

**Iterative Batch-to-Batch Input
Profile Determination for
Semi-Batch Processes**

By

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Iterative Batch-to-Batch Input Profile Determination for Semi-Batch Processes*

A Polymerization Example

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Abstract

Polymerization processes are very complex and high nonlinear. Their modeling often involves reaction mechanism analysis combined with empirical tests. Mismatch between the model and the industrial plant often exists and it can be the cause of bad performance when optimal input profiles computed for a particular model are applied to the actual plant. The approach followed in this paper is to directly modify the input profile from batch to batch, and it has been applied through computer simulations to the determination of the minimum-time temperature profile for the batch bulk polymerization of styrene under modeling error. The results demonstrate that the approach has strong robustness characteristics and fast convergence properties.

1 Introduction

Batch processing is becoming increasingly more important in the polymerization and biochemical industries. Since batch processes seldom operate at a steady state and the reaction system may be quite complex and not entirely known, model-plant mismatch always exists. Optimal profiles computed for a particular model are suboptimal when applied to the actual plant. Recently, Filippi et al [1,2] proposed a new approach for batch optimization, which uses tendency models to identify and modify actual kinetic models. In this approach, remodeling and reoptimization techniques are required. Since such techniques involve a significant effort in identifying many unknown parameters in complex reaction systems, this is often a very difficult task in industrial applications.

The approach followed in this paper is to directly modify the input profile during the course of successive batches [3]. The procedure uses information from previous batches to improve the operation of the next and no direct modification in the model is made. An approximate gradient, *based on both the model and the actual plant measurements*, is obtained and then used to compute the search direction. The search direction is used to compute the next input profile, until no further improvement in the objective function can be obtained. The proposed approach has been successfully applied through computer simulations on a cell mass production biochemical process [4]. Even though model error of 200% in a key coefficient in the yield expression was introduced to simulate the model-plant mismatch, the simulation results demonstrated that the proposed approach is very promising. The method also performed well when measurement error and error in the initial conditions was introduced. The methodology can be applied on any systems operated in semi-batch mode, regardless of their nature. This paper extends the procedure to the free batch time problem. The application

involves the determination of the minimum-time temperature profile for the batch bulk polymerization of styrene under unknown or poorly known reaction kinetic models.

2 Methodology

In this section, we briefly review the methodology [3]. The approach used in this paper is based on the analogy between the iterations during the numerical optimization of the appropriate cost functional (objective function) on one hand and the successive batches during the operation of the plant on the other. In gradient based numerical optimization, the gradient computation at every step involves two integrations. The first is forward integration of the differential equations describing the plant and the second is the backward integration of the adjoint system. We have developed a strategy in which the first integration is not carried out numerically, but rather it is carried out by the actual plant itself. Its result is the measurements from the previous batch. The second integration however requires the use of the process model. The result of the above procedure will be an estimate for the gradient $g(t)$, which is then used to compute the search direction that will result in the next input profile. In this way, a performance improvement will be obtained from batch to batch. This technique is shown in Table 1.

In batch or semi-batch polymerization processes, the objective is often to derive optimal temperature and initiator addition policies, which minimize the batch time to reach the specified conversion and other polymer properties. If the final time is not specified, the above gradient based optimization method requires the forward and backward integration on a time abscissa whose length changes with each iteration. However, a coordinate transformation method, proposed by Kwon [5] and Kwon and Evan [6], can convert the original fixed-end-point, free-end-time problem to a free-end-point, fixed-end-time problem. The transformed problem can be solved by gradient methods with penalty function techniques. In the following section, the coordinate transformation and our approach are applied to iterative batch-to-batch temperature profile determination for the bulk polymerization of styrene under modeling error.

