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# Step-wise sequential phase partition (SSPP) algorithm based statistical modeling and online process monitoring

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ABSTRACT

As batches operate at different statuses across different phases, it can be advantageous to partition the whole batch process into different phases and characterize them separately by multiple phase models. The conventional clustering-based division algorithm overlooks the time sequence of process phases and it is hard to capture the transition patterns between neighboring phases. In the present work, an automatic step-wise sequential phase partition (SSPP) algorithm is developed, which can capture the changes of process characteristics by checking their influences on monitoring system. Its theoretical support and the related statistical characteristics are analyzed. Using this algorithm, major phases are captured and also distinguished from transition patterns. The time-varying characteristics are thus described by different statistical models. For online application, the affiliation of each new sample can be realtime judged and its status can be checked by adopting the proper statistical model. Comparison is conducted between the proposed algorithm and clustering-based phase division algorithm. Comprehensive analyses are made regarding the influences of important parameters on monitoring performance. The proposed method is illustrated by a three-tank experimental system and an injection molding process which both present typical multiphase nature and transition characteristics.



## 1. Introduction

Batch and semi-batch processes play an important role in the processing of specialty chemical, semiconductor, food and biology industries for producing high-value-added products to meet today's rapidly changing market. Characterized by finite duration, batch process operation is in general carried out in different steps to produce products of desired quality at the lowest possible cost. Process disturbances, which may vary with time and from batch to batch, affect both process and product reproducibility. Hence proper process analysis and monitoring is important to not only quality improvement but also process safety [1–7]. Multivariate statistical analysis, such as principal component analysis (PCA) [8] and partial least squares (PLS) [9,10], have been widely used for data analysis and process monitoring. Bogomolov [11] gave a comprehensive review of chemometrics application to the process analysis that can be referred to as process chemometrics or multivariate process analysis. He mentioned that being a common attribute of all kinds of processes, the trajectory provides a useful point of view for the systematic consideration of various issues of the process analysis [11]. Several extensions of the conventional PCA/PLS to batch processes also have been reported. Most batch process monitoring methods are based on multi-way principle component analysis (MPCA) and partial least squares (MPLS) [12–16]. However, conventional MPCA method is difficult to reveal the changes of process correlations along time direction since it takes the entire batch data as a single object. Also, it is difficult for online application since the whole new batch data is not available up to the concerned time so that the unknown future values have to be estimated. Instead, time-slice models can be developed to capture the process characteristics at each time, which does not require future data estimation for online application. However, due to the complexity, it has not been widely used. Zhao et al. [17] proposed a moving-window based modeling approach for batch process monitoring. It does not have to estimate the unavailable future measurements, which, however, needs to develop different models at every time. Lee et al. [18] proposed a global modeling approach based on variable-wise unfolding of the entire batch data. It can be put into online application without fulfilling missing data, which, however, may not well reflect the time-varying dynamics.

Considering that the multiplicity of operation phases is an inherent nature of many batch processes, it is desirable to develop multiple phase models to capture different underlying behaviors [19–21]. Then each model represents a specific phase and explains the local process behaviors, which can effectively enhance process understanding and improve monitoring reliability. Kosanovich et al. [19] and Dong et al. [20] developed two MPCA/nonlinear MPCA models to analyze the phase-specific nature of a two-phase jacketed exothermic batch chemical reactor. The monitoring results show that the two phase-based models are more powerful than a single model. Their phase models,

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however, inherit the common weakness of the conventional MPCA model that the unavailable future data in an evolving batch should be estimated for on-line monitoring. Recently, clustering-based phase division and modeling method [22–25] has been well developed based on the fact that changes of the process correlations may relate to the phase shift in multiphase batch processes. This method doesn't require fulfilling missing process observations and can well capture the process dynamic. From another viewpoint, it can be regarded as a compromise between time-slice modeling method and variable-unfolding global modeling method. Also it can help to understand the operation process, revealing that the process characteristics stay similar within a certain time region. Since then, phase-based modeling methods have been widely used to handle different problems in multiphase batch processes. However, as one of drawbacks of clustering-based phase division algorithm, it did not consider the time sequence of operation phases. Therefore, time segments at different process time may be mixed as a single phase and some points in one phase may be assigned to other phases based on the evaluation of process similarity. Thus, the phase division results are not easy to read. Moreover, the phase partition algorithm neglects the phase-to-phase transition characteristics since each operation sample is strictly assigned to one specific phase. This may compromise the accuracy of phase-representative monitoring models. For readability, the clustering-based algorithm is simply shown in Appendix A. It is clear that the division results are also greatly influenced by the choice of input parameters, such as the initial centres, the number of clusters, minimal phase length and so on. Considering the between-phase transitional phenomena are very common in batch processes, it is significant and promising to investigate the transition characteristics. A soft-transition multiple PCA (STMPCA) modeling method [26] was proposed to overcome the hard-partition problem. Besides the identification of major phases associated with primary process operation behaviors, the smoothing transitions from one phase to another are separated by defining two kinds of radiuses for each cluster. However, it was in fact a post-processing performed on clustering-based hard division results, which can not avoid the disorder problem associated with clustering algorithm.

An automatic step-wise sequential phase partition (SSPP) algorithm is developed in the present article. The objective of SSPP is to automatically determine time segments in time sequence to present different variable correlations in order. Also, the transitions from one phase to another can be automatically separated as very short time segments or even single points. It addresses the phase nature of batch processes by tracking the influences of time-varying underlying process characteristics on the performance of monitoring models. After the division, the time-varying process behaviors can be well approximated by multiple simple and linear local models. Analyses are conducted, revealing the relationships between the proposed algorithm, the time-slice modeling method and variable-unfolding global modeling method. Online monitoring can be performed, where the specific affiliation of each new sample can be judged real time and its status is checked by projecting it onto the proper statistical model. Its feasibility and superior performance are illustrated by two typical multiphase batch processes.

## 2. Methodology

### 2.1. Automatic Phase Division

In each batch run (batch index $i = 1, 2, \ldots, I$), assume that $J$ process variables are measured online at $k = 1,2,\ldots, K$ time instances throughout the operation cycle. It forms the regular batch data set, denoted as $\mathbf{X}(K \times J)$. The data collected from $I$ batches are then arranged as $\mathbf{X}(I \times J \times K)$. As shown in Fig. 1, the three-way data array can be unfolded batch-wise or variable-wise, which keeps the batch dimension or variable dimension unchanged. The batch-wise modeling method focuses on the batch-wise variation and the variable correlations throughout the whole batch process. The variable-unfolding modeling method focuses on the variations throughout the whole batch processes and variable correlations are unified all the time. Clearly, both methods use one single model for the whole batch data which, however, focuses on different aspects. The advantages of the two methods can be effectively combined for the proposed division algorithm.

Using the proposed algorithm, both the similar characteristics within a local time region and the time sequence are considered for automatic phase and transition division. The landmarks are detected sequentially from the process beginning step by step so that it is termed step-wise sequential phase partition (SSPP) algorithm here. The basic analysis and modeling unit is time-slice data matrix. Variable-unfolding modeling unit is arranged iteratively by adding new time-slices sequentially and the resulting model's reconstruction power is checked based on the changes of confidence limits. Therefore, the phases and transition patterns are in fact identified by capturing their influences on monitoring performance.

The basic procedure is described as follows.

Step 1: Data preparation
Prepare the time-slice data matrix $\mathbf{X}_k(I \times J)$ from the process beginning. The variables at each time are then preprocessed to have zero mean and standard deviation. Input the normalized time-slice data matrix $\overline{\mathbf{X}}_k(I \times J)$.

Step 2: Time-slice based PCA modeling
Perform PCA algorithm on the time-slice data matrices and get the initial time-slice models. The number of principal components (PCs) is determined by cumulative explained variance rate [27] to keep most of the process variability (90% here). Then find the number of PCs that occurs most throughout the batch process and set it as the unified dimension of time-slice PCA models, $R$. Calculate the monitoring statistic values of squared prediction errors ($SPE$) after the explanation of PCA model at each time and determine the confidence limit, $Ctr_k$, by a weighted Chi-squared distribution [28]. It represents the reconstruction power of time-slice PCA model.

Step 3: Time-segment based PCA modeling
From the beginning of batch processes, add next time-slice one by one to the existing ones and variable-unfold them within the current time region, $\mathbf{X}_{v,k}(Ik \times J)$. Perform PCA on the rearranged data matrix and get the time-segment PCA model, $\mathbf{P}_{v,k}(J \times R)$. Calculate the $SPE$ values for each time-slice data matrix by the explanation of the current time-segment PCA model ($\mathbf{P}_{v,k}(J \times R)$) and determine the confidence limit, $Ctr_{v,k}$, by a weighted Chi-squared distribution [28]. It represents the reconstruction power of this time-segment PCA model to each time-slice within the concerned time region.

