

# Integrated Optimization Based on Transition Tracking Analysis for Batch Processes

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**Abstract:** Process optimization is an important issue for raising product quality and ensuring safety in batch processes and it is usually conducted at the point only when set-points are reached. In this work, process dynamic shift results from set-point tracking control, termed as transition tracking process, is detailedly analyzed in an integrated optimization framework, which also synthesizes the multi-stage characteristic of batch processes. Establishment of the regression relationship between process variables and quality indexes becomes possible and gradient directions are updated iteratively. The efficacy of the proposed scheme is illustrated on the injection molding, which is a typical multi-stage batch process.

**Keywords:** multi-stage batch processes, quality optimization, process control, iterative learning control, model predictive control

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## 1. INTRODUCTION

Batch and semi-batch processes play a significant role in the processing of a broad range of high-value-added products to meet the drastic competition market, such as specialty chemical products, semiconductor chips, plastics products. As a key factor of reducing production costs, improving product quality and meeting safety requirements, optimization technologies have been widely used in batch processes. In general, researchers in the field of automation refer to optimization operation by adjusting one or several key manipulated variables through some clever manner or on the basis of a process model (Bonvin, 1998). In practice, accurate mathematical process models are difficult to establish because of complicated physicochemical and mechanical characteristics. Thus, model-based optimization methods may not work effectively for the discrepancies between the simplified models and the real case.

In order to overcome the limitations, measurement-based optimization methods have been widely reported for batch processes. The essential relates to whether measurements are used on-line to drive the process towards the optimal strategy (Srinivasan et al., 2003). Bonvin et al. (2006) proposed an optimization scheme based on tracking appropriate reference through transforming the optimization problem into a control problem. It was an implicit model-based measurement optimization method had a close relationship with process knowledge or implicit model. Subsequently, model-free optimization methods have been developed in order to adjust process set-point just according to measurements. How to obtain accurate gradient information is an important issue to locate the optimum for model-free methods. DeHaan et al. (2005) gave an extremum-seeking control algorithm to

accelerate the convergence rate of finite difference method. Further, Srinivasan et al. (2007) utilized physical equipment to get multi-variable gradient information directly. However, this algorithm relied on the strong assumption that several identical units were available, which was unrealistic for industrial process. Further, Kong et al. (2011) presented an optimization method whose gradient information can be obtained through stochastic perpetuation of the process variables without the limitation of physical equipment.

However, the above methods are implemented under the implicit precondition that the set-point signal can be reached quickly. In fact, for a huge number of batch processes, the set-point tracking process will last for many batches mainly because of the unsatisfied controller performances. These batches see a gradual shift of variables steady-state from one to another and this dynamic procedure mainly caused by set-point tracking control is termed as transition tracking process (TTP) in this paper. During the TTP, to some intermediate batches, the real values of process variables have not reached the set-point yet. However, it may not necessarily mean poor-quality products. Sometimes these intermediate batches very likely bring better quality indexes because the initial set-points may not be the best. Unfortunately, all previous work conduct optimization only when the set-points are reached, which, however, did not explore the TTP information. Thus, we can make use of TTP to further explore the relationships between process variables and quality indexes to obtain better optimization results, i.e. to find more reasonable set-points. In addition, the multi-stage is an important characteristic of the batch processes. Here, the multi-stage is defined as steps occurring in different processing units and performing different unit operations (Undey, Cinar, 2002). Different stages have obviously different effects on product quality and

it is common that certain quality may be mainly affected by several key process variables in critical-to-quality stages (Lu, & Gao, 2006, Zhao, & Lu, 2014). Thus, stage based set-point adjustment may result in better quality control performance and less operation costs. To the best of the authors' knowledge, stage-based optimization method has not been explored.

Motivated by the aforementioned analysis, this paper devotes to develop an integrated optimization setting method by exploring TTP information and the multi-stage nature of batch processes. First, a process controller combing iterative learning control and model predictive control is designed to ensure every process variable in each batch run has a temporary steady-state during the TTP. And this makes it possible for the establishment of a regression relationship between process variables and quality indexes. Further, with the established regression relationship, a set-point iteration strategy is executed to obtain more accurate gradient information. Finally, the proposed method is validated in injection molding process.

