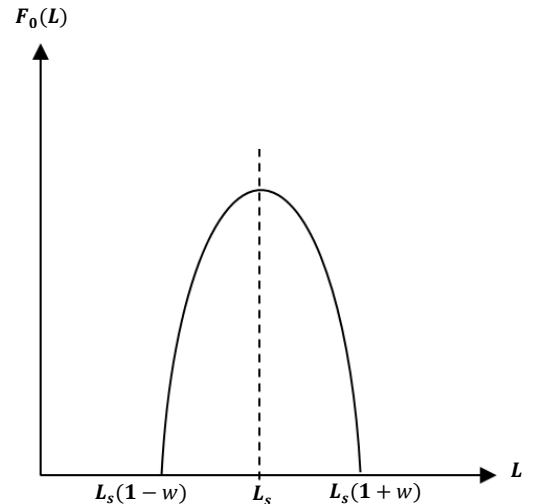

Figure 1. Size distribution of the seed crystals.

TABLE II.   PARAMETER SETTINGS OF OPTIMIZATION PROBLEM

Parameter	Value
Lower bound of temperature T_{min}(°C)	18
Upper bound of temperature T_{max}(°C)	30.5
Lower bound of initial temperature T_0^{min} (°C)	27
Saturation temperature of the feed T_{feed} (°C)	30
Final temperature T_{fin} (°C)	20
Lower bound of temperature changing rate R_{min}(°C/min)	-0.5
Upper bound of temperature changing rate R_{max} (°C/min)	0.5
Cooling duration t_{cool} (min)	120
Batch time t_{batch} (min)	150
N temperature point parameter	15
Seed loading ratio C_s (%)	2.0
Average seed size L_s (μm)	200
Distribution width w (μm)	50

B. Uncertain Optimization Problem

Since the product quality by the optimal trajectory can be very sensitive to small errors and fluctuations in the initial conditions of a batch, a robust optimal profile which has high tolerance is needed.

An uncertain optimization problem has been formed, by considering the initial operating perturbations. According to AVS and WCS, objective functions of both average and worst fitness evaluation are given as follows:

Minimize: $\frac{1}{m}\sum_{i=1}^{m} \mu_{3L\leq500\mu m}(T, s + \delta_i)$ (19)

Minimize: $\max \mu_{3L\leq500\mu m}(T, s + \delta_i)$ (20)

where δ_i $(i = 1,2,\cdots m)$ represents parametric perturbations of batch crystallization processes. The assumed uncertainties and their magnitudes are shown in Table III, which are all assumed as under uniform distribution. m is the number of samples evaluated for one solution.

TABLE III.   ASSUMED MODEL FLUCTUATIONS AND ERRORS

Parameter name	Magnitude (%)
Growth rate parameter k_g	±10
Dissolution rate parameter k_d	±10
Nucleation rate parameter k_b	±10
Solubility [a]	±2
Initial feed concentration	±2
Average size of the seed crystals	±10

a. ±2% of $C^*(T_{feed})$ is added as a bias to the solubility curve.

V. OPTIMIZATION RESULTS

A MATLAB MEX-file is used to solve the population balance equation. For the deterministic and uncertain optimization calculations, the particle swarm population is preset as 40, and it is determined as a general set that parameters are $c_1 = c_2 = 2$ and the inertia weight ω decreases linearly from 0.6 to 0.4 during the first 3500 iterations then keeps constant at 0.4 for 500 more iterations since this strategy proved to be able to give PSO algorithm better exploration and exploitation abilities to solve the optimization problem.

A. Deterministic Optimal Temperature Trajectory

Deterministic optimization simulation has been conducted for the nominal process parameters in Table 1. Figure 2 shows the obtained deterministic optimal temperature trajectory calculated by the standard PSO algorithm. Figure 3 shows the results of crystal volume distribution at the end time of the batch subject to the optimal temperature swing profile. The fitness value, the final third moment of the crystal size distribution whose size is smaller than 500μm, is minimized to 0.0437 m^3 , and an ideal monomodal crystal volume distribution of products is obtained, which implies that by using the optimal temperature swing trajectory almost all the final product crystals have grown from the seed crystals.

