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Stage-based soft-transition multiple PCA modeling and on-line monitoring strategy for batch processes

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Abstract

For the hard-partition and misclassification problems of stage-based sub-PCA modeling method, a new STMPCA (soft-transition multiple PCA) modeling method is introduced in this article to overcome these disadvantages. The method is based on the idea that process transition can be detected by analyzing changes in the loading matrices, which reveal evolvement of the underlying process behaviours. By setting a series of multiple PCA models with time-varying covariance structures, it reflects objectively the diversity of transitional characteristics and can preferably solve the stage-transition monitoring problem in multistage batch processes. The superiority of the proposed method is illustrated by applying it to both the real three-tank system and the simulation benchmark of fed-batch penicillin fermentation process with more reliable monitoring charts. Both results of real experiment and simulation clearly demonstrate the effectiveness and feasibility of the proposed method, which detects various faults more promptly with desirable reliability.

Keywords: Soft-transition multiple PCA (STMPCA) modeling; Soft partition algorithm; Transition regions; *A*-unfolding; Time-varying covariance

1. Introduction

Batch and semi-batch processes play an important role in the processing of specialty chemical, semi-conductor, 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 carried out to produce products of desired quality at the lowest possible cost. The common natures of non-steady, time-varying, finite duration and non-linear behaviors make batch processes more difficult to control than continuous processes. Process disturbances, which may vary with the development of time and from batch to batch, affect both process and product reproducibility. Hence proper process monitoring and diagnosis is important to not only quality improvement but also process safety [1–7]. Multivariate statistical methods such as principal component analysis (PCA) and partial least square (PLS) have been successfully used in modeling multivariate continuous processes. Several extensions of the conventional PCA/PLS to batch processes have also been reported, among which most batch process monitoring methods are based on multiway principle component analysis (MPCA) and partial least squares (MPLS) [8–13]. However, conventional MPCA method is difficult to reveal the changes of process correlations because it takes the entire batch data as a single object. Lee et al. [14] proposed a new statistical batch monitoring approach based on variable-wise unfolding and time-varying score covariance structures not having to estimating the unavailable future measurements whilst the dynamic characteristics of data are preserved. Chen et al. [15] integrated the time-lagged windows of process dynamic behavior with the principal component analysis and partial least square respectively for on-line batch monitoring, which can easily track the run progress and monitor the occurrence of upsets.

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Considering that the multiplicity of operation stages is an inherent nature of many batch processes and each stage exhibits significantly different underlying behaviors, it is desirable to develop stage-based models. Then each model represents a specific stage and focuses on the local behavior of the batch process, which can effectively enhance process understanding and improve monitoring reliability. Kosanovich et al. [16] and Dong et al. [17] developed two MPCA/non-linear MPCA models to analyze the stage-specific nature of a two-stage jacketed exothermic batch chemical reactor, where monitoring results show that the two stage-based models are more powerful than a single model. Their stage models, 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 which may distort the real dynamics of process correlations. Lu et al. [18,19] developed a stage-based sub-PCA modeling method based on the fact that changes of the process correlations may relate to its stages diversity in multistage batch processes, which does not require fulfilling missing process observations and preserves the dynamic relationships. However, their strict stage partition algorithm neglects the stage-to-stage transiting characteristics, which compromises the accuracy of sub-stage representative monitoring models. Considering the transitional phenomena are very common in industry batch processes, it is significant and promising to investigate the transiting characteristics.

These pioneer work have provided abundant theoretical bases for our following work. Here, a soft-transition multiple PCA (STMPCA) modeling method is introduced in the present article. Besides the division of major stages associated with primary process operation behaviors, the smoothing transitions from one stage to another are analyzed to overcome the drawbacks of strict division in stage-based sub-PCA modeling. This paper is organized as follows. In Section 2, different unfolding ways, the basis of MPCA, are outlined and sub-PCA modeling based on stage divisions is depicted, followed by a brief introduction to the proposed method (STMPCA). Consequently, details of the STMPCA strategy are explained and the monitoring statistics are established in Section 3. The superiority of process monitoring using the proposed method is illustrated by applying it to both the real three-tank system and the simulation benchmark of fed-batch penicillin fermentation process in Section 4. Finally, conclusions are drawn in Section 5.

2. MPCA modeling for batch processes

2.1. Different unfolding ways for batch data set

In each typical batch run, assume that J variables are measured at $k = 1,2,\ldots,K$ time instances throughout the batch. Then vast amount process data collected from similar I batches can be organized as a three-way array $X(I \times J \times K)$, which is the most popular batch data structure. Before performing PCA the three-dimension data array has to be unfolded in six possible ways, resulting in one of the following two-dimension forms: $A(KI \times J)$, $B(JI \times K)$, $C(IJ \times K)$, $D(I \times KJ)$, $E(I \times JK)$, and $F(J \times IK)$. Principal component analysis performed on these two-dimension matrices corresponds to checking different types of data variability. For industry applications, only matrix A and D have practical meanings. In the method of Nomikos and MacGregor [10,11], the three-way process data set is unfolded to matrix D: $X(I \times KJ)$, which is the most meaningful and popular rearrangement for batch analysis and monitoring. PCA performed on D unfolding focuses on covariance structure among process variables and variance variation along batch direction. However, it bears the drawbacks that missing future process observations should be predicted online. Recently, application have been presented in which the three-way process data set was unfolded to matrix A and scaled to zero mean and unit variance [3,12,20,21], where, each vertical slice $X_k(I \times J)$, a time-slice matrix is placed beneath one another, not beside. This approach does not require all batches to be of the equal length, nor does it require estimation of unknown future data for on-line monitoring. Mean centering directly on unfolding matrix A, however, simply subtracts a constant, the grand mean of each variable over all batches and all time, from the trajectory of each variable in each batch. Hence non-linear and time-varying trajectories are still left in the data matrix, which has little benefit to process monitoring as it focuses on the wrong source of variation in the data. Furthermore, the resulted PCA loadings can only reflect the process variable correlations and their relative importance from an ''overall'' perspective but lose their time dependency. Lee et al. [14] combined the merits of the two unfolding ways, in which the use of time-varying score covariance structures makes it able to better reflect the dynamic process relations. All the above clearly demonstrate the power and advantages of their methods in comparison to conventional MPCA. However, they suffer from the disadvantage that the loading vector is assumed to be constant throughout the entire duration, which loses the sensitivity to reflect the evolvement of process correlation characteristics from one stage to another.

2.2. Stage-based sub-PCA modeling

Generally speaking, many industrial batch processes are operated in different stages, in which input profiles, conditions, process characteristics and control strategy vary greatly from one stage to the next. To satisfy the monitoring requirements over different operation stages, Cenk Ündey and Ali Cinar [22] have argued that different stages in a process can also be included in the analysis by developing stage-based models, which allows one to unveil the correlation structure specific to each stage and increases our capability to identify events localized in stages. Louwerse and Smilde [4] developed a strategy to partition reference data into several time periods for improvement of on-line

monitoring. But their method, based on MPCA, requires also the unavailable process measurements be estimated when on-line monitoring. Adaptive batch monitoring strategy based on recursive multiblock PCA proposed by Rännar et al. [23] avoids the need of estimating or filling unknown data. Its computational demand, however, can be overwhelming. To overcome these disadvantages, a stage-based sub-PCA method has been developed by Lu et al. [18,19]. The key for their method is to divide a batch process into several stages and build a representative model for each stage. This idea assumes that stage nature can be reflected by statistical characteristics, such as the process variable correlations, by proper clustering. PCA is performed on these time-slice matrices X_k $(k = 1,2,\ldots,K)$ based on D-unfolding, generating K number of loading matrices, P^k, which represent the process correlations at K time intervals. Then an improved K-mean clustering algorithm is used to group the K patterns into C number of clusters, representing C different sub-stages. Within each of these "operation" stages, the process correlations are similar; and the representative sub-stage monitoring models can be built. This method allows two-way PCA to be "directly" applied for a batch process. It has significant benefits because the latent variable structure is allowed to change at each stage which reflects the dynamics of inherent process correlation nature [18,23,24]. Analysing the stage-based data model also allows for detecting more specific locations of faults in a process [4]. However, one major problem of sub-PCA modelling is that misclassification may occur during the transition region between two neighboring stages, because the adopted k-means clustering algorithm is a hard-partition method. They strictly separate sub-stages off from the process by clustering time-slice loading matrices into certain class with absolute memberships of 0 or 1. Another major deficiency is the representative stage models in [18,19] is simply obtained by averaging time-slice loadings within the same stages. Hence they only focus on the variance variation along batch axis isolated at each time interval without capturing the dynamic evolvement along time direction in every stage. Moreover, their covariance structures remain constant across each stage without taking into account the time varying of the scores. All the above mentioned will affect the accuracy of sub-stage representative monitoring models and inevitably result in false or missing alarms when online monitoring.