## 2. METHODOLOGY

### 2.1 Optimization setting problem description

For batch processes, product quality is commonly available offline after batch completion. And during each batch run, quality variables are influenced by many factors (e.g. material characteristic, machine performance, controller performance). For batch processes, set-point values and controller performance are more concerned.

Consider batch processes that are subject to process variable constraints described by

$$\begin{aligned} \min_{(y_1^*, \dots, y_p^*)} \gamma &= f(\mathbf{y}) \\ \text{s.t. } y_{i\_min} &\leq y_i^* \leq y_{i\_max} \quad i \in [1, p] \end{aligned} \quad (1)$$

The vectors  $\boldsymbol{\gamma} \in \mathbb{R}^q$ ,  $\mathbf{y} \in \mathbb{R}^p$  denote the quality variables and process variables, respectively. Function  $f(\cdot)$  denotes an unknown regression function about  $\mathbf{y}$  and  $\boldsymbol{\gamma}$ . Optimal process variables values  $\mathbf{y}^* = (y_1^*, \dots, y_p^*)$  are computed under the lower constraint  $y_{i\_min}$  and upper constraint  $y_{i\_max}$  with  $f(\cdot)$ . Process variables measurements are assumed to be sampled at every fixed time interval, whereas the elements of quality variables are only obtained after batch completion.

### 2.2 The idea of transition tracking process

In practice, process variables commonly can not arrive at the given set-points within a single batch. Naturally, the set-point tracking process results into a process state shift procedure, TTP, during which process variables are controlled to their set-points after certain batches. Injection molding, a typical batch process, is employed to better understand the TTP. In Fig. 1a, seven batches are needed for packing pressure to reach its set-point, 35bar. These batches constitute a TTP, during which packing pressure is controlled to approach its set-point as close as possible. Data information about corresponding quality index, weight, are plotted in Fig. 1b.

From the TTP, we can easily discover a very important property that a smaller packing pressure results into a lighter part weight. And the lightest part weight is obtained in an intermediate batch, namely 2<sup>nd</sup> batch, rather than the 7<sup>th</sup> batch, where the packing pressure is 35bar. Thus, TTP may provide a guidance for process optimization.

In batch processes, usually, control actions are imposed on plants both along batch-wise and time-wise. It helps to eliminate tracking error and obtains faster response. However, if real values of process variables are continuously adjusted by controller along the time-wise direction, it will be difficult to distinguish the steady value for a process variable in each batch. Under this situation, the existing optimization methods take effect until entire process enters into a steady state. As a result, if we expect higher optimization efficiency, TTP should be considered properly into the optimization process.

### 2.3 An Integrated Quality Optimization Method

Subsequently, how to get and utilize the TTP information is a crucial issue. Here, we introduce an integrated optimization method, presented in Fig. 2, which consists of two major parts: lower process control part and upper optimum seeking part.

Lower process control part: A controller is designed to achieve batch-wise temporary reference tracking and time-wise error elimination. The temporary reference is automatically calculated according to controller performance and is a temporary target evolved batch to batch which finally equals to set-point at the end of a TTP. Thus, with the help of temporary reference, every process variable will enter into a steady-state within each batch.

Without loss of generality, assume  $K_\eta$  batches data are collected in the  $\eta^{th}$  TTP, note the process variables data matrix  $\mathbf{Y}_\eta$  in the  $\eta^{th} (\eta=1, 2, \dots)$  TTP is:

$$\mathbf{Y}_\eta = \begin{pmatrix} \mathbf{y}_1^\eta \\ \vdots \\ \mathbf{y}_{K_\eta}^\eta \end{pmatrix} = \begin{pmatrix} y_{1\_1}^\eta & y_{2\_1}^\eta & \dots & y_{q\_1}^\eta \\ \vdots & \ddots & & \vdots \\ y_{1\_K_\eta}^\eta & y_{2\_K_\eta}^\eta & \dots & y_{q\_K_\eta}^\eta \end{pmatrix} \quad (2)$$

where  $y_{i\_k}^\eta$  is the steady value of  $i^{th}$  process variable in the  $k^{th}$  batch during the  $\eta^{th}$  TTP.