Step 4: Compare model accuracy
Compare $Ctr_{v,k}$ with $Ctr_k$ for each time slice within the concerned time region. Find the time $k^*$ from which consecutive three samples show $Ctr_{v,k} > \alpha^* Ctr_k$, where $\alpha$ is a constant attached to $Ctr_k$, termed relaxing factor here. It means that the addition of the current time-slice have imposed great influences on the time-segment PCA monitoring model and the resulting monitoring performance. The accuracy of time-segment model is thus significantly worse than that of time-slice models. So $\alpha$ determines how much the time-segment PCA model is allowed to be less representative than time-slice PCA models, i.e., insufficient reconstruction power than the same-dimensional time-slice PCA models. The time slices before $k^*$ are denoted as one sub-phase.

Step 5: Data updating and recursive implementation
Remove the first sub-phase and the left batch process data are now employed as the new input data in Step 3. Recursively repeat Steps 3–4 from the updated beginning of the batch process to find the following segments.
The output is a partition of process trajectory along time direction and the alternation of different sub-phases and transitions

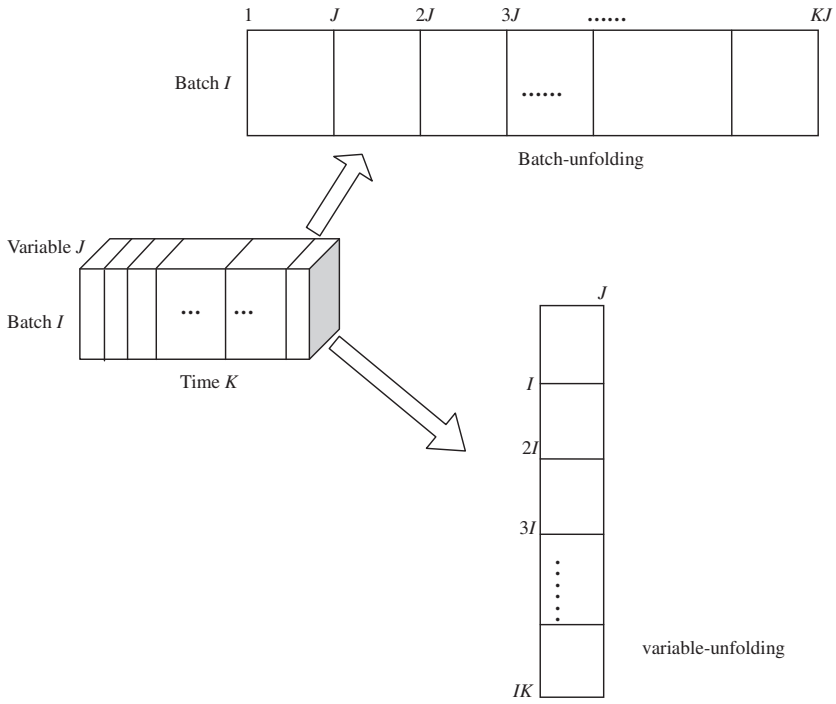


**Fig. 1.** Illustration of batch-unfolding and variable-unfolding batch data arrangement.

is consecutive along time direction. The basic idea is to find those samples with similar variable correlations so that they can be modeled by the same model while those different ones will be characterized by different models. The transition patterns, which are different from the operation patterns in steady phases, reveal faster changes of variable correlations and cannot be well accommodated by one representative monitoring model. Thus more models are required to reflect the process dynamics in transition region. It is quite possible that transition patterns can be separated at each time and represented by time-slice models.

### 2.2. Discussions and Analyses

#### 2.2.1. Relaxing Factor α

In the division algorithm, from the monitoring perspective, the modeling phases and transition patterns are separated by capturing their different influences on the monitoring system. The basic idea is that if the operation patterns are similar, they can be represented by the same monitoring system and the influences on the monitoring performance are negligible. A good monitoring model should present good reconstruction power and a close confidence limit. For time-slice modeling, the time-wise process dynamics are captured best, revealing the highest resolution to each time-slice operation pattern with the closest confidence limits. Comparatively, the number of time-segment models is clearly reduced. However, with more time-slices collected within the same phase, one single model has to cover more information simultaneously, which, thus, may lose high resolution to each time slice and result in looser confidence limits. Therefore, the relaxing extent of confidence limits is used to evaluate the jeopardized monitoring performance using time-segment model in comparison with that using time-slice models.

The relaxing extent is directly related with the predefined parameter, α. It determines the loss tolerance of reconstruction power of time-segment model in comparison with the associated time-slice models. From another viewpoint, the proposed method can be regarded as a smart combination of variable-unfolding global modeling strategy and time-slice modeling strategy. Larger α value means more reduction of reconstruction power is allowed, resulting in less number of monitoring models. Smaller α value means more accurate monitoring models to describe each time slice, which, however, requires more monitoring models.

There are two extreme cases:

- If α is set to be large enough, it will converge to variable-unfolding global modeling method;
- If α is set to be small enough, it will converge to time-slice modeling method.

Therefore, the parameter α reflects the compromise between model accuracy and model complexity. In general, the value of α can be set by trial and error so that each representative model will not cover too much process patterns to ensure the sensitiveness to the changes of process characteristics. However, up to now, there is no definite criterion or uniform standard to strictly quantify it. Therefore, its determination is inevitably affected more or less by artificial subjectivity factors. Discussions about α are shown in Illustration Section regarding its influences on the phase division results and the resulting monitoring performance.

#### 2.2.2. Number of PCs

The objective of step-wise sequential phase partition algorithm is to find segments, i.e., phases or transition regions, which can be well approximated by a linear PCA model, by performing an sequential iterative search based on a evaluation index *SPE*. So the same number of PCs can be kept in all models to find out whether the partition is acceptable by fairly comparing the confidence limits of *SPE* values. After the phase partition, the proper number of PCs retained within each representative model can be re-chosen.

#### 2.2.3. $T^2$ based Phase Division

The SSPP algorithm is presented for *SPE* monitoring statistic to track the changes of variable correlations. For $T^2$ statistic, the similar SSPP algorithm can be readily performed by simply replacing the evaluation index, which will focus on tracking the changes of systematic variations. However, it may not reflect the time-varying underlying process characteristics as sensitively as *SPE*. Also, from the illustration results, the division may not significantly influence the $T^2$ monitoring performance. Therefore, in the present work, only *SPE* is used for SSPP algorithm.

**Table 1**
Twelve fault cases in the three-tank system.

| Fault no. | Fault description |
|---|---|
| 1 | A leakage in Tank 1 by fully opening Valve 1 |
| 2 | A leakage in Tank 1 by half opening Valve 1 |
| 3 | A leakage in Tank 2 by fully opening Valve 1 |
| 4 | A leakage in Tank 2 by half opening Valve 1 |
| 5 | A leakage in Tank 3 by fully opening Valve 1 |
| 6 | A leakage in Tank 3 by half opening Valve 1 |
| 7 | A leakage in Tanks 1 and 2 by fully opening Valves 1 and 2 simultaneously |
| 8 | A leakage in Tanks 1 and 2 by half opening Valves 1 and 2 simultaneously |
| 9 | A leakage in Tanks 2 and 3 by fully opening Valves 2 and 3 simultaneously |
| 10 | A leakage in Tanks 2 and 3 by half opening Valves 2 and 3 simultaneously |
| 11 | A leakage in Tanks 1 and 3 by fully opening Valves 1 and 3 simultaneously |
| 12 | A leakage in Tanks 1 and 3 by half opening Valves 1 and 3 simultaneously |

### 2.3. Sub-phase Model Development

After phase and transition partition, the statistical analysis and modeling can then be performed for each local phase and different transition patterns. The phase-specific data matrix $\mathbf{X}_c(IK_c \times J)$ is obtained by variable-unfolding normalized time-slices which are partitioned into the same time region, where $K_c$ is the duration of the current time region $c$. It is possible that there may be only one time slice in some regions (they are actually transition patterns), i.e., $K_c = 1$. For simplicity, each transition region is also called phase here. The phase-representative relationship within each phase can thus be obtained by performing PCA on the phase-specific process data $\mathbf{X}_c(IK_c \times J)$:

$$\begin{aligned} \mathbf{T}_c &= \mathbf{X}_c\mathbf{P}_c \\ \hat{\mathbf{X}}_c &= \mathbf{T}_c\mathbf{P}_c{}^{\mathrm{T}} = \mathbf{X}_c\mathbf{P}_c\mathbf{P}_c{}^{\mathrm{T}} \\ \mathbf{E}_c &= \mathbf{X}_c - \hat{\mathbf{X}}_c \end{aligned} \tag{1}$$

where, $\mathbf{P}_c(J \times R_c)$, the sub-phase PCA loadings, reveal the major variation directions captured within the current phase and $R_c$ denotes the number of retained PCs; $\mathbf{T}_c(IK_c \times R_c)$ are the PCA scores derived from the measurement data $\mathbf{X}_c$ based on loadings $\mathbf{P}_c(J \times R_c)$. In this way,