As mentioned above, the MPCA based on A-unfolding as well as time-varying score covariance structures and stage-based sub-PCA methods both have advantages and drawbacks. In this paper, we propose a new online batch monitoring method which combines the advantages of the two approaches and extends the ideas of previous work [18,19]. Moreover, it gives a solution to the challenging modeling and monitoring for process transitional behaviors from one stage to another. The differences between the proposed method and previous ones lie in (1) this work separates the transition regions from neighbouring stages and gives them detailed descriptions focusing on process characteristics, which will be beneficial to process understanding and analysing; (2) In each sub-stage, representative sub-PCA models are rebuilt based on A-unfolding, which can capture the variance structures along both batch and time directions within the same stage. Moreover, it can reflect the process variable correlations and their relative importance from an "overall" perspective; and (3) during the transition regions, multiple monitoring models are established according to the changing membership trend, which cover more information connecting neighbouring stages and overcome the disadvantage of hard-partition. The concrete algorithm and procedure are shown in the next section.

3. Methodology

3.1. Nature of transition regions

Generally speaking, industry batch processes operate in a variety of states. Some of them are steady states while others including grade changes, startup, shutdown, and maintenance operations are transitional ones. For multistage process, transition regions between neighboring stages are very common, which show the gradual changeover between two neighboring patterns possibly with local fluctuation. Regularly, at the beginning they have the underlying characteristic more similar to the previous stage, while at the end more similar to the next one. Different transitions go through different trajectories from one steady pattern to another, where the underlying process characteristics dynamics along time direction are more frequent and complex than those within each stage. So they are more prone to abnormalities and become the crucial moment for the final product quality. Therefore valid process monitoring during transitions is important to guarantee stable and good transition performance. Duchesne et al. [25] focused on historical transition data analysis including the diagnosis of reasons for poor transitions to ensure that new transitions follow an acceptable path. Robin et al. [26] developed a methodology to improve the product quality in batch processes by reducing the transitional duration of the startup stage.

However, up to now very few investigations about transition modeling and monitoring have been reported to improve process transition performance using multivariate statistical methods based on PCA and PLS. In view of the close and changing relationships between transition regions and their neighboring stages, dynamic monitoring models covering process characteristics transiting trend seem to be an attractive approach.

3.2. Concept of division algorithm

Each vertical time slice from three-way matrix, $X_k(I \times J)$ is the basic unit in the stage division algorithm to obtain the process correlation at sampling time k. Although batch process variables are time-varying, fast or slow, the local

covariance structure will be largely similar within the same stage, indicating that the process is driven by the similar underlying characteristic.

As mentioned before, the time-slice loading matrices, P^k, represent the local covariance information and underlying process behaviours, which can be used to determine the operation stages by proper analyzing and clustering procedures. Generally, the process correlations show the developing trend along the time axis. In most cases, since all patterns are extracted along the sampling time of batch processes, the clustering result can be directly associated with the operation time, which makes the partition of the patterns well interpretable. Sometimes there may be some exceptions that several disjoint operation periods have the same underlying characteristics. Combined with actual process time, they are also divided into the different stages. So each separated process stage should contain a series of successive samples. Moreover, because of the variation complexity of process characteristics in transition regions between neighbouring stages, it is unsuitable to forcibly divide them respectively into the individual stages. On the one hand, the transiting alteration of process characteristics imposes disadvantageous effects on the accuracy of stage-based sub-PCA representative monitoring models; on the other hand, it deteriorates fault detecting performance just employing certain sub-PCA model to conduct monitoring of transition patterns. Consequently both will compromise the expected reliability of on-line monitoring.

Here, our proposed method separates the transition regions between neighboring stages after the initial stage division whose durations are determined through the detailed analysis of transition characteristics. Relying on the concept of membership grades, different monitoring models are established in transition regions which are the weighted sum of sub-models. The detailed modeling and monitoring procedure is shown in the next section.

3.3. Modeling and monitoring procedure

3.3.1. Clustering loading matrices

In PCA analysis, the loading matrix reveals the information of process correlations. The stage-based multiple PCA modeling begins with analyzing the loading matrix at each sampling interval. The time-slice matrices $X_k(I\times J)$ $(k=1,2,\ldots,K)$ are scaled firstly and PCA can be performed on these scaled ones $\overline{X}_k$, generating K number of loading matrices P_k. Then the loading matrices P_k are transformed into a weighted form after considering the importance of each column $\mathbf{p}_{j,k}$, which represent the process correlation pattern at each time interval

$$\begin{aligned}\breve{P}_k &= [\mathbf{p}_{1,k}\cdot g_{1,k}, \mathbf{p}_{2,k}\cdot g_{2,k}, \ldots, \mathbf{p}_{J,k}\cdot g_{J,k}] \\ &= P_k\cdot \mathrm{diag}(g_{1,k},\ldots,g_{J,k}) \end{aligned} \quad (1)$$

where $g_{j,k}=\lambda_{j,k}/\sum_{j=1}^{J}\lambda_{j,k}$, and $\lambda_{j,k}$ is the eigenvalue of the co variance matrix $(\overline{X}_k)^T\overline{X}_k$.

Since each loading matrix is actually vector space consisting of J column vectors, the square sum of difference between corresponding vectors, i.e. the Euclidean distance, is used to calculate the dissimilarity between two patterns. According to the prior process knowledge, the number of initial clusters can be chosen properly, which is commonly larger than the number of actual stages. But if there is no prior knowledge as the reference criterion, it can be assumed by experience to be $K/3\sim K/2$ (K is the whole process duration) uniformly distributed in the pattern set. Then a variant k-means algorithm [18,27] is adopted for automatically partitioning the K number of weighted loading matrices $\breve{P}_k$ $(k=1,2,\ldots,K)$ into the several clusters. The local squared error (for patterns within each cluster) and the global squared error (for all the patterns) are minimized by specifying a threshold θ of the minimal distance between two clusters' centres, or the maximal radius of a cluster. A step is added in the clustering algorithm to eliminate singular clusters that catch few patterns in the iterative clustering procedure to enhance the robustness and reliability of the partition algorithm. Finally, C number of clusters, representing C kinds of pattern features, can be used to define process stages, where the precise of stage representative models is determined how reasonably the data can be separated into different clusters so that each cluster can contain proper number of patterns. The modeling accuracy and complexity depend on the specification of the threshold. A larger threshold results in few clusters and more sampling points in each cluster, which contain stable and sufficient process characteristics but cannot more sensitively unveil the correlation varying between different patterns. Contrastively, a smaller one generates more clusters focusing on reflecting the evolvement of process correlation dynamics from one stage to another. However, less samples in each class cannot provide enough process operation information for each stage and induce the lack of model robustness. That is, larger threshold conforms to the capability of extracting stable and sufficient stage characteristics from each cluster and smaller value corresponds to the required ability to track the process varying dynamics between different stages along time. In conclusion, the threshold value should be determined by a tradeoff between the above two appealing abilities by trial and error. Moreover, the indicator variable technique [6] and prior process experience also can provide stage completion information that can help to divide stage instead of only depending on the clustering results.

3.3.2. Soft-transition partition algorithm

Each time-slice is partitioned into the corresponding sub-stage according to the above clustering algorithm mentioned in [18,19]. However, the cluster of data in real world mostly do not have so strict boundary, and there exists various transition regions between neighboring stages. The fuzzy rules proposed by Zedeh provide effective theory tools for solving the problem. Membership degree values with 0–1 are used to describe the partition problem with

ambiguous boundary, which can objectively reflect the process correlations changing from one stage to another. The values 0 and 1 indicate no membership and full membership respectively whereas grades between 0 and 1 indicate that the pattern has a partial membership in a cluster.