Correspondingly, note quality variables obtained from offline assay in each batch during  $\eta^{th}$  TTP as  $\boldsymbol{\Phi}_\eta$

$$\boldsymbol{\Phi}_\eta = \begin{pmatrix} \boldsymbol{\gamma}_1^\eta \\ \vdots \\ \boldsymbol{\gamma}_{K_\eta}^\eta \end{pmatrix} = \begin{pmatrix} \gamma_{1\_1}^\eta & \gamma_{2\_1}^\eta & \dots & \gamma_{q\_1}^\eta \\ \vdots & \ddots & & \vdots \\ \gamma_{1\_K_\eta}^\eta & \gamma_{2\_K_\eta}^\eta & \dots & \gamma_{q\_K_\eta}^\eta \end{pmatrix} \quad (3)$$

Then, steady-values of process variables  $\mathbf{Y}_\eta$  and the quality variables  $\boldsymbol{\Phi}_\eta$  can be used to establish the local regression function  $\Phi_\eta(\cdot)$  to substitute  $f(\cdot)$  in a local region. Through analyzing data distribution, linear or nonlinear relationship can be identified.

Upper optimum seeking part: Within the variation scope of  $\mathbf{Y}_\eta$ , a local quality optimum  $\gamma_\eta^*$  and the corresponding set-

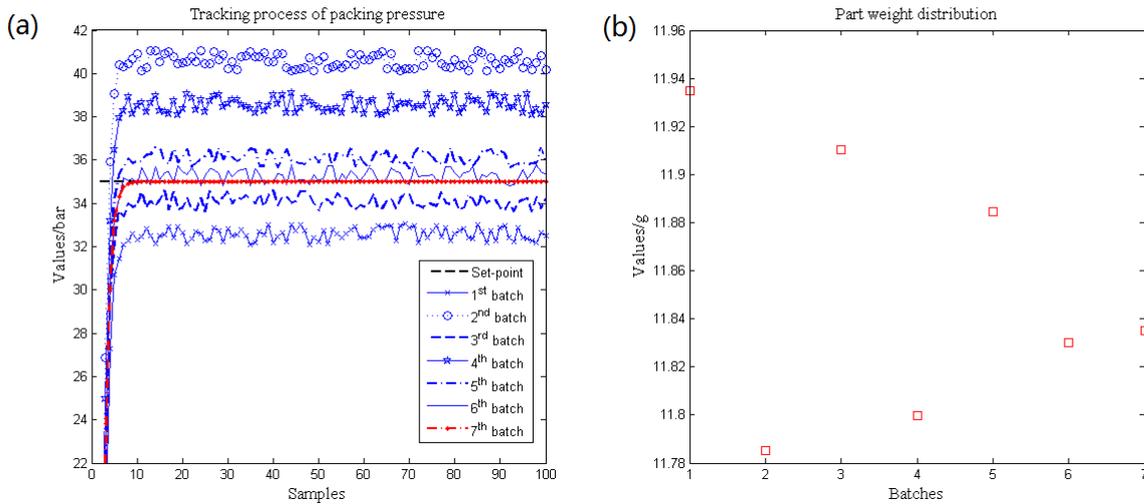


Figure 1. A TTP for injection molding proces

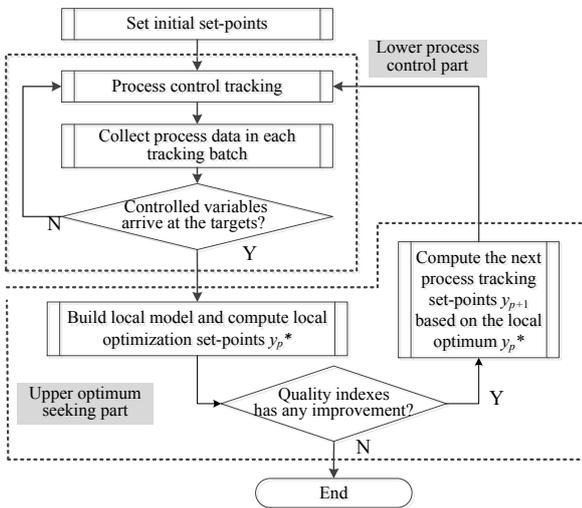


Figure 2. Flow chart of the optimization scheme

points  $\mathbf{y}_\eta^*$  can be computed via classical optimization algorithms. Calculate the gradient information at the local optimum point  $(\gamma_\eta^*, \mathbf{y}_\eta^*)$  based on the equation  $\gamma_\eta^* = \Phi_\eta(\mathbf{y}_\eta^*)$ . The next iteration point  $\tilde{\mathbf{y}}_{\eta+1}$  can be computed as