3 Iterative Batch to Batch Optimization

3.1 System Description and Optimization for the Model

Let us investigate the following thermally initiated bulk polymerization of a styrene process of industrial interest, modeled by Kwon and Evans through reaction mech-

Table 1: Analogy between Numerical Optimization and Plant Operation

NUMERICAL OPTIMIZATION	PLANT OPERATION
$\min_{u(t)} \phi(x(u(t)))$ where $\dot{x} = f(x, u)$, $x(0) = x_0$ (model)	$\min_{u(t)} \phi(x(u(t)))$ where $\dot{x} = \tilde{f}(x, u)$, $x(0) = x_0$ (plant)
1st iteration	1st batch
1. Forward integration of model $u_1(t) \rightarrow \dot{x} = f(x, u)$, (model) $\rightarrow x_1(t)$	1. Forward integration of plant $u_1(t) \rightarrow \dot{x} = \tilde{f}(x, u)$, (plant) $\rightarrow \tilde{x}_1(t)$ (off-line measurement)
2. Linearize model $f(x, u)$ at u_1, x_1 $u_1(t), x_1(t) \rightarrow$ Linearization $\rightarrow f_x(x_1, u_1)$ $u_1(t), x_1(t) \rightarrow$ Linearization $\rightarrow f_u(x_1, u_1)$	2. Linearize model $f(x, u)$ at u_1, \tilde{x}_1 $u_1(t), \tilde{x}_1(t) \rightarrow$ Linearization $\rightarrow f_x(\tilde{x}_1, u_1)$ $u_1(t), \tilde{x}_1(t) \rightarrow$ Linearization $\rightarrow f_u(\tilde{x}_1, u_1)$
3. Backward integration of adjoint system $\dot{\lambda} = -f_x(x_1, u_1)\lambda$; $\lambda(t_f) = \nabla_x \phi(x_1(t_f))$	3. Backward integration of approximate adjoint system $\dot{\tilde{\lambda}} = -f_x(\tilde{x}_1, u_1)\tilde{\lambda}$; $\tilde{\lambda}(t_f) = \nabla_x \phi(\tilde{x}_1(t_f))$
4. Gradient $g(t) = -f_u^T(x_1, u_1)\lambda(t)$	4. Estimate gradient $\tilde{g}(t) = -f_u^T(\tilde{x}_1, u_1)\tilde{\lambda}(t)$
5. Search $u_2(t)$ Search direction : $S(t) = -Pg(t)$ P : constraint projection matrix $u_2(t) = u_1(t) + \alpha S(t)$ $0 \leq \alpha \leq \alpha_{max}$ where α_{max} is the limit imposed by constraints.	5. Search $u_2(t)$ Search direction: $\tilde{S}(t) = -P\tilde{g}(t)$ P : constraint projection matrix $u_2(t) = u_1(t) + \alpha \tilde{S}(t)$ $0 \leq \alpha \leq \min[\alpha_e, \alpha_{max}]$ where α_{max} is the limit imposed by constraints; and α_e is a limit on maximum adjustment of actual plant.
2nd iteration	2nd batch
. .	. .

anism analysis and laboratory tests [5,6,7]:

$$\dot{x}_1(t) = f_1 = \frac{(r_1 + r_2 T_c)^2}{M_m} (1 - x_1)^2 \exp(2x_1 + 2\chi x_1^2) \left(\frac{1 - x_1}{r_1 + r_2 T_c} + \frac{x_1}{r_3 + r_4 T_c} \right) A_m \exp\left(-\frac{E_m}{T}\right) \quad (1)$$

$$\dot{x}_2(t) = f_2 = \frac{f_1 x_2}{1 + x_1} \left(1 - \frac{1400 x_2}{A_w \exp\left(\frac{B}{T}\right)} \right) \quad (2)$$

$$\dot{x}_3(t) = f_3 = \frac{f_1}{1 + x_1} \left(\frac{A_w \exp\left(\frac{B}{T}\right)}{1500} - x_3 \right) \quad (3)$$

where the state variables x_1, x_2, x_3 are conversion, dimensionless number average chain length (*NACL*) and dimensionless weight average chain length (*WACL*) respectively. A_w, B are coefficients in the relation between *WACL* and temperature, obtained from experiments; A_m, E_m are the frequency factor and activation energy of the overall monomer reaction; and $r_1 - r_4$ constants are density-temperature corrections. M_m, χ are the monomer molecular weight and polymer-monomer interaction parameter. T or T_c is the control variable (absolute temperature and temperature in $^{\circ}C$, respectively).