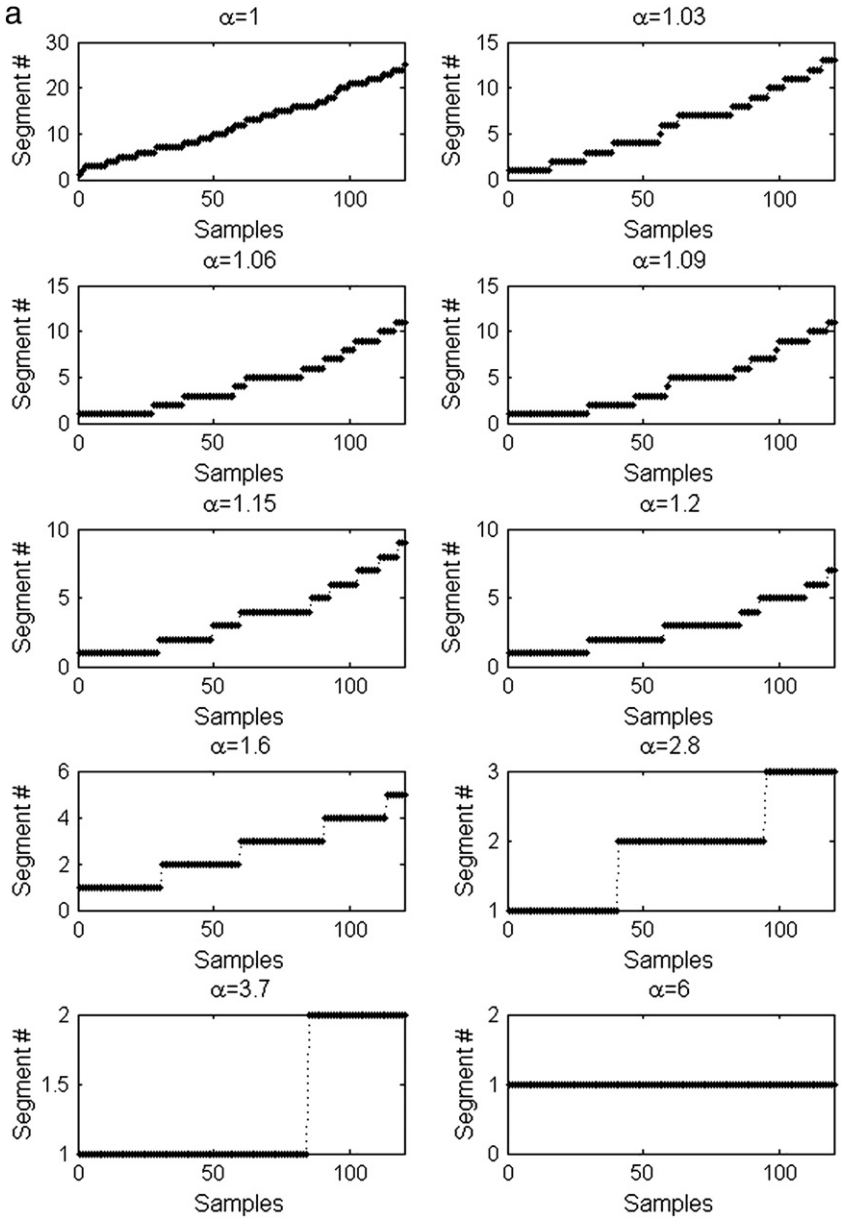


**Fig. 2.** Phase partition results for three-tank system using (a) the proposed SSPP algorithm and (b) clustering-based algorithm (bottom) regarding different values of division parameters.

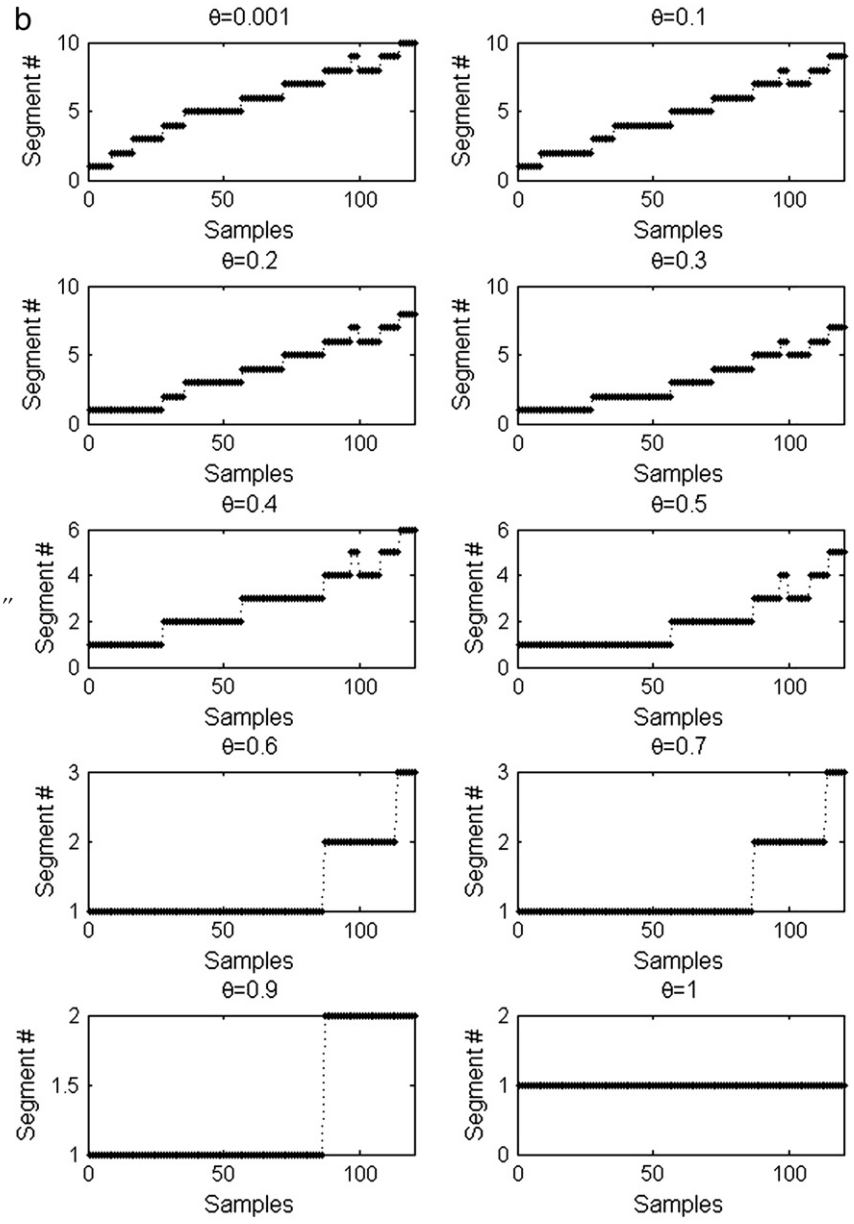


**Fig. 2** (*continued*).

the systematic information in $\mathbf{X}_c$ is characterized by $\mathbf{T}_c\mathbf{P}_c{}^{\mathrm{T}}$ and separated from the residuals $\mathbf{E}_c(IK_c \times J)$, which are deemed to be noises.