Figure 4 shows the time evolution of the third moment value of crystals whose size is less than 500μm, demonstrating how fine crystals are successfully eliminated by the temperature-swing operation. Figure 5 shows the total third moment value during the batch. The plot shows that although fine crystals dissolve because of the temperature rise, the entire crystal volume is enlarged to high-quality final product.
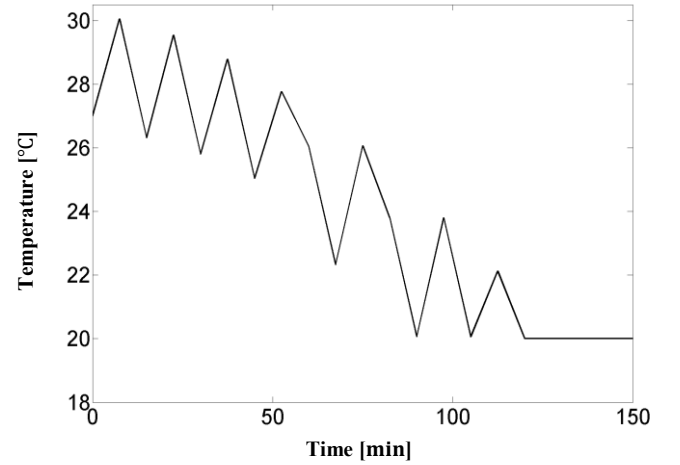

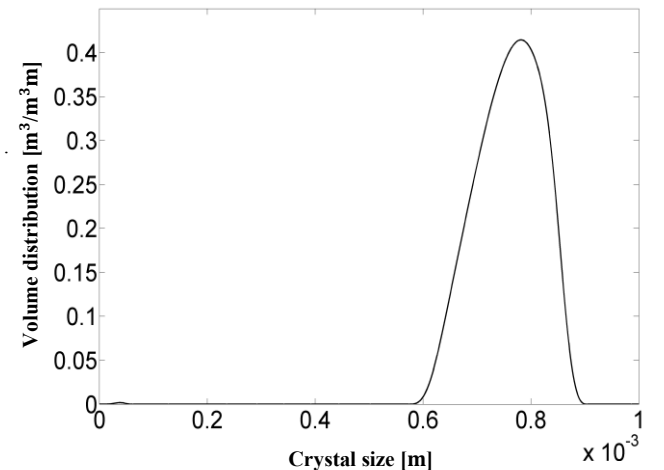Figure 2. Deterministic optimal temperature trajectory

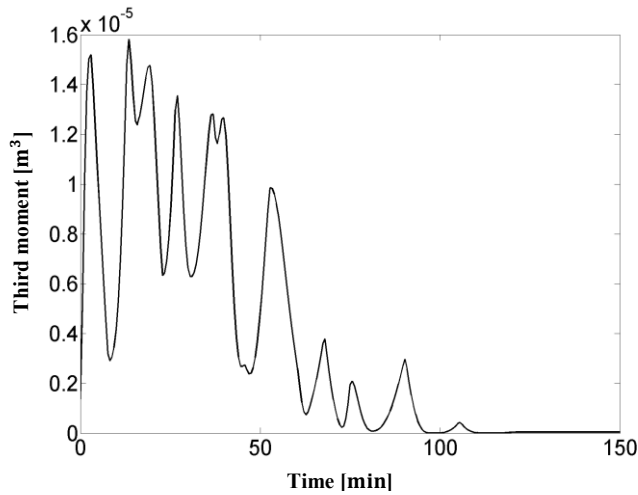Figure 3. Final crystal volume distribution calculated by the deterministic optimal trajectory

