

Lexicographic Optimization Control Method Based Phase Recognition for Multiphase Batch Polymerization Reaction

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Abstract: To solve the problem that the correlation of variables varies with phase in multiphase batch polymerization reaction, this paper proposes an optimization method for lexicographic order switching based on phase recognition. This method uses kernel principal component analysis to analyze the correlation of each sampling time in a sliding time window, and analyze the phase of each batch of historical data. According to the analysis results, each batch is divided into individual phases, and the sub-phase data is used to identify the phase of real-time production. This method sets the corresponding lexicographic control target according to the characteristics of each phase of the process. According to the lexicographical order of real-time phase location, the optimal operation variable trajectory is obtained, so as to achieve the best optimization effect in phase, and ensure the safe production process and continuous and stable product quality.

Keywords: Multiphase batch process, Kernel Principle Component Analysis(KPCA), Uneven-length, Phase Partition, Lexicographic Optimization

1. INTRODUCTION

Batch polymerization is a typical polymer industrial production method. To meet the market requirements of multiple varieties, multiple specifications, and high value-added, research on batch process optimization and quality control has become a hotspot. Due to the severe time-varying, dynamic, nonlinear, and other characteristics of the batch process, it is difficult to establish an accurate mechanism model to realize online monitoring, prediction, and control. On the other hand, data-driven multivariate statistical analysis methods require only the processing of historical data, severing a good way in process modeling, thanks to the rapid development of data acquisition systems and storage technologies. The Multilinear Principal Component Analysis (MPCA) (Nomikos et al., 1994) and Multivariate Partial Least-Square (MPLS) (Nomikos et al., 1995) extend the application range of traditional multivariate statistical analysis methods from continuous processes to multi-batch processes. However, most studies (Lu and Gao, 2005; Zhao and Sun, 2013) were based on the assumption that all batches have the same duration and phase change synchronization.

For multiphase batch processes, the problem of uneven length may occur at each stage, and the resulting irregular stage data cannot be directly used for statistical analysis and modeling. The solutions to the problem of uneven intermittent length in the intermittent process can be divided into two types: direct

signal synchronization methods (Ela and Ketan, 2013) and modeling method based on irregular phase division (Zhao and Gao, 2013). The direct signal synchronization method refers to the synchronization of the trajectory of intermittent production historical batches through some signal processing methods (Kourti, 2003). The missing data estimation method requires enough long batches for identifying the model to estimate the data. To overcome the limitation of requiring sufficiently long batches, Jian et al (2014) proposed an improved missing data estimation method for dealing with data synchronization problems. However, this method cannot describe the characteristics of multiphase intermittent process by a single model.

In order to overcome the limitations of direct signal synchronization method and consider the multiphase nature of batch processes, Lu et al. (2004) proposed a sub-phase segmentation method to deal with the uneven length of the historical batch process. This method uses an improved clustering algorithm combined with machine learning to directly divide irregular data. Zhao et al (2014) distinguished and analyzed two kinds of phase unevenness: the moderate case and the severe case. They proposed a method based on group division and subspace separation to model the different characteristics of irregular batches. Some scholars (Li et al., 2015) have studied a phase recognition algorithm for a single batch. This method compares the correlation of a single

batch with the established model, and uses the extended time slice method to align the irregular sub-phase data.

Due to changes in the process environment of the polymerization process, the results obtained by one-time optimization cannot meet the needs of long-term operation. On one hand, it is necessary to monitor the optimization model to prevent the distortion of the optimization results. On the other hand, it is necessary to formulate optimization strategies. For the polymerization reaction with very complex process mechanism, the optimization problem presents the characteristics of strong nonlinearity, multiple constraints, and multi-level values. The conventional optimization methods have limitations and deficiencies. This paper adopts a multi-objective control strategy based on lexicographic optimization (Rasekhipour et al., 2018). This method is based on a hierarchical optimization idea, which sorts multiple control targets from high to low in priority, and then starts with high priority targets and optimizes layer by layer to ensure the control performance of important targets.