The subspaces spanned by $\mathbf{P}_c(J \times R_c)$ and $\mathbf{E}_c$ are called the systematic subspace and the residual subspace respectively. Time-slices are then separated from them, revealing the time-slice information within the same phase. Based on the extraction of scores and their separation from residuals, one $T^2$-statistic for the systematic subspace and one $Q$-statistic (also known as *SPE*) for the residual subspace are calculated for each time slice within Phase $c$:

$$\begin{aligned} T_k{}^2 &= (\mathbf{t}_k - \bar{\mathbf{t}}_c)^{\mathrm{T}} \boldsymbol{\Sigma}_k{}^{-1} (\mathbf{t}_k - \bar{\mathbf{t}}_c) \\ SPE_k &= \mathbf{e}_k{}^{\mathrm{T}} \mathbf{e}_k \end{aligned} \tag{2}$$

where, $\mathbf{t}_k(R_c \times 1)$ is the PC vector separated from $\mathbf{T}_c(IK_c \times R_c)$ and $\bar{\mathbf{t}}_k$ is the mean vector of $\mathbf{T}_k(I \times R_c)$ which are time-slice scores separated from $\mathbf{T}_c(IK_c \times R_c)$. $\bar{\mathbf{t}}_k$ is actually zero vector resulting from the use of time-slice mean-centering in data preprocessing procedure; $\boldsymbol{\Sigma}_k$ is the variance-covariance matrix of time-slice PCs $\mathbf{T}_k$. In this way, the time-varying systematic variations are modeled to a certain extent. $\mathbf{e}_k(J \times 1)$ is the PCA residual vector which is in fact the row vector in residual matrix $\mathbf{E}_c(IK_c \times J)$. The $T^2$ -statistic describes the variations in systematic part captured by $\mathbf{P}_c$; the $Q$-statistic reveals the variations in the residual part unoccupied by $\mathbf{P}_c$.

Assuming the process variables follows a multivariate normal distribution, control limits can be defined at each time. $T^2$ can be obtained by the $F$ -distribution with $\alpha$ as the significance factor [29]:

$$T_k{}^2 \sim \frac{R\left(N^2-1\right)}{N(N-R)} F_{R,N-R,\alpha} \tag{3}$$

Similarly, in the residual subspace, the confidence limit of *SPE* can be approximated by a weighted Chi-squared distribution [28] at each time:

$$SPE_k \sim g_k \chi_{h_k,\alpha}{}^2 \tag{4}$$

where $g_k = v_k/2m_k$ and $h_k = 2(m_k)^2/v_k$, in which, $m_k$ is the average of all *SPE* values at each time calculated in Eq. (2), and $v_k$ is the corresponding variance.

### 2.4. Online Monitoring Strategy

For the new observation vector $\mathbf{x}_{new}(J \times 1)$, it is first normalized by the mean and variance of the current time obtained from training data. The corresponding systematic PCs and residuals are calculated by projecting $\mathbf{x}_{new}$ onto the current phase model ($\mathbf{P}_c$) as indicated by the process time:

$$\begin{aligned} \mathbf{t}_{new}{}^{\mathrm{T}} &= \mathbf{x}_{new}{}^{\mathrm{T}}\mathbf{P}_c \\ \mathbf{e}_{new}{}^{\mathrm{T}} &= \mathbf{x}_{new}{}^{\mathrm{T}} - \mathbf{t}_{new}{}^{\mathrm{T}}\mathbf{P}_c \end{aligned} \tag{5}$$

The new $T^2$ - statistic and new *SPE*-statistic are then calculated as:

$$\begin{aligned} T_{new}{}^2 &= (\mathbf{t}_{new} - \bar{\mathbf{t}}_k)^{\mathrm{T}} \boldsymbol{\Sigma}_k{}^{-1} (\mathbf{t}_{new} - \bar{\mathbf{t}}_c) \\ SPE_k &= \mathbf{e}_k{}^{\mathrm{T}}\mathbf{e}_k \end{aligned} \tag{6}$$

Online process monitoring is conducted by continuously comparing the two statistics with the predetermined control limits. If both of them stay well within the predefined normal regions, the current sample is deemed to be operating normally. Otherwise at least one of the two statistics will go beyond the control limits responding to some process abnormality.

## 3. Illustration and Discussion

The proposed phase division and process monitoring approach is tested with two typical multiphase processes, three-tank process and injection molding process. The first one is simpler and the second process is more complex. The superiority of the proposed algorithm is demonstrated regarding its feasibility of separating phases and transitions from the whole batch process and its online monitoring performance. Since phase-based modeling methods have been well demonstrated in previous work in comparison with multiway modeling method, the current work focuses on the comparison with the clustering-based phase partition and modeling method.

### 3.1. Three-tank System

In this subsection, a typical multiphase batch process, the three-tank system, is used to illustrate the performance of the proposed method. In this case, the illustration focuses on the comparison of phase and transition separation with clustering-based division algorithm. Also analyses regarding the influences of parameter α on fault detection performance are conducted for the proposed algorithm. For three-tank process, a detailed description can refer to [26]. The process experiences several operation steps with different underlying characteristics. It presents strong process dynamics and variable correlations.

120 points of historical data collected in each batch and nineteen normal experiments carried out under the same conditions are used to yield the modeling data matrix $\mathbf{X}(19 \times 5 \times 120)$. Fifteen normal batches are used to develop the monitoring system while the left are for model testing. Beside, twelve fault types are introduced to test the fault detection performance. The programmed faults are listed in Table 1, representing different disturbances with different magnitudes imposed on different measurement variables.

First, PCA is performed on each normal time-slice data and the number of PCs at different time is determined to keep 90% data variability. The PC number used for the proposed iterative partition procedure is unified to be 1. Using the proposed algorithm and clustering algorithm, the phase partition results are compared in Fig. 2 regarding different values of important parameters. For fair comparison, no post-processing is made for both methods. For clustering-based method as shown in Appendix A, parameter $\theta$ for cluster elimination is considered while for SSPP algorithm, the relaxing factor α attached to confidence limit is analyzed. Ten different values are tried for each parameter. For SSPP algorithm, with the increase of relaxing factor α, more samples are considered to be similar with each other and included in steady phases. In contrast, the transition patterns are missing since they are accommodated into neighboring phases due to the loose confidence limits. For example, when α is set to be 1.6, only five steady phases are separated from the batch process with no transition patterns. In comparison, when α is set to be 1.03, more transition patterns (short time segments or single points between neighboring phases, e.g., after 50th sampling interval) are separated from the batch process. Moreover, it is noted that without relaxing the confidence limits ($\alpha = 1$), some time-slices can be described by the model developed from other time-slices and thus they can be classified into the same phase. For clustering-based phase division method, it presents different division results from those using SSPP method although it shows similar converging trend as $\theta$ changes. Also, it is noted that because of the choice of initial centers, it can not converge to time-slice modeling method even the value of $\theta$ is set to be very small.

From the results, it is clear that using the proposed algorithm, the whole batch process is automatically partitioned into different time segments in time order. Besides the long time regions which are deemed to represent steady phases, the transitions between neighboring phases are separated. They are in fact short time segments or even single points resulting from their more dynamic process characteristics than steady phases. The division result using the proposed algorithm is more direct and easy to be understood and no extra post-processing has to be made.

Based on the SSPP results, different PCA monitoring models are developed for each phase and transition patterns by variable-unfolding data matrices assigned to the same segment. For different values of parameter α, the monitoring performance for both normal case and fault case is compared. False alarm ratio (FAR) is used to evaluate the monitoring performance for normal case where if three consecutive samples go out of control, it is deemed that alarming signals is falsely issued. Fig. 3 only shows the *SPE* monitoring results as evaluated by FAR index for four normal testing batches regarding six values of α. The FAR value is calculated based on four normal batches where the total number of false alarming signals is divided by the total number of measurement samples. For α10, it converges to global modeling method. Resulting from the use of 99% confidence limit, it is ideal if the FAR value is approaching 1%. $T^2$ monitoring charts do not issue any false alarming signals, revealing 0% FAR, which are not shown here. For different values of α, FAR values based on *SPE* monitoring results are not significantly different, revealing that the separation of phases is not influential for normal process monitoring. Fig. 4 presents the online monitoring performance of twelve faults using

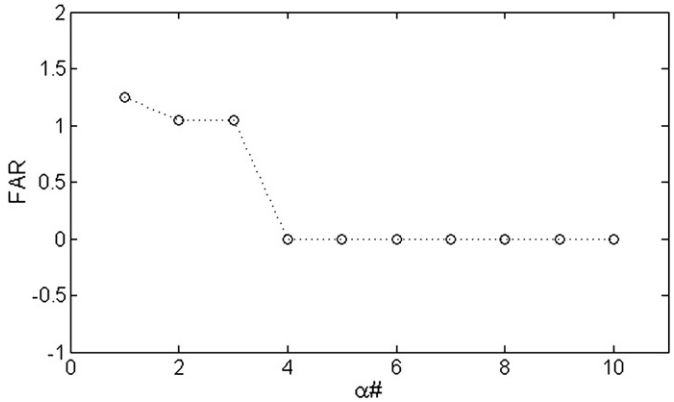


**Fig. 3.** Online *SPE* monitoring results evaluated by FAR index using SSPP algorithm for four normal batches in three-tank system regarding ten different values of α.