$$\tilde{\mathbf{y}}_{\eta+1} = \tilde{\mathbf{y}}_\eta - a_\eta \frac{\partial \Phi}{\partial \mathbf{y}} \Big|_{\mathbf{y}=\mathbf{y}_\eta^*} \quad (4)$$

where  $a_\eta$  is iterative step length in  $\eta+1^{th}$  TTP.

If product quality can not be improved after several TTP, the optimization procedure should be stopped. Otherwise, go to lower process control part for further iteration. Besides, some preparations are needed before the above contents, including select proper controller parameters, which will be given in section 2.4; analyze the critical stages and key variables about a certain product quality, the concrete algorithm can be found in Lu et al. (2006).

#### 2.4 Controller design based on iterative learning control and model predictive control

ILC is regarded as a close-loop control algorithm in batch-wise using last batch information to generate control outputs. However, it is still an open-loop controller in time-wise because no real-time feedback information is employed in current batch. In order to accelerate the convergence rate, predictive control algorithms have been integrated to improve the time-wise control performance within batch. As control performance is optimized along time-wise and batch-wise simultaneously, the real-value of process variables will be improved toward targets continuously. Under this situation, it is difficult to build a relationship between process variables and quality variables.

In this paper, we give a further consideration of controller design with the consideration of TTP. Set-point tracking error is mainly eliminated in batch-wise. Meanwhile, elimination rate is moderately adjusted to have more intermediate batches to obtain more process information. In time-wise, the concept "temporary reference (TR)" is introduced. TR is a temporary target in each batch during TTP and evolves batch to batch. It is an estimated value derived from the last batch and may have a great chance to be reached in the next batch. Thus, TR is a possible steady value for each batch during the TTP, which is

$$\tilde{y}_k^* = \frac{1}{N-1} \sum_{i=1}^N \hat{y}_k(t+i|t) \quad t \in [1, T_f] \quad (5)$$

where  $k$  denotes the batch index in batch-wise,  $N \in \mathbb{Z}^+$  is time-wise prediction horizon,  $t$  is the sample time and  $T_f$  is the termination times;  $\hat{y}_k(t+i|t)$  is process variable predictive value of  $y_k(t+i)$  at time  $t$  in  $k^{th}$  batch. If there are continuous three values  $(\Delta \hat{y}_k(t+i|t), \Delta \hat{y}_k(t+i+1|t), \Delta \hat{y}_k(t+i+2|t))$  fall into a small region, i.e. 0.05, we can say that the batch  $k$  enters into a steady state.

$$\Delta \hat{y}_k(t+i|t) = \hat{y}_k(t+i+1|t) - \hat{y}_k(t+i|t) \quad (6)$$

Consider a single-input single-out ARX model:

$$\begin{aligned} A(q^{-1})y_k(t) &= B(q^{-1})u_k(t) + v_k(t) \quad t \in [1, T], k = 1, 2, \dots \quad (7) \\ A(q^{-1}) &= 1 + a_1q^{-1} + a_2q^{-2} + \dots + a_nq^{-n} \\ B(q^{-1}) &= 1 + b_1q^{-1} + b_2q^{-2} + \dots + b_mq^{-m} \end{aligned}$$

where  $y_k(t)$ ,  $u_k(t)$  and  $v_k(t)$  present the output, input, and disturbance of the process at time  $t$  in  $k^{\text{th}}$  batch, respectively.

