Statistical batch process monitoring
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CHAPTER 6  ♦ STATISTICS AND MECHANISTICS*

* This chapter is based on the following publication(s):


H.J. Ramaker, E.N.M. van Sprang, J.A. Westerhuis and A.K. Smilde, The effect of the size of the training set and number of principal components on the false alarm rate in statistical process monitoring, Chemometrics and intelligent laboratory systems, accepted.

6.1 General introduction *

One of the pillars of statistical batch process monitoring is multivariate statistics. It is therefore interesting to investigate and understand these statistics. The most important statistical properties evaluated in this chapter, are the false alarm rate and action signal time. These indices are outlined in Chapter 4.2. The size of the false alarm rate is directly related to the definition of the null hypothesis. In this chapter, a new definition of the null hypothesis is presented.

Also, the effect of the number of samples in the dataset as well as the number of components for the PCA model on the false alarm rate is investigated.

Besides the false alarm rate and action signal time, other statistical properties are of interest. These properties, e.g. normality assumption of the scores/residuals, are discussed separately for the D and SPE-chart. Especially the D-chart is suspicious because of its poor performance in detecting faults compared to the SPE-chart. Several suggestions are given to improve the statistical properties of the control charts. The most important improvements are the Bonferroni adjustment and leave-one-out procedure.

The detection characteristics of the control charts can also be related to mechanistic properties of the control charts (e.g. how process faults manifest in the control charts). For this reason, these mechanistic properties are thoroughly discussed. It is shown how process faults with certain characteristics are distributed in both the control charts. These characteristics are coupled to correlation present in batch data. In order to understand these correlations better, a description of what types of correlation can be expected in batch process data is presented. Furthermore, it is shown how well different models are capable of modelling these correlations.

This chapter consists of four sections. The first section describes a performance assessment of control charts for statistical batch process monitoring. The statistical properties of the SPE and D-chart are discussed and tested for six different datasets. Improvements for these charts are presented and tested on the same datasets. In the second section, the effect of the number of components and size of the dataset is studied with respect to the false alarm rate. In the third section of

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this chapter, the mechanics of the $SPE$ and $D$-chart are thoroughly discussed. It is shown how process faults manifest themselves in these control charts for three different models: global, local and time evolving models. The fourth section of this chapter explains what different types of correlations are present in batch process data. It is shown how the different models are able to capture these correlations.
6.2 Performance assessment and improvement of control charts for statistical batch process monitoring*

6.2.1 SUMMARY
This section describes the concepts of statistical batch process monitoring and the associated problems. It starts with an introduction in process monitoring in general, which is then extended to batch process monitoring. The mechanics of the control charts for process monitoring are studied. Furthermore, the performance of the control charts is discussed by means of two performance indices: the overall type I error and the action signal time. The problems of the existing approach are highlighted and illustrated by e.g. the mechanistic properties of the control charts. Improvements are suggested and checked with the performance indices. To evaluate the effect of the proposed improvements as well as to assess the performance of the existing approach, six different data sets have been used: four industrial data sets, a simulated batch reaction and a laboratory spectral data set.

6.2.2 INTRODUCTION
The batch-wise production of chemicals, bio-chemicals, pharmaceutics and foods is common in industry. Monitoring such processes is necessary for several reasons: safety, improving product quality and a better understanding of the process. Besides knowledge based or state estimation approaches, multivariate statistical approaches can be used for monitoring batch processes.

The fundamentals of statistical process control were described by W.A. Shewhart [5]. H. Hotelling [8] was one of the first who introduced a multivariate approach to statistical process control to monitor a process of a multivariate nature. J.F. Jackson [9] then applied principal component analysis (PCA) to reduce the dimensionality of the multivariate data. B.M. Wise & N.L. Ricker [112] and J.V. Kresta

et al. [10] proposed a multivariate control technique that uses PCA or projection to latent structures (PLS). Finally, P. Nomikos & J.F. MacGregor [6] extended the concept of multivariate statistical process control using PCA to batch processes. This is called statistical batch process monitoring. Their approach to batch process monitoring is a purely data driven approach based on linear multivariate analysis methods. The relevant variation of a process is captured in a dimension-reduced model. This is advantageous in terms of interpretability and comprehensibility. Two test statistics are derived from this model and both are monitored using control charts.

In the paper by E.N.M. Van Sprang et al. [41] several problems were encountered with respect to batch process monitoring. In that paper the results of a comparison study between different approaches for on-line batch process monitoring is described. Two performance indices were used to qualify the different approaches. Also, the validity of assuming normality for the underlying distributions of the test statistics was checked. It was concluded that one of the control charts works reasonably well despite violating the normality assumption. The other control-chart has a poor performance with respect to the indices while the normality assumptions are reasonable.

The aim of the current section is to give an overview of the problems encountered with on-line batch process monitoring and propose possible solutions. It will be shown that the concepts from continuous process monitoring are not straightforwardly applicable to batch process monitoring. The mechanistic and statistical properties of the control charts for batch process monitoring are studied. Ideas are given on how to improve the performance of the control charts.

This section is organised in the following way. Section 6.2.3 gives a brief summary of the concepts of multivariate statistical process control considering the different phases in monitoring. In Section 6.2.4, the control charts for multivariate statistical process control are discussed for continuous processes with and without dimension reduction. An understanding of the type of process faults detected by the control charts is given. Also, the extension of multivariate statistical process control to post-batch analysis is discussed. This is followed by explaining on-line batch process monitoring. In Section 6.2.7, a description of the datasets used as examples is given. In Section 6.2.8, problems of statistical batch process monitoring are discussed and
possible solutions are applied to the examples. Finally, conclusions will be given in the last Section 0.

6.2.3 Multivariate Statistical Process Control: The Concepts

W.H. Woodall [48] stated that it is very important to improve communication between practitioners and researchers concerning statistical process control methods. One way to establish an improved communication is to assign phases to the concepts of multivariate statistical process control. In this section, multivariate statistical process control is carried out in three phases: The initial, training and application phase. Here, the training and application phase refer to respectively phase 1 and phase 2 from standard SPC terminology.

The initial phase consists of probably the most time consuming part: that is the collection and cleaning up of the process data. The goal of the training phase is to obtain a set of in-control observations, build a suitable model for these observations and derive control limits from this model. For this reason, the data must be checked for outliers that do not represent an in-control process. This step in the training phase resembles phase 1 from SPC terminology. The model can be further extended by incorporating existing knowledge of the process under consideration (A.K. Smilde et al. [113]). This can be helpful in terms of e.g. fault diagnosis or increased process understanding.

The application phase starts with monitoring new independent observations to detect abnormal deviations from the in-control process. This step refers to phase 2 from SPC terminology. Besides detection of process upsets, fault diagnosis has to take place in this phase. The detection and diagnosis of the fault may lead to an improvement of the process.

6.2.4 Multivariate Statistical Process Control: The Control Charts

The concepts of multivariate process control for batch processes are similar to multivariate process control for continuous processes. However, in transferring the methodology from continuous to batch processes, there are some issues that are not straightforward.
In the following an overview is given how the concepts of multivariate statistical process control for continuous process are extended to on-line batch process monitoring. This is done in a stepwise manner where the related problems for each step are highlighted and discussed.

Process measurements of \( J \) process variables are made at \( I \) time points on a continuous process and collected in the data matrix \( \mathbf{X} (I \times J) \). Preprocessing the process data \( \mathbf{X} \) is a first step. Generally, mean centering of the variables is a good idea and also scaling if the measured units of the process variables are different. In this section it is assumed that the data is properly pre-processed. Furthermore, \( \mathbf{X} \) is believed to represent in-control observations.

6.2.4.1 Continuous processes without dimension reduction

In 1947 Hotelling introduced the \( T^2 \) statistic as a method to apply statistical quality control to correlated data with a multivariate nature. This summary statistic enables easy control charting and allows for detecting faults of highly correlated measurements. In the case of continuous processes for chemical applications, at every time interval \( J \) process variables are measured and repeated \( I \) times. These measurements can be arranged in a matrix \( \mathbf{X} (I \times J) \). For a new measurement vector \( \mathbf{x}_{\text{new}} (J \times 1) \) the test statistic \( T^2 \) is calculated according to:

\[
T^2 = (\mathbf{x}_{\text{new}} - \bar{x})^T S^{-1} (\mathbf{x}_{\text{new}} - \bar{x})
\]

where \( S \) is the estimated covariance matrix of \( \mathbf{X} \) and \( \bar{x} \) is the target value or grand mean. Therefore, if the goal is to obtain an in-control set of observations as described in the training phase, the UCL is given by (N.D. Tracy et al. [14]):

\[
\text{UCL} = \frac{(I-1)^2}{I} B(\alpha; J/2; (I-J-1)/2)
\]

The UCL\text{\_new} to use for detecting deviations from the in-control process for new independent observations are given by:

\[
\text{UCL}_{\text{\_new}} = \frac{J(I^2-1)}{I(I-J)} F(\alpha; J; I-J)
\]
An important note here is that all the process variables are used and no dimension reduction takes place. With a large number of process variables this approach breaks down, e.g., $S$ might become (nearly) singular.

### 6.2.4.2 Continuous processes with dimension reduction

In a chemical process, the dimensions of $X$ may become large in the second mode $J$. It is therefore advantageous to reduce the dimensionality of the data. Principal component analysis (PCA) is a multivariate statistical method well suited for this purpose. J.E. Jackson & G.S. Mudholkar [24] was one of the first to apply a principal component analysis for this type of problem. Reducing the dimensionality of the data results in two test-statistics. The derivation of these test statistics will follow. In this section, these test statistics are referred to as $D$-statistic and Squared Prediction Error (SPE) statistic.

A PCA decomposes the matrix $X$ into the sum of $R$ outer products of scores $t$ and loadings $p$ plus a residual part $E$:

$$X = \sum_{r=1}^{R} t_r p_r^T + E = TP + E \quad (101)$$

where $T$ ($I\times R$) is the score matrix, $P$ ($J\times R$) contains the loadings and $E$ ($I\times J$) is the residual matrix. A geometrical representation of equation 101 is given in Figure 92.
Geometrical interpretation of PCA for a two component model in a three dimensional space. The $i$-th row of $X$ represents a sample and can be drawn as a vector $(x_i)$ in the space. The subspace where this sample is orthogonally projected on is spanned by the columns of $P$. The distance $(e_i)$ from $x_i$ to the model plane is given by the $i$-th row of $E$. The co-ordinates, or scores, of the projection in the plane are given by the $i$-th row of $T$.

Once the model plane, spanned by the columns of $P$, is defined, the scores and residuals for a new scaled measurement $x_{\text{new}}$ ($j\times 1$) are found by projection on the model plane. This is a regression problem:

$$x_{\text{new}} = Pt_{\text{new}} + e_{\text{new}} \quad (\text{102})$$

where $t_{\text{new}}$ is estimated using least squares. Two test statistics are defined. The $D$-statistic describes the Mahalanobis distance from the projection on the model plane to the centre of the model plane, and describes the systematic variation in the data. The centre of the model plane corresponds to average process behaviour. Therefore, the closer the projection of a sample is to this centre, the better. The $SPE$-statistic (Squared Prediction Error) represents the Euclidian distance between the
measurement vector and its projection on the model plane. This distance describes the variation in the data that is consistent with the model.

For the ease of illustration, the number of PCA components $R$ is chosen to be two. The test statistics are calculated according to:

\[
\begin{align*}
t_{m} &= \begin{bmatrix} t_1 \\ t_2 \end{bmatrix} = (P'P)^{-1}P'x_{mew} = P'x_{mew} \\
\Rightarrow \quad D &= t_{m}'S^{-1}t_{m} \\
e_{m} &= x_{mew} - \begin{bmatrix} t_1 \\ t_2 \end{bmatrix}P' \\
\Rightarrow \quad SPE &= e_{m}'e_{mew}
\end{align*}
\]

Here, $S$ is the variance-covariance matrix of the scores $T$ and is usually diagonal because the scores $T$ are orthogonal. Subsequently, the test statistics are plotted in the control charts and $x_{mew}$ is assigned to be statistically out of control if one of the control charts signals. The UCL for the $D$-statistic is the same as given in equation 100 except that $J$ is replaced by $R$. The UCL for the $SPE$-statistic resembles the critical value $SPE_{\alpha}$. The derivation of this critical value can be found in J.E. Jackson & G.S. Mudholkar [24].

6.2.5 MECHANISTICS OF THE $D$ AND $SPE$-CHART

Detecting a fault at the $D$ or $SPE$-chart is important, but of equal importance is the diagnosis of the fault found. There is a general believe that new events in a process not present in the NOC data are detected by the $SPE$-chart, e.g. sensor failure. The $D$-chart diagnosis abnormal variation in the process that still obeys the correlation structure of the process variables as reflected in the matrix $P$. Hence, this is a distinction between faults detected in the $SPE$ and $D$-chart. The following citations were found in the literature:

R. Dunia & S.J. Qin [114]: "A significant increase in $SPE$, which measures the distance from the principal component subspace, indicates a breakdown of the normal correlation". A significant increase in $T^2$ can be due to a fault or normal change in the process throughput that conserves the correlation structure ".

S. Albert & R.D. Kinley [43]: "... mainly in the direction of the $SPE$ statistic, which indicates that warnings were generated because of new events rather than changes in correlation structure ".
P. Nomikos & J.F. MacGregor [19]: "If a new batch is still operating in the same way as the batches in the reference database, but still has a larger than normal variation in its measurements, this will show up clearly as large deviations of the \( t \) scores from the origin of the reduces space. In the case in which a totally new fault that is not represented in the database occurs, the principal components will then not be able to describe correctly the variation. Thus the new observations will move off the MPCA plane, resulting in large values of the \( SPE \)."

Similar statements can be found in J.V. Kresta et al. [10], K.A. Kosanovich et al. [7], W. Lin et al. [115], B. Lennox et al. [37].

To investigate the diagnostic properties of the \( SPE \) and \( D \)-chart, a detailed analysis is made of the mechanics of the \( SPE \) or \( D \). Therefore, three types of faults are distinguished and discussed: i) a fault detected only in the \( D \)-chart ii) a fault detected only in the \( SPE \)-chart iii) a fault detected in both charts. It will be shown in the following how these faults can be envisioned and that it is not easy to establish a simple relationship between the type of fault and the detection signal.

i) Faults detected only in the \( D \)-chart: The vector \( x_1 \) (\( J \times 1 \)) in Figure 93 represents a sample of \( J \) process variables at time \( i \) under normal operating conditions, hence the control charts do not signal an error for \( x_1 \). The ellipse represents the UCL for the \( D \)-statistic.
At the next time point \(i+1\), the vector \(x_{i+1}\) represents a measurement containing a process fault at time \(i+1\). This sample has such properties that it only manifests itself in the D-chart. This implies that the length of the residual vector does not break the limit given by \(SPE_{\alpha}\), or \(e_{i+1} = e_i\). Only the coordinates after projection of \(x_{i+1}\), given by \([t_1, t_2]_{i+1}\), are significantly different for \(x_{i+1}\) as compared to the coordinates for \(x_i\). Thus, this particular fault only changes within the plane. Clearly, the projection of \(x_{i+1}\) results in a distance further away from the centre of the plane and now falls outside the confidence limits. It is insightful to 'translate' this information from the latent variable space to the raw process variables. In other words, if the scores change and the residuals stay constant due to a process disturbance, how can such an event happen in the original process variables? This is only possible if the new measurement moves in the process variable space parallel to the model plane. In Figure 94, the black bars (\(x_i\)) represent the scaled values for 15 process variables from a real process.
Let the white bars represent the process event $x_{i+1}$. This process event has been constructed. Considering $x_i$ and $x_{i+1}$, the correlation between the process variables is consistent and not broken, only more intense than under normal operating conditions. This is considered to be a multivariate fault where the correlation pattern is not disturbed and to be detected in the $D$-chart. Only if the process variables behave exactly as depicted in Figure 94, this will result exclusively in a change within the plane spanned by the loadings.

**ii) Faults detected only in the SPE-chart:** The vector $x_i$ ($j\times1$) in Figure 95 represents a sample of $j$ process variables at time $i$ under normal operating conditions, hence the control charts do not signal an error for $x_i$. 

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Again, the vector $x_{i+1}$ represents a measurement containing a process upset at time $i+1$. This sample has such properties that it only manifests itself in the SPE-chart. This implies that the scores have a limited space to vary. In this example $t_{i+1} = t_i$. Only the length of the residuals given by $\|e_{i+1}\|^2$ are significantly different for $x_{i+1}$ as compared to the residuals of $x_i$. Thus, this particular upset moves perpendicular from the plane. The translation of this fault in terms of raw process variables is given in Figure 96. The black ($x_i$) represents the scaled values for 15 process variables and the white bars represent the process upset $x_{i+1}$.
The fault is constructed in such a way that process variable 6 is erroneous. This becomes obvious from Figure 96. To detect this specific fault only in the $SPE$-chart, all the other process variables must change to some degree to be perpendicular to the model (i.e. in the direction $(I - PP')$). That is, some process variables are hardly affected, some are breaking the correlation structure and some will follow the correlation structure. To move perpendicular away from the plane in the same direction as $e_\gamma$, all process variables must contribute. Only if the process variables behave as depicted in Figure 96, the fault will be detected exclusively in the $SPE$-chart. The reason for all these changes in the process variables necessary for a fault only to show up in the $SPE$ is that the model plane is tilted meaning that all the process variables contribute to the model. In other words, none of the process variables are orthogonal to the plane.

It may become obvious that for a fault to show up exclusively in the $SPE$ or $D$-chart the process variables have to change in a very unique and specific way. That
is, to find a fault exclusively to be detected in the SPE or D-chart can be considered as extreme.

iii) Faults detected in both charts

From practice it is seen that most process upsets are detected in both the control charts. The reasoning for this observation will be illustrated in the following using an example.

For a fault to be detected exclusively in the D-chart all the process variables must behave as depicted in Figure 94. In reality there are dynamics in the process. This means that not all the process variables will immediately respond to such a process upset. As an example, suppose the cooling of a reactor is insufficient. Obviously, the area near the cooling jacket such as the wall of the reactor is heated faster than the area in the middle of the reactor. If the sensors are located in the middle, it will take some delay for the sensors to respond to the process upset. As a result, first the correlation pattern will be broken by the sensor located at the wall before the others get affected. A breakage of the correlation pattern is more likely to be detected in the SPE-chart.

For a fault to be detected exclusively in the SPE-chart all the process variables must behave as depicted in Figure 96. Hence, simple faults like a sensor failure will also always affect the D-chart, because not all the process variables change according to the pattern of Figure 96. This problem is known in the factor analysis literature as embedded error (E.R. Malinowski [125]).

In mathematical terms embedded error can be explained as follows. Let the vector \( \mathbf{x} \) represents the raw data defined as:

\[
\mathbf{x} = \mathbf{x}^* + \mathbf{e}
\]  \hspace{1cm} (105)

where \( \mathbf{x}^* \) is the pure data and \( \mathbf{e} \) is the true experimental error. The pure data \( \mathbf{x}^* \) can be written exactly according to the model, that is:

\[
\mathbf{x}^* = \mathbf{P}\mathbf{t}^*
\]  \hspace{1cm} (106)

Here, it is assumed that \( \mathbf{P} \) reflects the true underlying correlation in the process. The scores \( \mathbf{t} \) for \( \mathbf{x} \) are found by projecting \( \mathbf{x} \) according to:
\[ t = P'x = P'(x^* + e) = Px^* + Pe \neq t^* \]  (note that \( t^* = P'x^* = P'Pt^* = t^* \))  \hspace{1cm} (107)

Obviously, the scores \( t \) found are not the true scores \( t^* \) because part of the error mixes within the scores \( t \). Now, suppose the true experimental error looks like:

\[ e = [0\ 0\ 0\ 0\ 0\ \delta\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0\ 0] \]

where there is a sensor failure on the sixth process variable. It can be seen from equation 107, by projecting \( e \) on \( P \) that this projection is mixed within the scores \( t \). The \( SPE \)-chart is based upon the extracted error \( e^0 \) and is found by:

\[ e^0 = x - Pt = x - PP'x = x - PP'(x^* + e) = x^* + e - PP'(x^* + e) \]
\[ = (x^* - PP'x^*) + (e - PP'e) = (x^* - Pt^*) + (e - PP'e) = (x^* - x^*) + e - PP'e \]  \hspace{1cm} (108)

and thus \( e^0 \neq e \). As a result, the \( SPE \)-chart is based upon the part of the experimental error \( e \) that is not projected. Clearly, the fault is distributed in both the monitored scores \( t \) and residuals \( e^0 \) by nature. Therefore, as a consequence of applying PCA for monitoring purposes, the charts can only be complementary if \( e^0 = e \) (Figure 96). It was shown that this situation is extremely rare.