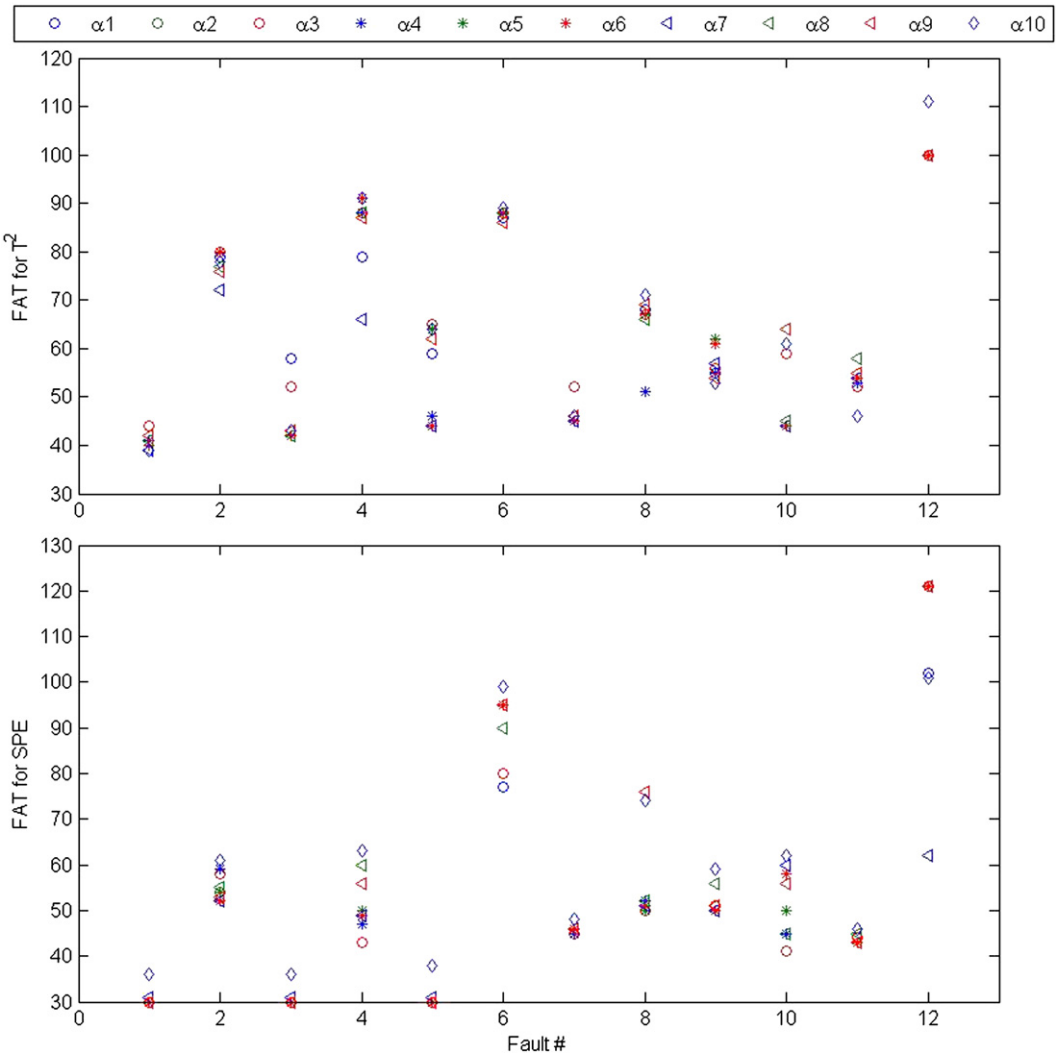


**Fig. 4.** Online monitoring results evaluated by FAT index using SSPP algorithm for twelve fault cases in three-tank system regarding ten different values of α.

SSPP algorithm for different values of α. The first alarming time (FAT) is used to evaluate the fault detection performance. It is defined as the first time when five consecutive monitoring statistic values go beyond the confidence limit, revealing the occurrence of some abnormality. Corresponding to the phase division results under different values of α shown in Fig. 2(a), *SPE* monitoring chart reveals that the global model ($\alpha 10 = 6$) is in general worse than the others for each fault, presenting larger FAT values. In comparison, $T^2$ monitoring chart does not show the superiority of multiple models over global model which may result from the use of time-varying covariance as shown in Eq. (2). This also agrees well with the recognition that *SPE* reflects the changes of variable correlations rather than $T^2$.

### 3.2. Injection Molding Process

In this section, a typical multiphase batch process, injection molding, is used to illustrate the performance of the proposed method which is a more complex process regarding the phase and transition division. Injection molding, a key process in polymer processing, transforms polymer materials into various shapes and types of products. A typical injection molding process consists of three major operation phases, injection of molten plastic into the mold, packing-holding of the material under pressure, and cooling of the plastic in the mold until the part becomes sufficiently rigid for ejection. Besides, plastication takes place in the barrel in the early cooling phase, where polymer is melted and conveyed to the barrel front by screw rotation, preparing for next cycle. It is a typical multiphase batch process and has been widely used in previous work for process monitoring and quality analysis [24,30]. It can be readily implemented for experiments, in which, all key process conditions such as the temperatures, pressures, displacement and velocity can be online measured by transducers, providing abundant process information. It provides an ideal candidate for application and verification of the proposed phase division algorithm and fault detection strategy.

The material used in this work is high-density polyethylene (HDPE). Nine process variables as shown in Table 2 are selected for modeling to reveal the process operation status. Totally 35 batch runs are conducted

**Table 2**
Nine process variables used in IM process.

| No. | Variable's descriptions | Unit |
|---|---|---|
| 1 | Pressure valve opening | % |
| 2 | Flow valve opening | % |
| 3 | Screw stroke | mm |
| 4 | Screw velocity | mm/s |
| 5 | Injection pressure | Bar |
| 6 | Nozzle temperature | °C |
| 7 | Barrel temperature zone 1 | °C |
| 8 | Barrel temperature zone 2 | °C |
| 9 | Barrel temperature zone 3 | °C |

under normal operation conditions. Beside, three types of faults are considered, each composed of forty-nine batches, including:

(1) Thermocouple fault simulation where the thermocouple measurement value of nozzle temperature is 90% of actual value. For this fault case, the real nozzle temperature will be higher than that in normal case since the nozzle will be heated until the measured temperature reaches the setting point.
(2) Heating fault simulation where the power to heat the nozzle is reduced to 80% of that under normal status. In this fault case, due to the sacrificed heating power, the real nozzle temperature is lower than the normal value.
(3) Material fault simulation where blue polypropylene (PP) is added to the original material HDPE. Due to their different properties, the corresponding process characteristics may be different.

All batches are unified to have even duration (526 samples in this experiment), which, thus, results in the three-way descriptor array $\mathbf{X}(I \times 9 \times 526)$ where $I$ denotes the number of batches for both normal and fault cases. Thirty normal batches are used for model development and the other five batches are used for model testing. For each fault case, 49 batches are available for testing.

First, PCA is performed on each normal time-slice data, the number of PCs at different time is determined to keep 90% data variability. The PC number used for the proposed iterative partition procedure is unified to be 4. Using the proposed algorithm and clustering algorithm, the phase partition results are comparatively shown in Fig. 5 for different values of parameters $\alpha$ and $\theta$. From the results, it is clear that using the proposed algorithm, the whole batch process is automatically partitioned into different time segments in time order. In comparison, based on the clustering-based partition algorithm, the affiliation of different time intervals shows quite frequent and chaotic changes. Also, different time regions are mixed together. A careful post-processing has to be carried out to manually identify the final phases. Comparatively, the division result using the proposed algorithm is more direct and easy to be understood and no extra post-processing has to be made.