For above process, a P-type ILC is introduced:

$$\begin{aligned} u_{ilc\_k}(t) &= u_{ilc\_k-1}(t) + r_k(t) \\ r_k(t) &= L e_k(t) = L(y_d(t) - y_k(t)) \end{aligned} \quad (8)$$

where  $y_d(t)$  is the set-point to be tracked;  $y_k(t)$  represents the measurements in  $k^{\text{th}}$  batch;  $L$  is learning factor which affects the convergence rate in batch-wise. Combining (7) and (8) results in (9)

$$A(q^{-1})\hat{y}_k(t) = B(q^{-1})u_{ilc\_k}(t) \quad t \in [1, T_f], k = 1, 2, \dots \quad (9)$$

The cost function in time-wise is designed as:

$$\begin{aligned} J(t, p, d, k) &= \sum_{i=1}^N \alpha(i) (\hat{y}_k^*(t+i) - \hat{y}_k(t+i|t))^2 \\ &+ \sum_{j=1}^d \beta(j) (u_k(t+j))^2 \quad t \in [1, T] \end{aligned} \quad (10)$$

where  $d \in \mathbb{Z}^+$  is time-wise control horizon;  $\alpha(i)$ ,  $\beta(j)$  are weighting factors in the cost function.

In (10), TR instead of  $y_d(t)$  is used as the reference for time-wise control. The main purpose of (10) is to track the estimated output, TR, and resist the possible disturbance. Time-wise penalty in cost function is adjusted by a small parameter  $\beta(j)$  because a better intra batch tracking control performance is expected. However, this may make the time-wise sensitive to high-frequency components of the control error and disturbance (Bonvin, 1998). And a small  $L$  in ILC may weak this disadvantage through sacrificing convergence rate in batch-wise. If  $L < 1$ , the original error is shrunk and the TTP will become longer. While if  $L > 1$ , the error will be amplified which results into a rapid but may be unstable control effects. When process variable enters into an preset expected region,  $y_d(t) \pm \Delta$ , we suggest a small  $L$  is switched and more transition batches will be produced.

### 2.5 Set-point iteration policy

Most batch processes are multi-stage processes. Process variables in different stages have different effects on final product quality. During the same phase, process variables have a similar relationship. Moreover, quality attribute depends on the whole operation performance within the same stage from an overall viewpoint, rather than individual time interval (Lu, & Gao, 2005). Inspired by the phase-specific average process trajectory method (Zhao, Wang, & Gao, 2009), which aims at the same operation conditions and involves many batches in a certain phase  $c$ , a modified method coping with a certain transition batch is given.

In each batch, assume that process variables, the number is  $J_{xc}$ , are sampled at  $k=1, 2, \dots, K_c$  time instances in a certain phase  $c$  and a set of product quality measurements, the

number is  $J_y$ , are collected at the end of the production. Then  $\bar{y}_{kc}(i)$ , the average of the process variable  $y_i$  in phase  $c$  in  $k^{\text{th}}$  batch can be calculated as:

$$\bar{y}_{kc}(i) = \frac{1}{K_c} \sum_{k=1}^{K_c} y_i(k) \quad (11)$$

After  $I$  batches, process measurements array  $\mathbf{Y}_c(I \times J_{xc})$  and a corresponding quality data array  $\boldsymbol{\phi}(I \times J_y)$  can be organized. With  $\mathbf{Y}_c$  and  $\boldsymbol{\phi}$ , a guidable model can be established to find local optimum. Intuitively, exploration of local optimum will help to approximate the optimal iterative gradient and accelerate the convergence rate. The linear or nonlinear relationship between matrix  $\mathbf{Y}_c$  and  $\boldsymbol{\phi}$  is difficult to tell at first. So we can plot  $\mathbf{Y}_c$  and  $\boldsymbol{\phi}$  in a same chart to identify the relationship preliminarily. Here, iterative steps in linear relationship are given, and the nonlinear relationship which has a similar procedure is omitted for brevity.

In linear relationship, PLS algorithm is performed on  $\mathbf{Y}_c$  and  $\boldsymbol{\phi}$  to obtain the following equations:

$$\begin{aligned} \mathbf{Y}_c(I \times J_{xc}) &= \mathbf{T}_c \mathbf{P}_c^T + \mathbf{E} \\ \boldsymbol{\phi}(I \times J_y) &= \mathbf{T}_c \mathbf{Q}_c^T + \mathbf{F} \\ \mathbf{T}_c(I \times A_c) &= \mathbf{Y}_c \mathbf{W}_c (\mathbf{P}_c^T \mathbf{W}_c) \end{aligned} \quad (12)$$

where  $\mathbf{P}_c(I \times A_c)$  is loading matrix of  $\mathbf{Y}_c$ ;  $\mathbf{Q}_c(J \times A_c)$  is loading vector of  $\boldsymbol{\phi}$ ;  $A_c$  is the retained number of latent variables,  $\mathbf{W}_c(J \times A_c)$  is weighting matrix. The final phase-based PLS regression model for quality prediction can be deduced as

$$\hat{\boldsymbol{\phi}}_c(I \times J_y) = \mathbf{Y}_c \mathbf{W}_c (\mathbf{P}_c^T \mathbf{W}_c) \mathbf{Q}_c^T = \mathbf{Y}_c \mathbf{A} \quad (13)$$