Summarised, a process disturbance very likely will not behave in such a way that it is detected in either the \( D \) or \( SPE \)-chart. The fault describing the insufficiently cooled reactor is only exclusively detected in the \( D \)-chart if the time is given to \( all \) the other process variables to be affected. The sensor failure can never cause the other process variables to change in such a way that it will be detected exclusively in the \( SPE \)-chart. As a result, faults will mix up or detected in both charts. This hampers the diagnostic capabilities of the control charts.

6.2.5.1 Post-batch analysis

So far the focus has been on continuous processes. The reason for this is that the arguments related to the \( SPE \) and \( D \)-chart as discussed in the previous chapters also apply to multivariate statistical process control for batch processes. The same
problems associated with the mechanistics of the control charts are encountered in batch process monitoring, and even more severe.

The objective of post-batch analysis is to assign a finished batch to be statistically in or out-of-control by comparing the batch to a reference distribution. In the initial phase of statistical batch monitoring, batch data are collected from the plant and are judged for their normal operating behaviour. Apart from using external information regarding producing on-spec products, batches are also subjected to post-batch analysis for judging whether or not they have been run under normal operating conditions (NOC). The selected batches are referred to as NOC batches.

Commonly batch process data is stored in such a way that it fits a three-way data array \( X \). Suppose \( J \) process variables are measured over \( K \) time points from a single NOC batch run, and stored in a matrix \( A (J \times K) \). A simple way of storing several batch runs is to stack each run in the three-way data array \( X (I \times J \times K) \) according to Figure 97.

![Figure 97](image)

**Figure 97**

*Arrangement of batch process data prior to model building.*

The different batches are denoted by \( i = 1,\ldots, I \) and form the first mode of \( X \). The process variables are denoted by \( j = 1,\ldots, J \) and form the second mode of \( X \). The time points at which measurements are taken form the third mode of \( X \) and are denoted by \( k = 1,\ldots, K \). Here, it is assumed that the NOC batches represent a process that is statistically in-control. Therefore, the variation between batches from the NOC data \( X \) is considered to be common cause variation that is to be modelled.

Since PCA is a linear technique, pre-processing of \( X \) is required because of the highly non-linear behaviour of batch processes. In this study \( X \) is column-centred and slab-scaled within each process variable (H.A.L. Kiers [49]), thereby removing the average trajectories of all the process variables and giving all process variables \textit{a priori} equal weight. The pre-processed data in \( X \) now represents deviations from the average trajectory and is therefore approximately linear (P. Nomikos & J.F. MacGregor [6]).
The data-array $\mathbf{X}$ is matricized (H.A.L. Kiers [49]) in the batch direction $I$ resulting in $\mathbf{X}^{i\times j\times K}$. Because of matricizing $\mathbf{X}$, the process variable mode and time mode are nested. The number of columns of this $\mathbf{X}$ is much larger as compared to data collected from continuous processes because all process variables at each point are considered as new variables. For an average number of process variables, this number may be as high as 2000. For a batch process monitored with spectroscopic instruments this number can easily be 100,000, thus, dimension reduction is necessary.

The derivation of the model and calculation of the test statistics is not different from the continuous processes with dimension reduction. Therefore, a PCA model on $\mathbf{X}^{i\times j\times K}$ is built according to

$$
\mathbf{X}^{i\times j\times K} = \mathbf{T}\mathbf{P}' + \mathbf{E}
$$

where $\mathbf{T}$ ($i\times R$) are the scores, $\mathbf{P}$ ($j\times K\times R$) the loadings and $\mathbf{E}$ ($i\times j\times K$) the residuals of the model. Every row of $\mathbf{X}$ represents a completed NOC batch.

The test statistics for a new independent finished batch $\mathbf{x}$ are calculated in the same manner as equation 103 and equation 104. For post-batch analysis the $SPE$-statistic is referred to as the $Q$-statistic (P. Nomikos & J.F. MacGregor [19]) to distinguish it from the on-line statistic $SPE$ (see later), that is the sum of squared residuals over all time periods.

The data structure of $\mathbf{x}$ has very important consequences for diagnosis and detection. Besides the correlation between the process variables for this type of batch data, there is also correlation in the time direction. That is, if a process fault that changes the correlation structure occurs somewhere halfway the batch run, it can never be detected uniquely in the $D$-chart. This situation has been illustrated in Figure 98.
The data represent the measurement of three temperatures in a batch reactor. The batch duration for this process is 15 time intervals. The x-axis of Figure 98 describes the vector $x_1^{15,15}$ when the 3 process variables are measured at the 15 time points. In the figure each measurement of the process variables is separated by small blank spaces. A process fault, indicated by the white bars, starts at time interval 12: the temperatures are simultaneously affected. The black bars represent the in-control situation where the process would still be normal. Clearly, the correlation pattern for the process variables at time interval 12-15 is the same. However, the test statistics are calculated by projection of the entire batch run, including time interval 1-11. Since for time interval 1-11 the correlation pattern is not changing, the projection of $x$ can never lead to detection of the fault exclusively in the $D$-chart. By projecting the entire batch run, the correlation pattern considering all the process variables is broken. Breakage of the correlation patterns is likely to be detected in the $Q$-chart.

Another consequence of the data-structure of batch data can be understood from the following. Figure 99 represents the same temperature measurements in the batch reactor.

Suppose one of the temperature sensors gives an abnormal signal due to a sensor failure at time interval 8. It was shown for the continuous case that these types of faults are only detected uniquely in the $SPE$-chart under specific conditions: all process variables must be changed in a certain way. To calculate the $Q$-statistic, the entire batch run is projected. Thus not only the two temperature measurements at time interval 8 have to change, but also the 'past' measurements from time interval 1-7 and future measurements of time interval 8-15 need to change. Clearly, this cannot happen. Therefore, this fault can never be detected uniquely in the $Q$-chart.

Figure 99
Construction of batch data including sensor fault.
Apart from the problems mentioned above, the problem of embedded error becomes even more pronounced in the $Q$-statistic. Moreover, since the $Q$-statistic takes a summation over the time direction, large squared residuals at a certain time point tend to be levelled out by small squared residuals at other time points (P. Nomikos & J.F. MacGregor [19]). This also hampers the detection performance of the $Q$-statistic.

Summarising, most process faults during a batch run will break the correlation pattern. This makes diagnosing difficult. The breakage of the correlation will show up mainly in the $Q$-chart and a portion of the fault creeps into the $D$-chart due to the embedded error. Hence, the diagnostic capabilities of the $Q$ and $D$-chart are limited. Moreover, due to the limited detection power of batch charts, using post batch analysis for deciding whether or not a training batch is an NOC batch is difficult.

6.2.6 Statistical performance of control charts for batch process monitoring

In the first part of this chapter, the problems associated with the mechanics of the control charts are discussed. In the following chapters, the statistical properties of these control charts are discussed. Problems related to these statistic properties are highlighted and suggestions for improvements are given. The properties of the charts are tested by means of performance indices. The effect of the improvements is tested using several batch process datasets.

6.2.6.1 On-line batch monitoring

It is also desirable to monitor a batch from the beginning until its completion. That is, at every time interval $k$, the batch $x_k$ is tested whether it is statistically in or out-of-control by comparing the test statistics $SPE$ and $D$ for $x_k$ with their reference distribution. This is called on-line batch monitoring.

A completely new batch independent from the NOC data will be denoted as $x_k$. This batch will be monitored on-line, $x_k$ will be projected on to the space generated by the model the model $P$ although the batch has not finished yet. The loadings $P$ are the loadings as given in equation 109. In order to project this particular batch on $P$ to obtain the scores needed for monitoring, the dimensions of $x_k$ must
match the dimension of the matrix \( P \). Therefore \( x_k \) is constructed as depicted in Figure 100.

![Diagram of Past, Current, Future Measurements](image)

**Figure 100**

The current deviations approach.

Suppose at time interval \( k \) a measurement of \( J \) process variables comes in. This will be denoted as the *current* part of \( x_k \). All the measurements until \( k \) are denoted as the *past* part of \( x_k \). Therefore, the *future* part of \( x_k \) from \( k+1 \) to \( K \) must be estimated somehow. Thus at each time interval \( k \), the vector \( x_k \) is constructed.

In this study the current deviations approach (P. Nomikos & J.F. MacGregor [6]) is chosen to fill in the future part of \( x_k \). E.N.M. Van Sprang et al. [41] used several performance indices and data-sets to show that this approach works reasonably well compared with other methods for on-line batch process monitoring.

The principle of batch process monitoring is comparing a new batch \( x_k \) (\( JK \times 1 \)) against a reference distribution. Therefore, in order to have a sensible comparison, the NOC batches are treated in a similar fashion as \( x_k \). Suppose there are 67 NOC batches and the batch duration \( K \) equals 58. To monitor the new batch \( x_k \), 58 different vectors are constructed (see Figure 100) according to the current deviations approach, each denoted by \( x_k \). Therefore, 58 different reference distributions are required, each denoted by \( X_k \) (\( J \times JK \)). The matrix \( X_k \) consists of three parts: a part with known process measurements, current measurements and a part with unknown process measurements. The latter part is estimated using the current deviations approach. This procedure is repeated for each time interval \( k \).
Once \( X_k \) is known for each time interval, the matrix is projected on the model using the loadings \( P \) in order to obtain the necessary scores \( T_k \) (I\( \times \)R) and residuals \( E_k \) (I\( \times \)K). These scores and residuals serve as reference distributions for the test statistics \( D_k \) and \( SPE_k \).

Thus, at every time interval \( k \), the batch \( x_k \) is tested whether it is statistically in or out-of-control by comparing the \( SPE_k \) and \( D_k \)-statistic for \( x_k \) with their reference distribution. The derivation of the test statistics is explained in the following.

**D-statistic for on-line batch monitoring**

The calculation of the \( D \)-statistic for a new batch \( x_k \) involves the scores. To calculate this test statistic, first the Mahalanobis distance (\( MD \)) is calculated to take variances and covariances between the scores into account (K.V. Mardia et al. [116]):

\[
t_k = (P'P)^{-1}P'x_k = P'S^{-1}t_k
\]

\[\rightarrow MD_k = t_k'S^{-1}t_k\]

where \( t_k \) (R\( \times \)I) are the scores calculated at time interval \( k \), and \( S \) (R\( \times \)R) is the variance-covariance matrix of \( T_K \). The scores are considered to be multivariate normally distributed due to the central limit theorem (P. Nomikos & J.F. MacGregor [19]). The \( D \)-statistic is the Mahalanobis distance times a correction factor, and is calculated according to (P. Nomikos & J.F. MacGregor [19]):

\[
D_k = MD_k \cdot \frac{I(I-R)}{R(I^2-1)}
\]

and follows an \( F(R, I-R) \) distribution, assuming that the new scores are drawn from the same distribution as the reference distribution (N.D. Tracy et al. [14]). Another assumption involving the \( D \)-statistic is that the covariance matrix \( S \) of the scores \( t_k \) of the new batch equals the covariance matrix \( S \) from the reference distribution \( T_K \). The limits for the control-chart are not time dependent. This is because the degrees of freedom for the \( F \)-distribution are determined by \( R \) and \( I \), which are fixed.

If a fault is not apparent from the beginning of a batch the correlation structure is always broken by definition and will therefore be detected in the \( SPE \)-chart. Thus after roughly 10% of the batch time the \( D \)-chart can be discarded as well. Furthermore, even if the fault is apparent from the start, it will break the correlation
structure due to the filling-in procedure. This is not true only for the current deviations approach, but holds for all filling-in methods. Only when the batch is completely finished, and the fault was apparent from the start until the end, a post batch analysis can result in a signal in the D-chart as explained earlier. It might become obvious from this that the D-chart is useless for on-line monitoring purposes using any filling-in procedure.

P. Nomikos & J.F. MacGregor [19] proposed also a third method to deal with the problem of an unfinished new batch. In this method $x_i$ is projected onto the part of $P$, which is associated to the measurements up to and including time point $k$. This is called the projection approach and it is recommended to use only after at least 10% of the new batch has finished, otherwise no reliable statistics will be obtained. From the analysis above it becomes clear that the projection approach will not solve the problems of the D-statistic either. There is only one specific situation where the D-chart, using the projection approach, is useful. Suppose there is abnormal variation within the process variables from the start of the batch that behaves according to the correlation structure. Since the projection approach does not use a filling-in procedure, such a fault will indeed show up in the D-statistic.

**SPE-statistic for on-line batch monitoring**

The calculation of the SPE-statistic for a new batch $x_k$ involves the residuals. Using the scores $t_k$ from equation 110, the residuals $e_k$ ($Jx1$) are calculated according to:

$$e_k = x_k^{j=1:J_1} - t_k P^{j=1:J_1} \rightarrow SPE_k = e_k^T e_k$$  \hspace{1cm} (112)

Notice that the residuals are calculated using only the current part of $x_k$ and $P$ indicated by $R,(Jk - J + 1):Jk$. That is, only the part corresponding to the $J$ process variables measured at time interval $k$ are of interest. This notation is read as follows. Suppose the matrix $P'$ has 5 rows and 100 columns. The process variables ($J = 10$) are nested within the time mode ($K = 10$) as a result of matricizing. Suppose only the measurements at e.g. time $k=3$ are of interest. In other words only the columns 21 until 30 are of interest. This is denoted as $5, 21:30$.

The effect of the embedded error is decreased for the $SPE_k$ by the fact that only the current part of the calculated residuals is used. Moreover, the $SPE_k$ only
considers squared residuals at a certain time point, and does not sum-up over time as does the Q-statistic. This increases the detection power of the $SPE$-chart. The $SPE_k$ follows approximately a weighted $\chi^2$ distribution (P. Nomikos & J.F. MacGregor [19]) with $b$ degrees of freedom and weight $g$ to account for the magnitude of $SPE_k$, assuming that $x_k$ is multivariate normally distributed with expectation zero (G.E.P. Box [23]). These parameters $g$ and $b$ are estimated (J.E. Jackson & G.S. Mudholkar [24]) at each time interval $k$ using the proper reference distribution: the current part of $E_k$ of size $1 \times J$. The limits for a $SPE$-chart will vary in time. This is because the degrees of freedom at each time interval are estimated from the residuals.

6.2.6.2 Further comments on-line statistical batch monitoring
The batch is statistically out of control when the limits are broken in one of the control charts. If the limits are broken the cause of the deviation from the normal operating conditions can be located using contribution plots (P. Miller et al. [16], J.A. Westerhuis et al. [17]). Since the (on-line) control charts $D$ and especially $SPE$ are much more sensitive than the post-batch versions ($D$ and $Q$), it is recommended to use on-line monitoring to judge whether training batches are NOC batches or not (apart from a priori information concerning on-spec products).

6.2.7 Description of the data
In order to evaluate the effect of improvements (see Section 6.2.8) for on-line monitoring, six different types of data sets are used as an example. The different features of these data sets will be explained in the following. The NOC data sets are used to compute the overall type I error and disturbed batches are used to compute the action signal time (AST). The definition of these performance indices is given in Section 6.2.8. The time of disturbance occurrence for the erroneous batches and the dimensions of the three-way data array $X$ for the studied data sets are given in Table 12.
### Table 12

<table>
<thead>
<tr>
<th>Data set (I×J×K)</th>
<th>Disturbed batches</th>
<th>Fault start</th>
</tr>
</thead>
<tbody>
<tr>
<td>Data set 1 (67 × 15 × 58)</td>
<td>Batch 14</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td>Batch 17</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td>Batch 24</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td>Batch 49</td>
<td>46</td>
</tr>
<tr>
<td>Data set 2 (27 × 67 × 271)</td>
<td>Batch 2*</td>
<td>138</td>
</tr>
<tr>
<td>Data set 3 (47 × 3 × 100)</td>
<td>Wrong batch 1</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td>Wrong batch 2</td>
<td>1</td>
</tr>
<tr>
<td>Data set 4 (50 × 9 × 200)</td>
<td>Batch 106*</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td>Batch 99*</td>
<td>100</td>
</tr>
<tr>
<td>Data set 5 (47 × 8 × 116)</td>
<td>Batch 41</td>
<td>58</td>
</tr>
<tr>
<td></td>
<td>Batch 46</td>
<td>94</td>
</tr>
<tr>
<td></td>
<td>Batch 47</td>
<td>94</td>
</tr>
<tr>
<td>Data set 6 (36 × 10 × 100)</td>
<td>Batch 50</td>
<td>56</td>
</tr>
</tbody>
</table>

*Description of the data sets and disturbed batches. * = exact occurrence of fault known*

**Data set 1**

This data set is from an industrial (Shell) suspension polymerisation of polyvinylchloride (PVC) in a batch reactor. The data set consists of 67 batch runs obtained under NOC and 15 process variables were measured for a period of 58 time intervals. The NOC batches are aligned using the conversion as a maturity variable. A more detailed description is given by A.A. Tates et al. [2]. The process variables consist of: temperatures inside reactor and cooling water, condenser duty, agitator speed and power supply, two mass streams of cooling water, batch reactor level and pressure. Four disturbed batches are available to calculate the AST. Analysis of univariate plots of the process variables is used to study the disturbed batches. Batch 14 shows a disturbance in the amount of cooling water through the condensor. Batch 17 indicated a disturbance in the temperature of the top condensor. Batch 24 suffers from disturbances for the batch level and the power supply to the agitator. Analysis of batch 46 revealed that the temperature and the amount of refrigerant water to the jacket are disturbed at the end of the batch run.
Data set 2
Data set 2 consists of a laboratory spectroscopic batch process of a two-step biochemical conversion reaction described in S. Bijlsma et al. [66], and can be obtained from the Process Analysis & Chemometrics website of the University of Amsterdam. The data set consists of 27 NOC batch runs measured at wavelengths 300-500 nm during 271 time intervals. One batch run with a pH disturbance at time interval 138 is available to compute the AST.

Data set 3
This data from Loders Croklaan concerns a food manufacturing batch process during which the catalysed hydrogenation of bean oil is performed. Three process variables (H₂ flow, pressure and temperature) are measured over 100 time points for 50 batches. These batches were synchronised based on the rate of conversion. A more detailed description of the process can be found in A.K. Smilde & H.A.L. Kiers [39]. Two disturbed batches are used in this study: the first batch has a too high H₂ flow as well as a too high rising pressure, the second batch suffers from a deactivated catalyst. Besides, the product quality of the first batch was of low quality.

Data set 4
Data set 4 consists of a simulated emulsion co-polymerisation of styrene-butadiene. This process is well described in the paper by P. Nomikos & J.F. MacGregor [6]. The data set consists of 50 NOC batches for which 9 process variables are measured during 200 time intervals. The following process variables are measured: two flow-rates, four temperatures, density, conversion and rate of energy release. Two disturbed batches are available to test the detection power of the methods. The first (batch 106) has an initial organic impurity contamination in the butadiene feed. The second (batch 99) has a similar, but larger, contamination halfway the batch run.

Data set 5
Data set 5 consists of an industrial (DuPont) polymerisation in an autoclave with 47 NOC batch runs. This data set is also referred to by K.A. Kosanovich et al. [7]. The polymer is produced through five stages during these stages the process is monitored by 8 process variables (six temperatures and two pressures) during 116 time intervals. Prior to model building, the raw data is linearly interpolated to align the NOC
batches, this is described in R. Boqué & A.K. Smilde [40]. Three disturbed batches are available to calculate the AST. A univariate analysis of batch 41 shows clear disturbances for two pressure and four temperature measurements halfway the batch run. This is probably due to an upset in the first stage of the process. For batch 46 and batch 47, univariate analysis shows disturbances at the end of the batch run for one pressure and five temperature measurements.