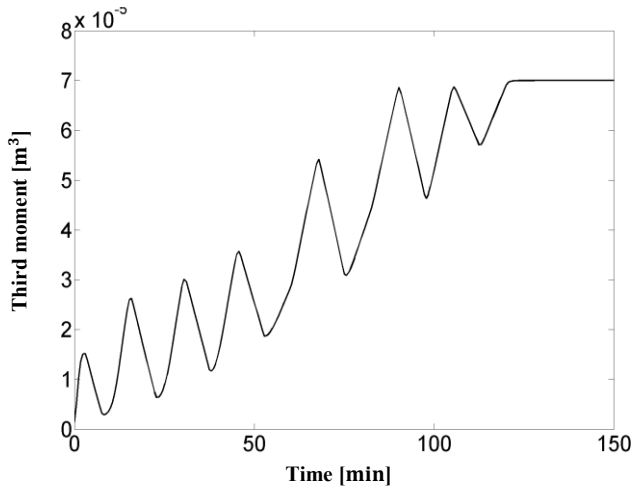Figure 4. Time evolution of the third moment value for crystals smaller than 500 μm.

Figure 5. Time evolution of the third moment value for all-size crystals

B. Robust Optimal Temperature Trajectory

Crystal size distribution of final product is sensitive to initial operating perturbations. Hence, to improve robust performance of batch operations, proposed AMPSO and WMPSO are applied to the uncertain problem subject to the assumed parametric perturbations given in Table III. For both AVS and WCS cases, multiple-time m in (19) and (20) are set to $m = 3$ and 10. Since calculation of the population balance model through evolutionary algorithm is extremely time-consuming, here we use the Parallel Computing Toolbox in MATLAB to shorten the computing time. However, it is still computationally expensive, e.g. it takes about 120h solving AVS ($m = 10$) case by incorporating 12 parallel workers on Intel Core i7-3930k (OC to 4GHz). Figure 6 compares robust optimal trajectories through AMPSO and WMPSO.

It is worth noting that the worst case of seeded batch crystallization process is complete dissolution of crystals during temperature swing operation; all seed-growth crystals

disappear and intractable primary nucleation should appear, which usually leads to extremely low quality product. In this study, when the complete dissolution occurred, the simulation calculation was terminated since primary nucleation is not modeled.

Figure 7 compares the robustness of both deterministic and robust optimal trajectories by 50000-case Monte Carlo experiments. The Latin hypercube sampling method is used to generate the random uncertainties. The abscissa represents the base 10 logarithm value of the third moment of crystals which are smaller than 500μm. Comparing with deterministic result, AMPSO and WMPSO results showed high-tolerance towards initial process perturbations. The best improvement by robust trajectories is that the worst case of complete crystal dissolution is completely avoided while it occurs 2781 times under the deterministic trajectory. Statistical results are given in Table 4 which shows AMPSO solutions can produce smaller amounts of fines with higher possibility than WMPSO solutions.

Surprisingly, the performances by m=3 are already comparable with those by m=10 for both AMPSO and WMPSO. AMPSO($m = 3$) appears to be the most appropriate method in our case studies because it is the least time-consuming profile to calculate and it yields the smallest mean value of final fine crystal.

VI. CONCLUSION

This paper describes an approach to optimize seeded batch crystallizer operations with size-dependent crystallization kinetics. Temperature-swing operation which minimizes the amount of fine crystals is obtained through optimization calculations.

Since final properties of crystals are very sensitive to initial operating condition changes such as shifts in solubility and feed concentration, uncertain optimization problems are also considered. After investigating the deterministic optimal trajectory, robust optimal profiles are calculated through AMPSO and WMPSO, and their performances are compared. The temperature trajectories obtained by solving the uncertain optimization problems are found to be insensitive against operating condition perturbations and capable of eliminating the worst case of complete crystal dissolution, which is not possible with the deterministic optimal trajectory.