According to the above-mentioned, a soft-transition multiple PCA (STMPCA) modeling method is introduced in this article to treat the problem. It presents the definition of class radius and kernel radius to determine the range of transition region between different stages. Meanwhile, it introduces the membership grades to evaluate quantitatively the similarity between transitional and sub-stage patterns. Then sub-PCA models for sub-stages and weighted sum models for transition regions are established respectively which greatly improves the accuracy of sub-stage models and strengthens the robustness of transition models. Moreover, it reflects objectively the changing diversity during transition regions between different neighboring stages. Here, the previous stage division results provide an important precondition for the following separation of transition regions.

Firstly define P_c^* $(c = 1, 2, \ldots, C)$ as the initial representative center for the cth stage:

$$P_c^* = \frac{1}{n_c} \sum_{k=1}^{n_c} \breve{P}_k \tag{2}$$

where n_c is the number of time-slices belonging to the cth stage.

For K number of weighted loading matrices $\breve{P}_k$, the Euclidean distance between them and the corresponding initial centre P_c^* of sub-class is defined as follows:

$$d_{k,c} = \|\breve{P}_k - P_c^*\| \quad (k = 1, 2, \ldots, K,\ c = 1, 2, \ldots, C). \tag{3}$$

Then they are used as dissimilarity index to quantitatively evaluate the changing trend of process correlation characteristics. In our work, the distance is defaulted as Euclidean distance without special statement.

Class radius r_c and kernel radius r_c' for each centre P_c^* lay the crucial foundation for soft-transition partition algorithm, which are defined respectively by (4) and (5), where γ_c and γ_c' are adjustable parameters.

$$r_{c,i} = \gamma_{c,i} \cdot \|P_i^* - P_c^*\|, \quad 0.5 < \gamma_{c,i} < 1 \tag{4}$$

$$r'_{c,i} = \gamma'_{c,i} \cdot \|P_i^* - P_c^*\|, \quad 0 < \gamma'_{c,i} < 0.5 \tag{5}$$

where $i = c - 1,\ c + 1$. P_c^* is the current centre and P_i^* stands for the adjacent centers around the current one. It can be seen that for the current center, its class or kernel radiuses may be asymmetric with distinct adjustable parameters when approaching different neighbouring stages before and after it.

After that, we can concretely differentiate the fuzzy membership regions from strict ones in the following two aspects:

Firstly, when $\breve{P}_k$ is within the kernel radius r_c' of P_c^*, or $\breve{P}_k$ lies within the class radius r_c of centre P_c^*, and outside those of any other centre, it is said that $\breve{P}_k$ only belongs to centre P_c^*. That is to say, the sampling data lies in the cth sub-stage with full membership. The definition can be simplified as Eq. (6):

$$\mu_{k,c} = 1,\ \mu_{k,j} = 0 \ \text{ if } d_{k,c} < r_c' \text{ or } (d_{k,c} < r_c \text{ and } d_{k,j} > r_j),\quad j = 1, 2, \ldots, C,\ j \neq c \tag{6}$$

Otherwise, the others are set to be within the transition regions between neighbouring stages, i.e. when $\breve{P}_k$ lies at the conjoint regions between p_{c-1}^* and P_c^*, define the membership grades as follows:

$$\mu_{k,c} = d_{k,c-1}/(d_{k,c} + d_{k,c-1}), \mu_{k,c-1} = 1 - \mu_{k,c}, \mu_{k,j} = 0 \quad (j = 1, 2, \ldots, C,\ j \neq c, c-1) \tag{7}$$

At last, each time-slice loading matrix $\breve{P}_k$ is partitioned into the corresponding sub-stage or transition region with its membership grade to C number of sub-stages, which can be summarized as $\mu_k = [0, 0, \ldots, \mu_{k,c-1}, \mu_{k,c}, \ldots, 0]$.

To help understand the meaning of soft-partition, without losing generality, we simply take an ideal two-stage process with K-duration as example to visually explain the above definitions and descriptions. The sketch map of soft-transition partition algorithm as well as the two radiuses is illustrated in Fig. 1. Based on the clustering procedure in [14], the whole process duration is initially partitioned into two strict clusters along time, i.e. span OS is cluster 1 and span SK is cluster 2. Then for each individual sampling pattern at time interval k, there are two distance metrics, $d_{k,1}$ and $d_{k,2}$, respectively corresponding to centers 1 and 2, where P is the dividing point of the two sub-stages. The change of process correlations evaluated by Euclidean distance along time reveals the real existence of transition region midst the two stages and shows the gradual transiting from one stage to another. From the plot, the patterns around point "P" have the underlying process characteristics similar to the two neighboring sub-stages. So it is improper to strictly partition them into the individual stages assigned with full membership. Based on the definition of kernel and class radiuses, the transition region is naturally separated from the strict sub-stages, which are depicted in Table 1. Consequently, according to Eqs. (6)

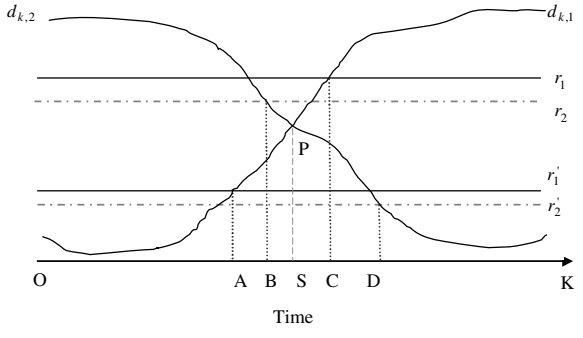

Fig. 1. Sketch map of soft-transition partition algorithm for a two-stage process ($d_{k,1}$ and $d_{k,2}$ are the distance dissimilarity of every pattern $\breve{P}_k$ to centers 1 and 2, respectively; r and r' denote class and kernel radiuses of the two centers).

Table 1
Separation of sub-stages and transition regions

Span	Membership classes	Warranty conditions
OA	Sub-stage 1	$d_{k,1} < r'_1$
AB	Sub-stage 1	$d_{k,1} < r_1$ and $d_{k,2} > r_2$
BS	Transition region	Otherwise
SC	Transition region	Otherwise
CD	Sub-stage 2	$d_{k,2} < r_2$ and $d_{k,1} > r_1$
DK	Sub-stage 2	$d_{k,c} < r'_2$

and (7), the patterns during the transition region can be attributed with partial membership grades to the two sub-stage so as to cover the two different modes of process nature.

From the above illustration analysis, it can be seen that the transitional duration has a close relationship with the determination of class and kernel radiuses. Their relaxation or tightness will lead to the corresponding span variety of transition regions and affiliation memberships, and then indirectly impose on the accuracy and sensitivity of on-line monitoring models. For the case illustrated in Fig. 1, generally larger class radius and smaller kernel radius can generate broader span of transition regions sensitive to reflect the evolvement of process states from one stage to another. However, shorter sub-stages duration after the separation of transition regions may not contain sufficient points to establish a more robust sub-stage monitoring model covering more comprehensive stage-specific characteristics. Moreover, some patterns during transition regions with larger membership grades very close to 1 indicates that it is unnecessary to calculate their fuzzy sub-ordinate relationship with calculation complexity and burden. In contrast, if class radius is set smaller, main sub-stages will contain more sampling patterns, which reveal rich process operation conditions and help to establish more stable sub-stage models. However, the transition regions between two neighboring stages will shrink correspondingly. Some patterns characterized with transiting nature are forcibly partitioned into sub-stages, which may impact the accuracy of sub-stage modeling. In conclusion, the key point is how to properly choose the size of kernel and class radiuses so that process sampling patterns can be separated properly into different sub-stages and transition regions to make sure that each stage can contain proper number of patterns and the duration of transition regions can flexibly and rationally reflect real process characteristic changeover from one mode to another.

Here, we give the following guideline for the determination of the two radiuses:

Firstly, according to the physical meaning of the two radiuses, the specific transition circumstance in actual process, such as transiting speed and duration, can provide important reference standard. For the case depicted in Fig. 1, long transition duration and slow transiting speed correspond to larger class radius and smaller kernel radius; otherwise short and fast transition conform to smaller class radius. The real transition circumstantiality can be obtained through various means: the distances calculated from Eq. (3) provides quantitatively the changing trend of process correlations along time, especially during the transition regions, which will help to evaluate the dynamic transition characteristics. Moreover, the indicator variable technique can help to capture process progress state and the prior expertise of process operation can also offer general reference information about transiting nature.