Data set 6
Data set 6 is an industrial (Du Pont) two-stage polymerisation process with 36 selected NOC batches for which 10 process variables are measured during 100 time intervals. One disturbed batch is available to calculate the performance indices, which was known to have a poor end-product quality. This data set is described in P. Nomikos & J.F. MacGregor [19].

For all data sets except the sixth one, three principal components are chosen to describe the data in $X^{p \times k}$. For data set six, two components were sufficient. The data is cleaned from outliers, and thus represents NOC batches.

6.2.8 RESULTS
All the issues and problems discussed in the previous section will be discussed and illustrated using the case study of the several batch processes. The problems and issues, however, hold in general for all batch process monitoring problems.

6.2.8.1 Performance indices
The next two indices are used to test the performance of the proposed methods: the overall type I error and the action signal time (AST). The definition of these indices will be discussed in this section.

6.2.8.1.1 The overall type I error
On-line monitoring of batches can be regarded as a sequential hypothesis-testing problem. A batch is tested at every time interval $k$ whether it is statistically in-control by comparing the test statistics $SPE$ and $D$-statistic to their reference distribution. There are two hypotheses of importance, that is, the null hypothesis $H_0$ (the batch is in-control) and the alternative hypothesis $H_1$ (the batch is out of control). The
rejection of $H_0$ while it is true is called a type I error. The size of this error, or the probability of a type I error, is given by:

$$\alpha_{\text{imposed}} = P(\text{type I error}) = P(\text{reject } H_0 | H_0 \text{ is true})$$ (113)

In this study $\alpha_{\text{imposed}}$ is set to 5%. It is useful to check the performance of the control charts by comparing the actual $\alpha$ to $\alpha_{\text{imposed}}$. This is done by sequentially monitoring all NOC batches. Since an NOC batch is by definition statistically in-control, a signal in one of the charts can be regarded as a type I error. The performance of a chart is good if $\alpha_{\text{actual}}$ is close to $\alpha_{\text{imposed}}$.

### 6.2.8.1.2 Action Signal Time

The action signal time (AST) is used to study the efficiency of a control chart to detect faults. A more commonly used criterion in SPC for investigating the type II error is the average run length (ARL) curves. However, theoretical derivations of ARL statistics are difficult if not impossible in the case of batch process monitoring. An alternative might be an extensive and rigorous simulation study but that is not the

The action signal time is defined as the time between the introduction of an error and the out-of-control signal. Three consecutive points outside the control limit defines the out-of-control signal. This definition of the out-of-control signal is common practice since process operators will unlikely undertake action after just one crossing of the limit.

### 6.2.8.2 Accounting for the difference between NOC and new batches

In on-line batch monitoring, the scores and residuals of new batches are compared with their reference distributions for NOC batches. The NOC model is calculated in such a way as to minimise the residual variance of the NOC batches. This is reflected in the NOC scores and residuals. As the NOC model is based upon a limited number of batches, it is likely that the projection of independent new batches from the same population will give higher residuals (and lower scores) than the NOC batches, because the new batches did not take part in the fitting of the model to the data.

In order to make the NOC scores and residuals more comparable to those of new batches, a leave-batch-out procedure is used, where each NOC batch is treated as a new independent batch. This approach was suggested by D.J. Louwerse & A.K.
Statistical batch process monitoring

Smilde [38]. A model is built on the remaining NOC batches, and the NOC batch left out from the model is projected to calculate the scores and residuals. This is repeated for all NOC batches, and all of the calculated scores and residuals form the new NOC distribution of scores and residuals. This procedure is explained in more detail in Appendix E. Clearly, these scores are not calculated in order to minimise the residuals variance. Therefore, this distribution of scores and residuals is better comparable to those found by projection of a new independent batch.

6.2.8.3 Hypothesis testing

A crucial point in calculating $\alpha_{actual}$ is the definition of the null hypothesis. The null hypothesis used by Nomikos and MacGregor is specified for each separate time point $k$: a batch is in-control at a certain time point $k$ if it does not exceed the limit for that time point. Such a hypothesis is then tested sequentially for all $K$ time points during the batch run (P. Nomikos & J.F. MacGregor [19]). Every rejection of the null hypothesis for an NOC batch is a false alarm. When all $I$ NOC batches are subjected to monitoring then $\alpha_{actual}$ can be calculated by:

$$\alpha_{actual} = P(\text{overall type I error}) = \frac{\sum \text{false alarm}}{IK} \quad (114)$$

E.N.M. Van Sprang et al. [41] used the same method to calculate $\alpha_{actual}$. They found an $\alpha_{actual}$ which is slightly too high for the $SPE$-chart and too low for the $D$-chart for various datasets.

A more realistic null hypothesis is: a batch is in-control if it is in-control during the entire batch run. As soon as an NOC batch breaks the limits of the control chart somewhere along the time trajectory, the null hypothesis that the batch is in-control is rejected and a false warning has occurred. After breaking the limits no further testing is required. This $H_0$ is more realistic than the one used by P. Nomikos [21] because it tests whether a whole batch is in-control and not only at a certain point. Moreover, the $H_0$ used by Nomikos and MacGregor will result in 5% false alarms in the whole batch trajectory of each new batch (by construction). Stated otherwise, the $H_0$ of P. Nomikos [21] will result in too tight control limits, which will be a problem in practice, which has indeed already been observed (P. Nomikos [21]).
The \( \alpha_{\text{actual}} \) for the new null hypothesis can be calculated as follows. Let the indicator variable \( \sigma_i \) be 1 if this event is true, and zero if the limits are not broken somewhere along the time trajectory. Then, the calculation of \( \alpha_{\text{actual}} \) is now given by:

\[
\alpha_{\text{actual}} = P(\text{overall type I error}) = \frac{\sum_{i=1}^{l} \sigma_i}{l}
\]

The new null hypothesis is a composite hypothesis and testing this hypothesis comes down to testing sequentially individual hypotheses: one hypothesis for each point in time. The difference between this new way of testing and the previous approach is that the latter does not consider the overall (composite) hypothesis, which causes the problems as indicated above.

6.2.8.4 The Bonferroni adjustment

Because of the sequential testing problem in both the D and SPE-chart, the probability of improperly rejecting the null hypothesis is given by:

\[
P(\text{Overall type I error}) = 1-(1-\alpha)^K
\]

where \( \alpha = \alpha_{\text{inprod}} \). For example, if \( \alpha = 0.05 \) and if \( K \) is equal to 58, then the probability that an overall type I error occurs is equal to 0.95. Clearly, the magnitude of 0.95 \( \gg \alpha \). In other words, the probability of rejecting a batch while it produced a good product is 95%. This is a problematic situation for practical purposes. Setting the limits according to the Bonferroni correction (G.W. Snedecor & G.C. Cochran [68]) solves this problem. To assure the probability of an overall type I error reflects the value of \( \alpha \), \( \alpha_{\text{inprod}} \) is calculated according to:

\[
\alpha_{\text{inprod}} = 1-(1-\alpha)^{1/K} = \frac{\alpha}{K}
\]

For the previously example, where \( K = 58 \), the overall type I error is now approximately equal to 0.05. Clearly this is more in agreement with the expectations of choosing the confidence limit of 95%.
In chemical engineering practice it is common to set $\alpha_{\text{impred}}$ to 99.9% to reduce the number of false alarms (D.J. Louwers & A.K. Smilde [38]). Applying Bonferroni and the leave-one-out approach motivates this practical usage from a theoretical background.

The Bonferroni procedure ignores important information about the data, that is the correlation structure of the test statistics. This will result in too conservative limits leading to decreased detection speed of erroneous batches. Therefore the critical level of $\alpha_{\text{impred}}$ can be adjusted according to the Dubey and Armitage-Parmar (D/AP) procedure (A.J. Sankoh et al. [117]). They suggest to use the following adjusted value for $\alpha_{\text{impred}}$:

$$\alpha_{\text{impred}} = 1 - (1 - \alpha)^{1/m} \quad \text{where} \quad m = K^{1-\bar{r}_i} \quad (118)$$

Here, $\bar{r}_i$ is the mean correlation coefficient between the $K$-test statistics and is calculated as follows. Each NOC batch is monitored in a leave-one-out procedure, and the test statistics $D$ and $SPE$ are stored in the $i$-th row of the matrices $D$ ($I \times K$) and $SPE$ ($I \times K$). Then for both matrices the correlation matrix is calculated. Now, $\bar{r}_i$ is the mean value for the off-diagonal elements and can be calculated for both the $SPE$ and $D$-chart. If the correlation is zero, the D/AP procedure is equal to the Bonferroni adjustment. The average value over all datasets of $\bar{r}_i$ for the $SPE$ fluctuates around 0.26 and for the $D$-statistic around 0.71. These numbers are reasonable since the residuals describe non-systematic variation and therefore behave erratic. At the other hand the $D$-statistic captures the structured variation in the data and will therefore be correlated.

6.2.8.5 Further comments on the D-statistic

The following general improvements to the $D$ and $SPE$-chart were suggested in the foregoing: the leave-one-out approach, a new definition of $H_0$, the Bonferroni correction and the D/AP procedure. Although the $SPE$-chart is clearly improved, these suggestions will not improve the performance of the $D$-statistic since the performance of this statistic is severely hampered due to its construction. The problem of the $D$-statistic resulting from filling-in procedures, however, can be solved partly by changing the way to calculate the $D$-statistic. This is described in Section
6.2.8.5.1. The rest of this Section gives some auxiliary comments with respect to the D-statistic.

**6.2.8.5.1 Time dependent covariance matrix**

As can be seen from equation 110, the calculation of the D-statistic is based upon the covariance matrix \( S \) (\( R \times R \)) and \( S \) is assumed to be constant over time. This covariance matrix is calculated according to:

\[
S = \frac{1}{I-1} \left( T_k \right)' \left( T_k \right)
\]  

(119)

P. Nomikos & J.F. MacGregor [7] already mentioned in their paper that it is possible to calculate a covariance matrix for each point in time. This would result in:

\[
S_k = \frac{1}{I-1} \left( T_k \right)' \left( T_k \right)
\]  

(120)

The impact of using a time varying covariance matrix is shown Figure 101.

**Figure 101**

*Dynamic behaviour over time for the covariance matrix.*

Here, \( S_k \) is calculated for the first dataset. The dotted lines in Figure 101 represent the R diagonal elements (variances) of \( S_k \) where the solid line represents the squared sum.
of the off-diagonal elements (covariance) $S_k$. This figure shows that the covariance matrix cannot be assumed to be constant over time. Moreover, it can be seen from Equation 120 that the scores $T_k$ are used to calculate $S_k$. These scores $T_k$ are affected by the filling-in procedure. Using $S_k$ compensates to some extent for the problems of using the filling-in procedure. Therefore, it is strongly recommended to use a time dependent $S_k$.

The fault detection is clearly improved as can be seen from an example in Figure 102. Here, the $D$-statistic using $S_k$, marked by the star, clearly shows a more dynamic behaviour compared to the situation where $S$ is used. Moreover, the faulty batch goes unnoticed in the $D$-chart when $S$ is used!

![Figure 102](image)

**Figure 102**

* $D$-chart using time independent and time varying covariance matrix.

### 6.2.8.5.2 Non-normality

The scores used to calculate the $D$-statistic are assumed to be normally distributed. P. Nomikos & J.F. MacGregor [19] use the central limit theorem to justify the assumption of normality. N.D. Tracy et al. [14] stated in their paper that the results for deriving control limits depend on the validity of this assumption. That is, the rejection of the null hypothesis using a F-test is very sensitive to small departures
from normality. The effect on the limits increases when the distributions are either heavy-tailed or light-tailed (G.M. Miller [118]).

In this study, normality plots of the scores $T_k$ indicated that the scores did not depart severely from normality. It can be concluded that the normality for all data sets is acceptable, and therefore the F-test is allowed.

6.2.8.5.3 Violation of the $\Sigma_f = \Sigma$ assumption

If the variation of the process variables in the application phase can be described by the covariance matrix $\Sigma_f$, then it is be assumed that $\Sigma_f = \Sigma$. An estimate of the true population value $\Sigma$ is given by S. S.J. Wierda [119] investigated the effect of the violation of this assumption, that is, $\Sigma_f \neq \Sigma$. Wierda concluded the following: the true probability that the $D$-chart produces a false alarm can be considerably smaller or larger than $\alpha$ if the above stated assumption is violated.

Reasons for violation of this assumption are twofold. First, the estimate $S$ or $S_k$ is inaccurate because of the limited number of batches. Secondly, a new monitored batch will always differ from the NOC batches and therefore the assumption is violated to a certain extent by definition. S.J. Wierda [119] suggest to use a control chart that tests the hypothesis that the covariance remains stable over time. In fact, this is exactly what the SPE-statistic is testing. In other words, as soon as the SPE-chart detects a breakage of the correlation pattern, the $D$-statistic becomes unreliable. The argumentation does not only apply to on-line monitoring, but also for post-batch analysis.

6.2.8.5.4 Degrees of freedom in process monitoring

In the current methodology for deriving control limits for the $D$-statistic (for on-line monitoring of batch/continuous processes as well as for post-batch analysis), the degrees of freedom in the F-distribution considers the number of components ($R$) and the number of batches ($I$). This is a direct generalization of the degrees of freedom for Hotellings $T^2$ statistic derived from normally distributed variables. The $D$-statistic, however, uses $t$-scores, which are constructed variables based on the original measured variables in the dataset. It is unclear whether the generalization of the degrees of freedom for Hotellings $T^2$ to the $D$-statistic is valid. The $t$-scores can also be derived from other models of the three-way dataset $\mathbf{X}$, e.g. using PARAFAC or Tucker models (H.G. Law et al. [120], R. Coppi & S. Bolasco [121]). The $t$-scores of,
e.g., a PARAFAC model might have completely different statistical properties as the t-scores from the unfold PCA model. These differences are not reflected in the direct generalization of degrees of freedom going from $T^2$ to $D$. This is still an open question. Note that for the $SPE$-statistics things are different. The residuals (as a result from either an unfold PCA or PARAFAC model) are used to calculate approximate degrees of freedom for the $\chi^2$-distribution and the problem as mentioned above does not exist. The observation is that indeed the degrees of freedom for the $SPE$ of an unfold PCA or PARAFAC model are different.

6.2.8.6 Further comments on the squared prediction error (SPE)

The second test statistic, which will be discussed in further detail, is the squared prediction error. E.N.M. Van Sprang et al. [41] already showed that the performance for this test statistic is generally good. The overall type I error is satisfying ($\alpha_{\text{desired}} = \alpha_{\text{actual}}$) and the detection power is good as will be shown in the following. Therefore the performance of this chart is not alarmingly poor.

6.2.8.6.1 Non-normality

Assuming normality of the residuals, the $SPE$ statistic is distributed according to a weighted sum of $\chi^2$ distributions. This weighted sum can be approximated by a $\chi^2$ distribution $g\chi^2(h)$ where $g$ is a size factor and $h$ the degrees of freedom (G.E.P. Box [23]). Inspection of normality plots for the residuals showed a deviation from normality for all datasets. Therefore, the residuals are not normally distributed and the assumption of normality is violated.

The $g$ and $h$ parameters are obtained by fitting the $g\chi^2(h)$ distribution to the reference distribution of the SPE from the NOC data. Experience has shown that, although the residuals are not exactly distributed according to a normal distribution, the $g\chi^2(h)$ distribution approximates the behaviour of the $SPE$-statistic quite well. Hence, it appears that in practice small deviations from normality are not troublesome.
6.2.8.7 Results adjustments to the control charts

The results of the previously discussed adjustments to the control charts are presented in Table 13.

<table>
<thead>
<tr>
<th></th>
<th>Standard</th>
<th>Bonferroni</th>
<th>Bonferroni + Leave one out</th>
<th>Bonferroni + Leave one out + $S_k$</th>
<th>D/AP + Leave one out</th>
</tr>
</thead>
<tbody>
<tr>
<td>SPE</td>
<td>OT1</td>
<td>73.87</td>
<td>9.95</td>
<td>5.79</td>
<td>5.79</td>
</tr>
<tr>
<td></td>
<td>OT1 N&amp;M</td>
<td>7.85</td>
<td>-</td>
<td>5.23</td>
<td>5.23</td>
</tr>
<tr>
<td></td>
<td>AST</td>
<td>18</td>
<td>38</td>
<td>44</td>
<td>44</td>
</tr>
<tr>
<td></td>
<td>OT1</td>
<td>19.46</td>
<td>3.12</td>
<td>11.58</td>
<td>2.57</td>
</tr>
<tr>
<td></td>
<td>OT1 N&amp;M</td>
<td>2.68</td>
<td>-</td>
<td>3.85</td>
<td>3.88</td>
</tr>
<tr>
<td></td>
<td>AST</td>
<td>56</td>
<td>91</td>
<td>72</td>
<td>76</td>
</tr>
</tbody>
</table>

Overall type I (OT1) error and action signal time (as the percentage of the batch length).

The overall type I error is calculated using the definition of the null hypothesis by Nomikos and MacGregor (OT1-N&M), as well as the definition given in this study (OT1). Also, the overall type I error is calculated as the average for all six data sets. The action signal time (AST) for the faulty batches have been calculated as a percentage of the batch length. No detection of the fault results in an action signal time of 100%. The average AST for the 13 faulty batches is presented in Table 13.

The first column of Table 13 represents the results found for the standard approach for batch monitoring, that is the current deviations approach as described by Nomikos and MacGregor in their paper. The second column represents the results of the standard approach extended with the Bonferroni adjustment, the third column the standard approach extended with Bonferroni plus the leave-one-out method, the fourth column shows the results for the standard approach extended with Bonferroni, leave-one-out and $S_k$ and the fifth column describes the results for the standard approach extended with leave-one-out, $S_k$ and D/AP. It should be noted that the last extension, that is $S_k$, has no effect on the SPE-chart. The residuals are already

-241-
calculated using the time varying scores as can be seen from Equation 112. Also, the Bonferroni and D/AP correction do not apply to OT1-N&M.

**Overall type I error**

It can be seen that the OT1-N&M is slightly too high for SPE and too low for D. These results are in agreement with E.N.M. Van Sprang et al. [41]. The OT1 error in combination with the standard approach is far too high for both charts. This is expected considering the sequential nature of hypothesis testing; $K$ hypothesis tests are performed each having a probability of 5% rejecting $H_0$. This results in high probability of a false alarm. Adjusting the limits according to the Bonferroni theory obviously improves the results for both the SPE and D-chart.

The results for the standard approach extended with the Bonferroni limits and leave-one-out approach improve the OT1 for the SPE even further. Using a time varying covariance matrix (column four in Table 13) to calculate the $D$-statistic improves the OT1 error to an acceptable number. Therefore, it may be concluded that the standard approach extended with Bonferroni limits, leave-one-out approach and a time varying covariance matrix gives satisfying results in terms of the OT1.

If the limits are based on the D/AP procedure, the leave-one-out approach and a time varying covariance matrix, the OT1 error increases. This is a disappointing result since the D/AP procedure is intuitively an appealing method. A reason might be that the OT1 error is calculated based on a total of 275 batches, which might be too limited, a number to obtain a reliable estimate of the OT1 error.

Also for the OT1-N&M method the leave-one-out approach is helpful. The average value found for the actual $\alpha$ is in good agreement with the expected value for $\alpha$. Note that in this case, the OT1 error is calculated over approximately 23000 samples (sum over batches for all datasets $\times$ sum over all time points for all datasets). Hence, these results are more reliable than the ones obtained for the D/AP procedure above.

*Action Signal Time*

It may be expected that the action signal time is related to the overall type I error. This is also what is concluded from the results given in Table 13. For example, the increment of the OT1 after applying the Bonferroni adjustment increases the average AST for both charts. In fact, three wrong batches were unnoticed after applying the Bonferroni adjustment. Also, it may be concluded that the average AST for the SPE-
chart is better than the average AST of the $D$-chart. This is in agreement with the theory described in Section 6.2.4, as well as the observations from several researchers:

E.B. Martin et al. [27]: "An abnormal batch will exhibit significant deviations in an SPE plot ".

B.M. Wise et al. [62]: "It is our experience that most process faults show up in Q. Very few faults are detected by $T^2$ alone ".