2. MULTIPHASE PARTITION ALGORITHM OF BATCH POLYMERIZATION

2.1 Multiphase characteristics of batch polymerization process

Poly tetra fluoroethylene (PTFE) is a polymer compound made of tetrafluoroethylene polymerization. The complex polymerization production process includes raw material mixer, polymerization reactor, separation tower, drying tower, packaging and other unit operations. This paper takes only the polymerization reactor as the research object, specifically focusing on the material changes, temperature changes and molecular length changes in the reactor. During the polymerization process, three main reactions (chain initiation, chain growth and chain termination) are taking place simultaneously.

The polymer production process usually consists of different production stages such as heating, feeding, high temperature polymerization and other sub-stages. The primary reaction of the polymer produced during polymerization: chain initiation and chain growth are endothermic. For free radical polymerization, when the temperature is higher, the small molecular chains will be more active and the rate of polymerization will be faster. Therefore, in order to ensure the yield of the product, the temperature of the reactor needs to be increased as soon as possible with minimal cost. When the temperature enters the first stage and stabilizes for a period of time, it is necessary to accelerate production through entering the second stage of high-temperature polymerization. In this high-temperature stage, due to the temperature rises, the output of the polymer increases and the average molecular length of the polymerization reaction decreases. Each time period has its specific control goal with different process dominant variables and process characteristics. In different phases, the correlation between variables is different, but the correlation of internal data in the same phase and its process characteristics are similar.

2.2 Data preprocessing and statistical model establishment

Historical batch process data usually contain similar manufacturing processes in different batches. Assume that $X(I \times K \times J)$ is the regular three-dimensional sample data, where I is the number of batches, J is the number of variables, and K is the number of samples. For the convenience of analysis, the three-dimensional data $X(I \times K \times J)$ are unfolded in the variable-wise direction into two-dimensional data. The method keeps the dimension of variables unchanged, and the data are arranged in columns from top to bottom to form a $(KI \times J)$ dimensional matrix.

The kernel principal component analysis (KPCA) method is needed when performing time series correlation analysis. For the samples $x_i \in \mathcal{R}^J$ in the data matrix X , there is a non-linear mapping $\Phi(\cdot)$ to map the sample to the feature space. The principal component information can be decomposed by the following feature problems.

$$\begin{cases} \lambda \mathbf{v} = \mathbf{C}^F \mathbf{v} \\ \mathbf{C}^F = \frac{1}{N} \sum_{i=1}^N \Phi(x_i) \Phi(x_i)^T \end{cases} \quad (1)$$

where λ is the eigenvalue, \mathbf{v} is the eigenvector, and $\mathbf{C}^F(N \times N)$ is the covariance matrix of X in the high-dimensional space. Define an $N \times N$ kernel matrix $K_{ij} = \langle \Phi(x_i), \Phi(x_j) \rangle$, where the vector inner product mapped to the high-dimensional space can be calculated with the kernel function $k(x,y)$ in the low-dimensional space, and the kernel function takes the form of a radial basis kernel function:

$$K_{ij} = k(x_i, x_j) = \exp(-\|x_i, x_j\|^2 / \gamma) \quad (2)$$

where γ is a custom parameter. Solve the eigen decomposition problem to get the eigenvalue $\lambda_1 \geq \dots \geq \lambda_N > 0$ and the eigenvector β_1, \dots, β_N by equation (3). Use $\lambda_j \langle (\beta_i), (\beta_j) \rangle = 1$ to normalize eigenvector.

$$N \lambda \beta = \mathbf{K} \beta \quad (3)$$

Calculate the number of retained pivots p ($p < J$) by determining the cumulative variance contribution ratio of pivots η (usually between 85% and 99%). η can be calculated by equation (4):

$$\frac{\sum_{j=1}^p \lambda_j}{\sum_{j=1}^N \lambda_j} > \eta \quad (4)$$

For a new sample data $x^* \in \mathcal{R}^J$, the corresponding pivot is recorded as $t^* \in \mathcal{R}^p$, then t^* can be calculated by equation (5).