Secondly, since the present method is oriented to improve the transition smoothness between neighboring stages and strengthen the reliability of online monitoring, the radiuses should be chosen by trial and error through cross-validation so that the number of online false and missing alarms is minimized.

At present there is still no related pioneer guideline about how to determine the starting time and span of transition regions. Moreover, in practical process, due to the complex process operation characteristics, it is difficult to establish the definite criterion or uniform standard to strictly quantify transition course. Accordingly, the determination of kernel and class radiuses is inevitable to be affected a certain extent by artificial subjectivity factors. According to our trial and error, the proposed method has better robust adaptability to the mild variation of the two parameters. Although the optimal values of the two parameters cannot be acquired strictly and determinately, their variety within small range cannot affect severely the monitoring effectiveness and reliability. In fact, the soft-partition idea and concept have been an improvement and progress with the stage-based sub-PCA modelling [14] by yielding more reliable monitoring results which will be illustrated further in the later Section 4. Therefore, the proposed soft-partition algorithm and transition analysis has provided a significative attempt to solve the modeling and monitoring problem of transition regions between neighboring stages. The rational analysis and separation of transition regions will be promising and deserve further attention and investigation in future.

3.3.3. Develop multiple PCA models

A representative sub-model should cover the major variance/covariance information across both time and batches and reflect the correlations of the process variables from an overall perspective. And transition models should be able to reflect the changeover trend from one stage to another with the development of process time. Here, according to the above soft clustering results shown in Fig. 2a, multiple PCA models are designed respectively for sub-stages and transition regions.

(1) In each sub-stage, scaled time-slice data matrices $\overline{X}_k(I \times J)$ are rearranged into the form of A-unfolding $\overline{X}_c(n_c I \times J)$, where n_c is the number of time-slices belonging to stage c. Then using PCA method, the loading $P_c(J \times R_c)$ and score $T_c(n_c I \times R_c)$ are extracted, which preserve simultaneously the major dynamic relations along both time and batch direction within each stage from the viewpoint of

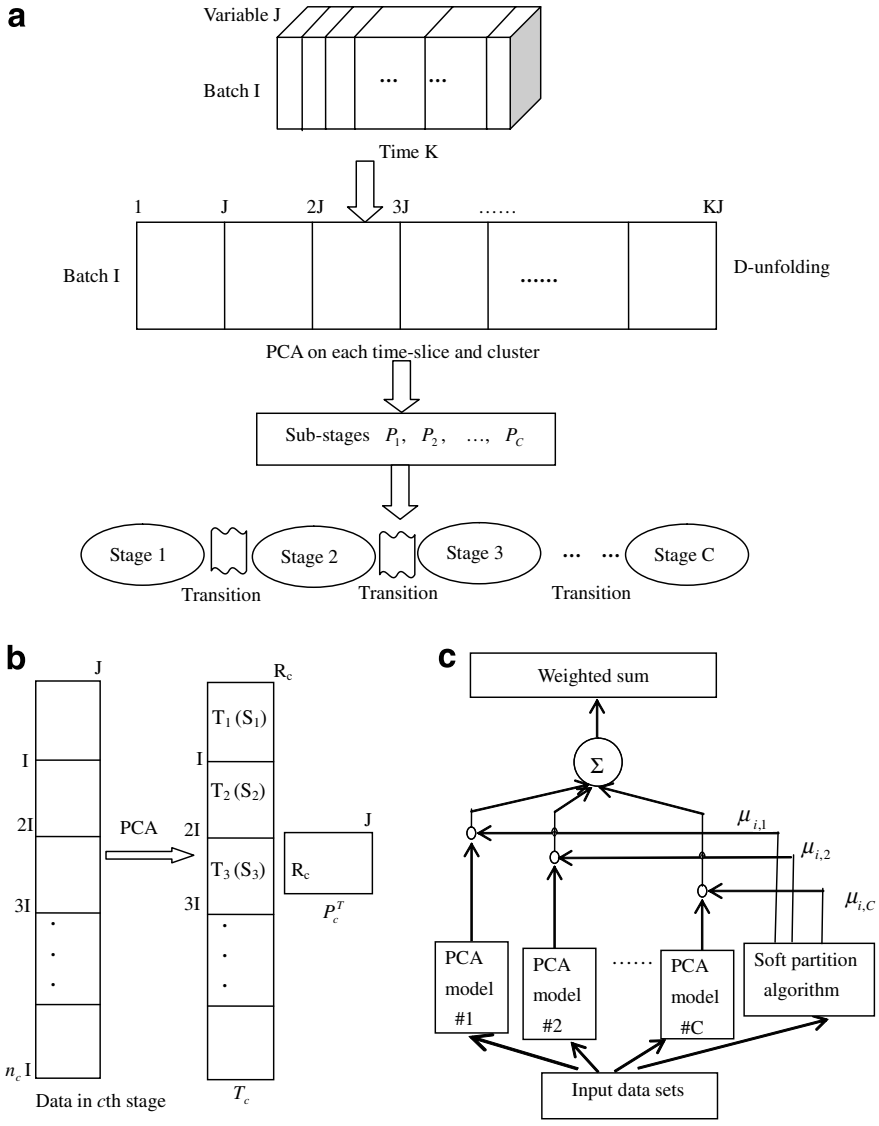

Fig. 2. Illustration of STMPCA algorithm. (a) Soft-partition, (b) sub-stage model, (c) transition model.

process development. Here, R_c is the number of retained principal components determined by the cumulative explained variance rate $\sum_{j=1}^{R_c}\lambda_{j,c}/\sum_{j=1}^{J}\lambda_{j,c}$. Next, the score at each time k, $T_k(I\times R_c)$ is separated from T_c and the covariance matrix at each time, $S_k(R_c\times R_c)$, is calculated from the score matrix at each time. The algorithm can be seen in Fig. 2b.

(2) During transition regions, every time interval obtains its membership grades to sub-models as the dissimilarity index which determine the degrees in which the corresponding sub-stage models contribute to the transiting model. From Fig. 2c, the transition monitoring models at every time interval can be obtained by the weighted sum of sub-PCA models $P_k = \sum \mu_{k,c}P_c^*$, which properly reflect the time-varying transiting development. The number of retained principal components is set equal to the larger one of its two neighbouring sub-PCA models to keep the uniform dimensions. Also the score T_k as well as the covariance matrix $S_k(R_k\times R_k)$ can be obtained at each time instance.

3.3.4. Process monitoring with STMPCA models

By the above algorithm, STMPCA model P_k is obtained corresponding to every time interval in sub-stages and transition regions. In this study, we have made such an important premise that the batches should be of equal length that the specific process time can be used as an indicator to judge which representative model should be employed to calculate the two statistics, Hotelling-T^2 and SPE. The control limit trajectories for the two statistics are estimated from the sufficient successful reference batches. For error sub-space, adopting the works of Box [28] and Jackson and Mudholkar [29], the SPE statistic can be approximated by a weighted Chi-squared distribution, $g\chi_h^2$, where the weight g and the freedom degree h can be obtained following the same approach of Nomikos and MacGregor [10].

The distribution parameters at time k are estimated from $\mathrm{SPE}_k = \{\mathrm{SPE}_{1,k}, \mathrm{SPE}_{2,k}, \ldots, \mathrm{SPE}_{I,k}\}$, $g^k = v^k/2m^k$ and $h^k = 2(m^k)^2/v^k$, where m^k is the average of the SPE_k and v^k is the corresponding variance. Thus, SPE control limit at time k can be approximated by

$$\mathrm{SPE}_\alpha^k = g^k \chi^2_{h^k,\alpha} = (v^k/2m^k)\chi^2_{2(m^k)^2/v^k,\alpha} \tag{8}$$

For principal component sub-space, Hotelling-T^2 is defined as follows and its confidence limits can be obtained to describe the average variability of process variables from the indicted F-distribution.

$$T^2_{i,k} = (t_{i,k} - \bar{t}_k)S_k^{-1}(t_{i,k} - \bar{t}_k)^T \sim \frac{R_k(I-1)}{I-R_k} F_{R_k, I-R_k,\alpha} \tag{9}$$

where R_k is the number of retained principal components. $t_{i,k}(1\times R)$ is the ith row and $\bar{t}_k(1\times R)$ is the mean of the score matrix $T_k(1\times R)$ at k time sampling. S_k is the covariance matrix of T_k. Here, it should be noted that compared with conventional A-unfolding, it satisfies $\bar{t}_k = 0$ in the proposed approach since the time-slice data is mean centered in the preprocessing procedure.