The AST for the $SPE$-chart using the D/AP procedure is hardly improved compared to the Bonferroni adjustment. This is caused by the weak correlation of the sequential $SPE$ test-statistic. In this case, the factor $m=K$ and there is hardly any difference between the Bonferroni limits and D/AP limits.

Sometimes an action signal in the $SPE$-chart is followed by an action signal in the $D$-chart. This is mainly due to the embedded error and therefore has no diagnostic value.

**6.2.9 CONCLUSIONS**

In this study the performance of control charts used for on-line monitoring of batch processes have been assessed and suggested improvements have been evaluated using six data sets. The mechanistics of the control charts are discussed. It seems that most faults break the correlation structure. This is due to the mechanistic properties of the $D$ and $SPE$-chart. The current deviations approach of Nomikos and MacGregor has been used for on-line batch process monitoring because it was found to work reasonable (also in earlier work), but the conclusions have a more general applicability. The main conclusions are the following:

- Most process faults show up in the $SPE$-chart and hardly any in the $D$-chart. This is mainly due to the construction of the model and mechanistic properties of the control charts where by definition all faults break the correlation structure and therefore are detected in the $SPE$. From an engineering point of view, all faults show up in the $SPE$, not only new events such as sensor failures. This hampers the diagnostic capability of the $SPE$-statistic.
• The $D$-chart is hardly of practical use in detecting process faults. This is due to the construction of this test statistic. A small improvement of the $D$-statistic is obtained by using a time varying covariance matrix of the scores.

• A new null-hypothesis has been formulated which improves the performance of the control charts in terms of false alarms. This new null-hypothesis necessitates the use of Bonferroni or similar types of corrections. A consequence of this adjustment is that the Action Signal Time (the time it takes to signal a fault) increases.

• A leave-one-out approach is suggested to use in calculating the control limits. This works very well in practice. Moreover, on-line aap monitoring of all available batches using this leave-one-out approach is well suited for selecting NOC batches.
6.3 The effect of the size of the training set and number of components on the false alarm rate in statistical process monitoring*

6.3.1 SUMMARY
This paper describes the sensitivity of false alarm rate to misspecification of the number of PCA components in multivariate statistical process control (MSPC) models. Using simulations, it is shown that choosing an incorrect number of components in monitoring models may seriously affect the false alarm rates of the control charts. Furthermore, the false alarm rate becomes worrisome when the size of the training set is small. Using a leave-one-out procedure for building the control charts partly solves this problem.

6.3.2 INTRODUCTION
One way of monitoring continuous or batch processes is to use multivariate statistical process control (MSPC) models (P. Nomikos & J.F. MacGregor [6], J.V. Kresta et al. [122]). To establish this monitoring tool, an empirical model is built on the data. From these models, control charts are derived: the $SPE$ and $D$ charts. The performance of these charts is reflected in probability of the type I and type II errors. In this paper the focus will be on the probability of the type I error or false alarm rate.

MSPC models are often based on principal component analysis. In PCA a number of components must be chosen and this number can therefore be considered as a metaparameter. The number of components is important with respect to the type I and type II errors (H.W. Tong & C.M. Crowe [87], H.H. Yue & S.J. Qin [110]). The choice of the number of components is often supported by cross validation. However, the goal of cross validation methods is to find an optimal model in terms of the model fit. This is not the objective of e.g. statistical batch process monitoring.

* HJR in: H.J. Ramaker, E.N.M. van Sprang, J.A. Westerhuis and A.K. Smilde, The effect of the size of the training set and number of principal components on the false alarm rate in statistical process monitoring, Chemometrics and intelligent laboratory systems, accepted.
where the objective is to minimize the probability of the type II error for a given probability of the type I error. To select the number of components according to this objective, large datasets and numerous faulty batches are required. In practice, ad-hoc methods are used to select the number of components supported by process knowledge.

A. Nijhuis et al. [123] showed the robustness of MSPC to the number of significant PC's in the sense that out-of-control situations are correctly flagged going from one principal component to three components. For all choices of components, the out-of-control situations were flagged in the control charts. The robustness against the false alarm rate was not determined. However, the speed of detection of real out-of-control situations and the false alarm rate are closely related. That is, an increase in the speed of detection might be coupled to an increased false alarm rate. It is therefore worthwhile to investigate the possible causes for a high false alarm rate. In the paper by H.W. Tong & C.M. Crowe [87] it is mentioned that the false alarm rate can become unacceptably high even for moderate size problems. Furthermore, it is an often heard remark that multivariate charts such as the SPE chart suffer from too many false alarms. A common way to overcome this problem is to set the limits of the control charts rather high e.g. 99.9% (P. Nørskov [45]). However, this reduces the capability of the control chart to detect wrong batches.

Therefore, the question is how sensitive the probability of the type I error is to misspecification of selecting the number of components. A simulation study is performed to show two things i) the sensitivity of the $D$ and $SPE$ charts to the number of components in terms of the false alarm rate ii) improvement of the false alarm rate using a leave-one-out procedure.

It will be shown that problems related to over- or underfitting can be substantially reduced for the $SPE$ chart by using a improved leave-one-out procedure. However, this leave-one-out procedure is not helpful for the $D$ chart.

### 6.3.3 Simulation of a Continuous Process

A continuous process is simulated with some special features. Fifteen process variables ($J = 15$) were measured each minute for a period of $K$ time points. The process has been designed in such a way that there are only three underlying phenomena. The data set has been constructed as follows:
\[ \mathbf{X} = \mathbf{z}_1 \mathbf{y}_1 + \mathbf{z}_2 \mathbf{y}_2 + \mathbf{z}_3 \mathbf{y}_3 + \mathbf{S} \]

where the vectors \( \mathbf{y}_1, \mathbf{y}_2 \) and \( \mathbf{y}_3 \) form the columns of an arbitrary orthogonal matrix \( \mathbf{Y} (15 \times 3) \). The vectors \( \mathbf{z}_1, \mathbf{z}_2 \) and \( \mathbf{z}_3 \) have been constructed using a moving average model. The element of e.g. vector \( \mathbf{z}_1 \) at time interval \( k \) is constructed as follows:

\[ z_1(k) = a_1(k) - \theta \cdot a_1(k-1) \]

where \( a_1 \) is a random shock with a standard deviation of 0.7. The \( \theta \)'s for \( \mathbf{z}_1, \mathbf{z}_2 \) and \( \mathbf{z}_3 \) are 0.8, 0.5 and -0.2 respectively. These numbers for the random shock and \( \theta \) were chosen in order to reflect different types of dynamics in the data. The matrix \( \mathbf{S} \) represents white noise with mean zero and standard deviation of 0.36. \( \mathbf{S} \) comprises approximately 40% of the total variation in \( \mathbf{X} \). An amount of 60% explained variance is commonly encountered in MSPC models (E.N.M. Van Sprang et al. [41]).

### 6.3.4 Multivariate Statistical Process Control

Once the data \( \mathbf{X} \) has been obtained, MSPC can be applied to the data (B.M. Wise & N.I. Ricker [112], J.V. Kresta et al. [122]). The approach used in this paper is based on MSPC and will be explained in the following. Two different datasets are simulated. The first dataset \( \mathbf{X}^{\text{training}} \) represents normal operating conditions (NOC) and is referred to as the training set. The NOC training set is statistically in control by definition and is used for model building. The other dataset \( \mathbf{X}^{\text{test}} \) represents the test set and will be used for monitoring. This test set is independent from the NOC data but also represents NOC conditions.

First, a PCA model is built on the NOC data. A PCA decomposes the matrix \( \mathbf{X}^{\text{training}} \) into the sum of \( R \) outer products of scores \( \mathbf{t} \) and loadings \( \mathbf{p} \) plus a residual part \( \mathbf{E} \):

\[ \mathbf{X}^{\text{training}} = \sum_{r=1}^{R} \mathbf{t}_r \mathbf{p}_r^\top + \mathbf{E} = \mathbf{T} \mathbf{P}^\top + \mathbf{E} \]
where $T (K \times R)$ is the score matrix, $P (J \times R)$ contains the loadings and $E (K \times J)$ is the residual matrix. Here, $K$ denotes the number of samples, $J$ the number of process variables and $R$ the number of principal components.

Secondly, the monitoring of a sample is performed. The scores $t_{new}$ and residuals $e_{new}$ for a new scaled measurement $x_{new} (J \times 1)$ from the test set $X^{test}$ are found by projection on the model plane. From these scores and residuals, the $D$ and $SPE$ statistics are calculated:

\[
\begin{align*}
    t_{new} &= (P'P)^{-1} P'x_{new} = P'x_{new} 
    \quad \rightarrow \quad D = t_{new}'A^{-1}t_{new} \\
    e_{new} &= x_{new} - Pt_{new} 
    \quad \rightarrow \quad SPE = e_{new}'e_{new}
\end{align*}
\]

(124) \hspace{1cm} (125)

Here, $A$ is the variance-covariance matrix of the scores $T$. The $D$ statistic follows an $F$-distribution and the control limits (CL) for the control chart can be calculated according to (N.D. Tracy et al. [14]):

\[
    CL_{new} = \frac{R(K^2 - 1)}{R(K - R)} F(\alpha; R; K - R)
\]

(126)

The $SPE$ statistic follows a weighted chi-square distribution ($\sim gX^2_h$). The parameters $g$ and $b$ are estimated according to J.E. Jackson & G.S. Mudholkar [24]. From this chi-square distribution, the CL for the $SPE$ chart can be calculated.

The probability of the type I error is calculated for the test set. The type I error describes the event that a NOC sample is flagged out-of-control while it is in control. The limits for $D$ and $SPE$ statistics are derived assuming in-control data. In the ideal case, the probability of the type I error reflects the choice of the significance level $\alpha$. In this work, the significance level $\alpha$ of the $SPE$ and $D$ charts is set to 0.05. In other words, there is a probability of 5% that the type I error occurs. The false alarm rate for the test set is calculated as follows. Every sample is projected to calculate the $SPE$ and $D$ statistic. Since in this simulation the test set is in control by definition, a crossing of the control limits in the control charts is regarded as a false alarm. The number of false alarms is counted and divided by the number of observations of the test set. This number is the false alarm rate.
In order to obtain reliable results, the construction of the training and test set is repeated hundred times. As a result, the average false alarm rate over the hundred test sets is calculated.

6.3.4.1 Size of the NOC data
It is expected that the statistical properties of the control charts are directly related to the number of samples in the training set. That is, the estimation of the reference distribution parameters will be increasingly accurate if the number of samples increases. The number of samples in the training set can be regarded as another meta parameter. To study this behavior, the training set is constructed for different lengths of the time \( K \). The difference in length is as follows: 20, 30, 50, 75, 100, 150, 250, 500, 1000, and 2000 time points. That is, a training set is constructed for e.g. 20 time points. This is repeated 100 times and the average false alarm rate is calculated. Then, the next training set with a length of 50 time points is constructed, etc. The length of the test set is fixed and contains 250 time points.

6.3.4.2 Number of principal components
Due to the construction of the dataset, only three underlying phenomena are present in the data. If the number of principal components is less than three, the model underfits the data. The opposite is true if the number of principal components is higher than three: the model overfits the data. In order to study the effect of over and underfitting on the false alarm rate, every model is calculated for 1 to 14 principal components. The fifteenth component is omitted since there are no residuals in \( \text{SPE} \) when fifteen components are selected.

6.3.5 FALSE ALARM RATE RESULTS
The results for the false alarm rate have been presented in Figure 103. In this Figure, only the datasets with a number of 20, 50, 100, 500 and 2000 observations are presented. The resulting curves of the other datasets (30, 75, 250, 1000) are in-between the curves as shown in Figure 103:
The following conclusions can be drawn for the false alarm rate for the SPE chart (Figure 103a):

- The false alarm rate is too high for the smaller datasets \((K = 20 - 100)\).
- If the data is overfitted, the false alarm rate is too high. This is especially true for the smaller datasets \((K = 20 - 100)\). The residuals that result from the projection of a new independent sample will on average always be larger than the residuals of the NOC model (H.-J. Ramaker et al. [71]). This is true because the new sample did not take part in fitting the NOC samples. The difference in size between the residuals of the new sample and NOC samples increases when the data is overfitted. The chi-square distribution does not capture this as reflected in the false alarm rate. Overfitting the large datasets \((K = 500 - 2000)\) will not further increase the false alarm rate.
- For very large datasets \((K = 500 - 2000)\), the false alarm rate will be slightly higher than \(\alpha (0.05)\) in the case of overfitting the data, although not disturbingly.
- Underfitting is not a problem for reasonably sized datasets (\( K > 50 \))
- The false alarm rate is in perfect accordance with \( \alpha (0.05) \) if the sample size is large enough (\( K > 100 \)) and the model is chosen correctly (\( R = 3 \))

The false alarm rate calculated for the \( D \) chart leads to the following conclusion (see Figure 103b):

- Underfitting the data is not really a problem. In case of overfitting, the false alarm rate will always be too small. This problem is less pronounced for large datasets (\( K > 500 \)). While the residuals are on average larger for a new sample compared to the test set, the scores are on average smaller. This effect is reflected in the false alarm rate that is too small.
- The false alarm rate is in perfect accordance with \( \alpha (0.05) \) if the sample size is large enough (\( K > 100 \)) and the model is chosen correctly (\( R = 3 \))

6.3.5.1 Discussion about the false alarm rate results

The \( D \) statistic as defined by Nomikos and MacGregor is different from the Hotelling \( T^2 \) statistic. The difference is that the \( D \) statistic is based on scores obtained from a latent variable model while the \( T^2 \) statistic is based on the originally measured process variables. For manifest variables the distributions of calibration samples and test samples are the same. This is not true for the \( D \) statistic where the scores and residuals obtained from a component model are different for test samples and calibration samples. A direct generalization of Hotellings \( T^2 \) statistic to component models is not valid.

The Hotelling \( T^2 \) statistic uses the correlation between a set of manifest variables and these are stored in the covariance matrix \( \mathbf{B} \). The \( T^2 \) statistic is defined as follows, where \( \mathbf{x} \) is a measurement vector (\( J \times 1 \)) of \( J \) manifest variables:

\[
T^2 = (\mathbf{x} - \bar{x}) \mathbf{B}^{-1} (\mathbf{x} - \bar{x})
\]

Tracy et al (N.D. Tracy et al. [14]) derived exact distributions for \( T^2 \) statistic values for calibration samples used to calculate the mean and covariance matrix and for new samples. The \( T^2 \) values of the calibration samples follow a Bêta-distribution while the \( T^2 \) of the new samples follow an \( F \)-distribution. However, an important assumption is
that the calibration samples and the new samples come from the same multivariate normal distribution. This assumption is violated when a latent variable model is used.

If a latent variable model (e.g. PCA) is developed from the calibration data, the total variance of all $J$ process variables is distributed in a specific way over the scores. As an example, hundred PCA models are developed for hundred different sets of manifest variables (20 samples $\times$ 15 variables). The average variance of all scores (white bars) is plotted in Figure 104:

![Figure 104: Variance of the scores](image)

Hundred test sets (250 samples $\times$ 15 variables) obtained from the same distribution as the calibration sets are projected to the PCA models. The average variance of the test scores is plotted in the same figure (black bars). The first score value of the calibration set is much larger than the first score of the test set. Each sample in the calibration set tries to draw the PCA model and specifically the first principal component towards itself. Furthermore, the variance of the latter scores are low and go to zero. This is not the case for the test set scores. In this example, three scores were sufficient to describe the data. It is easy to imagine that the covariance matrix from the calibration scores does not represent the covariance of the test scores. Stated otherwise, the
expected value of the estimated covariance matrix of the calibrations scores ($\Sigma_{\text{train}}$) differs from its counterpart in the testset ($\Sigma_{\text{test}}$) where in both cases the expectations are over repeats of the calibration and test set. Wierda S.J. Wierda [119] already showed that the performance of the $T^2$ chart is very sensitive to violating the assumption that $\Sigma_{\text{true}} = \Sigma_{\text{test}}$.

In the same way as for the scores, the residuals of the calibration samples will be different from the test set samples. The residuals for the test set are expected to be higher because the test set did not take part in the fitting of the model in the data. For the same example set, the variance of the residuals is calculated and plotted in Figure 105:

![Figure 105: Variance of the residuals](image)

Clearly, the variance of the projected residuals (black bars) is higher compared to the variance of the model (white bars). This can be understood in the same way as explained above.
6.3.6 5. LEAVE-ONE-OUT METHOD

In order to overcome the difference in distribution between the calibration set and the test set, an additional test set should be used. This additional test set is projected on the calibration model and scores and residuals are obtained. The scores and residuals from this additional test set represent scores and residuals from new samples and therefore should be used to calculate the covariance matrix (for the $D$ statistic) and the control limits for $SPE$ statistic.

Unfortunately problems arise when the number of calibration samples is only small. In that case it is not possible to split the calibration samples over the calibration set and the additional test set. However, to simulate the additional test set, a leave one out projection procedure (D.J. Louwerse & A.K. Smilde [38]) is applied.

The leave-one-out method works as follows (H.-J. Ramaker et al. [25]). From the training set $X^{\text{training}}$ the first sample $k = 1$ is removed. A model $P_{k=1}$ is built for the remaining $K - 1$ samples. The left out sample is projected onto the model $P_{k=1}$ to find the scores $t_{k=1}$ and residuals $e_{k=1}$. Then, the next sample $(k = 2)$ is left from $X^{\text{training}}$ and in the same manner, the scores $t_{k=2}$ and residuals $e_{k=2}$ are calculated for the left out sample. This is repeated for all samples. The score matrix $\mathbf{T} = [t_{k=1} \ t_{k=2} \ \ldots \ t_{k=K}]$ and residual matrix $\mathbf{E} = [e_{k=1} \ e_{k=2} \ \ldots \ e_{k=K}]$ form the reference distributions for the $SPE$ and $D$ charts and are called the leave-one-out scores and residuals. The leave-one-out scores and residuals are better comparable to the scores and residuals of a completely new and independent sample since they are calculated in a similar manner. This is illustrated in Figure 104 and Figure 105 where the variance for the scores $\mathbf{T}$ and residuals $\mathbf{E}$ are presented (gray bars) using the same calibration and test set as discussed in Section 4.1. As a result of the leave-one-out procedure, the variance of the leave-one-out scores and residuals are better comparable to the monitored score and residuals.

6.3.6.1 Results for the leave-one-out method

The results found for the false alarm rate where the reference distributions from the control charts are based on the leave-one-out method are presented in Figure 106:
The following conclusion from the SPE chart based on leave-one-out residuals can be drawn (see Figure 106a):

- When overfitting the data, a far better false alarm rate is obtained as compared to the case with fitted residuals (see Figure 103a). Therefore the performance of the control chart based on the leave-one-out residuals is more robust against overfitting.
- The false alarm rate is slightly higher than \( \alpha (0.05) \) for the smaller datasets \((K = 20 - 50)\) in the case of underfitting. However, if a reasonable model is chosen \((2 < R < 10)\), the false alarm rate is satisfying.
- The false alarm rate is in perfect accordance with \( \alpha (0.05) \) for all sample sizes if the model is chosen correctly \((R = 3)\)
The last case describes the false alarm rate for the $D$ chart calculated in a leave-one-out manner (see Figure 106b):

- Underfitting the data gives satisfying results for the false alarm rate for large datasets ($K = 500\rightarrow 2000$) while the smaller datasets ($K = 20\rightarrow 100$) have a false alarm rate that is slightly higher than $\alpha (0.05)$. Therefore the leave-one-out method is only more robust against underfitting for the largest datasets ($K > 100$).
- The false alarm rate is in perfect accordance with $\alpha (0.05)$ if the sample size is large enough ($K > 500$) and the model is chosen correctly ($R = 3$)
- The problems associated to overfitting and underfitting small datasets ($K < 2000$) are not solved (compare with Figure 103b).

It was already explained that the generalization going from the Hotelling $T^2$ statistic to the $D$ statistic is problematic. These problems are not solved by using a leave-one-out method. Researchers in the field of MSPC are challenged to investigate alternative test statistics for the scores that have better statistical properties.