The selection of a new set of state variables (y_1 dimensionless reaction time, y_2 *NACL*, y_3 *WACL*) and conversion τ as an independent variable (fictitious time) lead to a fixed-end-time and free-end-point problem. The new optimization problem statement for the reaction time minimization with final specific *NACL* and *WACL* equal to 1 and conversion (τ_f) at 80% [6] can be mathematically described as follows:

$$\min_{T_c(\tau)} J = y_1(\tau_f) + \gamma_k [(y_2(\tau_f) - 1)^2 + (y_3(\tau_f) - 1)^2] \quad (4)$$

$$\dot{y}_1(\tau) = M_m / [5.0(r_1 + r_2 T_c)^2 (1 - \tau)^2 \exp(2\tau + 2\chi \tau^2) \left(\frac{1 - \tau}{r_1 + r_2 T_c} + \frac{\tau}{r_3 + r_4 T_c} \right) A_m \exp\left(-\frac{E_m}{T}\right)]$$

$$y_1(0) = 0 \quad (5)$$

$$\dot{y}_2(\tau) = \frac{y_2}{1 + \tau} \left(1 - \frac{1400 y_2}{A_w \exp\left(\frac{B}{T}\right)} \right) \quad y_2(0) = 1 \quad (6)$$

$$\dot{y}_3(\tau) = \frac{1}{1 + \tau} \left(\frac{A_w \exp\left(\frac{B}{T}\right)}{1500} - y_3 \right) \quad y_3(0) = 1 \quad (7)$$

$$100^{\circ}C \leq T_c \leq 200^{\circ}C \quad (8)$$

In the numerical optimization of the model, the penalty coefficient γ_k is specified as follows [6]:

$$\gamma_k = \min [10^4, 0.77(\frac{10^4}{0.77})^{0.1k}] \quad (9)$$

and the initial guess $T_c = 133^\circ C$. The optimal reaction time t_f^* is 5.22hr and performance index $J(T_c^*(\tau))$ is 1.1 ($NACL(t_f) = 1.000$ and $WACL(t_f) = 0.999$). The optimal temperature profile is shown in Fig. 2. This operating policy is defined as nominal for the remainder of the paper.

3.2 Batch to Batch Temperature Profile Determination in the Presence of Modeling Error

The reaction mechanisms for the thermally initiated bulk polymerization of styrene described in the previous section are not well understood [7]. Some parameters in the model may vary with concentration, temperature, viscosity etc. Certainly, a mismatch between the developed model and the industrial plant often occurs in practice. In order to investigate the proposed approach, we introduce error in the parameters of the empirical relation between the properties of newly formed increment of polymer and temperature [6,7] as shown in Table 2.

The steepest descent with constraints search direction used for batch to batch iteration is defined as follows:

$$\tilde{S}^m(\tau) = -P\tilde{g}(\tau) \quad (10)$$

where $\tilde{g}(\tau)$ is the estimate for the gradient $g(\tau)$ [3,4], as briefly described in section 2, and P is the constraint projection matrix. Because of the line search, one will occasionally have a temporary increase in the objective function for certain batches. In an attempt to make the proposed approach more appropriate for industrial application, a limit on the maximum adjustment from batch to batch for the control variable T_c is introduced so that steady improvement can be obtained. This limit corresponds to α_e . In our example, the maximum adjustment $(\Delta T_c)_{max}$ is selected $5^\circ C$, and decreases with increasing batch number. The line search

$$T_c^m(\tau) = T_c^{m-1}(\tau) + \alpha\tilde{S}^m(\tau) \quad 0 \leq \alpha \leq \min [\alpha_e, \alpha_{max}] \quad (11)$$

terminates as soon as improvement is obtained [4], where α_{max} is the limit imposed by the constraints.