When a new batch run is available, the new sampling data, $x_{\mathrm{new}}(1\times J)$, should firstly be normalized before calling the corresponding STMPCA model to obtain the two statistics. Then the principal component score, estimated value, T^2 and SPE can be calculated by the following model:

$$\begin{aligned} t_{\mathrm{new}} &= x_{\mathrm{new}} P_k \\ \tilde{x}_{\mathrm{new}} &= t_{\mathrm{new}} P_k^T \\ T^2_{\mathrm{new}} &= (t_{\mathrm{new}} - \bar{t}_k)S_k^{-1}(t_{\mathrm{new}} - \bar{t}_k)^T \\ \mathrm{SPE}_{\mathrm{new}} &= (\tilde{x}_{\mathrm{new}} - x_{\mathrm{new}})(\tilde{x}_{\mathrm{new}} - x_{\mathrm{new}})^T \end{aligned} \tag{10}$$

Process monitoring is conducted by comparing the two statistics with the predetermined control limits. If the new batch follows the similar operation sequence and variable trajectories of the reference, it can be considered as a usual batch and should be archived in a normal database. Otherwise when the statistics go beyond the control limit responding to an abnormality, the contribution plot [30,31], a commonly used diagnosis tool, can be used to check the variables impacted by the occurred process failure.

3.3.5. Outline of STMPCA for batch processes

We has developed stage-based soft-transition multiple PCA modelling and on-line monitoring strategy to supervise the progressing of a batch process. Now the modelling and monitoring procedure can be summarized as follows:

3.3.5.1. Developing the online monitoring models.

1. Unfold three-way batch data matrix $X(I\times J\times K)$ into two-way matrices $X(I\times KJ)$, and then normalize each time-slice data matrices $X_k(I\times J)$.
2. Perform PCA on these time-slice matrices and get K weighted loading matrices $\breve{P}_k$, which represent the process correlation at each time interval.
3. Cluster the loading matrices $\breve{P}_k$ to get C clusters P_c^*, representing C different stages.
4. By the determination of class radius r_c and kernel radius r_c', the transition regions are properly separated from the neighboring sub-stages.
5. Establish STMPCA models respectively for each sub-stage based on A-unfolding, $P_c(J\times R_c)$, and each time interval of transition regions, $P_k = \sum \mu_{k,c} P_c^*$, using the concepts of membership grade $\mu_k = [0, 0, \ldots, \mu_{k,C-1}, \mu_{k,c}, \ldots, 0]$.
6. Calculate the confidence limits of T^2 and SPE statistics at each time k.

3.3.5.2. On-line monitoring.

1. For new sampling data at time k, $x_{\mathrm{new},k}(1\times J)$, normalize it using the same mean and standard deviation obtained from the modeling procedure.
2. According to process time, project the new data $x_{\mathrm{new},k}$ onto the corresponding STMPCA model P_k to calculate the score vector $t_{\mathrm{new},k} = x_{\mathrm{new},k}P_k$.
3. Calculate the T^2 and SPE statistics:

$$T^2_{\mathrm{new},k} = (t_{\mathrm{new},k} - \bar{t}_k)S_k^{-1}(t_{\mathrm{new},k} - \bar{t}_k)^T$$
$$\mathrm{SPE}_{\mathrm{new},k} = (x_{\mathrm{new},k} - x_{\mathrm{new},k})(x_{\mathrm{new},k} - x_{\mathrm{new},k})^T$$

4. Conduct process monitoring by judging whether T^2 or SPE statistics exceeds its confidence limit.
5. If an abnormality is detected, identify the variables relevant to the fault by contribution plot.

The whole algorithm of STMPCA is illustrated in Fig. 2.

4. Illustration and discussion

The proposed process monitoring and diagnosis approach is tested with two processes. One is the experimental application on a typical multistage process, three-tank system, and the other is the well-known benchmark simulation of fed-batch penicillin production. The previous focuses on analyzing the rationality and necessary of separating the transition regions from sub-stages in detail whereas the latter emphasizes particularly on verifying the superiority of the proposed method over some other PCA modeling strategies when online monitoring and fault diagnosing for a relatively complex batch process. In the present work, for MPCA, the missing future process values are filled with zero deviation approach [10].

4.1. Three-tank system

A three-tank system, as shown in Fig. 3, is used to simulate a multistage batch process and demonstrate the proposed approach. In the typical multivariable experiment equipment, five process variables including two float inputs Fl, F2 and three liquid levels L1, L2, L3, are measured every second. The two levels are brought from their initial

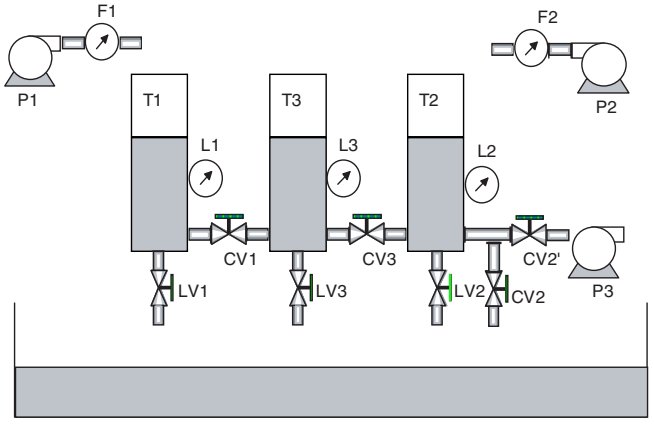

Fig. 3. Structure of the three-tank plant.

conditions to the set points of L1 = 300 mm and L2 = 200 mm. The level of L3 is left to float to reflect the interaction between Tank 1 and 2. The process finishes after the three levels stabilize over a period of time. The raising water levels in the tanks result in a time-varying process dynamics and closed-loop control implemented for the levels of the two tanks cases the strong coupling relationships between variables. One hundred and twenty points of historical data collected in each batch and 24 normal experiments carried out under the same conditions are used to yield the modeling data matrix $X(24 \times 5 \times 120)$.

The weighted loading matrices calculated from the time-slice matrices are fed to the clustering algorithm and we can get a typical four-stage process combined with the whole measurement trajectory shown in Fig. 4: Stage I, in which manipulated variables F1 and F2 are both at saturation, and three levels are increasing steadily; Stage II, in which F1 is at saturation while F2 starts decreasing, and three levels keeps increasing; Stage III, in which both manipulated variables are decreasing whereas all levels continue increasing. Meanwhile, the decreasing or increasing trend gradually becomes slow until saturation when approaching the boundary regions with stage IV; Stage IV, in which all five variables tend to level off after mild fluctuation and both LI and L2 reach their set points. It is clear that agreeing well with the actual correlation variation, the clustering algorithm generates reasonable stage division results, which can indeed enhance the process understanding.

Since loading matrices represent the process correlations, we can use the distance calculated from Eq. (3) to analyze quantitatively the changing trend of process nature along time, which is displayed in Fig. 5. Corresponding to the development of process nature, it is clear that the distance, $d_{k,c}$ shows the gradually changing trend: smaller approaching to the current stage, stable duration, and larger when farther away. When the process has successfully achieved a new steady state, in which the distance $d_{k,c}$ has been stable, one can deem that the transition completes and the process is operating in a new state. After choosing class radius and kernel radius using Eqs. (4) and (5) the duration of transition region can be determined properly midst neighboring stages. The distance dissimilarity proves obviously that the actual existence of transition regions and their separation from sub-stages are significative. So the proposed soft-partition algorithm focusing on the transition regions is attractive and provides a feasible attempt to solve such problems. Then to more clearly reveal the gradual and various changeovers of process characteristics from one state to another, the concept of membership grade is used to quantitatively express the sub-ordinative relationships. As shown in Fig. 6, it gives us a visual description of transiting trend. Although the limited sampling data cannot take all the transiting details into account perfectly, the above analyses and investigations still provide comprehensive information for time-varying transiting nature.

For each monitoring model, only two or three principal components are needed to explain over 90% variations.