After establishment of local model, the optimal minimum can be found by solving (1) (a typical linear programming algorithm or nonlinear programming problem) under the variation region of the process variables. Note  $\mathbf{y}_\eta^*$  as the local optimum, the next iteration point  $\mathbf{y}_{\eta+1}$  can be calculated according to (14).

$$\mathbf{y}_{\eta+1} = \mathbf{y}_\eta^* - a_\eta \mathbf{g}_\eta(\mathbf{y}_\eta^*) \quad (14)$$

where  $\mathbf{g}_\eta(\mathbf{y}_\eta^*)$  is the estimated gradient value of the iterative point  $\mathbf{y}_\eta$ . In the linear case, from (14),  $\mathbf{g}_\eta(\mathbf{y}_\eta^*)$  is  $\mathbf{A}$ .  $a_\eta$  is the iterative step in  $\eta^{\text{th}}$  TTP. Based on the local optimum exploration process, the next iterative direction will be more accurate because more information is used. Repeat this process until the product quality have few improvements.

## 3. EXPERIMENT RESULTS

### 3.1 Process description and optimization objective

A simplified diagram of a typical reciprocating-screw injection molding machine is showed in Fig. 3 (Yang, 2004). Injection molding is a multistage process, which mainly consists of filling, packing-holding, plastication and cooling stage. Initially with filling stage, the plastic melt is injected into the mold cavity at a certain speed. After the mold is filled with the plastic melt, injection is stopped, initiating the packing-holding stage. During this stage, melt flows are prevented out of the mold and the additional material is compacted into the mold to make up the shrinkage

associated with cooling and solidification. The process then switches to cooling stage during which the material is cooled inside the mold until it is rigid enough to be ejected. Happened in the early stage of the cooling, plastication stage sees the polymer melts and is conveyed to the front of the barrel by screw rotation. The machine is then ready for the next cycle.

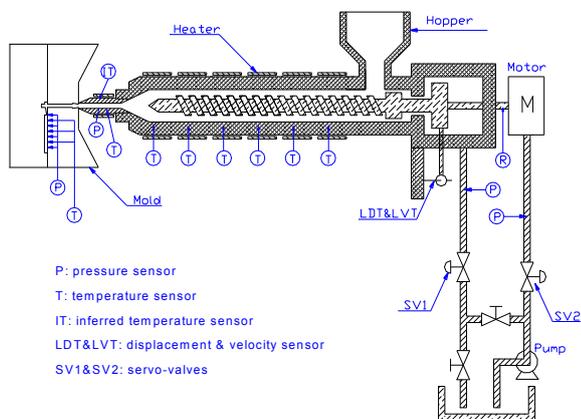


Figure 3. Injection molding process

In our study, part weight is used as optimization objective, which is an important factor affecting economic benefits and other product qualities. A light part weight under the insurance of other part quality indexes brings cost savings. Material used in the experiment is high-density polyethylene and the mold tested is an iPhone 4s case. As part weight is mainly determined by packing-holding stage, key controlled process variables, packing pressure  $y_1 \in [25, 30]$  and packing time  $y_2 \in [3, 7]$ , are selected. The operating conditions are set as follows: three-band barrel temperatures are set to be (200, 200, 200)°C; mold temperature is 60°C; cooling time is 15s; inject velocity equals 24mm/s.

### 3.2 Selection of tuning parameters

Through experiments, the identified model of packing pressure control in the packing-holding stage is:

$$Y(Z)/U(Z) = 12/(Z^2 - 0.8Z) \quad (15)$$

Considering experiment cost, 10-15 batches are expected during each TTP. According to rules in MPC algorithm given by Xi (2013), as the dynamic part of packing stage lasts almost 10 sample points, the prediction horizon  $N$  is selected as 20 steps and the control horizon  $d$  is 5 steps. According to the real tracking control performance, 0.2 is given to learning factor  $L$  at first several tracking batches. If the steady value of packing pressure enters into the region of  $y_d \pm \Delta$ , a smaller  $L$  is preferred. In this paper, the value of  $\Delta$  is defined to be two. In order to keep a fast convergence rate during each TTP and have a larger cover range, the iterative step-size is set to be  $2\Delta$ .