Besides, as briefly described in section 2, since the measurements are not used for on-line control, samples of the product can be collected and sent for analysis during the course of the batches.

Table 2:

	Parameter Error	True Optimum Value	Plant Operation	
			initial batch	after 20 batches
case 1	$A'_w = 1.50A_w$ $B' = B$	$t_f = 2.08$ $NACL(t_f) = 1.000$ $WACL(t_f) = 1.000$	$t_f = 5.22$ $NACL(t_f) = 1.174$ $WACL(t_f) = 1.222$	$t_f = 2.09$ $NACL(t_f) = 1.000$ $WACL(t_f) = 1.000$
case 2	$A'_w = 0.50A_w$ $B' = B$	$t_f = 23.96$ $NACL(t_f) = 0.999$ $WACL(t_f) = 0.989$	$t_f = 5.22$ $NACL(t_f) = 0.693$ $WACL(t_f) = 0.778$	$t_f = 23.83$ $NACL(t_f) = 0.950$ $WACL(t_f) = 0.985$
case 3	$A'_w = A_w$ $B' = 1.05B$	$t_f = 1.64$ $NACL(t_f) = 1.000$ $WACL(t_f) = 1.000$	$t_f = 5.22$ $NACL(t_f) = 1.224$ $WACL(t_f) = 1.316$	$t_f = 1.65$ $NACL(t_f) = 1.000$ $WACL(t_f) = 1.001$
case 4	$A'_w = A_w$ $B' = 0.95B$	$t_f = 18.18$ $NACL(t_f) = 1.000$ $WACL(t_f) = 0.993$	$t_f = 5.22$ $NACL(t_f) = 0.763$ $WACL(t_f) = 0.815$	$t_f = 18.53$ $NACL(t_f) = 0.996$ $WACL(t_f) = 0.996$

The results of plant operation under model plant mismatch are compared to the optimal for the true plant (if the true plant were known) in Table 2 (for $\gamma_k = 10^4$; the initial guess $T_c(\tau)$ is the optimal for the nominal case).

Let us look in more detail in case 1 of the Table 2. Fig. 1 demonstrates that the proposed algorithm can provide significant improvement in the performance index (computed for the actual plant) even if a mismatch between the model ($A'_w = A_w$) and the plant ($A'_w = 1.50A_w$) exists.

For plant operation in the initial batch, the performance index $J(T_c(\tau))$ is 792 and the reaction time t_f is 5.22hr; $NACL(t_f)$ and $WACL(t_f)$ are 1.174 , 1.221 respectively. Note that the desired value for both $NACL(t_f)$ and $WACL(t_f)$ is 1. After 20 batches, the performance index $J(T_c(\tau))$ decreases to 0.18 and the reaction time t_f is 2.09hr; $NACL(t_f)$ and $WACL(t_f)$ are 1.000,1.000 respectively. If the plant were known, the optimal performance index $J(T_c^*(\tau))$ is 0.17, and the reaction time t_f is 2.08hr; and $NACL(t_f)$ and $WACL(t_f)$ are 1.000,1.000, respectively. For comparison, the initial temperature profile, the profile reached after 20 batches and the true optimal temperature profile are shown in Fig. 2. The results of the batch to batch profile determination during the plant operation definitely approach the true optimal solution. Also, from the convergence of the terminal states as shown in Fig. 3, one can see desired values were essentially reached within 6-8 batches, a fact that makes the proposed algorithm is attractive for industrial applications.