$$t_j^* = \sum_{i=1}^N \beta_{i,j} \langle \Phi(x_i), \Phi(x^*) \rangle = \sum_{i=1}^N \beta_{i,j} k(x_i, x^*) \quad (5)$$

where $\beta_{i,j}$ is the i th component of the vector β_j . Calculate the principal component and residual error by equation (6):

$$\begin{cases} \hat{x}^T = t^T P^T \\ e^T = x^T - \hat{x}^T \end{cases} \quad (6)$$

where P is the load matrix transformed from the eigenvector, t is the principal component, and e is the residual vector. The deviation between the tested sample and the modeled sample is calculated by calculating the Q statistic. The statistic Q is

also called the Squared Prediction Error (SPE), and SPE is defined as follows:

$$SPE = e^T e = \sum_{j=1}^J (x_j - \hat{x}_j)^2 \quad (7)$$

The control limit of SPE is calculated by equation (8).

$$SPE_{\mu} \sim g \cdot \chi_{\tau, \mu}^2 \quad (8)$$

where $g = \nu/2m$, $\tau = 2m^2/\nu$, m and ν are the mean and variance of SPE.

2.3 Phase partition algorithm for unequal length batch process

As shown in Figure 1, during the phase transition period, some batches have entered the next stage, while some are still in the previous stage. In the time period between the shortest length k_s^1 and the longest k_l^1 in phase 1, the same sampling time will contain two phases at the same time.

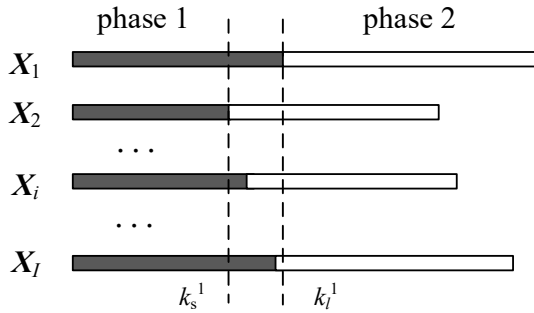


Figure 1. Schematic diagram of unequal length phase

The basic idea of phase partition is to test for changes in the correlation of process variables sequentially in the order of sampling time. The initial phase model is built based on the first w time slices, and then it can be used to determine whether the samples of the next time slice are in the same phase as the current ones. The specific steps are as follows:

Step 1: Data processing. Expand the three-dimensional data $X(I \times J \times K_i)$ according to variables to get $X_V(\sum_{i=1}^I K_i \times J)$.

Step 2: The establishment of the initial model. Starting from the first time slice matrix, create an initial window matrix slice $X_w(wI \times J)$ and normalize X_w along the variable direction. Calculate the kernel matrix $K_w(wI \times wI)$, apply the KPCA algorithm to X_w , and obtain the first statistical model load matrix $P_w(J \times p)$ by solving the characteristic problem.

Step 3: Relevance assessment. P_w is used as the load matrix of the window statistical model to evaluate the correlation of each row of each internal time slice. Calculate the kernel matrix $K_k(I \times I)$ of each time slice $X_k(k=1, \dots, w)$ of the current window, and use the window kernel matrix K_w to normalize the kernel matrix K_k of each time slice. Then use the window statistical model load matrix P_w to calculate the $Ctrl_w^k$, SPE and residual of each row of each time slice. $Ctrl_w^k$ is the control limit of the time window. Except for the first window, each subsequent window only needs to calculate the KPCA statistics of the last three time slices. The statistics are calculated as follows:

$$\begin{cases} t_i = P_{w,1} x_i^* \\ e_i = x_i^* - P_{w,1}^T t_i \\ SPE_i = e_i^T e_i \end{cases} \quad (9)$$

Step 4: Phase division. If the SPE_i at three consecutive time points k calculated by the current monitoring model P_w exceeds the control limit, the phase switching time k_c^i of the i th batch is recorded. The batch data before the k_c^i of the i th batch is expressed as a sub-phase. When all batches of all time slices in a window have no phase division, it means that the data in the time window has to slide by a time k as a whole. And when there are batches that meet the phase division requirement in this window, this row is temporarily fixed and the data row x_k^j (The j th batch of data at the k th time) whose phase is not determined phase division is slid back by one sampling time. Update the KPCA model K_w and P_w of the window.

Step 5: Repeat steps (2), (3), (4) until all data are divided.

3. OPTIMAL CONTROL OF MULTI-OBJECTIVE LEXICOGRAPHIC ORDER BASED ON PHASE RECOGNITION

3.1 Lexicographic order switching based on phase recognition

Optimization flowchart of lexicographic order switching based on phase recognition is shown in Figure 2.