The results found for continuous processes can be extrapolated to batch processes. The sample size of NOC data for batch processes lies typically around $20\rightarrow 50$ batches. It was shown that such small datasets may suffer from poor false alarm rate properties. Overfitting this data will enlarge this problem of the false alarm rate even further. It is the experience of the authors that the false alarm rate for batch process monitoring models is on average too high. It is recommended to use a parsimonious model together with a proper leave-one-out procedure.

6.3.7 Conclusions

Various datasets have been constructed to examine the effect of over/underfitting to the false alarm rate. The results of these simulations lead to the following conclusions. First, the performance of the $SPE$ chart in terms of the false alarm rate is too high for small datasets ($K < 100$) in case of overfitting. This problem is substantially reduced by using leave-one-out residuals. Secondly, the false alarm rate for the $D$ chart is troublesome for small datasets ($K < 500$) if the data is overfitted. This effect is not
improved by using a proper leave-one-out procedure to calculate the scores. Finally, it is recommended to use a parsimonious model together with a proper leave-one-out.

The performance of the $D$ chart is troublesome. More research is required to investigate alternative test statistics for the scores that have better statistical properties.
6.4 Fault manifestation in the $D$ and $SPE$-chart for different batch monitoring strategies*

6.4.1 SUMMARY

The manifestation of process faults in the control charts is studied for three methods of batch process monitoring. These methods are discussed in Chapter 4.3. For batch process monitoring, two test statistics are monitored in-control charts: the $SPE$ and $D$-statistic. The diagnosing capabilities of the $SPE$ and $D$-statistics are established by a mathematical analysis of their underlying structure. This analysis shows that the diagnosing capabilities are severely hampered by the problem of embedded error, always present in projection methods (like PCA). These problems are, to some extent, present in all the process monitoring models considered, i.e. global, local and time evolving models.

6.4.2 INTRODUCTION

Many products originating from the chemical, pharmaceutical and food industry are produced in a batch-wise manner. The flexibility of batch processes can be regarded as an important advantage as compared to e.g. continuous processes. For this reason, the usage of batch processes is expected to increase in the coming years. Batch processes can be characterized by their finite duration, non-linear behavior of the process variables and high conversions. Most batch processes are recipe driven and this recipe is often based on expert knowledge. Monitoring of batch processes is preferable for several reasons. Monitoring schemes may lead to e.g. shorter batch durations, improved product quality, better process understanding, more consistent batches, reduced waste streams or improved safety circumstances. One way of monitoring batches is to develop fundamental models and use these models to predict the ongoing of a batch process. However, the chemistry of most batch processes such as biochemical or polymerization reactions is very complex. Therefore, it is not

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possible to construct a detailed model because too limited knowledge is available. Furthermore, model development of such complex processes is time-consuming.

One of the alternatives is to measure and collect important process variables during a batch run. These measurements from as many batches as possible are stored in a well organized database. Such a collection of data contains valuable information about the batch process under consideration. Multivariate statistical methods are well suited to deal with this type of data. These methods offer the possibility to capture the underlying phenomena present in the data in terms of easily interpretable latent variables.

The application of multivariate statistical methods to batch processes is referred to as statistical batch process monitoring. The concept of statistical batch process monitoring was introduced in the mid-ninety's by P. Nomikos & J.F. MacGregor [6]. Many applications and extensions of statistical batch process monitoring have followed since (S. Rannar et al. [31], S. Wold et al. [53], D. Neogi & C.E. Schlags [44], D.J. Louwerse & A.K. Smilde [38], J.A. Westerhuis et al. [42], A.K. Smilde & H.A.L. Kiers [39]). The statistical performance of a batch is monitored in two multivariate control charts, namely the SPE and D-chart.

This chapter discusses alternative models, which can be used in statistical batch process monitoring. From these models, control charts are derived. This series focuses on the fault detection (Part I) and the diagnostic capabilities (Part II) of these charts. The goal of this chapter is twofold. First, the mechanics and non-complementarity of the control charts are shown. This is done by attempting to construct process faults that only manifest themselves in one of the two charts. Secondly, the diagnosing characteristics of typical faults are shown for all three models.

The chapter is organised as follows: in Section 6.4.3 a description of the SPE and D-statistic is given. In Section 6.4.3.1, Section 6.4.4 and Section 6.4.5 the mechanistic of the SPE and D-statistic are thoroughly discussed. Section 6.4.7 explains the problem of the embedded error. Section 6.4.9 describe the simulation of a series of faults. These faulty batches are then monitored using three models for statistical batch process monitoring. The diagnosing capability for each model is discussed.
6.4.3 A QUALITATIVE DESCRIPTION OF THE D AND SPE-STATISTIC

A description of the global, evolving and local model is given section 4.3. Section 4.3 describes the performance of these three models in terms of the overall type I error (OT1) and action signal time (AST). The OT1 is an indicator for the number of false alarms where the AST is a measure of the speed of detecting process upsets. It was concluded from Part I that in terms of detection all three models gave satisfying results. Another issue besides fault detection is the diagnosis of a fault. This part deals with the issue of the diagnostic properties of the three models. The distinction between the failure of a sensor or the drift of a process might be beneficial. The broken sensor might be easily repaired and there is no need to stop the reaction.

In part I it was explained that the statistical behavior of a batch is monitored in the SPE and D-chart. These two charts are the tools for diagnosing a fault. If e.g. a fault manifests itself only in the SPE-chart, certain characteristics can be described to this process fault. The same is true if a process fault manifest itself only in the D-chart. The construction of the SPE and D-statistic is explained briefly in the following. This has already been explained in part I, however, the mechanics are explained in more detail now. Also the physical representation of the SPE and D-statistic is discussed.

6.4.3.1 Mechanics of the SPE and D-statistic

The mechanics of the SPE and D-statistic is explained using a simple example. A set of 10 samples, denoted as $X$ (10x2), is used to perform a principal component analysis (PCA) on. First, the columns of $X$ are mean centered and scaled to unit variance. The pre-processed data is represented by the diamonds in Figure 107.
A PCA model with only one component is chosen to describe the data. The PCA model that describes \( \mathbf{X} \) looks like:

\[
\mathbf{X} = \mathbf{t} \mathbf{p}^T + \mathbf{E}
\]  \hspace{1cm} (128)

The scores are given by \( \mathbf{t} \) \((10\times1)\), the model loadings by \( \mathbf{p} \) \((2\times1)\) and the residuals by \( \mathbf{E} \) \((10\times2)\). The graphical interpretation of the PCA model is given in Figure 107. The samples of the matrix \( \mathbf{X} \) are orthogonally projected onto the line represented by the loadings. As can be seen from Figure 107, these projections onto this line are given by the dots. In this picture, two important distances can be distinguished.

First, consider the vector that connects e.g. sample 1 to the line. This connection is described by the vector given by the first row of the residual matrix \( \mathbf{E} \). The sum of the squared elements of this vector makes up the \( \text{SPE} \)-statistic for this sample:
Secondly, consider the distance from the projection of the sample (given by the dots) to the centre of the line given by [0,0]. The centre of the model represents the average of all samples. The coordinates of e.g. the first sample on the line are given by the first row of the score vector \( t \). The sum of the squared elements of this row of \( t \) scaled by the covariance matrix is referred to as the \( D \)-statistic. This covariance matrix is calculated from the scores. In this example, the covariance matrix of the scores \( t \) will result in a scalar \( s^2 \) because \( t \) is a vector. The equation to calculate the \( D \)-statistic that represents the distance from the projected sample towards the model centre is given by:

\[
D_i = t_i \cdot s^2 \cdot t_i
\]

Summarizing, the samples can be described by scores and residuals. The residuals are a measure of the distance towards the model where the model is described by the loadings. This is expressed in the \( SPE \)-statistic. The scores are a measure of the distance within the model represented by the \( D \)-statistic. That is, the distance from the orthogonally projected sample towards the centre of the model. A sample can be assigned as non-representative if the \( SPE \) or \( D \)-statistic or both statistics are above control limits.

6.4.3.2 Relationship between real process disturbances and the \( SPE \) and \( D \)-statistic

In statistical batch process monitoring, fault detection plays an important role. The result of the projection of a sample is expressed in terms of the \( SPE \) and \( D \)-statistic. The question therefore is: what kind of relation is there between a real process fault and the result of this fault in the \( SPE \) and \( D \)-statistic? To answer this question, the process measurements are categorised in two groups: i) faults that break the correlation structure or ii) faults that obey the correlation structure but have a more than normal variation. As an example, think of the following situation. The temperature \( T_{m} \) and pressure \( P_{m} \) are measured in the inner tube of a car tyre, as well as the ambient temperature \( T_{a} \). Now, as \( T_{a} \) increases because of the sunny
weather, so will $T_{yn}$ and $P_{yn}$. Thus, the process variables are correlated and behave according to simple physical laws. This is depicted in Figure 108:

The white bars represent situations where the process behaves under normal conditions. The black bars show the reaction of $T_{yn}$ and $P_{yn}$ when $T_s$ is changing. This is according to the correlation between the process variables. Besides, the variation of the increment of the process variables is considered as acceptable.

The first group of faults is the following. In extreme situations where the surrounding temperature may become very high (e.g. the car has been parked in the burning sun), the pressure and temperature in the inner tube might become dangerously high. Although the process variables behave according to the law of physics, the variation of the process variables is more intense than under normal circumstances. This is illustrated in Figure 108 as the patterned black bar. Such a fault where the variation of the process variables is abnormal high but the correlation between the process variables remains intact will be denoted as intensified correlation throughout this chapter.

The other group of faults is of the following. Suppose the temperature in the inner tube $T_{yn}$ increases as a result of $T_s$, but because the valve of the car tube is leaking, the pressure $P_{yn}$ remains constant. This is not according to the correlation between the process variables, and therefore the correlation is broken. This can be seen from Figure 109. Such disturbances will be denoted as breakage of the correlation.
Often heard remarks in the literature (R. Dunia & S.J. Qin [114], S. Albert & R.D. Kinley [43], J.V. Kresta et al. [10], P. Nomikos & J.F. MacGregor [6]) are that abnormal variation that still obeys the correlation structure of the process variables is described by the scores ($D$-statistic) while new events not present in the NOC data will represent itself in the residuals ($SPE$-statistic). It is believed that an intensified correlation (e.g. process shifts) is detected in the $D$-statistic while the breakage of the correlation (e.g. sensor failure) is detected in the $SPE$-statistic. This implies that the control charts are complementary. That is, certain type of process faults can be detected in either the $SPE$-chart of $D$-chart. Assessing this complementarity gives better understanding how the $SPE$ and $D$-chart work together to detect faulty batches. Therefore, it is investigated what type of faults form the base of the complementarity. That is, faults are constructed that generate a pure $SPE$-signal or a pure $D$-signal. These faults can be regarded as extreme faults. If both type of faults cannot be constructed for a model, the complementarity of the control charts does not exist. It will be shown that for some models pure $D$- or $SPE$-signals cannot be constructed. Furthermore, the issue of embedded error is discussed. Due to the embedded error, most faults will be distributed over both charts.

6.4.4 Construction of Pure $D$-Faults

The performance of monitoring charts based on the $D$-statistic are troublesome (E.N.M. Van Sprang et al. [41]). The reason for this poor performance can be understood from the nature of a $D$-fault. This section describes the construction of a fault that only affects the scores (and thus $D$-statistic) in an on-line monitoring scheme.
Consider the measurement of a pure sample $x^*$ depicted as the black dot in Figure 110:

In the ongoing of this chapter a sample $x^*$ refers to the ideal measurements of the process variables from a batch process. This sample is free from any experimental noise and can be written exactly according to the model $p$. Therefore this sample can be expressed in the coordinate system described by the latent variables as:

$$x^* = p t^*$$ \hfill (131)

There are no residuals because $x^*$ can be written exactly according to the model. The scores $t^*$ found for $x^*$ are the true scores \((t^* = p' x^* = (p' p)t^* = t^*)\). However, in a real situation the pure sample $x^*$ is affected by process faults denoted as $e$. A real sample $x$ affected by process fault is written as:

$$x = x^* + e = p t^* + e$$ \hfill (132)

In this Section, the process fault $e$ is chosen in such a way that it only affect the scores. This requires a specific direction of the process fault $e$. It can be seen from
Figure 110 that the direction of the process fault is given by $\varepsilon^D$. The uppercase $D$ refers to the case where the process fault represents a pure $D$-fault. It can be seen that $\varepsilon^D$ moves along the line given by $p$. This process fault $x^D (= x^* + \varepsilon^D)$ only affects $D$, and thus, represents a pure $D$-fault and generates a pure $D$-signal. The required behaviour of the process variables to generate a pure $D$-signal is given by $x_1^D$ and $x_2^D$ from Figure 7. Only those combinations of $x_1^D$ and $x_2^D$ that sum up to $x^D$ will result in a pure $D$-fault.

For practical reasons, it was assumed in the preceding that $x^*$ could be written exactly according to the model. Therefore the residuals $e^*$, describing random error sources such as measurement error, were zero. This is not a very realistic assumption, therefore $e^*$ is non-zero in the remaining of this chapter. This has the following consequences: compared to the situation where the residuals $e^*$ are zero, the vector $e^D$ is not lying on the line described by $p$ anymore. Instead, it moves somewhere in the space described by $x$ in a direction perpendicular to the model $p$. This situation is depicted in Figure 111:

![Graphical interpretation of a pure D-signal with nonzero residuals.](image)
In this picture, \( \mathbf{x}' ( = \mathbf{p}' + \mathbf{e}' ) \) is represented by the black dot. In this case the nature of \( \mathbf{x}' \) is different compared to Equation 131 where the residuals \( \mathbf{e}' \) are zero. The vector \( \mathbf{x}^D \left( = \mathbf{x}' + \mathbf{e}^D = \mathbf{p}' + \mathbf{e}' + \mathbf{e}^D \right) \) represents the measurement of process variables that contain a process fault \( \mathbf{e}^D \).

The extracted residuals \( \mathbf{e}^0 \) for \( \mathbf{x}' \) are given by:

\[
\mathbf{e}^0 = \mathbf{x}' - \hat{\mathbf{x}}' = (\mathbf{p}' + \mathbf{e}^* ) - \mathbf{p}' = \mathbf{e}^*
\]

For a pure \( D \)-fault, the extracted residuals \( \mathbf{e}^0 \) resulting from the projection of \( \mathbf{x}' \) or \( \mathbf{x}^D \) should be the same if the process fault represents \( \mathbf{e}^D \) where the extracted residuals for \( \mathbf{x}^D \) are given by

\[
\mathbf{e}^0 = \mathbf{x}^D - \hat{\mathbf{x}}^D = (\mathbf{p}' + \mathbf{e}^* + \mathbf{e}^D ) - (\mathbf{p}' + \mathbf{e}^D ) = \mathbf{e}^*
\]

The extracted residuals are the same for both cases. This should hold for every pure \( D \)-signal.

From Figure 111, an important remark can be made. Suppose that the process fault \( \mathbf{e}^D \) was of opposite direction and moved towards the origin of the line. This will still result in a pure \( D \)-signal, but the value for the \( D \)-statistic will decrease. Such a process fault will never lead to detection in conventional \( D \)-charts. N.D. Tracy et al. [14] mentioned to use a control chart with upper and lower control limits. This, however, suggests a two sided test with null hypothesis of the form \( D = 0 \) on target (target > 0) and alternative hypothesis \( D \neq 0 \) on target. A \( D \)-value of zero might then indicate wrong behaviour, but this contradicts intuition since the best batch should have a \( D \)-value of zero (S.J. Wierda [129]). This suggestion is therefore not followed.

### 6.4.4.1 General construction of a pure \( D \)-fault:

This section describes the general way to construct a sample \( \mathbf{x}^D \). Therefore the theory and figures presented in the ongoing of this chapter are applicable to higher dimensional problems. The machinery to construct such a sample can be easily extrapolated to the more specific case where a sample \( \mathbf{x}^D \) is constructed using local,
evolving or global models. The general equation to construct such a sample $x^D$ reads as:

$$x^D = \frac{Pt^* + e^* + \varepsilon^D}{x^*}$$  \hspace{1cm} (135)$$

The process fault $\varepsilon^D$ can be written according to:

$$\varepsilon^D = \delta \cdot Pt^* \rightarrow x^D = Pt^* + e^* + \delta \cdot Pt^* = (1 + \delta)Pt^* + e^*$$  \hspace{1cm} (136)$$

Equation 136 means that $\varepsilon^D$ moves along or parallel to the line in Figure 110. The positive value of $\delta$ will increase the $D$-statistic and therefore moves away from the model centre. This process fault describes the situation where the process variables behave exactly according to the correlation. In other words, the projection of $x^D$ or $x^*$ results in exactly the same residuals $e^*$ according to Equations 133 and 134. This was referred to as an intensified correlation. The construction of such process faults that generate a pure $D$-fault in an on-line manner using local, evolving and global models will be discussed in the following sections.

6.4.4.2 Construction of a pure $D$-fault for the local model

For a local model, $K$ different models are constructed at every time interval. These models are denoted as $\hat{P}_k$ ($k = 1, \ldots, K$). The measurement of the process variables that is taken at every time interval is denoted as $\hat{x}_k^*$. The general Equations 135 and 136 applied to a local model to construct a pure $D$-signal at time interval $k$ are as follows:

$$\tilde{\varepsilon}_k^D = \delta \cdot \hat{P}_k \tilde{t}_k^* \rightarrow \tilde{x}_k^D = (1 + \delta) \cdot \hat{P}_k \tilde{t}_k^* + \tilde{\varepsilon}_k^*$$  \hspace{1cm} (137)$$

An example of the process variables that represent this fault is given in Figure 112.
In this figure, the white bars represent the measurements of a new independent batch without a process fault ($x^*_{k}$) and therefore represents NOC conditions. At time interval $k + 3$, the process fault appears. This is represented by the black bars. It can be seen that at this time interval the process variables react according to the correlation. It must be stressed that the situation shown in Figure 112 represents very specific and unique process conditions. Only the slightest deviation from this situation will distribute the process fault over both the scores and residuals and the instantaneous correlation is broken. In this case, both the SPE and D-statistic are affected. This will be further discussed in the Section about the embedded error.

It can be concluded that it is possible to construct a process fault that results in a pure $D$-signal in an on-line monitoring scheme for the local model. For this reason it is also possible that real process faults can be detected exclusively in the $D$-chart using a local model.
6.4.4.3 Construction of a pure D-fault for the evolving model

This section describes the construction of a fault that is detected exclusively in the $D$-chart for the time evolving model. Let the batch operated at NOC be depicted in Figure 113a as the white bars.

![Figure 113](image)

*Overall correlation breakage due to the evolving model.*

This batch has a duration of seven time intervals. Suppose that the first measurement of the process variables is carried out and the future measurements of the batch are unknown. This measurement corresponds to the white bars that represent NOC conditions at time interval $k = 1$ and is denoted as $\bar{x}_1^*$. This part of the batch is modelled as follows:

$$\bar{x}_1^* = \bar{P}_1 \bar{t}_1^* + \bar{e}_1^*$$

(138)
Now, let the batch resulting in a pure $D$-signal after projection at the first time interval be denoted as $\tilde{x}_1^D$. This fault is constructed according to the following equation (see general Equation 136):

$$\tilde{x}_1^D = (\delta + 1) \cdot \tilde{P}_1 \tilde{t}_1^* + \tilde{e}_1^*$$  \hspace{1cm} (139)

The process fault is depicted as the black bars in Figure 113a. The evolving model at the first time interval equals a local model for the same time interval. Therefore, considering the first time interval, the same conclusions can be drawn as for a local model.