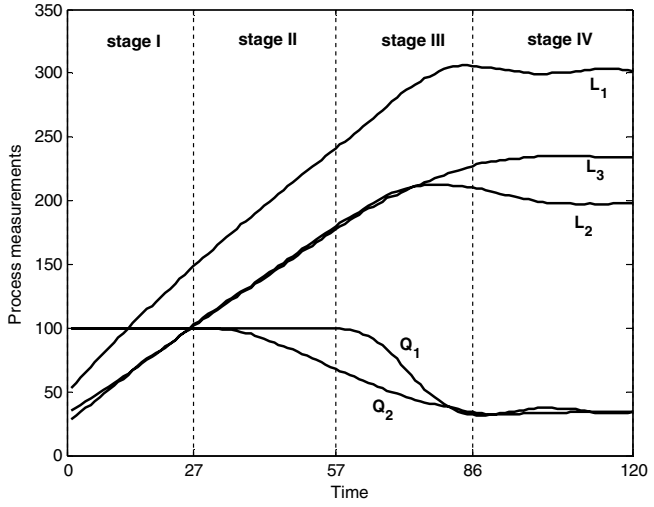

Fig. 4. Stage division result with process trajectory for three-tank system.

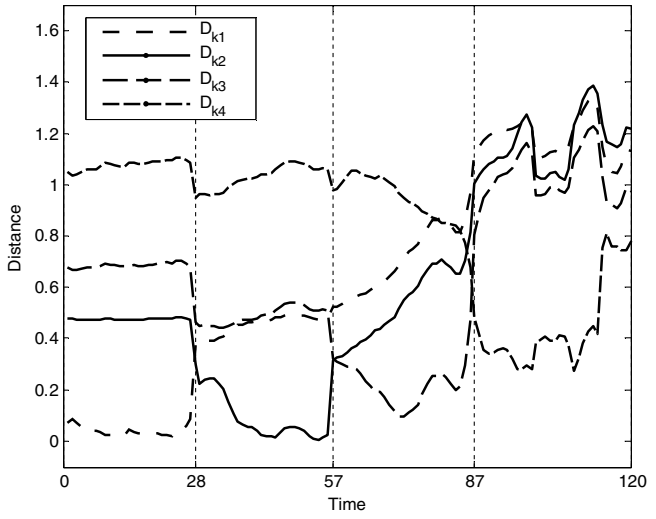

Fig. 5. Changing trend of process nature evaluated with Euclidean distance (D_{kc}, the distance between loading matrix at time k and center of cth sub-stage).

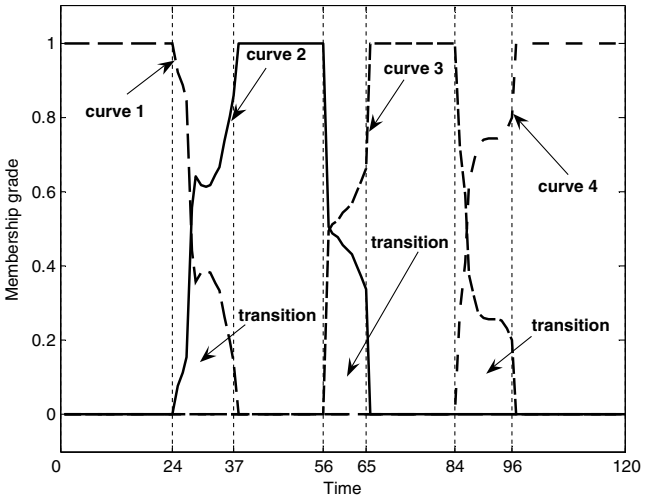

Fig. 6. Sketch map of membership grades (Curve 1, membership grade values of each time-slice loading to stage I, and curves 2, 3, 4 are the corresponding values to stages II, III, IV, respectively).

After modeling, the proposed approach is put into on-line monitoring tests by judging whether the Hotelling-T^2 and SPE values of the coming measurements in a running batch are below the control limits. Fig. 7a shows the monitoring of a normal batch using the proposed method, where time trajectories of the two statistics stay well below the confidence limits, indicating that the whole batch is free of any process upset. In contrast, the SPE monitoring values of MPCA shown in Fig. 7b display wrong alarms especially at the beginning of the process. Hence, the reliability of MPCA monitoring model has been compromised compared with the multistage modelling.

For the second case, a fault is introduced at 50th sampling time to simulate a leakage of both Tanks 2 and 3 by opening valve LV2 and LV3 synchronously. From the monitoring results shown in Fig. 8a, the abnormality can be clearly detected as SPE values go beyond the control limit almost immediately and T^2 with a delay of only several samplings after the occurrence. In contrast, monitoring charts using sub-PCA and MPCA methods in Figs. 8b and c show obvious detection delay and false alarms

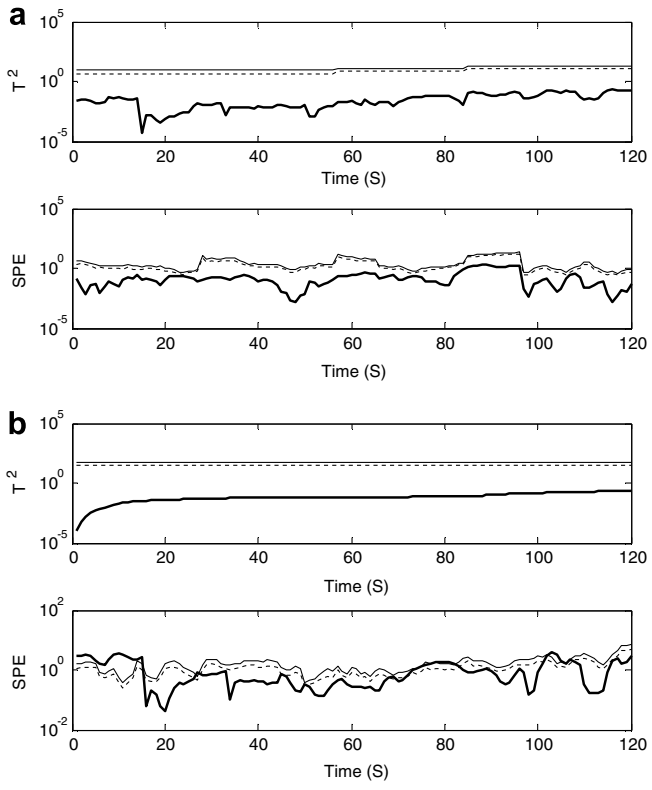

Fig. 7. Monitoring plots of a normal batch using (a) STMPCA and (b) MPCA (solid line, 99% control limit; dashed line, 95% control limit; solid line, on-line T^2 or SPE statistic).

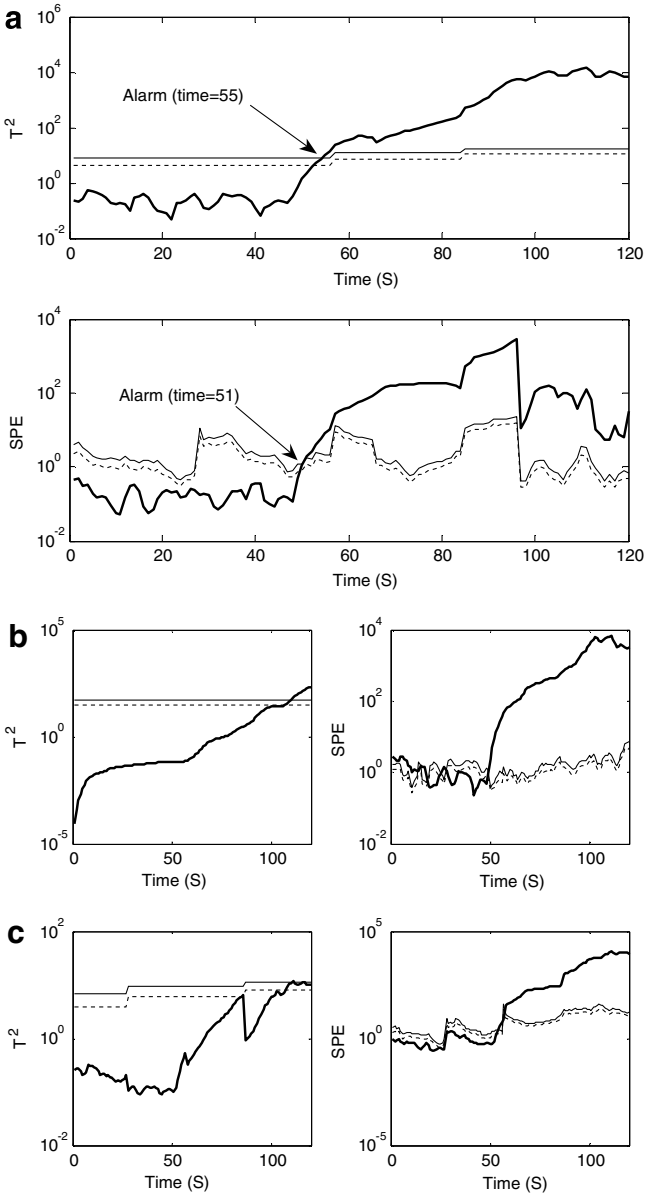

Fig. 8. Monitoring plots of (a) STMPCA (b) MPCA and (c) sub-PCA for an abnormal batch (solid line, 99% control limit; dashed line, 95% control limit; solid line, on-line T^2 or SPE statistic).