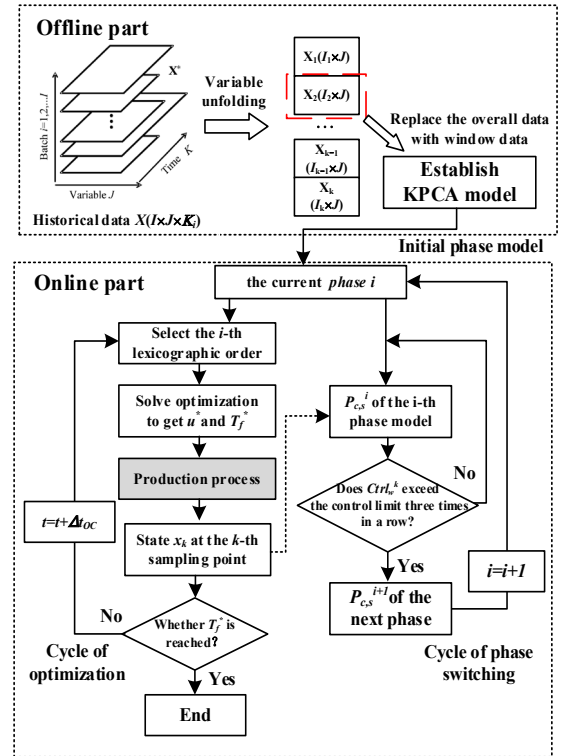


Figure 2. Optimization flowchart of lexicographic order switching based on phase recognition

The specific steps of the algorithm are as follows:

Step 1: The historical data $X(I \times J \times K_i)$ were expanded by variable unfolding method. Establish a time window $X_w(wI \times J)$, normalize the data in the window along the variable direction and obtain K_w and P_w . Use the window model to calculate the control limit $Ctrl_w^k$ of each time slice matrix X_k in the window, and calculate the SPE and residual of each row. Determine the phase division point according to the statistics

and the control limit, and update the data in the window according to the method of sliding a single batch of data. After dividing all the data, the phase division accuracy was analyzed based on the process and experience information.

Step 2 : Establish PCA models in different phases and calculate control limits for standard statistics T^2 and SPE

Step 3: Perform batch production according to the optimization control described above and apply the optimization target dictionary of the first phase. Measure the variable data of the current sampling time. Apply the PCA model from the first phase model, use the sub-phase data to normalize the measured data at each sampling time, and calculate its T^2 and SPE. If the statistic exceeds the control limit, go to step 4. Otherwise, repeat this step.

Step 4: Apply the PCA model of the next phase. Apply the model information of the next phase to re-normalize the current sampled data and calculate the statistics. If the control limit is still exceeded, repeat this step. Otherwise, it means that the process has entered the next phase and the priority setting of the objective function of the next phase is applied.

Step 5: Repeat steps 3 and 4 until the control of this batch of production is completed.

3.2 Establishment of the phase identification statistical model

In the establishment of the sub-phase identification statistical model, each phase of each batch is strictly distinguished, and the data in the same phase have greater correlation. It is necessary to consider the characteristics of each batch to establish a more accurate sub-phase model. Therefore, a method of normalization in the batch direction is adopted to capture the time change of each batch.

In the first phase, assuming that the shortest phase length is k_s and the longest is k_l , the time slices between $[1, k_s]$ are regular time slices. For those irregular time slice matrices between $[k_s+1, k_l]$, the generalized time slice matrix $X^G(\sum_{k_s+1}^{k_l}(I_k \times J))$ is constructed by variable expansion method.

After aligning the irregular time slices, the regular time slice and the generalized time slice are normalized along the batch direction. Then the regular time slice $X_k(I_k \times J)$ ($k=1, \dots, k_c-1$) and the extended time slice X^G are arranged into a matrix $X_c(\sum_{k=1}^{k_c}(I_k \times J))$ by the method of variable expansion. Perform PCA operation on X_c :

$$\begin{cases} T_c = X_c P_{c,s} \\ X_c = T_c P_{c,s}^T + E_c = X_c P_{c,s} P_{c,s}^T + E_c \\ \hat{X}_c = T_c P_{c,s}^T = X_c P_{c,s} P_{c,s}^T \\ E_c = X_c - \hat{X}_c = X_c P_{c,e} P_{c,e}^T \end{cases} \quad (10)$$