Now consider the measurement at the second time interval of the batch. The mathematical construction of a pure $D$-signal is discussed first. Let the vector $\tilde{x}_2^*$ consist of the white bars measured at time interval $k = 1$ and $k = 2$ from Figure 113a. This vector can be partitioned and is given by:

$$\tilde{x}_2^* = \begin{bmatrix} \tilde{x}_{21}^* \\ \tilde{x}_{22}^* \end{bmatrix}_{(2 \times 1)}$$  \hspace{1cm} (140)

where the first part corresponds to time interval one and the second part to time interval two. The fault that will result in a pure $D$-signal is given by $\tilde{x}_2^D$ and is calculated as:

$$\tilde{x}_2^D = (\delta + 1) \cdot \tilde{P}_{21} \tilde{t}_2^* + \tilde{e}_2^* = (\delta + 1) \cdot \begin{bmatrix} \tilde{P}_{21} \\ \tilde{P}_{22} \end{bmatrix}_{(j \times k)} \begin{bmatrix} \tilde{t}_2^* \\ \tilde{e}_2^* \end{bmatrix}_{(j \times 1)} = \begin{bmatrix} \tilde{x}_{21}^D \\ \tilde{x}_{22}^D \end{bmatrix}_{(j \times 1)}$$  \hspace{1cm} (141)

The only fault that results in a pure $D$-signal is represented by the grey bars in Figure 113b. Notice that the correlation is intensified over both time intervals in order to generate a pure $D$-signal. This is referred to as an intensification of the overall correlation.
However, this pure mathematical construction cannot be realized because of the following. The mathematical part of $\tilde{x}_2^D$ corresponding to the first time interval ($\tilde{x}_{21}^D$) will be replaced with the real fault given by $\tilde{x}_1^D$. These parts are different as can be seen from the following:

$$\tilde{x}_{21}^D \neq \tilde{x}_1^D \quad \text{because} \quad (\delta + 1) \cdot \tilde{t}_2^P \tilde{P}_{21}^P + \tilde{e}_{21}^* \neq (\delta + 1) \cdot \tilde{t}_1^P \tilde{P}_1^P + \tilde{e}_1^* \quad (142)$$

since the loadings $\tilde{P}_{21}^P$ and $\tilde{P}_1^P$ are different. The vector that results is depicted in Figure 113c. In this picture $\tilde{x}_{21}^D$ is replaced with $\tilde{x}_1^D$. If the vector $\tilde{x}_2^D$ is projected, the extracted residuals found for $\tilde{x}_2^D$ will not be the same as $\tilde{x}_1$ according to Equation 133 and 134. This is required for a pure $D$-signal. The overall correlation pattern is broken instead. The same arguments hold for the following time points.

It was shown that a batch generating a pure $D$-signal couldn't be constructed for evolving models in an on-line monitoring scheme except for the first time interval. The overall correlation is always broken while it can still be true that the instantaneous correlation is intensified. The only exception to this is the first time interval of a batch where indeed it is possible to intensify the correlation in such a way that a pure $D$-signal is generated. From a practical point of view the first measurement can be used to check the initial conditions of a batch. A shift in these conditions might lead to an intensified correlation.

6.4.4.4 Construction of a pure $D$-fault for the global model

This section will show that for on-line batch monitoring the overall correlation modeled by a global model will always be broken. This situation is easier to explain as compared to the evolving models. Consider the normal batch $x^*$ represented as white bars from Figure 114.
Assume this figure represents the whole batch run. To construct a faulty batch $\bar{x}_K^D$ in such a way that only the $D$-chart is affected because the correlation is intensified, only one situation is possible. This situation is given by the following equation:

$$\bar{x}_K^D = (\delta + 1) \cdot \bar{l}_k^* + \bar{e}_k^*$$  \hspace{1cm} (143)

The batch $\bar{x}_K^D$ has been illustrated in Figure 114a. It is required that the overall correlation is intensified during the entire batch run from time point one until seven. Projection of this batch indeed will lead to the desired pure $D$-fault. Projection of $\bar{x}_K^D$ resembles a post-batch analysis, and for such an analysis it is possible to detect a fault only in the $D$-chart. Another situation of a post batch analysis is depicted in Figure 114c. This picture represents a completed batch. The instantaneous correlation has been changed from $k = 4$ until the end of the batch. The post batch projection of this batch will break the correlation structure, because the first part of the batch did not take part in the intensification of the correlation. This batch does not resemble the
required correlation intensification from Figure 114a. Deviations from this ideal correlation intensification will result in a breakage of the overall correlation.

The situation where the correlation indeed is intensified but the batch has not finished yet is illustrated in Figure 114b. The batch has evolved until $k + 3$. The instantaneous correlation is intensified on the time intervals 1, 2, 3. This is illustrated as black bars. Because of the filling in procedure, the process variables at $k = 4,5,6$ and 7 resemble the last measurement from $k = 3$. These measurements are given in Figure 114b as grey bars. Clearly, this batch deviates from the batch depicted in Figure 114a although the first measurement $k = 1$ until $k = 3$ fulfill the requirements. The required overall correlation intensification is disturbed because of the filling in procedure. This disturbance will be higher if the batch has just started because relatively large future parts of the batch need to be filled in. In other words, the overall correlation will always be broken using the global model for on-line batch monitoring. This is not only true for the current deviations approach but for any filling-in procedure.

It can be stated that pure $D$-signals cannot be constructed for global models for on-line monitoring. This statement holds for every filling in method. The overall correlation is always broken when global models are used for on-line batch monitoring.

6.4.5 Construction of Pure $SPE$-Faults

This section describes the construction of process faults that generate a pure $SPE$-signal in an on-line monitoring scheme. The line of reasoning will be the same as for the construction of faults that generate pure $D$-signals. Like a pure $D$-signal, the direction of the vector $s$ that describes a process fault needs to be very specific in order to only affect the residuals. Assume that $x^*$ can be written exactly according to the model. To visualize this situation, the residuals $e^*$, describing random error sources such as measurement error, are zero for the moment. The vector of interest is denoted as $e^{SPE}$ and is illustrated in Figure 115:
Figure 115

Graphical interpretation of a pure SPE-signal.

The superscript SPE refers to the case where the process fault represents a pure SPE-fault. It can be seen that \( e^{SPE} \) moves perpendicular from the model given by \( p \). The projection of the sample \( x^{SPE} \) leads to unaffected scores compared to the scores calculated for the sample \( x^* \). The uppercase SPE refers to a vector or number that will generate a pure SPE-signal. The residuals after projection \( (e^0) \) of \( x^{SPE} \) equal the process fault \( (e^{SPE}) \). In this situation, the SPE is based upon a completely projected process fault.

The necessary behaviour of the process variables to generate a pure SPE-fault is given by \( x_1^{SPE} \) and \( x_2^{SPE} \). If \( x_1^{SPE} \) and \( x_2^{SPE} \) do not describe a situation such that \( x^{SPE} \) lies somewhere in the line of \( e^{SPE} \), the scores will be affected and the residuals are based upon the part of \( e^{SPE} \) that is not projected. The construction of a vector \( x^{SPE} \) will be discussed in the following using local, global and evolving models.
6.4.5.1 General construction of a pure SPE-fault:

Let the sample $\mathbf{x}' = (\mathbf{p}^* + \mathbf{e}')$ represents NOC conditions. For the same reasons as for the pure $D$-signal, the residuals $\mathbf{e}'$ are now chosen to be non-zero. The scores for $\mathbf{x}^{\text{SPE}}$ are calculated according to:

$$
\mathbf{t}^{\text{SPE}} = \mathbf{P} \mathbf{x}^{\text{SPE}} = \mathbf{P} (\mathbf{x}' + \mathbf{e}' + \mathbf{e}^{\text{SPE}}) = \mathbf{P} \mathbf{x}' + \mathbf{P} \mathbf{e}' + \mathbf{P} \mathbf{e}^{\text{SPE}}.
$$

(144)

Since these scores $\mathbf{t}^{\text{SPE}}$ are not allowed to be affected compared to the scores $\mathbf{t}'$, it should hold that $\mathbf{t}^{\text{SPE}} = \mathbf{t}'$. Therefore the following should hold:

$$
\mathbf{t}' = \mathbf{t}^{\text{SPE}} \rightarrow \mathbf{P} \mathbf{x}' + \mathbf{P} \mathbf{e}' = \mathbf{P} \mathbf{x}' + \mathbf{P} \mathbf{e}' + \mathbf{P} \mathbf{e}^{\text{SPE}} = \mathbf{t}' + 0 + \mathbf{P} \mathbf{e}^{\text{SPE}} \rightarrow \mathbf{P} \mathbf{e}^{\text{SPE}} = 0.
$$

(145)

Since the residuals $\mathbf{e}'$ are perpendicular to the loadings $\mathbf{P}'$, the term $\mathbf{P}' \mathbf{e}'$ equals zero. Equation 145 holds if the vector $\mathbf{e}^{\text{SPE}}$ lies in the null-space of the loadings $\mathbf{P}'$ because

$$
\mathbf{N} (\mathbf{P}') = \left\{ \mathbf{e}^{\text{SPE}} : \mathbf{P} \mathbf{e}^{\text{SPE}} = 0, \mathbf{e}^{\text{SPE}} \in \mathbb{R}^r \right\}.
$$

(146)

The vector $\mathbf{e}^{\text{SPE}}$ is constructed as follows. Consider the singular value decomposition of some matrix $\mathbf{X}$ that can be written according to:

$$
\mathbf{X} = \mathbf{U} \mathbf{S} \mathbf{V} = \mathbf{U}_1 \mathbf{S}_1 \mathbf{V}_1 + \mathbf{U}_2 \mathbf{S}_2 \mathbf{V}_2 = \mathbf{T} \mathbf{P} + \mathbf{E}.
$$

(147)

where

$$
\mathbf{U} = [\mathbf{U}_1 : \mathbf{U}_2], \mathbf{S} = \begin{bmatrix} \mathbf{S}_1 & 0 \\ 0 & \mathbf{S}_1 \end{bmatrix}, \mathbf{V} = [\mathbf{V}_1 : \mathbf{V}_2].
$$

(148)

Thus,

$$
\mathbf{P}' = \mathbf{V}_1 \rightarrow \mathbf{P} = \mathbf{V}_1.
$$

(149)

Since $\mathbf{V}$ is an orthogonal matrix it holds that $\mathbf{V}_1 \mathbf{V}_2 = 0$. Hence, $\mathbf{V}_2$ is a basis for the null space of $\mathbf{P}'$ and $\mathbf{e}^{\text{SPE}}$ can be constructed using that basis.
6.4.5.2 Construction of a pure SPE-fault for the local model

The construction of a pure SPE-fault for a local model is fairly easy. First, the null space of the loadings \( \hat{P}_k \) is calculated at time interval \( k \). An arbitrary combination of the columns of the null space is taken to be the vector \( \hat{e}_k^{\text{SPE}} \). The measurement that describes the pure SPE-signal is like:

\[
\hat{x}_k^{\text{SPE}} = \hat{x}_k^{*} + \hat{e}_k^{*} + \hat{e}_k^{\text{SPE}}
\]

The scores at time interval \( k \) are calculated according to:

\[
\hat{t}_k^{\text{SPE}} = \hat{P}_k' \hat{x}_k^{\text{SPE}} = \hat{P}_k' \left( \hat{x}_k^{*} + \hat{e}_k^{*} + \hat{e}_k^{\text{SPE}} \right) = \\
\hat{P}_k^{*} \hat{x}_k^{*} + 0 + 0 = \hat{t}_k^{*}
\]

where the residuals \( \hat{e}_k^* \) are nonzero. The projection of the residuals \( \hat{e}_k^* \) on the model \( \hat{P}_k \) is zero due to the model and the projection of the process fault \( \hat{e}_k^{\text{SPE}} \) is zero by construction. Obviously the scores \( \hat{t}_k^{\text{SPE}} \) are not affected and equal the pure scores \( \hat{t}_k^{*} \). The extracted error \( \hat{e}_k^0 \) is calculated as:

\[
\hat{e}_k^0 = \hat{x}_k^{\text{SPE}} - \hat{x}_k = \left( \hat{x}_k^{*} + \hat{e}_k^{*} + \hat{e}_k^{\text{SPE}} \right) - \hat{P}_k \hat{t}_k = \\
\left( \hat{x}_k^{*} + \hat{e}_k^{*} + \hat{e}_k^{\text{SPE}} \right) - \hat{P}_k \left( \hat{P}_k' \left( \hat{x}_k^{*} + \hat{e}_k^{*} + \hat{e}_k^{\text{SPE}} \right) \right) = \\
\left( \hat{x}_k^{*} + \hat{e}_k^{*} + \hat{e}_k^{\text{SPE}} \right) - \hat{P}_k \left( \hat{t}_k^{*} + 0 + 0 \right) = \left( \hat{x}_k^{*} + \hat{e}_k^{*} + \hat{e}_k^{\text{SPE}} \right) - \hat{t}_k^{*} - \hat{e}_k^{*} + \hat{e}_k^{\text{SPE}}
\]

This pure SPE-signal can be constructed at every time interval. Since the construction of such a fault is possible, it could also happen in a real life process. The process variables describing this fault break the correlation in a very specific way. The likelihood of meeting these process conditions in a real life situation can be considered as very low.

6.4.5.3 Construction of a pure SPE-fault for the evolving model

The SPE-statistic, using an evolving model, calculated at time interval \( k \) is based upon the current part of the residuals \( e_k^0 \) (P. Nomikos & J.F. MacGregor [6]). For this reason, only the null space of the current part of the loadings \( \hat{P}_k' \) needs to be calculated. The current part \( \langle \text{cp} \rangle \) of e.g. the loadings \( \hat{P}_k' \) is denoted as \( \hat{P}_k^{\text{cp}} \) while the
past part (pp) is referred to as $\tilde{P}_k^{pp}$. Thus the matrix $\tilde{P}_k^*$ can be partitioned according to:

$$
\tilde{P}_k^* = \begin{bmatrix}
\tilde{P}_k^{pp} \\
\tilde{P}_k^{pp'} \\
\end{bmatrix}
$$

(153)

Consider the batch $\tilde{x}_k$ ($J \times 1$) at time interval $k$. The current part $\tilde{x}_k^{sp}$ ($J \times 1$) of this batch is replaced with:

$$
\tilde{x}_k^{sp} = \tilde{x}_k^{cp} + \tilde{e}_k^{cp} + \tilde{e}_k^{sp}.
$$

(154)

where the vector $\tilde{e}_k^{sp}$ ($J \times 1$) is constructed from the null space of the loadings $\tilde{P}_k^{pp'}$. The batch where the current part has been replaced according to Equation 154 will be referred to as $\tilde{x}_k^{sp}$ ($J \times 1$). The scores $\tilde{t}_k^{sp}$ for this particular batch are calculated according to:

$$
\tilde{t}_k^{sp} = \tilde{P}_k^{pp'} \tilde{x}_k^{sp} = \begin{bmatrix}
\tilde{P}_k^{pp} \\
\tilde{P}_k^{pp'} \\
\end{bmatrix} \tilde{x}_k^{sp} = \\
\tilde{P}_k^{pp'} \tilde{x}_k^{pp*} + \tilde{P}_k^{pp'} \tilde{x}_k^{cp*} + \tilde{P}_k^{pp'} \tilde{x}_k^{sp*}.
$$

(155)

where the projection of residuals $\tilde{e}_k^{pp*}$ and $\tilde{e}_k^{cp*}$ are zero due to the model properties and the process fault $\tilde{e}_k^{sp*}$ is zero by construction. It can be seen from Equation 155 that the scores $\tilde{t}_k^{sp}$ are equal to $\tilde{t}_k^*$ and therefore the scores are not affected by the process fault $\tilde{e}_k^{sp*}$. The extracted error and thus $SPE$-statistic at time interval $k$ is based upon the current part of $\tilde{x}_k^{sp}$.
It can be seen that the extracted residuals are based upon the complete projection of the process fault $\bar{e}_{SP}^k$ since they are in the same direction. Then, the extracted residuals are used to calculate the $SPE$-statistic. The construction of pure $SPE$-signals in an on-line monitoring scheme can be repeated for every time interval using an evolving model. Since a pure $SPE$-signal is possible by construction, it could also happen in practice. However, like the pure $SPE$-signal constructed for a local model, the process variables require a very specific breakage of the correlation. It is unlikely that the process conditions will be exactly similar to this required behaviour of the process variables.

6.4.5.4 Construction of a pure SPE-fault for the global model

The main difference between the global model on the one hand and the local and evolving model on the other hand is that for the global model the future part of the batch needs to be estimated. The filling-in procedure using the current deviations approach makes it impossible to construct a pure $SPE$-signal. It will be sufficient to show that the scores are affected. The loadings $\hat{P}$ and the batch $\hat{x}_k$ of a global model can be partitioned in three parts: past part (pp), current part (cp) and a future part (fp). The current part of the batch $\hat{x}_k$ is replaced according to the following equation:
Statistical batch process monitoring

\[
\frac{X_{k}^{\text{P}}}{j=1} = \tilde{X}_{k}^{\text{P}} + \tilde{e}_{k}^{\text{P}} + \tilde{e}_{k}^{\text{SP}}
\]  \hspace{1cm} (157)

Here, the process fault given by \(\tilde{e}_{k}^{\text{SP}}\) \((j=1)\) is constructed from the null space of the loadings \(\tilde{P}^{\text{op}}(RxJ)\). The batch that is to be projected is given by \(\tilde{X}_{k}^{\text{SP}}\) \((JK\times1)\) where the current part of this batch is replaced according to equation 157. The future part \(\tilde{X}_{k}^{\text{SP}}(j=1)\times1\) of this batch is estimated with the current deviations approach:

\[
\frac{X_{k}^{\text{SP}}}{j=1} = \left[ \begin{array}{c}
X_{k}^{\text{SP}} \\
\vdots \\
X_{k}^{\text{SP}} 
\end{array} \right]  \hspace{1cm} (158)
\]

The scores at time interval \(k\) are calculated according to:

\[
\begin{align*}
\tilde{r}_{k}^{\text{SPE}} &= \tilde{P}^{\text{op}} \tilde{X}_{k}^{\text{SP}} \\
\tilde{P}_{\text{op}} X_{k}^{\text{SP}} + \tilde{P}_{\text{op}} X_{k}^{\text{SPE}} + \tilde{P}_{\text{op}} \tilde{X}_{k}^{\text{SPE}} &= \\
\tilde{P}_{\text{op}}(\tilde{X}^{\text{op}} + \tilde{e}_{k}^{\text{op}}) + \tilde{P}^{\text{op}}(\tilde{X}^{\text{op}} + \tilde{e}_{k}^{\text{op}} + \tilde{e}_{k}^{\text{SPE}}) + \tilde{P}_{\text{op}} \tilde{P}_{\text{op}} &= \\
\tilde{P}_{\text{op}} \tilde{X}^{\text{op}} + 0 + \tilde{P}_{\text{op}}(\tilde{X}^{\text{op}} + \tilde{e}_{k}^{\text{op}} + \tilde{e}_{k}^{\text{SPE}}) &= \\
\tilde{P}_{\text{op}} \tilde{X}^{\text{op}} + \tilde{P}_{\text{op}}(\tilde{X}^{\text{op}} + \tilde{e}_{k}^{\text{op}} + \tilde{e}_{k}^{\text{SPE}}) &=
\end{align*}
\]  \hspace{1cm} (159)

It can be seen from Equation 159 that the scores found after projection of the batch are not the same as the true scores. The process fault was taken from the null space of the loadings \(\tilde{P}^{\text{op}}\). The projection of the process fault to the future part of the loadings will not be zero according to Equation 145. As a result, it can be concluded that for on-line monitoring using a global model it is impossible to construct a pure \(\text{SPE}\)-signal. If this is not possible by construction, it will also not happen in a real process.

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The only case where a pure SPE-signal can be constructed is for a finished batch. This is also true for the pure D-signal. A process fault $\tilde{e}_{k}^{SPE}$ ($J \times 1$) can be constructed from the null space of the complete loading matrix $\bar{P}$. The finished batch that results in a pure SPE-signal after projection looks like:

$$\tilde{x}_{k}^{SPE} = \tilde{x}_{k}^{*} + \tilde{e}_{k}^{*} + \tilde{e}_{k}^{SPE}$$

(160)

6.4.6 CONCLUSIONS FROM SPE AND D-MECHANICS

The following conclusions can be drawn from the preceding Sections. Pure SPE and D-faults were constructed to show the complementarity of the D and SPE-chart for the different models. In case of on-line monitoring, pure SPE and D-signals can be constructed only for the local model. For the evolving model, only pure SPE-signals can be constructed. No pure SPE or D-signal can be constructed for the global model. Therefore, for evolving and global models the control charts cannot be complementary due to model construction. This hampers the diagnostic value of the control charts. The control charts of the local model are indeed complementary by construction.