Packing time is another important variable affecting the part weight. However, packing time is a special process variable because it is controlled by an electronic timer which does

not have a transition process. Thus, we use finite difference method to obtain gradient information for packing time.

### 3.3 Results and Discussion

Using the scheme developed in previous parts, in this section, we present the results from implementing the part weight optimization and compare its performance against a method proposed by Kong et al. (2011). Under this situation, initial set-point of packing pressure and packing time are randomly set to 38bar and 5s, respectively.

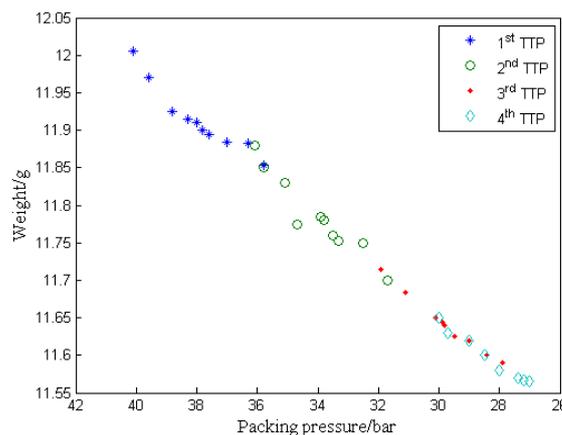


Figure 4. Part weight optimization with the proposed method

Table 1. The optimization results during TTP

No.	Part weight local optimum	Packing pressure local optimum
1	11.854	35.8
2	11.700	31.7
3	11.580	27.9
4	11.565	27.0

In Fig. 4, the initial part weight yielded from the proposed method decreased with the reduction of packing pressuring. It is a linear relationship which can be observed just from the first TTP. Thus, tools for solving linear programming problems can be employed to obtain the 1<sup>st</sup> local optimum. The local optimal part weight and corresponding set-point are presented in Table 1. In the 4<sup>th</sup> TTP, the part surface quality has some problems if the packing pressure less than 27bar. Though mainly determined by filling stage, surface quality is still influenced by other stages more or less. The steady value of packing pressure is presented in Fig. 5. In the bottom chart, the packing time is perturbed based on the finite difference algorithm. Through experiment, a small packing time will generate a smaller part weight. Finally, the obtained optimal part weight is 11.565g, which is 0.335g less than the initial part weight.

In Fig. 6, the part weight results obtained from the proposed scheme is compared with Kong's algorithm (Kong et al., 2011). With Kong's method, the optimal part weight is 11.73g. Obviously, there is an improvement in the proposed method. As the method proposed by Kong et.al adopts the standard SPSA algorithm (Spall, 1998), the iterative step will become smaller with the increase of batch number.

Thus, the method may be easy to fall into the local optimal value because it may not be able to jump out of the local region.