Based on the SSPP result, different PCA monitoring models are developed for each phase and transition region by variable-unfolding data matrices within the same affiliation. The developed monitoring system is then applied for online monitoring. Fig. 6 shows the monitoring

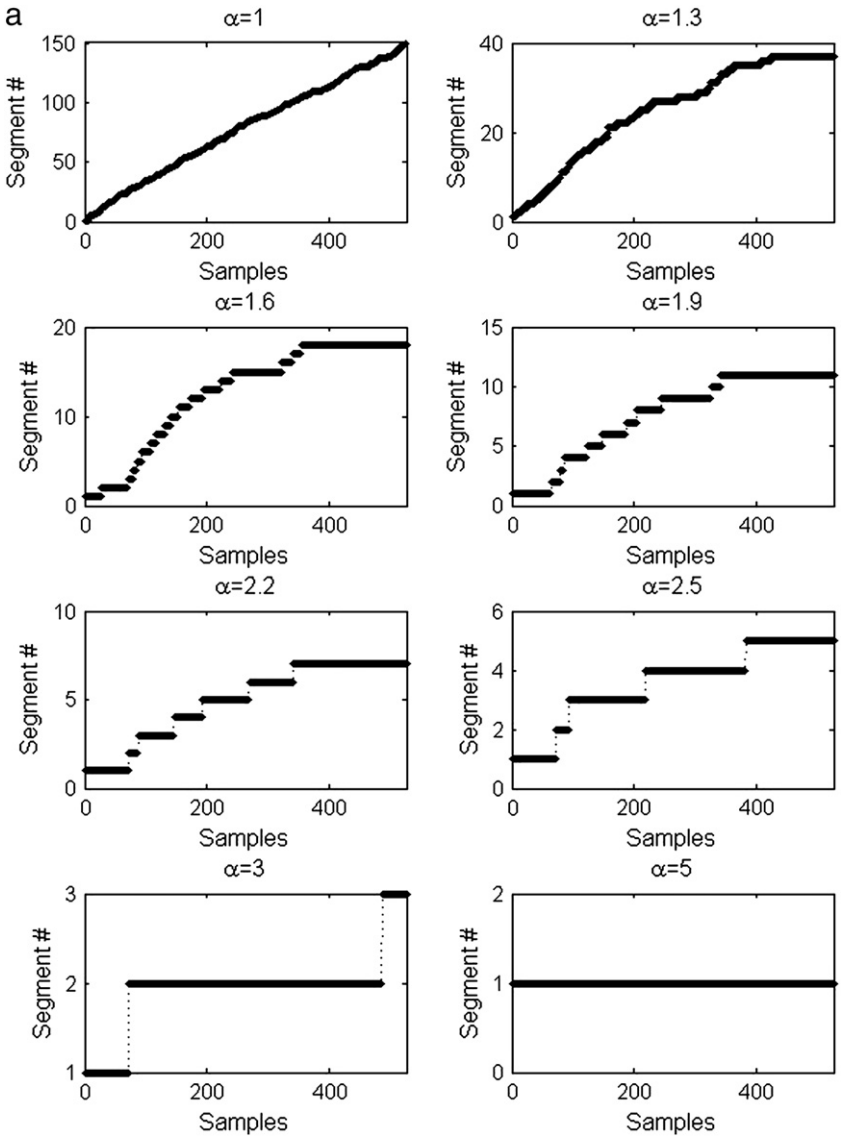


**Fig. 5.** Phase partition results for IM process using (a) the proposed SSPP algorithm and (b) clustering-based algorithm regarding different values of division parameters.

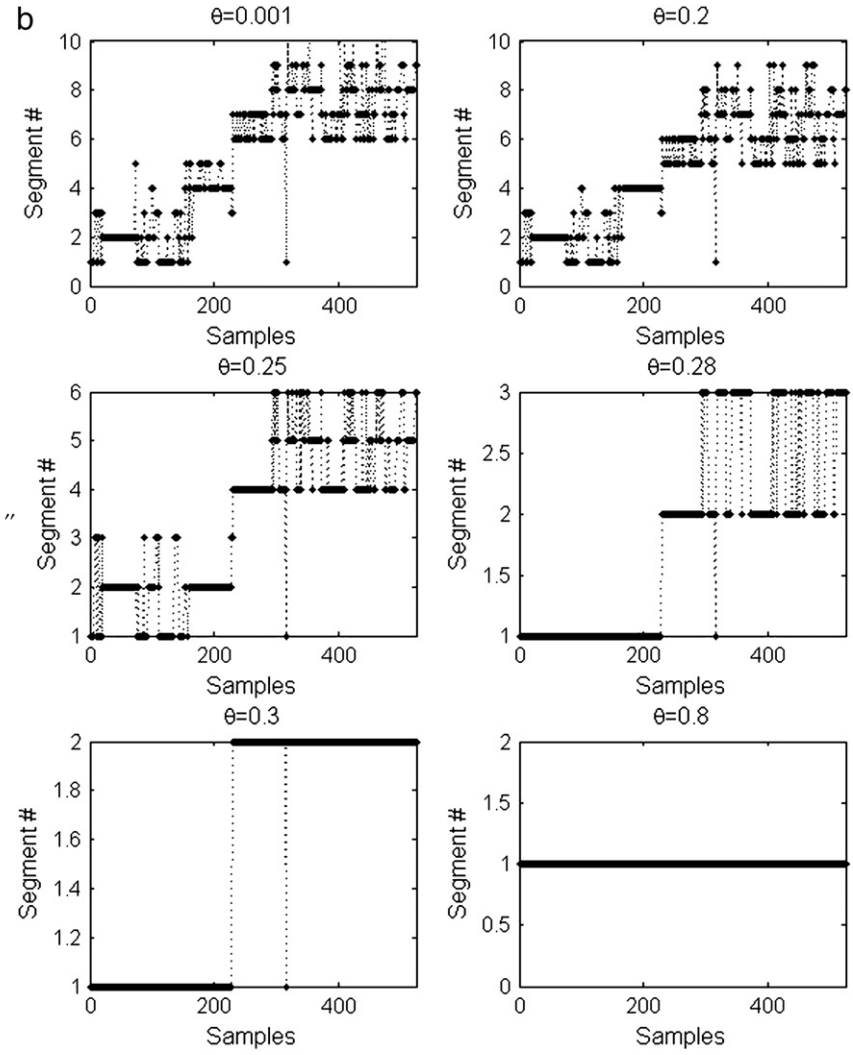


**Fig. 5** (*continued*).

performance of $T^2$ and *SPE* as evaluated by FAR index for five normal batches regarding eight values of α. FAR is calculated for five normal batches where the total number of false alarming signals is divided by the total number of measurement samples. For α10, it converges to global modeling method. In general, they do not show significant difference for different values of α except the results based on α1. Fig. 7 presents the online monitoring performance of SSPP algorithm for different values of α and three fault cases where 49 batches are tested for each fault. As evaluated by FAT index, $T^2$ monitoring chart does not obviously show the superiority of multiple models over global model which may result from the use of time-varying covariance as shown in Eq. (2). In comparison, *SPE* monitoring results present that they are more seriously influenced by α. The above conclusions are similar with those from three-tank process. Besides, for different faults, the monitoring results may be different to a certain extent. As shown in Fig. 8, the online monitoring results for one normal case and Fault 3 are presented using the proposed algorithm (SSPP) with the parameter $\alpha = 1.3$. In general, no clear false alarms are issued in normal case for both monitoring statistics. For Fault 3, the out-of-control *SPE* signals are more obvious and clear throughout the batch process in comparison with $T^2$ monitoring chart.

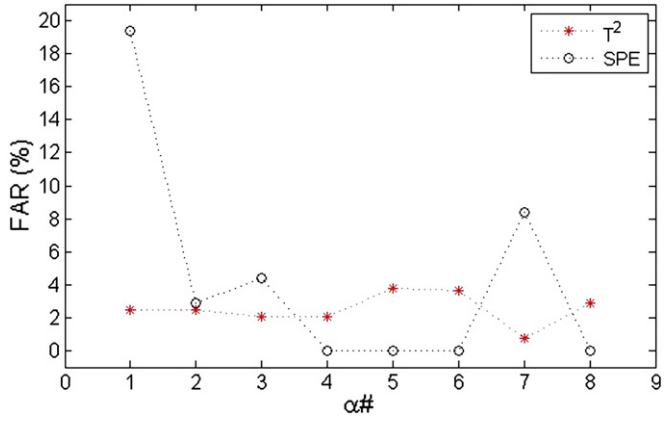


**Fig. 6.** Online monitoring results evaluated by FAR index using SSPP algorithm for five normal batches in IM process regarding eight different values of α.