The pure D-signal and pure SPE-signal may be considered as the two extremes. The pure D-signal requires an intensified correlation between the process variables where the pure SPE-signal requires a very specific breakage of the correlation. The process conditions necessary for a pure D and SPE-signal can be regarded to be very rare and will not happen in practice. The consequence of this will be discussed in the following sections.

In case of post-batch analysis for the global and time evolving models ($k = K$), pure SPE and D-faults can be constructed by projection of the entire batch run to the model corresponding to time interval $K$. In fact, the time-evolving model is exactly the same as the global model for $k = K$, that is $\bar{P}_{k} = \bar{P}$. Such pure SPE or D-faults need to describe disturbances that occurred during the whole batch run. Process faults that occurred e.g. somewhere halfway the batch can never result in pure D or SPE-faults for a post-batch analysis. The faults representing pure D or SPE-faults for post-batch analysis are non-realistic and will not happen in practice.
There is no overall local model available at time interval $K$ as is the case for the global and evolving model. This makes the post-batch analysis different for the local model. The measurements at every time interval from a finished batch are projected onto the corresponding models. This is not different from on-line monitoring of a new batch as described for the local model. Therefore the same arguments as explained for on-line monitoring using local models hold for the pure $SPE$ and $D$-fault.

6.4.7 THE PROBLEM OF THE EMBEDDED ERROR

It was shown in the previous Section that the complementarity does not exist by construction in some cases. This is not true for e.g. the local model where the complementarity does exist. However, the process faults that represent pure $SPE$ or $D$-faults are considered to be rare. It is more likely that the process conditions deviate from this rare behaviour. This Section describes the effect of this deviation in terms of the embedded error.

The measurement of a sample will always be affected by a measurement error. In this chapter, this is referred to as a process fault. It will be shown that this process fault affects both the $SPE$ and $D$-statistic. This problem is known in factor analysis as the embedded error (E.R. Malinowski [124]). The embedded error problem will be illustrated with the same simple example from the previous section. Consider the measurement of a pure sample $x^*$ depicted as the black dot in Figure 116:

![Graphical interpretation of the embedded error.](image)

Figure 116

*Graphical interpretation of the embedded error.*
This sample can be written exactly according to the model and there are no residuals. Let the sample $x$, depicted as the checkerboard dot in Figure 116, be affected by a process fault. This sample is considered to be very likely in a real process. The next step is to project $x$ on the model $p$ to find the scores and the residuals. The projected sample is depicted in Figure 110 as the striped dot. The scores are calculated according to:

$$ t = p'x = p'(x^* + \varepsilon) = p'x^* + p'\varepsilon = t^* + p'\varepsilon \neq t^* $$

(161)

The score $t$ found for $x$ consists of two mixed parts. The first part represents the distance of the centre of the plane towards the black dot. This was given by the true scores $t^*$. The distance of the black dot to the striped dot gives the second part. This results from the projection of the process fault $\varepsilon$.

It can be seen from Figure 110 that the extracted residuals for $x$ are given by $e^0$. These extracted residuals represent the distance towards the model and result from the projection. Clearly, the extracted residuals are not the same as the process fault $\varepsilon$.

This can be shown mathematically from Equation 162:

$$ e^0 = x - p't = x - pp'x = x - pp'(x^* + \varepsilon) = x^* + \varepsilon - pp'(x^* + \varepsilon) $$

$$ = (x^* - pp'x^*) + (\varepsilon - pp'\varepsilon) = (x^* - p't^*) + (\varepsilon - pp'\varepsilon) = (x^* - x^*) + \varepsilon - pp'\varepsilon $$

(162)

$$ = (1 - pp')\varepsilon \neq \varepsilon $$

Summarizing, the calculated scores $t$ and residuals $e^0$ are different from the true underlying scores $t^*$ and $\varepsilon$. Therefore, general faults will most likely be distributed over the scores and residuals. This is schematically depicted in Figure 117:
The process fault may differ in direction and size, and therefore will determine the magnitude of $e^0$ and $p\varepsilon$. However, there are directions of $\varepsilon$ (indicated in Figure 117 as the bold arrows) that define special cases as explained before. The directions of these vectors can be considered as rare. It is much more likely that the vector describing a process fault lies in other directions as depicted in Figure 117. It was shown that these vectors would result in the embedded error (Figure 116).

It can be concluded that the complementarity of the SPE and D-chart by the local model is very likely to be destroyed because of the embedded error. Most process faults will therefore be distributed over both control charts. This hampers the diagnostic properties of these charts.

6.4.8 Case study: Temperature control of a PVC batch reactor

The data set used for illustration purposes consists of 67 NOC batches where 7 process variables have been measured over 58 time intervals. The main reason to use this dataset is that the correlation between the process variables is fairly predictable.
from an engineering point of view. This makes it possible to simulate certain classes of faults that are realistic. The reactor has been illustrated in Figure 118 and the location of the measured process variables in the reactor is given:

PVC is produced on large scale at Pernis-Holland. At the start of a batch, a 50/50 w/w % mixture of water and monomer are mixed together with some stabilisers to affect the particle morphology. When the mixture reaches a specified temperature, initiator is added. Energy is provided to the mixture until the reaction has started to supply enough heat by itself. This polymerisation reaction is highly exothermic and therefore the withdrawal of heat is very important. This cooling is realised by a cooling jacket and condenser. At 70% conversion, the liquid monomer is no longer present and the remaining vinyl chloride is present in the gas-phase. Vinyl chloride is then absorbed from the gas phase and the reaction is killed when 80-90% conversion is reached. The process variables, which are considered to be of most importance, are the measured temperatures. Therefore, temperature control of the reactor is very important. The subset of process variables comprises the cooling system of the reactor. In order to give an impression of these process variables, the trajectories are given in Figure 119:
Figure 119
Univariate charts of the trajectories of the measured process variables.

The dotted line represents the average value for all the batches. The solid lines are 3σ limits and the dots represent the measurements of a new independent batch.

6.4.9 PROCESS FAULTS FOR BATCH PROCESSES
In this section the different models are illustrated by examining the detection of a certain type of fault in the Shell PVC process. Besides the relative action signal time (RAST), diagnosing the fault will also be discussed. For this purpose, four different type of faults are constructed. A new batch operated at NOC is denoted as \( \mathbf{x} \). The batch \( \mathbf{x} \) contains no process faults and consists of the seven process variables measured at 58 time intervals as described in section 6.4.4.2. The measurements from time interval \( k \) until \( k + 6 \) are depicted in Figure 120.
Every white bar in this picture represents the NOC value of a scaled process variable measured at a certain time interval $k$. This Figure will be used in the following to explain the construction of 4 different process faults. For every class of faults a description is given how the faults are constructed. The ability of the models to detect the faults will be discussed. A batch that contains a process fault will be denoted in the ongoing of this chapter as $x^*$.

### 6.4.9.1 Breakage of the correlation: sensor faults

A distinction can be made for two different situations. The first situations deals with process variables that are integrated in a control loop. In this case a sensor fault will distort the controller action and process control of the reactor is affected.
The other situation is given by process variables that are not part of a control loop. A failure of such a sensor does not always have to disturb the process. The main disadvantage of such a fault is that complete information about the process under consideration cannot be obtained anymore. For simplicity, the case when the process variable is not part of the control loop is considered. Such a sensor fault is illustrated in Figure 121.

At time interval $k + 2$ the sensor measuring process variable five fails indicated by the black bar. The remaining process variables do not respond to this failure since the broken sensor is not incorporated in a control loop. At time interval $k + 2$ itself the correlation between the process variables is broken.

**6.4.9.1.1 Construction**

Since the action signal is defined as three consecutive points outside the control limits, the actual sensor fault was constructed slightly different as depicted in Figure 121. That is, the fault lasted four time intervals from $k$ until $k + 3$ instead of one. The original value of the process variable was multiplied by a factor $\delta$ and was tuned such that the process variable just exceeds the $3\sigma$ limits from Figure 119 and sometimes stays within these limits. Note that such faults would have been undetected using univariate charts like the ones in Figure 119. The sensor fault was constructed at
different periods during the batch run. This procedure is repeated for every process variable. For example, the first wrong batch $x^*$ contains a sensor fault for process variable number one at time interval 10 until 13 where the value for $\delta$ is 1.01. The actual values for $\delta$ and relevant time intervals are given in Table 14.

<table>
<thead>
<tr>
<th>$j$ = 1</th>
<th>$\delta$</th>
<th>Global model</th>
<th>Evolving model</th>
<th>Local model</th>
</tr>
</thead>
<tbody>
<tr>
<td>$k = 10 : 13$</td>
<td>1.01</td>
<td>-</td>
<td>-</td>
<td>$SPE$-chart</td>
</tr>
<tr>
<td>$k = 30 : 33$</td>
<td>1.004</td>
<td>-</td>
<td>-</td>
<td>$SPE$-chart</td>
</tr>
<tr>
<td>$k = 50 : 53$</td>
<td>1.0266</td>
<td>-</td>
<td>-</td>
<td>$SPE$-chart</td>
</tr>
<tr>
<td>$j = 2$</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$k = 10 : 13$</td>
<td>1.01</td>
<td>-</td>
<td>-</td>
<td>$SPE$-chart</td>
</tr>
<tr>
<td>$k = 30 : 33$</td>
<td>1.0045</td>
<td>-</td>
<td>-</td>
<td>$SPE$-chart</td>
</tr>
<tr>
<td>$k = 50 : 53$</td>
<td>1.03</td>
<td>-</td>
<td>$SPE$-chart</td>
<td>-</td>
</tr>
<tr>
<td>$j = 3$</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$k = 10 : 13$</td>
<td>1.01</td>
<td>-</td>
<td>-</td>
<td>$SPE$-chart</td>
</tr>
<tr>
<td>$k = 30 : 33$</td>
<td>1.0037</td>
<td>-</td>
<td>-</td>
<td>$SPE$-chart</td>
</tr>
<tr>
<td>$k = 50 : 53$</td>
<td>1.0265</td>
<td>-</td>
<td>$SPE$-chart</td>
<td>-</td>
</tr>
<tr>
<td>$j = 4$</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$k = 10 : 13$</td>
<td>1.18</td>
<td>-</td>
<td>-</td>
<td>$SPE$-chart</td>
</tr>
<tr>
<td>$k = 30 : 33$</td>
<td>1.055</td>
<td>-</td>
<td>-</td>
<td>$SPE$-chart</td>
</tr>
<tr>
<td>$k = 50 : 53$</td>
<td>1.35</td>
<td>-</td>
<td>$SPE$-chart</td>
<td>-</td>
</tr>
<tr>
<td>$j = 5$</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$k = 10 : 13$</td>
<td>1.18</td>
<td>-</td>
<td>-</td>
<td>$SPE$-chart</td>
</tr>
<tr>
<td>$k = 30 : 33$</td>
<td>1.06</td>
<td>-</td>
<td>-</td>
<td>$SPE$-chart</td>
</tr>
<tr>
<td>$k = 50 : 53$</td>
<td>1.38</td>
<td>-</td>
<td>$SPE$-chart</td>
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<td></td>
<td></td>
</tr>
<tr>
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<td>1.45</td>
<td>-</td>
<td>-</td>
<td>$SPE$-chart</td>
</tr>
<tr>
<td>$k = 30 : 33$</td>
<td>1.7</td>
<td>-</td>
<td>-</td>
<td>$SPE$-chart</td>
</tr>
<tr>
<td>$k = 50 : 53$</td>
<td>6.15</td>
<td>-</td>
<td>$SPE$-chart</td>
<td>-</td>
</tr>
<tr>
<td>$j = 7$</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$k = 10 : 13$</td>
<td>1.02</td>
<td>-</td>
<td>-</td>
<td>$SPE$-chart</td>
</tr>
<tr>
<td>$k = 30 : 33$</td>
<td>1.011</td>
<td>-</td>
<td>-</td>
<td>$SPE$-chart</td>
</tr>
<tr>
<td>$k = 50 : 53$</td>
<td>1.08</td>
<td>-</td>
<td>$SPE$-chart</td>
<td>-</td>
</tr>
</tbody>
</table>

Detection results of faults that break the correlation.

From this table it can be seen that a total of 21 wrong batches containing a sensor fault are simulated.
6.4.9.1.2 Results
For every fault, it was checked i) which model detected the fault first and ii) in which of the two control charts. The detection of the sensor faults gives a very consistent outcome. The global model did not detect any fault. This model seems to suffer from the embedded error where the fault is spread over the scores and residuals. For all the faults at time interval 10 – 13 and 30 – 33, the faults are detected by the local model in the SPE-chart. The faults at time interval 50 – 53 are detected in the SPE-chart by the evolving model except for the sensor fault for process variable one. It must be stressed that the moment of detection in the SPE-chart is very close for the local and evolving model at the time intervals 50 – 53. A more realistic sensor fault would have been detected in the SPE-chart by both models with the same RAST.

It can be concluded that the local model is well suited to detect breakage’s of the correlation due to e.g. sensor faults over the whole batch run. These faults are detected exclusively in the SPE-chart enhancing the diagnostic properties of this model. The global model is not capable to detect the small sensor failures fast enough.

6.4.9.2 Instantaneous intensification of the correlation
One of the main advantages of using multivariate methods instead of univariate methods is the fact that multivariate methods can be utilized to model the correlation between the individual process variables. Intensification in this correlation can be caused by an upset in the reactor. As discussed in Section 2, the variance of the process variables increases but the correlation between the process variables remains intact (not the case for e.g. sensor failures). An example is given in Figure 112. At time interval \( k + 3 \) the process variables significantly change according to the correlation between the process variables. This may be referred to as an instantaneous intensification of the correlation since it only occurs in one point in time. This type of fault will be further discussed in the following.

6.4.9.2.1 Construction
The construction of the process fault has some specific properties. First, the new batch \( \mathbf{x} \) from Figure 120 is monitored and for each time interval \( k \) the scores \( t_k^* \) and residuals \( e_k^* \) are calculated. To simulate the faulty batch at time interval \( k \) from Figure 112 (denoted by \( \mathbf{x}_k^* \)), the following calculation is used:
\[ \tilde{x}_k^* = (\delta + 1) \cdot \tilde{i}_k^* \hat{P}_k^* + \tilde{e}_k^* \]  \hfill (163)

Here, \( \delta \) is chosen in such a way that the raw process variables from Figure 119 stay within or move slightly outside the 3\( \sigma \) limits depending on the time interval. Then, \( \tilde{x}_k^* \) is re-scaled using the scaling constants from the NOC data. Note that \( \tilde{x}_k^* \) is constructed specifically to keep the correlation constant by using the loadings \( \hat{P}_k \).

If this batch \( \tilde{x}_k^* \) is to be monitored using a local model, the embedded error is zero. This is true because \( \tilde{e}_k^* \), \( \hat{P}_k \) and \( \tilde{x}_k^* \) are simultaneously calculated. Projection of \( \tilde{x}_k^* \) to the loadings \( \hat{P}_k \) in order to calculate the scores leads to:

\[ \tilde{x}_k^* \hat{P}_k = ((\delta + 1) \cdot \tilde{i}_k^* \hat{P}_k^* + \tilde{e}_k^*) \hat{P}_k = (\delta + 1) \cdot \tilde{i}_k^* \hat{P}_k + \tilde{e}_k^* \hat{P}_k = \]  \hfill (164)

Which show that the embedded error is zero. As a result, the fault at time interval \( k \) is expected to be detected exclusively in the \( D \)-chart by the local model. For the same reasons as the sensor fault, the fault lasted four time intervals instead of just one from \( k \) until \( k + 3 \). The value of \( \delta \) is chosen in such a way that the process variables from Figure 119 stay within or move slightly outside the 3\( \sigma \) limits depending on the time interval. The actual values for \( \delta \) and relevant time intervals are given in Table 15.

**Table 15**

<table>
<thead>
<tr>
<th>( k )</th>
<th>( \delta )</th>
<th>Global model</th>
<th>Evolving model</th>
<th>Local model</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 : 4</td>
<td>4</td>
<td>-</td>
<td>SPE-chart</td>
<td>-</td>
</tr>
<tr>
<td>10 : 13</td>
<td>6.5</td>
<td>-</td>
<td>SPE/D-chart</td>
<td>D-chart</td>
</tr>
<tr>
<td>20 : 23</td>
<td>10</td>
<td>SPE-chart</td>
<td>SPE-chart</td>
<td>D-chart</td>
</tr>
<tr>
<td>30 : 33</td>
<td>8</td>
<td>SPE-chart</td>
<td>SPE-chart</td>
<td>D-chart</td>
</tr>
<tr>
<td>40 : 43</td>
<td>14.5</td>
<td>SPE-chart</td>
<td>SPE-chart</td>
<td>D-chart</td>
</tr>
<tr>
<td>55 : 58</td>
<td>4.47</td>
<td>-</td>
<td>-</td>
<td>D-chart</td>
</tr>
</tbody>
</table>

Detection results of faults that intensify the correlation

From this table it can be seen that a total of 6 wrong batches containing a change of the correlation are constructed.
6.4.9.2.2 Results

For all the faults, except for time interval 1 – 4 that gave no detection, the fault was detected uniquely in the $D$-chart for the local models. This was expected since the fault is constructed in such a manner that the embedded error is zero. Apparently $\delta$ is not large enough to generate a signal at time interval 1 – 4.

The faults at time interval 1 – 4, 10 – 13, 20 – 23, 30 – 33 and 40 – 43 were detected by the evolving model in the $SPE$-chart. At time interval 10 – 13 also the $D$-chart detected the fault. The global model detected only the faults at time interval 20 – 23, 30 – 33 and 40 – 43 in the $SPE$-chart. The fault at time interval 55 – 58 goes unnoticed by both models. The detection of these faults mainly in the $SPE$-chart for the global and evolving models can be expected from the Theory Section because the overall correlation pattern is broken by definition and such faults are generally picked up by the $SPE$-fault.

Thus, it can be concluded that a fault, which intensifies the instantaneous correlation between the process variables, at a certain time interval, is not always detected by the models. For the global and evolving models this fault is detected in the $SPE$-chart, sometimes followed by a signal in the $D$-chart.

6.4.9.3 Realistic intensification of the instantaneous correlation between the process variables

This fault resembles a more realistic fault as compared to the previous fault. It was assumed in the previous fault that all process variables react instantaneously to a process upset. This is only possible if the dynamics of the process are fast or the time between the process measurements is sufficiently large to let all process variables respond. A more realistic situation is depicted in Figure 122.
At time interval $k$ the process is in-control. At time interval $k + 1$ three process variables are somehow affected by a process upset. This does not intensify the instantaneous correlation at $k + 1$ but breaks it instead. At time interval $k + 2$ almost all process variables respond to the process upset. Now, at time interval $k + 3$ all process variables are affected according to the correlation. This time interval resembles the instantaneous change of the correlation previously described. After this time interval the process returns gradually to the NOC region at $k + 7$. So, this fault breaks the correlation first until $k + 3$ where the process variables behave according to the correlation. After $k + 3$ the instantaneous correlation is broken again until the process is back on target.

6.4.9.3.1 Construction
The simulated fault describes the heating of the reactor as a result of an unwanted side reaction. The fault has a duration of 7 time intervals and reads as:
Statistical batch process monitoring

Time $k$: the temperatures and pressure in the reactor are increasing as a result of an unwanted side reaction that generates extra heat.

Time $k+1$: the temperatures and pressure have increased further. As a result of the controller action, the amount of refrigerant is increased. This decreases the temperature of the refrigerant medium and this is detected in the temperatures of the inlet and outlet of the cooling jacket.

Time $k+2$: the fault remains and the temperatures in the reactor and the pressure are becoming dangerously high. The amount of refrigerant water is further increased to cool the reactor.

Time $k+3$: the temperatures and pressure in the reactor have reached a maximum now. The control action of the opening valve is adjusted to further increase the amount of refrigerant.

Time $k+4$: the temperature in the reactor is decreasing again as a result of the control action. Therefore, the amount of refrigerant is decreased. This slightly increases the temperatures of the inlet and outlet of the cooling jacket.

Time $k+5$: The temperatures and pressure in the reactor have reached their nominal values while the temperature of the cooling medium still increases. The control action of the opening valve is adjusted to decrease the amount of refrigerant further.

Time $k+6$: the temperature drift in the reactor has been successfully controlled and the process is back on NOC.