TABLE IV. STATISTICAL RESULTS OF MONTO CARLO EXPERIMENTS

Trajectory	Mean value	Standard deviation	Worst cases occurred time
PSO [a]	-0.7669	1.5756	2781
AMPSO(m =3)	-0.5873	1.0160	0
AMPSO(m =10)	-0.3455	0.9546	0
WMPSO(m =3)	0.3836	0.8340	0
WMPSO(m =10)	0.2272	0.7080	0

a. Analysis of deterministic PSO trajectory does not include the crystal complete dissolution cases.

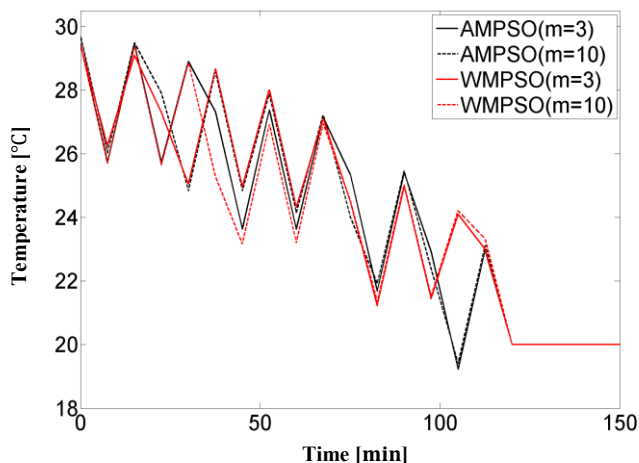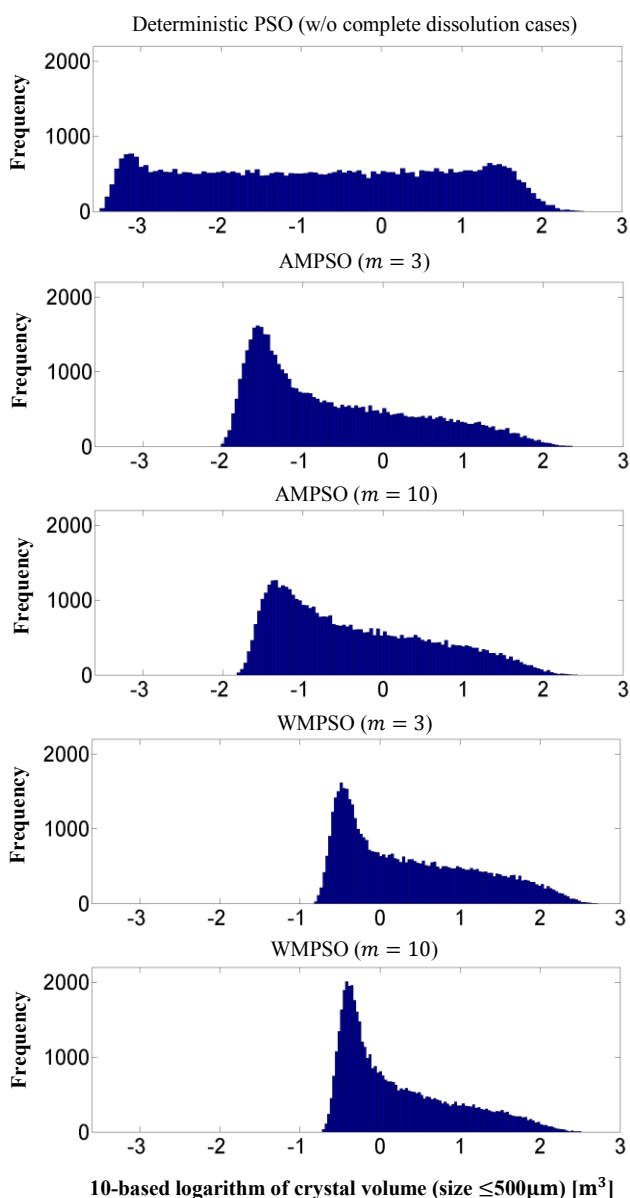Figure 6. Robust optimal temperature profiles

10-based logarithm of crystal volume (size ≤500μm) [m³]

Figure 7. Robustness evaluation of proposed optimal trajectories. 5000 cases.

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