Table 3 summarizes the comparison of monitoring performance between the proposed algorithm and clustering-based method for three fault types, which are evaluated using FAT index. For $T^2$ monitoring results, they do not show significant difference, which are thus not shown here. Only *SPE* based monitoring results are focused on. The mean and mean absolute deviation (MAD) values of FAT index are calculated based on forty-nine batches for each fault case. First, the best

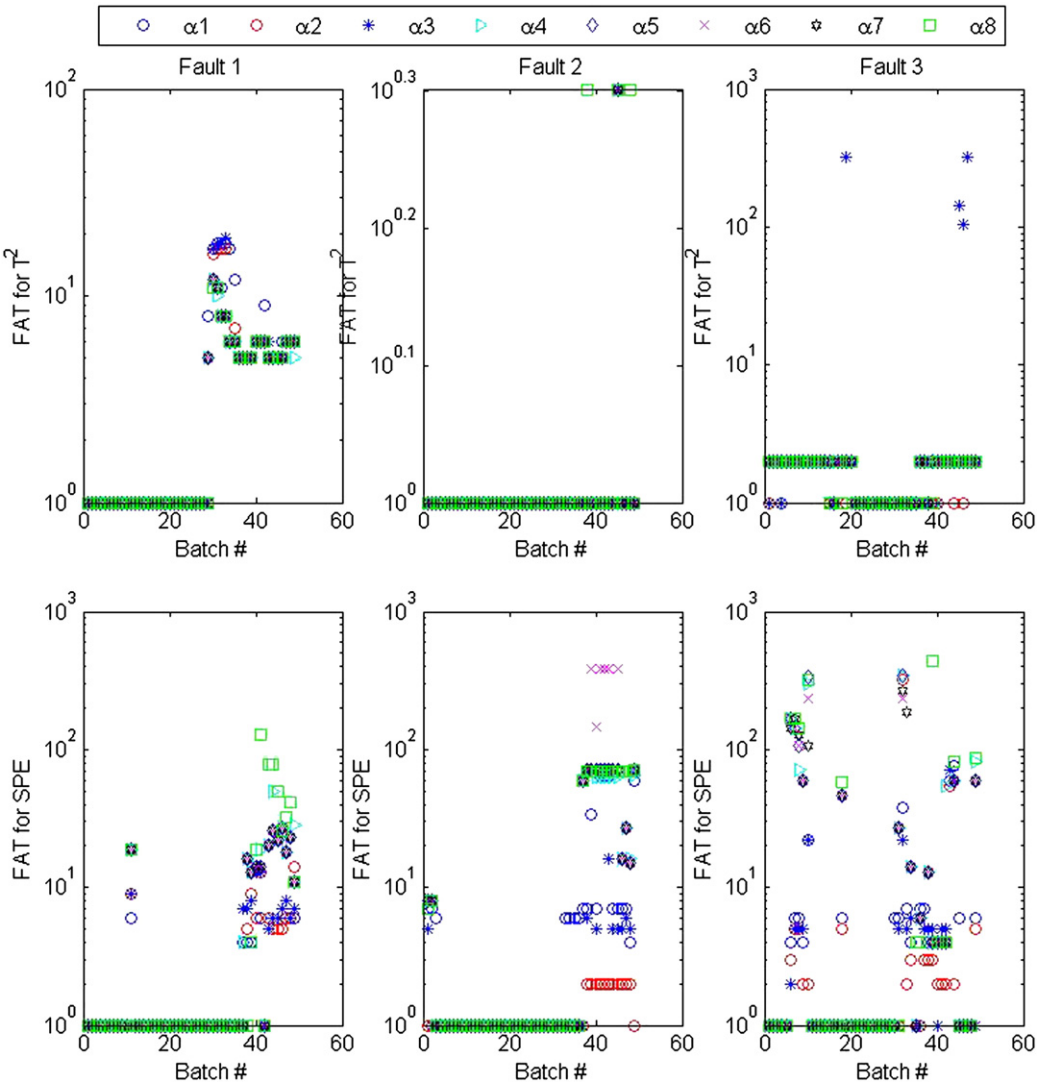


**Fig. 7.** Online monitoring results evaluated by FAT index using SSPP algorithm for three fault cases in IM process regarding eight different values of α.

monitoring results are picked up for clustering-based algorithm regarding different settings of $\theta$. For different faults, the best performance results from different choices of $\theta$. Then for the proposed algorithm, the *SPE* monitoring results that are better than the best clustering-based results (called concerned SSPP results here) are presented as well as the corresponding values of α. Finally, from the concerned SSPP results, the best one and the worst one are displayed. It reveals that there are more alternative choices of α by the proposed algorithm that can generate better results than those by clustering-based method. In general, the proposed algorithm shows comparable and even better monitoring performance especially for *SPE* monitoring statistic.

Based on the above results, clearly, by the consideration of time sequence and monitoring performance index in the phase division, the changing process characteristics can be better captured and separated for the purpose of online monitoring. By simply adjusting the value of parameter α, the proposed algorithm can converge to global and time-slice modeling methods respectively. Also the phase division results are easy to be understood.

## 4. Conclusion

In this article, a step-wise sequential phase partition algorithm is proposed to capture the time-varying underlying characteristics for multiphase batch processes. Steady phases and dynamic transitions are separated and modeled separately for online monitoring. The case study on two typical multiphase processes, three-tank system and injection molding process, shows the feasibility of the proposed algorithm for both process understanding and online monitoring. The proposed method was presented in the context of PCA monitoring system, which can be readily extended to a broad range of statistical monitoring methods. Although the division algorithm is developed for batch processes, it can also be readily modified and applied to continuous processes with time-varying characteristics that can not be well represented by a single model.

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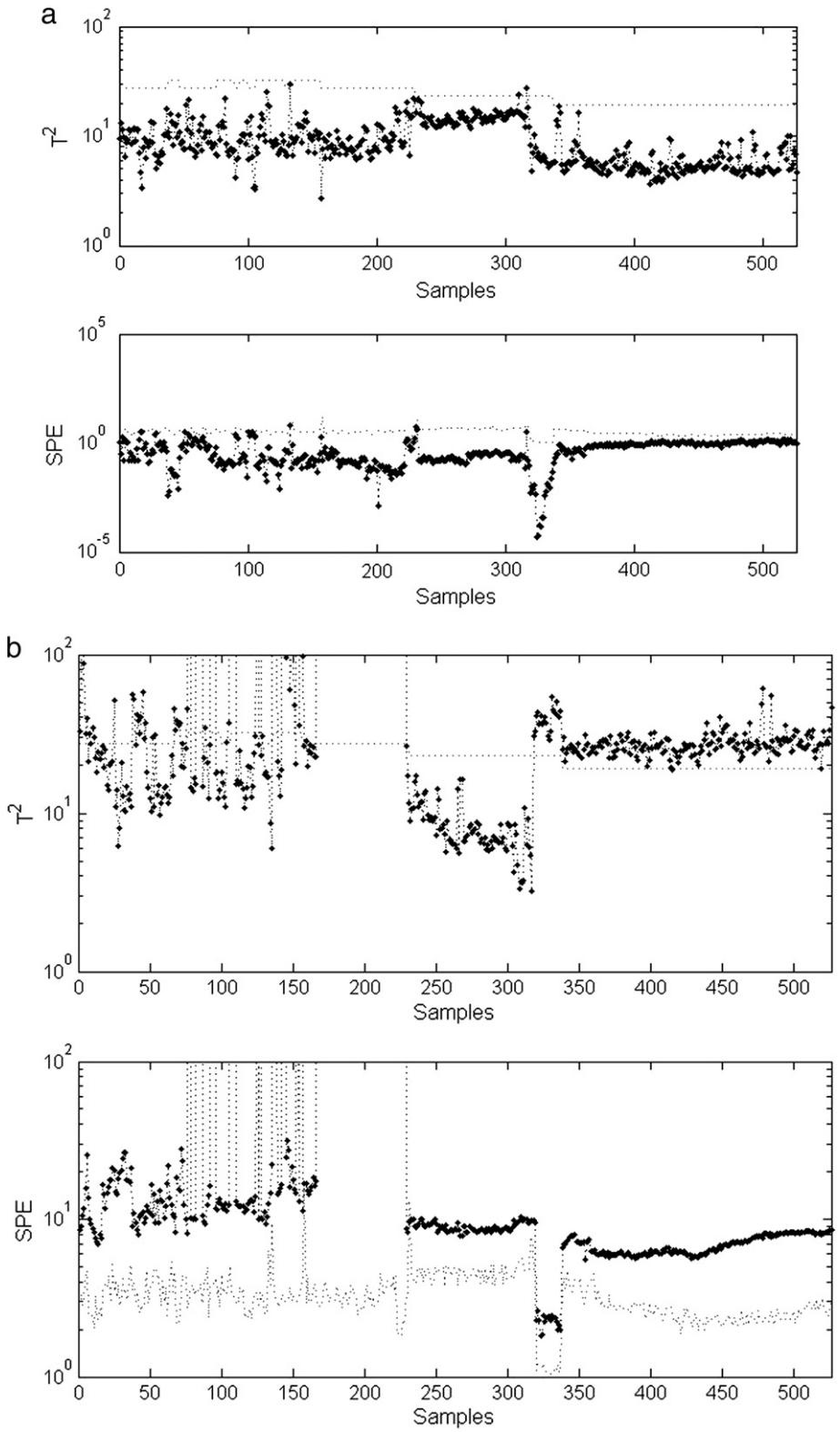


**Fig. 8.** Online monitoring results for (a) one normal case and (b) Fault 3 in IM process using SSPP algorithm ($\alpha = 1.3$) (Red dashed line: 99% control limits; black dot line: the online monitoring statistics).