The construction of this fault is almost similar to the fault described in the previous section (Equation 163). The reason to simulate the fault at time interval 31 – 37 was supported by the fact that loadings $P_k$ of all variables are very similar on these intervals and only differ in intensity. The fault is constructed in such a way that not all the process variables contribute to the disturbance at some time intervals. Also, the magnitude of this contribution is not equal for all process variables but can be set individually. Therefore, the loadings of the local model at time interval $k$ are constructed:

$$\tilde{P}_k^o = \Delta P_k$$

(165)
where $\Delta$ ($J \times J$) is a diagonal matrix. For example, the matrix $\Delta$ at time interval $k = 31$ where only the temperatures and pressures (process variables 1, 2, 3 and 7) are increasing looks like:

$$
\Delta = \begin{bmatrix}
13 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 13 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 13 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 12
\end{bmatrix}
$$

Then, the batch $\hat{x}^*_k$ is constructed using Equation 163 and monitored as described in the previous section. This batch is projected using all three models.

### 6.4.9.3.2 Results

For the local model this fault is detected in the $D$-chart only (RAST = 3). This is a promising result considering the fact that not all the process variables take place in the intensification of the modelled correlation at time intervals 31 and 36. In other words, although the instantaneous correlation is broken and the embedded error is not zero, the fault leads to detection only in the $D$-chart. Probably the effect of the embedded error is very small. This enhances the diagnostics of this fault that clearly is more related to an increase of variance of constant correlation instead of a breakage of the correlation. For the time evolving model as well as the global model the fault is detected in the $SPE$-chart (RAST = 3) while the $D$-chart does not detect at all. This detection in the $SPE$-chart is just as fast as for the detection in the $D$-chart for the local model. These types of fault that break the overall correlation structure seemingly manifest themselves in the $SPE$-chart for the global and time evolving model. The RAST for all the models is the same. The fact that the process fault was detected only in the $D$-chart by the local model enhances the diagnostic properties of such models. Fault detection in this chart can be ascribed due to an intensified correlation, which is indeed true at a certain time interval.
6.4.9.4 Small change breaking the correlation over time

This typical fault describes a minor drift of the process where the instantaneous correlation among the process variables remains intact, however the correlation over time is broken. This time correlation is implicitly modelled and captured in the time evolving and global models. Therefore these models are believed to detect these type of faults. A figure representing the disturbed variables is not helpful in understanding and is therefore not shown like the other faults.

6.4.9.4.1 Construction

The fault that describes this behaviour is modelled as follows. In contrast to the earlier constructed faults, the latent variables from the global models instead of the local are used to construct this faulty batch because the time correlation is captured by a global model. A normal operating batch $x$ is projected onto the model resulting in scores $\tilde{t}_k$ and residuals $\tilde{e}_k$. These will be used to construct a fault as described above by calculating:

$$\hat{x}_k = \delta_k \cdot \tilde{P}^T \tau + \tilde{e}_k$$

where $\delta_{k-1} < \delta_{k-2} < \ldots < \delta_{k-K}$

The vector $\tau$ (R×1) represents a direction within the model plane (together with $\tilde{P}^T$) in which the scores will evolve. This vector is chosen randomly from the rows of the NOC scores $\tilde{T}_k$. Consider the first time interval $k = 1$. The loadings $\tilde{P}^T$ (J×R) represent the instantaneous correlation between the process variables at time point 1. Notice that only the current part (cp) from the loadings $\tilde{P}$ are taken. Pre-multiplying this $\tilde{P}^T$ with $\delta$ and $\tau$ simply moves $\hat{x}_k$ to a certain amount ($\delta$) in a direction ($\tau$) within the model plane ($\tilde{P}^T$). The value of $\delta$ is steadily increasing over time with small steps of 0.3. After time interval 30 the process is assumed to be in-control again. At time point 2, the current part (cp) of $\tilde{P}$ differs form the part at time point 1. Because of this, the correlation in time is changing. The above described procedure is repeated for every time interval until $k = 30$. The direction within the model plane in which the scores will evolve is kept constant but the model plane changes at each time interval.

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6.4.9.4.2 Results

The fault was detected by the time evolving model and global model in the SPE-chart both with an RAST of 21 time intervals. It is expected that this fault breaks the correlation between the process variables when using global and time evolving models.

The local model also detected the fault in the D-chart with an RAST of 21 time intervals. This fault is detected in the D-chart because the loadings of the local model \( \tilde{\mathbf{P}}_s \) at time interval 19, 20 and 21 are very similar to corresponding current parts of the loadings \( \mathbf{P}^{sp} \) from the global model. It was already shown that these faults are detected in the D-chart by the local model.

It can be concluded that faults, which break the correlation over the time, while at the same time the correlation between the process variables is maintained, are detected by all models with the same RAST. The local model is also capable of detecting this fault although time correlation is not explicitly modelled for this type of model.

6.4.10 Conclusions

In this chapter the diagnosing performance of the SPE and D-chart based on local, global and evolving models is assessed. The required behaviour of the process variables to generate either an pure SPE or D-signal is thoroughly discussed. These pure signals are represented by very specific process conditions that are not realistic in a real process. The conclusions can be split into two parts concerning the control charts and the models.

For the control charts, a summary of the results is given in Table 16:

| Table 16 |
|------------------|------------------|
| **POSSIBILITY TO CONSTRUCT PURE SPE OR D SIGNALS** | **DETECTION OF REALISTIC PROCESS FAULTS** |
| **6.4.10.1.1 On-line monitoring** | **6.4.10.1.2 Post batch analysis** | **6.4.10.1.1 On-line monitoring** | **6.4.10.1.2 Post batch analysis** |
| Global | SPE & D | SPE |
| Evolving | SPE & D | SPE |
| Local | SPE & D | SPE & D |
| **DETECTION OF REALISTIC PROCESS FAULTS** | **COMPLEMENTARITY PROPERTIES FOR THE VARIOUS MODELS** |
| Global | SPE | SPE & D |
| Evolving | SPE | SPE & D |
| Local | SPE & D | - |

*Complementarity properties for the various models.*
It can be seen from the upper part of this table that pure $SPE$ and $D$ signals can only be constructed for local models in the case of on-line monitoring. This is favourable for diagnosing faults. Considering the post-batch analysis, pure $SPE$ and $D$ can be constructed for all three models. However, the process conditions representing these pure signals are very unlikely to occur. This results in embedded error that complicates the diagnostics of the control charts. It can be seen from the lower part of Table 16 that despite the construction and embedded error problem, the local model still detects faults in either the $D$ or $SPE$-chart. This is not true for the global and evolving model where realistic process faults are detected in the $SPE$-chart. This hampers the diagnostic properties of these control charts.

The conclusions concerning the models are as follows. It is better to use local or time evolving models for on-line monitoring of batch processes instead of global models. The local models are able to detect pure $SPE$ and $D$-faults, where the time evolving model only can detect pure $SPE$-faults. Therefore, in terms of diagnosing capabilities the local model is slightly favoured.
6.5 Correlations in batch process data*

6.5.1 SUMMARY
A repeatedly returning discussion in modelling batch process data using component models is the effect of the matricizing direction of the three-way array $X$ and the selection of a model for capturing the auto-and cross-correlations of the process variables. To study the capture of the correlations by the models, a benchmark dataset is matricized and modelled accordingly. Next, an independent batch run is fitted to the models and the recovery of the correlations is analysed. The results show that all models are well able to capture the correlations in the data and the matricizing direction only affects the distribution of the correlations among the model parameters.

6.5.2 INTRODUCTION
Since the introduction of multivariate statistical batch process monitoring by Nomikos and MacGregor in 1992 (J.F. MacGregor & P. Nomikos [22]), there has been a lot of research done in this field. This has led to numerous applications and many extensions of the original proposed ideas (S. Rannar et al. [31], S. Wold et al. [53], D. Neogi & C.E. Schlags [44], D.J. Louverse & A.K. Smilde [38], J.A. Westerhuis et al. [42], A.K. Smilde & H.A.L. Kiers [39]).

A discussion, which regularly returns is the effect of the matricizing directions of the three-way array $X$ ($I \times J \times K$) and which model to use to capture the auto- and cross-correlations present in the data (T. Kourt [18]). In the following, a preliminary assessment is given of correlations in batch process data and how the different models capture these correlations.

Batch processes can be thought of as non-linear, non-stationary multivariate time series of limited length. By removing the average trajectory of a process variable during a batch run, the main non-linear behaviour of the process data is removed and the remaining variation around this average batch trajectory can be modelled using a linear model such as PCA. The model captures auto- and cross-correlations present in

the data. In statistical batch process monitoring the aim is to detect deviations from normal operating conditions such as process upsets, sensor failure or impurities. This reflects itself in correlation breakage or intensification of the existing correlations. With an intensified correlation is meant a process, which has similar correlations as under normal operating conditions, with the difference, that the correlations have a much higher or lower magnitude.

The procedure is to collect a historical reference set of batches from which it is known that these batches operated under normal operating conditions (NOC). Next, a model is constructed of the historical dataset, which captures the correlations under normal operating conditions. After that, a new batch is monitored by checking if the new batch has similar correlations as the reference data. Therefore, understanding how the models capture these correlations is relevant.

After removal of the average trajectory of the process variables, the main non-linearity in the data is removed. Now, the data can be seen as non-stationary linear multivariate time series with a zero mean. From this, auto and cross-correlations between the process variables can be computed and analysed. The recovery of correlations by the models is studied by fitting new data to the models, compute auto and cross-correlations from the fitted data and compare these with the auto- and cross-correlations from the original data.

6.5.3 Theory

6.5.3.1 Structure of the data
A single batch run is represented by a matrix \( X (K \times J) \) where \( J \) process variables are measured during \( K \) time intervals. Since a batch process is recipe driven, it is assumed that a batch behaves according to a certain trajectory. Batches that are repeated according to the recipe are stacked in a three-way array \( X (I \times J \times K) \) where \( I \) is the number of batches. Having three-way data in \( X \), there are three possible ways to arrange the data. The first is to keep the three-way array as is and use multi-way models such as PARAFAC and Tucker3. Next, there are two sensible directions of matricizing the three way array (P. Nomikos [21]), leading to two different Tucker1 models. The first is matricizing in the batch direction and forming matrix \( X_{NM} (I \times JK) \). The second is matricizing in the process variable direction and form the matrix \( X_{PI} (KI \times J) \). Note that the mean time trajectory of all process variables has been
removed before hand. This makes matricizing to $X_{PV}$ rather different from the
approach described by S. Wold et al. [53], where the mean trajectories are not
removed. As a result, the non-linearity is still present in the data. This will have an
effect on the correlations in the data (T. Kourt [18]). Nevertheless, both approaches
have shown to work well in practice (E.N.M. Van Sprang et al. [41]).

In order to compare the different models, a similar preprocessing is required
for each model. For that reason, the average batch trajectory of the process variables
is removed prior to matricizing and modelling. That is, each column in $X$ is mean
centered and scaled to unit variance (auto scaling).

6.5.4 Correlation in batch process data
To understand how the models capture the correlations, an overview of correlations
present in the data is given. Within a batch, there are time dependent correlations due
to the underlying chemistry of the process. Furthermore, seasonal influences or
process history may cause correlations between the batches e.g. carry-over effects.
The following correlations can be distinguished (see Figure 123):

1. correlation within a batch
   a) Autocorrelation of process variables
   b) Correlation between process variables at the same time interval
   c) Cross-correlation between process variables over time

2. correlation between batches
   d) Correlation between batches

Figure 123
Correlations in batch process data.
1. **Within batch run correlations**
   a) Autocorrelation of a process variable over time
   b) Correlation between the process variables at the same time interval
   c) Cross-correlation between the process variables over time

In addition, in the three-way array there are between correlations such as:

2. **Between batch run correlations**
   d) Correlation between the batches

Since batch processes are recipe driven, it is expected that all the batches have a similar within correlation structure over time (1). In principle, since each batch is considered as an independent sample from a population of batch runs, there should be no correlation between the batches.

### 6.5.5 MATRICIZING AND MODELLING THE THREE-WAY ARRAY

The two sensible directions of matricizing also define two extremes in modelling the correlations in the data. The first way is matricizing in the batch direction and forming matrix $X_{NM}$. The second way is matricizing in the process variable direction and form the matrix $X_{PV}$. The arrangement of the three-way array also defines the distribution of the auto- and cross-correlations in the data and therefore, also defines the distributions of the correlations among the model parameters. In here, the distributions of the correlations among the model parameters of three modelling strategies will be discussed: the global model, the local model and the $p^*$-model. The first strategy is proposed by P. Nomikos & J.F. MacGregor [6]. With this strategy $X$ is matricized in the batch direction and a model is constructed on $X_{NM}$.

$$X_{NM} = T_{NM} P_{NM}^T + E^{T_{NM}}$$

This model forces all within batch-run variations (1.a, 1.b, 1.c) into the model loadings $P_{NM} (JK x R)$. The differences between the batches are captured by the scores $T_{NM} (I x R)$. 

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A geometrical representation of the global model is given in Figure 124. In this figure, it is shown that a point in the $JK$ dimensional space represents a batch (Batch 1, Batch 2 and Batch 3). The model plane ($P_{NM}$) captures the principal direction of the process, which is defined by the underlying physics and chemistry. It is easy to understand that the differences between the batches are given by the projections (reflected in the scores $T_{NM}$) of the batches on the model plane. Actually, in process monitoring, it is tested how much within variation is present in a new batch against a historical reference. If the new batch has a similar within variation as the reference batches, the new batch will be in the model plane somewhere between the reference batches. The position in the model plane is monitored using a $D$-chart (P. Nomikos & J.F. MacGregor [6]). If a batch does not have a similar within variation, the correlations break down and the batch will deviate from the model plane. The level of dissimilar within variations is monitored with $Q/SPE$-chart (P. Nomikos & J.F. MacGregor [6]).

If this model is used for on-line process monitoring, it is well known that assumptions have to be made about the future unknown observations (P. Nomikos & J.F. MacGregor [6]). As a result of these assumptions, the projection of a batch will "walk" a certain trajectory on the plane during the evolution of the batch (see the running batch in Figure 124). Moreover, because assumptions are made, the batch data will not have a similar within variation compared to historical reference batches and the distances to the model will vary a lot for each time interval $k$. To overcome this feature, a historical reference distribution is estimated for each time interval $k$. As
the batch approaches the end of its trajectory, the projection reaches its true point on the model plane.

The second strategy is suggested by P. Nomikos [21] and further elaborated by H.-J. Ramaker et al. [125]. With this strategy the process is approximated by $K$ independent local models and does not include previous process measurements in the model. With batch processes, it is assumed that each batch run is operated in an identical manner according to a recipe. Therefore, each batch run has the same underlying phenomena as a driving force implying that the present auto and cross-correlations in the data are not arbitrary but fixed for each time interval. Given that the correlations in the data are fixed, also the direction of the process for each time interval is fixed and can be captured with a multivariate model valid for time interval $k$.

$$X^{[s]}_k = T_k P_k^T + E^{[s]}_k \quad (169)$$

The auto-and cross correlations (1.a, 1.b and 1.c) are implicitly captured by the plane direction of the loadings $P_k$. A geometrical interpretation is given in Figure 125. The underlying dynamics will determine the principal direction of the process for time interval $k$ and the model plane will be different for each time interval.
The last strategy is matricizing in the batch direction and model $X_{pv}$ with a bilinear model. This was suggested by P. Nomikos [21] and elaborated by Wold and co-workers (S. Wold et al. [53]). This model will be referred to as the pv-model.

$$X^{K_{ef}} = T_x P_x^T + E^{K_{ef}}$$ (170)

This strategy uses a common model $P_{pv} (J \times R)$ consisting of the overall correlations between the process variables. The behaviour of the process is now expressed on a common base defined by the model $P_{pv}$. That is, the path followed by the projections determines the dynamic behaviour of the process. This is different from the other approaches where the model plane captured the dynamics of the process. A geometric representation is given in Figure 126. A new batch follows a path in the model plane. In process monitoring, the historical batches follow a similar path in the model plane. Next, control limits are placed around this trajectory and new batches are compared if they follow the same path.

![Figure 126](image_url)

**Correlations captured by a pv-model.**
6.5.6 CASE STUDY
To show that all previously mentioned models capture the correlations in the data, a benchmark dataset is modelled using the previously mentioned models. Next, a new independent batch is fitted and the obtained model parameters are used to reconstruct the original data. From this, auto- and cross-correlations of the process variables are computed and compared with the correlations computed from the original data.

6.5.6.1 Description of the data
The dataset is described by P. Nomikos & J.F. MacGregor [126] and consists of 50 batches for which 9 process variables are measured during 200 time intervals. The following process variables are measured: two flow-rates of the monomers, four temperatures, the density of the latex, the conversion and an estimate of the rate of energy release.

6.5.6.2 Preprocessing
To compare the models, the main non-linear behaviour is removed by column centering in the $I$ mode and scaled to unit variance by dividing each column by its standard deviation after centering. Next, the data cube is matricized in the desired direction and modelled by the three models. The number of components for each model is determined using cross-validation and scree-plots (S. Wold [137]).

6.5.6.3 Results
Because the results were similar for all batches, one batch is selected for analysis. As a result, 49 batches are used to construct a reference model and the selected batch (normal batch) is used for analysis (batch 43). Furthermore, to avoid an abundance of results, a selection is made of two process variables based on their different nature of correlation during the batch run.

For this analysis, the normal batch is preprocessed and rearranged accordingly. Next, the normal batch is fitted to the different models. The first step is to reconstruct the original signal from the models.
The explained variance by each model is given in Table 17. The global model explains a relative small amount of the variance in the data compared to the pv-model and local model. This is easy to understand since the pv-model and local model use much more model parameters to model the batch data. Using a similar number of parameters with a global is not possible since this will lead to over fitting.

The low amount of explained variance by the global model does give information about the nature of the process data, namely that the data does not have an ideal batch structure. That is, the underlying common phenomena suffer from relative large variations, which are different from batch-to-batch.

The recovered signals after fitting the new batch are given in Figure 127. Each graph represents one process variable with the results from the models and the original signal. From the reconstructed signals auto- and cross-correlations are computed.

![Figure 127](image)

Reconstruction of the original signal for batch 43.
The autocorrelations of two process variables are given in Figure 128 and Figure 129: Process variable 2 (pv2) and process variable 6 (pv6). Process variable 2 is a flow rate and is expected to be less autocorrelated than process variable 6, which is a temperature measurement. From top to bottom, the graphs correspond to the autocorrelation of original (preprocessed) signal, the local model, the pv-model and the global model.
The cross correlations between the two variables are given in Figure 130. In the original signal of process variable 2 an autocorrelation is observed for the first seven lags. The same correlation structure is observed for the fitted local and pv-model. The global model shows a more correlated structure compared to the original correlation structure, implying that the reconstructed signal is more smoothed than the original. The autocorrelation for process variable 6 is well captured by all models except for the global model, which shows a slightly different correlation structure.
The cross-correlation between the two process variables reveals that the local and pv-model capture more or less the same correlation structure whereas the global model shows a more smoothed structure which is expected since the reconstructed signal of process variable 2 is smoothed by the global model. The smoothed signal obtained with the global model can simply be understood since the model considers the entire process at once whereas with the W-model and local model the process is approximated by successive multiple models. The global model captures the principal directions in a JK-dimensional space. That is, only the main directions are considered. Because of this smoothed loadings are obtained and certain local variations are less described than others. This is also reflected by the explained variance in Table 17.
In short:

- The global model captures the general direction of the auto-and cross-correlations in the model loadings. However, because of its construction and nature of the data, it has a smaller amount of explained variance.
- With the pv-model, the correlations are captured by a combination of scores and a common model plane. The path of scores on the model plane represents the dynamics of the process.
- The local model is a fingerprint of the process correlations at time interval $k$.

6.5.7 CONCLUSIONS

It is shown that the data arrangement or the matricizing direction of $X$ determines the distribution of the correlations among the model parameters. Hence, there is no loss of correlation structure as a result of the model choice. However, the important difference between the different modelling strategies is the number of acceptable model parameters necessary to capture the underlying phenomena. Therefore, it is a priori not clear which model will give the best monitoring performance.