where $P_{c,s}(J \times p_c)$ is the load matrix of the recognition model, and p_c is the number of principal elements retained in the current stage. The principal component matrix $T_c(\sum_1^{k_c}(I_k \times p_c))$ is extracted from the process variables and represents the main amount of change. \hat{X}_c is the sub-phase data reconstructed from T_c , which contains most of the information of X_c . E_c is the residual of the reconstructed sub-phase data, which includes irregular and irrelevant information. $P_{c,e}(J \times J - p_c)$ reflects the

direction of minor changes in addition to the main changes. The subspaces constructed by $P_{c,s}$ and $P_{c,e}$ are called system subspace and residual subspace respectively. Two monitoring statistics can be calculated in each subspace. T^2 is used for system subspace, and SPE is used for residual subspace.

$$\begin{cases} T_k^2 = (t_k - \bar{t}_k)^T S_c^{-1} (t_k - \bar{t}_k) \\ SPE_k = e_k^T e_k \end{cases} \quad (11)$$

where $t_k(p_c \times 1)$ is the principal component vector separated from T_c and S_c is the covariance matrix of T_c . \bar{t}_k is the mean vector of T_k , which is actually almost the zero vector. $e_k(J \times 1)$ is the residual vector from E_c . T^2 describes the system change captured by the system subspace load matrix $P_{c,s}$, while SPE reveals the change occupied by $P_{c,e}$ in the residual part. The change of these two statistics reflects the correlation of the variables.

After the monitoring model of the irregular phase is built, the real-time data generated in the production process can be identified online. For multiphase intermittent processes with unequal length periods, only after the production process is positioned to the correct phase, the appropriate control strategy can be made for the operation state of new samples.

3.3 Lexicographic optimization of multiphase process

The reaction process considered in this article is given by ordinary differential equations(ODEs) in the following form:

$$\begin{cases} \frac{dx(t)}{dt} = F(x(t), u(t), d(t)) \\ x(t_0) = x_0 \end{cases} \quad (12)$$

where t_0 is the initial moment, $x(t) \in \mathbb{X} \subseteq \mathbb{R}^{n_x}$ is the state variables, $u(t) \in \mathbb{U} \subseteq \mathbb{R}^{n_u}$ is the control input variables of the process, and $d(t)$ is the disturbance variable. $F: \mathbb{R}^{n_x} \times \mathbb{R}^{n_u} \times \mathbb{R}^{n_d} \rightarrow \mathbb{R}^{n_x}$ represents the mapping relationship of the nonlinear dynamic model of the process, where n_x , n_u , and n_d are the dimensions of process state variables, control input variables, and disturbance variables.

The control objectives of the lexicographic order are sorted to form a hierarchical structure, the first objective in the hierarchical structure is the most important, and the last objective function has the lowest priority. The most important objective function is minimized under the original constraint. The second most important objective function adds a new lexicographic constraint in addition to the original constraints, ensuring that the most important objective function is kept near the optimal value. Using the lexicographic ordering method, different Pareto optimal solutions can be obtained by modifying the hierarchical structure of the objective function. This strategy does not need to adjust and weigh the weight of the control target.

In general, the form of multiple goals can be expressed as:

$$\min_{u, T_f} J(x, u) = (J_1(x, u), J_2(x, u), \dots, J_N(x, u)) \quad (13)$$

the initial constraints of the process:

$$\text{s. t. } \begin{cases} \frac{dx}{dt} = F(x, u) \\ x(t_s) = x_s \\ x, u \in \Omega \end{cases} \quad (14)$$

For lexicographic optimization, the following N optimization problems are solved sequentially in each iteration:

$$\begin{aligned}
& \text{Opt 1: } \min_u J_1 \\
& \text{s.t. } \text{constraints}(14) \\
& \dots \\
& \text{Opt N: } \min_u J_N \\
& \text{s.t. } \begin{cases} \text{constraints}(14) \\ J_i \leq J_i^*, i = 1, 2, \dots, N-1 \end{cases}
\end{aligned}$$

Some important goals are inevitably convex functions. Therefore, in order to improve the $(i+1)$ th optimization problem, the following form of slack variable δ is added to replace $J_i \leq J_i^*$ in the lexicographic order constraint:

$$J_i \leq J_i^* + \delta, i = 1, 2, \dots, n \quad (15)$$

The optimization problem should not only consider the economic cost, but also consider safe and smooth operation of equipment. The economic goal is usually to maximize profit. To guarantee smooth batch reaction, it is important to prevent sudden jumps in control inputs. This means the derivatives of control input variables should be limited. This issue can be tackled by restricting the cumulative differences of important system state variables. The derivative test and the difference accumulation method are introduced as extra penalties into the original objective function.