## Appendix A*Clustering-based phase division algorithm*

Inputs: the patterns to be partitioned, $\{\breve{P}_1, \breve{P}_2, \ldots, \breve{P}_k\}$, and the threshold $\theta$ for cluster elimination, the minimal phase length, $L_{\min}$.

Outputs: the number of clusters $C$, the cluster centers $\{\mathbf{W}_1, \mathbf{W}_2, \ldots, \mathbf{W}_C\}$, and the strict membership of $\breve{P}_k$ to $C$ centers, $m(k)$.

The index variables are the iteration index $i$, and the pattern index, $k$.

(1). Choose $C^0(i = 0)$ cluster centers $\mathbf{W}_c^0(c = 1, 2, \ldots, C^0)$ from the $K$ patterns along the time direction. Practically, the initial cluster centers can be assumed to be uniformly distributed in the pattern set.

**Table 3**
*SPE* based monitoring performance comparison (FAT index) in IM process between the proposed algorithm and clustering-based algorithm for three fault cases.

| Fault No. | The proposed algorithm | | | Clustering-based algorithm | |
|---|---|---|---|---|---|
| | Alternative choices of α[a] | Best *SPE* based results[b] | Worst *SPE* based results[c] | θ | Best *SPE* based results |
| 1 | α1–α7 | 2.14 ± 1.68 | 6.30 ± 7.89 | θ1 | 6.39 ± 8.23 |
| 2 | α1–α5, α7, α8 | 1.22 ± 0.35 | 19.39 ± 26.49 | θ6 | 19.39 ± 26.49 |
| 3 | α1–α3, α6 | 5.41 ± 6.24 | 24.78 ± 36.25 | θ5 | 25.92 ± 40.15 |

[a] This indicates all the alternative choices of α for the proposed algorithm which show better *SPE* monitoring results than those using clustering-based algorithm.
[b] It shows the best *SPE* monitoring results chosen from the alternative choices of α.
[c] It shows the worst *SPE* monitoring results chosen from the alternative choices of α.

(2). Merge pairs of clusters whose inter-centre distance, $dist(\mathbf{W}_{c1}^{i-1}, \mathbf{W}_{c2}^{i-1})$, is below the predetermined threshold $\theta$.
(3). Calculate the distances from each pattern $\breve{P}_k$ to all of the centers, $dist\left(\breve{P}_k, \mathbf{W}_c^{i-1}\right)$, assign $\breve{P}_k$ to the nearest center $\mathbf{W}_{c^*}^{i-1}$, and denote its membership as $m(k) = c^*$.
(4). Eliminate the clusters that catch few patterns (less than the minimal phase length, $L_{\min}$) after a set number of iterations to avoid singular clusters.
(5). Update the number of cluster centers to be $C^i$, recompute the new cluster centers $\mathbf{W}_c^i(c = 1, 2, \ldots, C^i)$, using the current cluster membership, $m(k)$.
(6). Go back to Step 2 if a convergence criterion is not met. Typical convergence criteria are minimal changes in the cluster centers and/or minimal rate of decrease in squared errors.

## References


[1] T. Kourti, J.F. MacGregor, Process analysis, monitoring and diagnosis, using multivariate projection methods, Chemometrics and Intelligent Laboratory Systems 28 (1995) 3–21.
[2] K.A. Kosanovich, K.S. Dahl, M.J. Piovoso, Improved process understanding using multiway principal component analysis, Industrial and Engineering Chemistry Research 35 (1996) 138–146.
[3] S. Wold, N. Kettaneh, H. Friden, A. Holmberg, Modelling and diagnosis of batch processes and analogous kinetic experiments, Chemometrics and Intelligent Laboratory Systems 44 (1998) 331–340.
[4] D.J. Louwerse, A.K. Smilde, Multivariate statistical process control of batch processes based on three-way models, Chemical Engineering Science 55 (2000) 1225–1235.
[5] E.N.M. Sprange, H.J. Ramaker, J. Westerhuis, A. Smilde, Critical evaluation of approaches for on-line batch process monitoring, Chemical Engineering Science 57 (2002) 3979–3991.
[6] C.H. Zhao, Y.X. Sun, F.R. Gao, A Multiple-Time-Region (MTR)-Based Fault Subspace Decomposition and Reconstruction Modeling Strategy for Online Fault Diagnosis, Industrial and Engineering Chemistry Research 51 (2012) 11207–11217.
[7] T. Kourti, Multivariate dynamic data modeling for analysis and statistical process control of batch processes, start-ups and grade transitions, Journal of Chemometrics 17 (2003) 93–109.
[8] S. Wold, K. Esbensen, P. Geladi, Principal component analysis, Chemometrics and Intelligent Laboratory Systems 2 (1987) 37–52.
[9] A.J. Burnham, R. Viveros, J.F. MacGregor, Frameworks for latent variable multivariate regression, Journal of Chemometrics 10 (1996) 31–45.
[10] B.S. Dayal, J.F. MacGregor, Improved PLS algorithms, Journal of Chemometrics 11 (1997) 73–85.
[11] A. Bogomolov, Multivariate process trajectories: capture, resolution and analysis, Chemometrics and Intelligent Laboratory Systems 108 (2011) 49–63.
[12] P. Nomikos, J.F. MacGregor, Monitoring of batch processes using multi-way principal component analysis, AICHE Journal 40 (1994) 1361–1375.
[13] J.A. Westerhuis, T. Kourti, J.F. Macgregor, Comparing alternative approaches for multivariate statistical analysis of batch process data, Journal of Chemometrics 13 (1999) 397–413.
[14] P. Nomikos, J.F. MacGregor, Multiway partial least squares in monitoring batch processes, Chemometrics and Intelligent Laboratory Systems 30 (1995) 97–108.
[15] B.M. Wise, A comparison of multiway principal components analysis, tri-linear decomposition and parallel factor analysis for fault detection in a semiconductor etch process, ICRM98, International Chemometrics Research Meeting, Veldhoven, 1998.
[16] A.K. Smilde, Comments on three-way analyses used for batch process data, Journal of Chemometrics 15 (2001) 19–27.
[17] C.H. Zhao, F.L. Wang, M.X. Jia, Dissimilarity analysis based batch process monitoring using moving windows, AICHE Journal 53 (2007) 1267–1277.
[18] J.-M. Lee, C.K. Yoo, I.-B. Lee, Enhanced process monitoring of fed-batch penicillin cultivation using time-varying and multivariate statistical analysis, Journal of Biotechnology 110 (2004) 119–136.
[19] K.A. Kosanovich, M.J. Piovos, K.S. Dahl, Multi-way PCA applied to an industrial batch process, Process of American Control Conference, 1994, pp. 1294–1298.
[20] D. Dong, T.J. McAvoy, Multi-stage batch process monitoring, Process of American Control Conference, 1995, pp. 1857–1861.
[21] C. Ündey, A. Cinar, Statistical monitoring of multistage, multiphase batch processes, IEEE Control Systems Magazine 22 (2002) 40–52.
[22] C.H. Zhao, F.L. Wang, Z.Z. Mao, N.Y. Lu, M.X. Jia, Improved knowledge extraction and phase-based quality prediction for batch processes, Industrial and Engineering Chemistry Research 47 (2008) 825–834.
[23] C.H. Zhao, F.L. Wang, Z.Z. Mao, N.Y. Lu, M.X. Jia, Improved batch process monitoring and quality prediction based on multiphase statistical analysis, Industrial and Engineering Chemistry Research 47 (2008) 835–849.
[24] C.H. Zhao, F.L. Wang, F.R. Gao, N.Y. Lu, M.X. Jia, Adaptive monitoring method for batch processes based on phase dissimilarity updating with limited modeling data, Industrial and Engineering Chemistry Research 46 (2007) 4943–4953.
[25] C.H. Zhao, F.L. Wang, F.R. Gao, Improved calibration investigation using phase-wise local and cumulative quality interpretation and prediction, Chemometrics and Intelligent Laboratory Systems 95 (2009) 107–121.
[26] C.H. Zhao, F.L. Wang, N.Y. Lu, M.X. Jia, Stage-based soft-transition multiple PCA modeling and on-line monitoring strategy for batch processes, Journal of Process Control 17 (2007) 728–741.
[27] J.E. Jackson, A user's guide to principal components, Wiley, New York, 1991.
[28] C.A. Lowry, D.C. Montgomery, A review of multivariate control charts, IIE Transactions 27 (1995) 800–810.
[29] P. Nomikos, J.F. MacGregor, Multivariate SPC charts for monitoring batch processes, Technometrics 37 (1995) 41–59.
[30] C.H. Zhao, F.L. Wang, Z.Z. Mao, N.Y. Lu, M.X. Jia, Quality prediction based on phase-specific average trajectory for batch processes, AICHE Journal 54 (2008) 693–705.