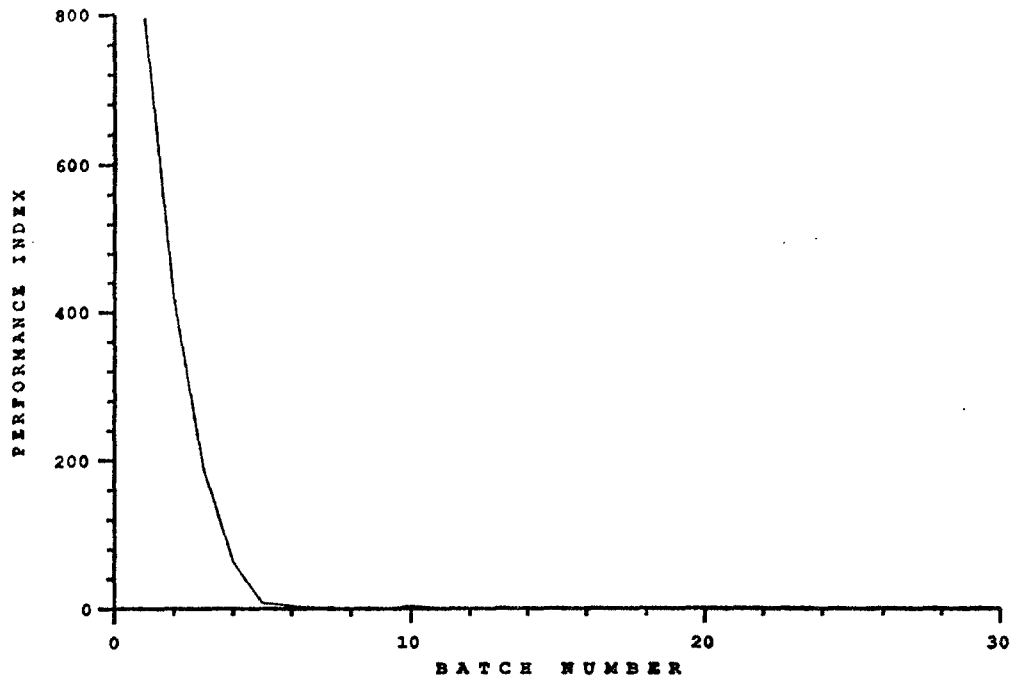


Figure 1: Performance Index J as a Function of Batch Number

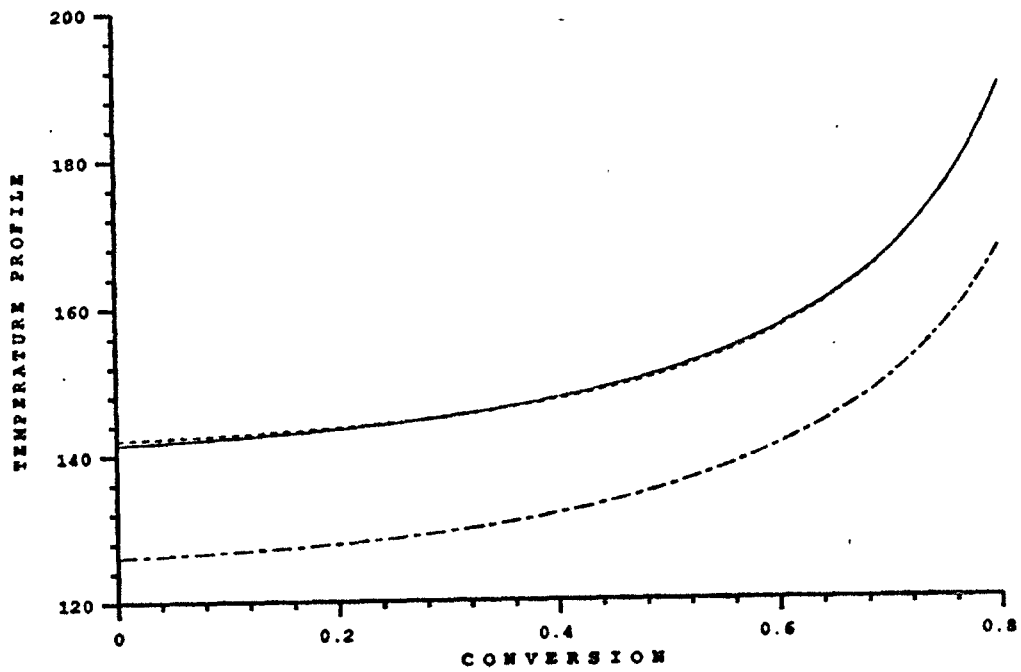


Figure 2: Temperature Profiles Solid Line: T Profile by Batch to Batch Iteration after 20 Batches; Dash-dot Line: Initial (nominal) T Profile; Dotted Line: True Optimal T Profile

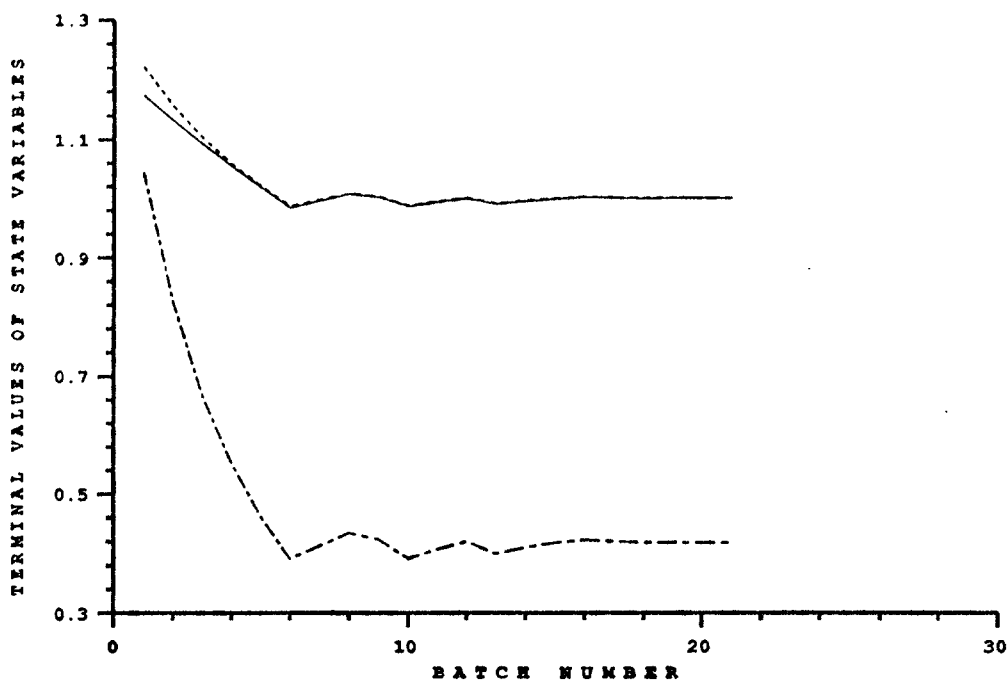


Figure 3: Value of Terminal State Variables as Function of Batch Number Dash-dot Line: $y_1(\tau_f)$; Solid Line: $y_2(\tau_f)$; Dotted Line: $y_3(\tau_f)$

4 Conclusion

The reaction mechanisms for polymerization processes are often not well understood and the systems are occasionally described with inaccurate models. Need definitely exists for an effective method that modifies the input profile during the course of successive batches without requiring extensive remodeling between batches. Our approach combines information from previous batches with the existing model to improve the operation of the following batches. This method has been successfully applied through computer simulations on the batch bulk polymerization of styrene. The results coupled with a previous case study on a biochemical process [4], indicate that the proposed approach has strong robustness characteristics for large model-plant mismatch and fast convergence properties during plant operation. However, in order to have a descent property (that guarantees improvement) during successive batches, the effect of model uncertainty on the convergence needs to be further investigated. We are currently looking at ways for obtaining a better estimate of the gradient as well as gradient approximation error bounds after each batch. Such information will be used to check whether a steady improvement can be accomplished during successive batches.

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