at the beginning of the process respectively. Moreover, combined with the use of time-varying score covariance structures, the T^2 monitoring chart also shows the superiority of STMPCA. T^2 statistics of the other two methods indicate gradual increase and do not exceed the control limits until approaching the end of batch run although an abrupt increase appears upon the occurrence of upset. This delay may lead to rejected batches. Comparatively, it can be seen that STMPCA outperforms the other two methods. The experiment demonstrates the swiftness and effectiveness of the proposed method for detecting faults, which will benefit the accuracy of fault diagnosis and the final quality control.

4.2. Fed-batch penicillin fermentation

In this section, the proposed method is applied to the monitoring of a well-known benchmark process, fed-batch penicillin fermentation process [32,33]. A flow diagram of the penicillin fermentation process is given in Fig. 9. The production of secondary metabolites such as antibiotics has been the subject of many studies because of its academic and industrial importance. Here, we focus on the process to produce penicillin, which has non-linear dynamics and multiphase characteristics. In typical operating procedure for the modeled fed-batch fermentation, most of the necessary cell mass is obtained during the initial preculture phase. When most of the initially added substrate has been consumed by the microorganisms, the substrate feed begins. The penicillin starts to be generated at the exponential growth phase and continues to be produced until the stationary phase. A low substrate concentration in the fermentor is necessary for achieving a high product formation rate due to the catabolite repressor effect. Consequently, glucose is fed continuously during fermentation instead of being added one-off at the beginning.

In the present simulation experiment, a total of 60 reference batches are generated using a simulator (PenSim v1.0 simulator) developed by the monitoring and control group

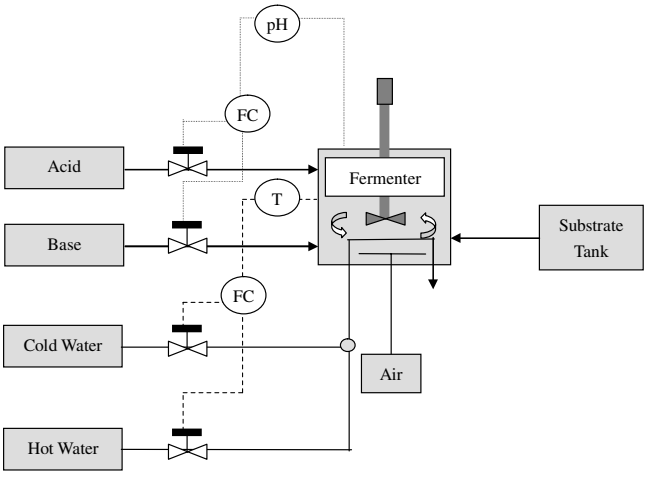

Fig. 9. A flow diagram of the penicillin fermentation process.

Table 2
Variables used in the monitoring of the benchmark model

Number	Variables
1	Aeration rate (l/h)
2	Agitator power (W)
3	Substrate feed rate (l/h)
4	Substrate feed temperature (K)
5	Dissolved oxygen concentration (g/l)
6	Culture volume (l)
7	Carbon dioxide concentration (g/l)
8	pH
9	Generated heat (kcal)

of the Illinois Institute of Technology. These simulations are run under closed-loop control of pH and temperature, while glucose addition is performed open-loop. Small variations are automatically added to mimic the real normal operating conditions under the default initial setting conditions. The variables monitored in this work are shown in Table 2. The duration of each batch is 400 h, consisting of a preculture phase of about 45 h and a fed-batch phase of about 355 h. Using the proposed partition method, the real fed-batch phase is sub-divided into three main stages, so the whole process is divided into four primary stages as well as corresponding transition regions. The more elaborate stage partition results emphasize the changes of process correlations rather than the physical operation, which will benefit making more detailed analyses of underlying process behaviours and establishing more appropriate monitoring models.

The models constructed using the proposed method, sub-PCA and MPCA are then tested against monitoring of fault batches. Fault 1 is implemented by introducing a 15% step decrease into the substrate feed rate at 100 h and retaining until 220 h. Fig. 10 shows the on-line monitoring charts using three methods respectively for the fault batch. In the case of STMPCA, the SPE statistic values increase sharply beyond confidence limit and the T^2 values also exhibit an abrupt rise right at time 100 h when the fault is introduced, which will enable the operator to respond rapidly as soon as the abnormality occurs. Although the fault ends at time 220 h, the process correlation nature has been deteriorated and cannot return to the normal trajectory. So the SPE values yield the decreasing trend around 220 h but still outside the normal boundary until the end of batch. Contrastively, sub-PCA delivers some alarm mistakes in the initial period while MPCA presents consecutive process failure before the real onset of disorder.

For fault 2, a linear decrease of slope 0.2% is imposed on the agitator power from time 100 h until the end. Fig. 11 shows the monitoring results of T^2 and SPE, respectively. In SPE monitoring charts, the proposed method reveals the abnormality almost immediately since SPE values go abruptly outside their desired level at time 100 h. However, SPE values by sub-PCA show some false alarms ahead of the occurrence of fault and MPCA exhibits fault

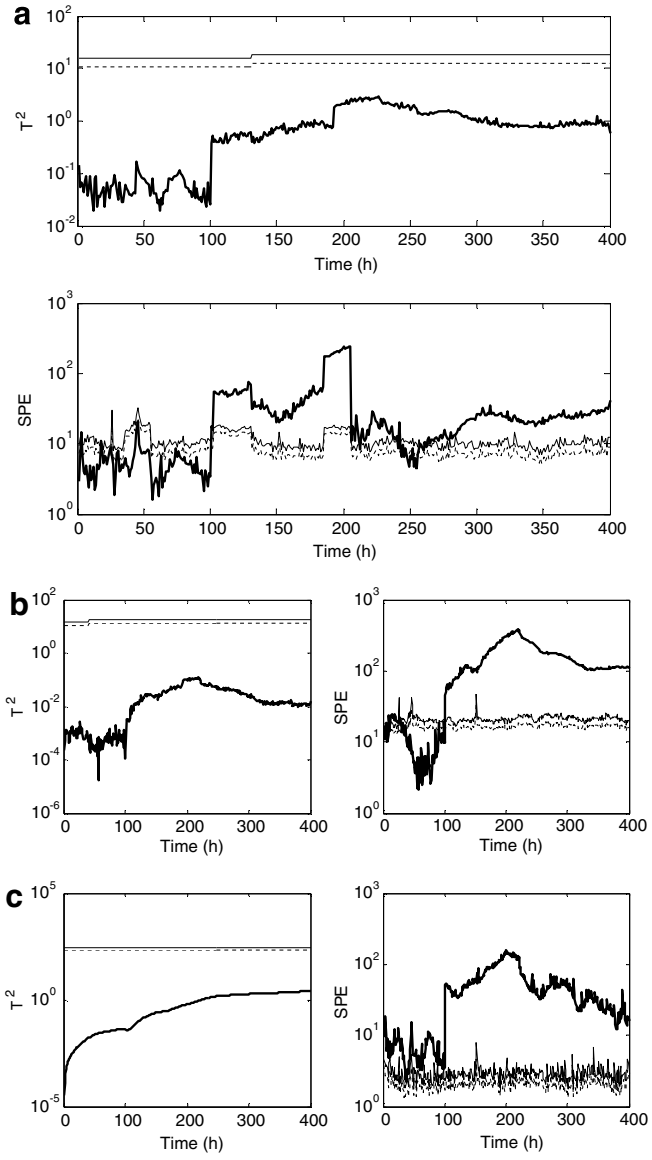

Fig. 10. Monitoring charts of fault 1 using (a) STMPCA, (b) sub-PCA and (c) MPCA (solid line, 99% control limit; dashed line, 95% control limit; solid line, on-line statistic).