The objective function is as follows:

$$\begin{aligned}
J = & V_{Economic}(\hat{x}(t|t_s), u(t), d(t), T_f, t_s) \\
& + g_{control}(\hat{x}(t|t_s), u(t), T_f, t_s) + g_{state}(\hat{x}(t|t_s), u(t), T_f, t_s) \\
= & \int_{t_s}^{T_f} V_1(\hat{x}(t|t_s), u(t), d(t))dt - V_2(\hat{x}(T_f|t_s)) + V_3(x(t_0)) \\
& + \sum_{k=1}^{n_x} \alpha_k \left(\frac{\hat{x}_k(t_s + \Delta t_{OC}|t_s) - x_k(t_s)}{\Delta t_{OC}} \right)^2 \\
& + \sum_{i=1}^{n_u} \frac{\beta_i}{(N_{u_i}^i + 1)(\Delta T_u^i)^2} \left(\sum_{j=2}^{N_{u_i}^i + 1} (u_{i,j} - u_{i,j-1})^2 \right) \\
\text{s.t. } & \begin{cases} \frac{dx(t)}{dt} = F(x(t), u(t), d(t)) \\ x(t_s) = x_s \\ x, u \in \Omega \\ T_f^{min} < T_f < T_f^{max} \end{cases} \quad (16)
\end{aligned}$$

where t_s is each sampling point, T_f is the terminal time, which is also the predictive horizon of the optimization control, V_1 , V_2 and V_3 are process cumulative cost function, product revenue function and dimensionless function of one-time cost before production. V_3 is usually a constant. α and β are weighting factor vectors with dimensions n_x and n_u . The k th component of α and the i th component of β correspond to the degree of gentle change of the system state variable and the intensity of the control input variable or steady change.

The optimization problem in Eq. (16) can be solved directly by transforming it into a nonlinear programming (NLP) problem through the Control Variables Parameterization (CVP) method. To reduce the solution difficulty, the ODE solution integration and the barrier function method are used to solve the equation and inequality constraints respectively, and the constrained NLP problem is transformed into an unconstrained NLP problem to be solved. The optimal batch production time T_f and the optimal sequence of manipulated variables U are

obtained by solving the NLP problem with boundary constraints by the interior point optimizer (IPOPT) method.

From the general objective function (16), it can be concluded that there are three main objectives for the polymerization process, namely $V_{Economic}$, $g_{control}$ and g_{state} . $V_{Economic}$, $g_{control}$ and g_{state} represent economic benefit objectives, control stability objectives and state smoothing objectives. Mark them as J_1 , J_2 and J_3 to form a multi-objective optimization problem. Its specific lexicographical order is as follows.

In the first phase, the state smoothing target J_3 is set to the third level of the lexicographic order. J_1 is set to the second priority and J_2 is set to the first priority so that the process state is stable within a cell without large fluctuations. In the second phase, J_1 is set to the first priority. Due to the high temperature, J_3 needs to be set to the second priority to control the temperature and control smoothing J_2 is set to the third priority.

4. EXPERIMENT RESULTS

The method proposed in this paper is tested by Matlab simulation. The sampling time is set to 300s and the optimization problem is solved sequentially according to the above lexicographic ordering at each sampling time. The relationship between the temperature curves of the two batches and the highest statistics at each moment and their respective control limits after normalization is shown in the Figure 3 and 4.