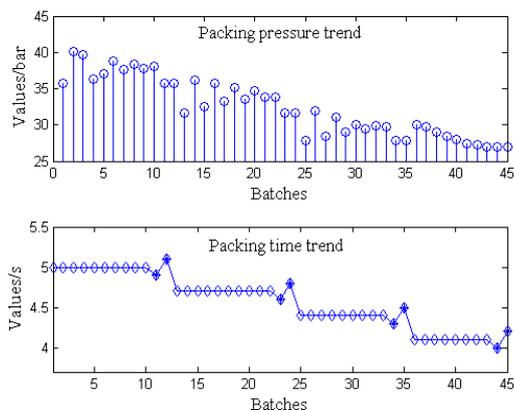


Figure 5. Packing pressure and packing time trend

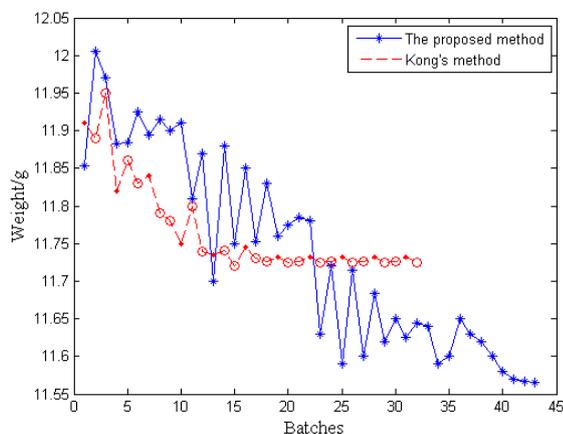


Figure 6. Comparison of part weight optimization results between two methods

#### 4. CONCLUSIONS

In this work, an integrated statistical optimization setting scheme has been proposed for batch processes by analysis of transition tracking process and inherent multi-stage characteristic. Via the controller combing the iterative learning control and the model predictive control, the transition tracking process is explored to establish regression relationship between process variables and quality indexes. The convergence rate is adjusted batch-wise by iterative learning control. The time-wise control performance is ensured by model predict control. A direct gradient exploration method is given based on the transition tracking process information. Guidelines on how to select parameters in controller configuration and set-point iteration procedure are also provided. Experiment results on injection molding demonstrated the performance of the proposed method.

#### 5. ACKNOWLEDGEMENT

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#### REFERENCES

- Bonvin, D. (1998). Optimal operation of batch reactors-a personal view, *Journal of Process Control*, 8(5), 355-368.
- Bonvin, D., Srinivasan, B. and Hunkeler, D. (2006). Control and optimization of batch processes, *IEEE Control System Magazine*, 26(6), 34-45.
- DeHaan, D. and Guay, M. (2005). Extremum-seeking control of state-constrained nonlinear system, *Automatica*, 41(9), 1567-1574.
- François, G., Srinivasan, B. and Bonvin, D. (2012). Comparison of six implicit real-time optimization schemes, *Journal Européen des Systèmes Automatisés*, 46(2-3), 291-305.
- Kong, X.S., Yang, Y., Chen, X., Shao, Z.J. and Gao, F.R. (2011). Quality control via model-free optimization for a type of batch process with a short-cycle time and low operation cost, *Industrial Engineering Chemical Research*, 50(5), 2994-3003.
- Lu, N.Y., Gao, F.R. (2005). Stage-based analysis and quality prediction for batch processes, *Industrial Engineering Chemical Research*, 44(10), 3547-3555.
- Lu, N.Y. and Gao, F.R. (2006). Stage-based online quality control for batch process, *Industrial Engineering Chemical Research*, 45(7), 2272-2280.
- Shi, J., Gao, F.R. and Wu, T.J. (2003). Single-cycle and multi-cycle generalized 2D model predictive iterative learning control (2D-GPILC) scheme for batch processes, *Journal of Process Control*, 17(9), 715-727.
- Spall, J.C. (1998). An overview of the simultaneous perturbation method for efficient optimization, *Johns Hopkins APL Technical Digest*, 19(4), 482-492, 1998.
- Srinivasan, B., Bonvin D., Visser, E. and Palanki, S. (2003). Dynamic optimization of batch processes II. Role of measurements in handling uncertainty, *Computers and Chemical Engineering*, 27(1), 27-44.
- Srinivasan, B. (2007). Real-time optimization of dynamic systems using multiple units, *International Journal of Robust and Nonlinear Control*, 17(13), 1183-1193.
- Undey, C., Cinar, A. (2002). Statistical monitoring of multistage, multiphase batch processes, *IEEE Control Systems Magazine*, 22(5), 40-52.
- Xi, Y.G. (2013). *Predictive control*, Chapter 5. National Defence Industry Press, Beijing, China.
- Yang, Y. (2004). Injection molding control: from process to quality, Ph.D. dissertation, Chemical Engineering, Hong Kong University of Science and Technology, Hong Kong.
- Zhao, C.H., Wang, F.L. and Gao, F.R. (2009). Improved calibration investigation using phase-wise local and cumulative quality interpretation and prediction, *Chemometrics and Intelligent Laboratory Systems*, 95(2), 107-121.
- Zhao, C. H., Lu, N. Y. (2014). *Statistical monitoring and quality analysis for batch processes*. Science Press, Beijing, China.