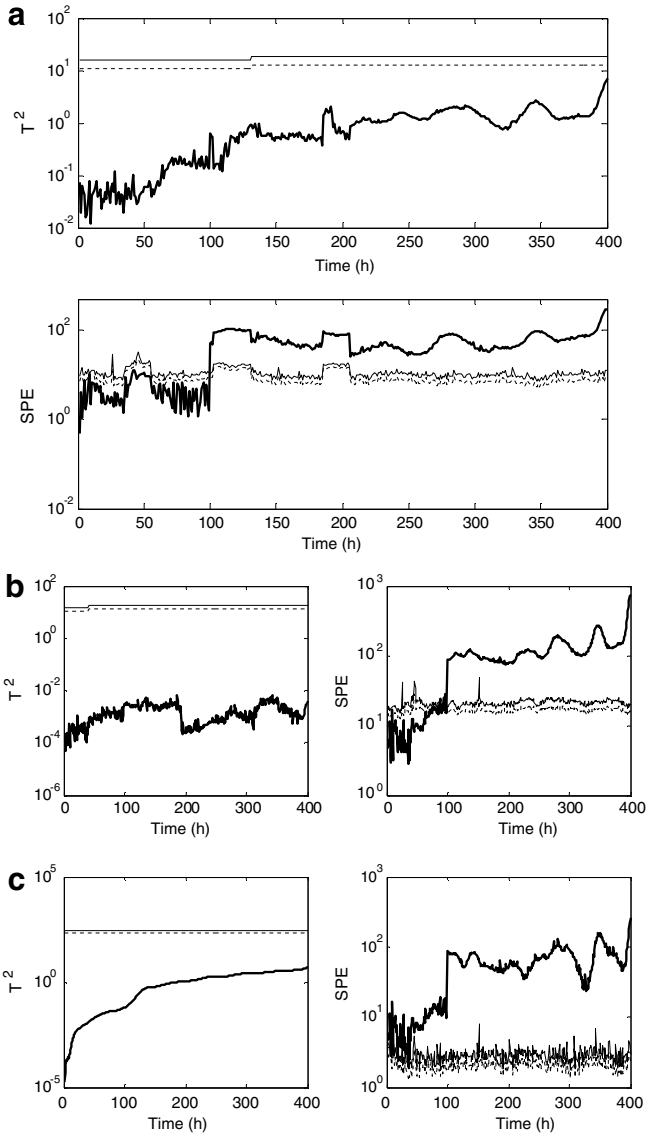

Fig. 11. Monitoring charts of fault 2 using (a) STMPCA, (b) sub-PCA and (c) MPCA (solid line, 99% control limit; dashed line, 95% control limit; solid line, on-line SPE statistic).

throughout the process duration. In contrast to the T^2 values from sub-PCA, which stay within the normal coverage throughout, those of STMPCA display a significant deviation at 100 h and begin to increase gradually since then. This is attributed to the fact that the adoption of time-varying score covariance structures in the proposed method makes the T^2 monitoring chart able to reflect the dynamic relations and reveal those even smaller changes in the systematic part of the process variation.

Once the abnormal condition is detected by the monitoring charts, the contribution plot is used to analyze the fault cause, which can indeed enhance the process understanding and improve the ability of fault detection and diagnosis. For fault 1, the contribution plots shown in Fig. 12a display that the primary fault cause variable is variable 3 (substrate feed rate), which is well agreed with the real state. For fault 2, Fig. 12b indicates that variable 2 (agitator power) makes the greatest contributions to the fault. However, since the processes variables tend to complicatedly related to each other in practical batch industry, the deviation of one variable will influence the others from different aspects and to various extents. With the development of time, if fault detection is delayed, the fault diagnosis will be more complicated and deceptive when various variables indicate significant deviations from their expected values. Hence, fast fault detection is important for correct fault diagnosis and will help the operator to check the fault cause in time and reduce the economic loss. The monitoring and diagnosis results clearly show that after the onset of the fault, the disturbance changes the residual parts of the process variation and its effect is propagated causing other variables to depart from their expected trajectories.

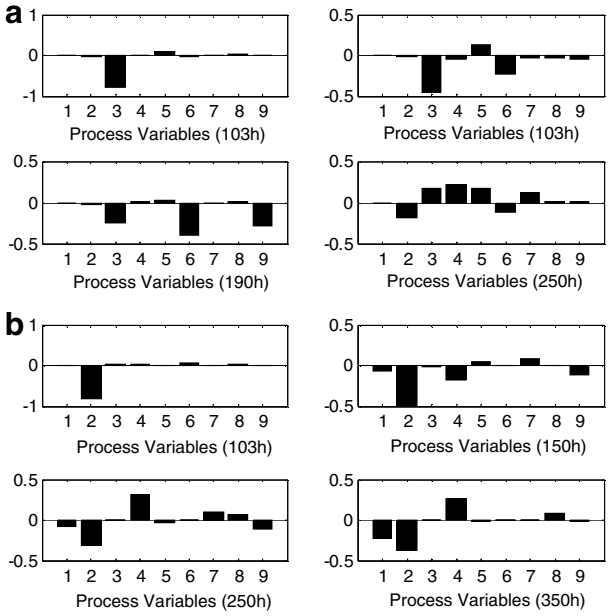

Fig. 12. Contribution plots to SPE statistic in case of (a) fault 1 and (b) fault 2.

From the above simulation results, the proposed method shows superiority over sub-PCA because the changes of transitional process correlations exert influence upon sub-stage monitoring models. Also it outperforms MPCA using specific-to-stage models to unveil the local correlation structures, where the latter has the major problem that during the early period most of the process measurements are unavailable. The estimated values of missing future data are obviously different from the real process observations and thus yield unreliable monitoring results. The illustration results demonstrate effectively that STMPCA method is more reliable and sensitive for on-line monitoring and significantly decreases the frequency of false alarms and missing alarms.

5. Conclusions

A statistical process monitoring method termed STMPCA has been proposed based on soft-partition algorithm for multistage batch processes. Focusing on stage-to-stage transition behaviors, the process correlations are analyzed in detail to quantitatively evaluate the transition duration and overcome the disadvantage of hard-partition strategy. Accordingly the whole process can be properly divided into different sub-stages and the relevant transition regions. They properly reflect the gradual developing trend of underlying behaviors from one state to another along time direction and can enhance the understanding of real process operation. The proposed approach has been demonstrated to be effective and reasonable by two kind of typical batch processes, indicating that it is competent to detect process faults with greater reliability and reduce false alarms and missing alarms used as an on-line monitoring tool for multistage batches.

In conclusion, the proposed STMPCA algorithm has provided a significative attempt to the transition modeling and monitoring. A process monitoring system based on analyses of transition dynamic characteristics will be promising and should be devoted more efforts in future.

Acknowledgments

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Appendix. The modified *k*-means clustering algorithm

Inputs: The patterns to be partitioned, $\{\breve{P}_1, \breve{P}_2, \ldots, \breve{P}_k\}$, and the threshold θ for cluster elimination.

Outputs: The number of clusters C, the cluster centers $\{W_1, W_2, \ldots, W_c\}$, 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 W_c^0 $(c=1,2,\ldots,C^0)$ from the K patterns along the time series. Practically, the initial cluster centers can be assumed to be uniformly distributed in the pattern set.
2. Merge pairs of clusters whose inter-centre distance, $\mathrm{dist}(W_{c1}^{i-1}, W_{c2}^{i-1})$, is below the predetermined threshold θ.
3. Calculate the distances from each pattern $\breve{P}_k$ to all of the centers, $\mathrm{dist}(\breve{P}_k, W_c^{i-1})$, assign $\breve{P}_k$ to the nearest center $w_{c^*}^{i-1}$, and denote its membership as $m(k)=c^*$.
4. Eliminate the clusters that catch few patterns after a set number of iterations $i > I_{\_\mathrm{num}}$ to avoid singular clusters.
5. Update the number of cluster centers to be C^i, recompute the new cluster centers 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.

Remark: The detailed programs for implementing STMPCA algorithm and the experimental data are not included in the present work due to lack of space. A more complete document will be provided upon request addressed to the first author.

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