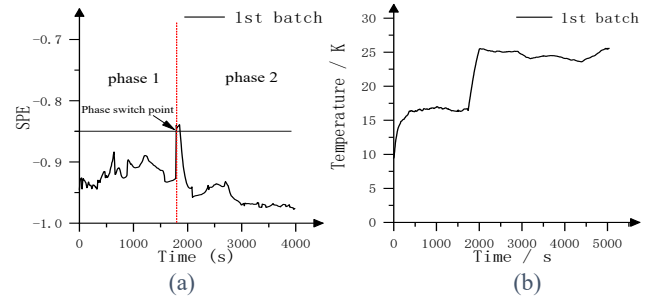


Figure 3. The first batch (a) the relationship between the highest statistics and their respective control limits (b) temperature curve

As shown in the temperature curve of Figure 3(b), in the first phase, since the first control target is smooth control, the temperature curve in the first phase is relatively gentle. After entering the second phase, the primary goal is to complete the economic indicators, so the control fluctuations at this stage are relatively large.

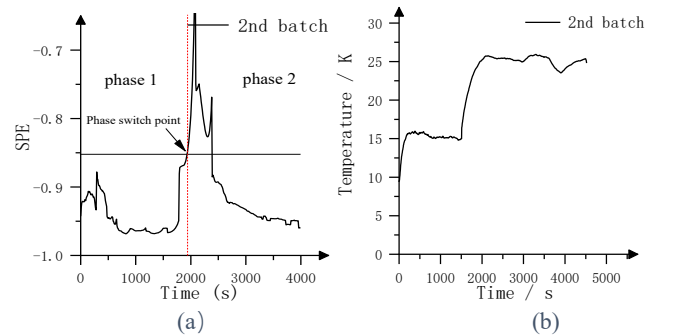


Figure 4. The second batch (a) the relationship between the highest statistics and their respective control limits (b) temperature curve

As shown in Figure 4(a), the phase transition time of the second batch is longer. This is due to the unpredictable measurement white noise disturbance in the second batch, the measured state variables are not accurate and the setting of control variables is relatively unreasonable. Since the control variable is recalculated every 300 s, the phase has an optimized cycle transition time. As shown in Figure 4(b), although the phase switching transition time is longer due to noise disturbance, the temperature profile of the second batch is similar to the first batch.

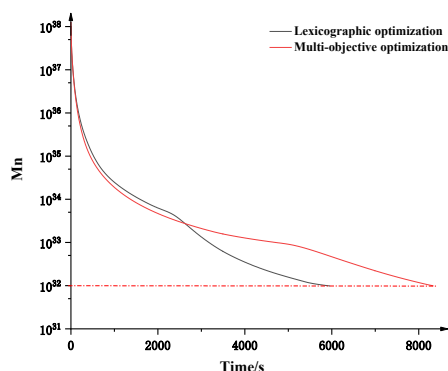


Figure 5. Average molecular weight of polymer

The production goal of the polymerization reaction can be expressed by the average molecular weight of the polymer. The average molecular weight of the polymer reaching the level of 10^{32} means that the reaction reaches the production index. As shown in Figure 5, in the first phase, the average molecular weight of the polymer is not much different. After adopting the method proposed in this paper, the method recognizes that the phase has changed, and adjusts the optimization target according to the situation. In the second phase, the first control target is the economic target. At this time, the temperature will rise steadily. In the process of the temperature rising steadily, the smooth state and the control stability target will keep the production running smoothly. The acceleration of the polymerization rate caused by this period of temperature rise enables the production to reach the required production target in advance. The economic conditions under the two types of control are shown in Table 1. Using the control method proposed in this article can greatly reduce the production time and have a higher average profit.

Table 1. Economic Profitability of two kinds of control

	Lexicographic optimization	Multi-objective optimization
T_f / s	5380	7930
Mn	1.22×10^{32}	1.21×10^{32}
Total output / kg	394.34	560.45
Total revenue / ¥	5.91×10^4	8.40×10^4
Average earnings/ ¥·s ⁻¹	10.9	10.5

5. CONCLUSIONS

Polymer production process usually has different production modes, and the optimization and control strategies corresponding to each production mode are not consistent. Since the phase switching point is usually not determined in

the actual process, this paper proposes the lexicographic optimization control method based on phase recognition. The corresponding control target lexicographic sequence is set for different phases, and the optimal manipulated variable trajectory is obtained according to the lexicographic sequence of real-time phase positioning. The simulation results show that the method can obtain more gains.

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