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STATISTICAL PROCESS CONTROL OF MULTIVARIATE PROCESSES

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Abstract: With process computers routinely collecting measurements on large numbers of process variables, multivariate statistical methods for the analysis, monitoring and diagnosis of process operating performance have received increasing attention. Extensions of traditional univariate Shewhart, CUSUM and EWMA control charts to multivariate quality control situations are based on Hotelling's T^2 statistic. Recent approaches to multivariate statistical process control which utilize not only product quality data (Y), but also all of the available process variable data (X) are based on multivariate statistical projection methods (Principal Component Analysis (PCA) and Partial Least Squares (PLS)). This paper gives an overview of these methods, and their use for the statistical process control of both continuous and batch multivariate processes. Examples are provided of their use for analysing the operations of a mineral processing plant, for on-line monitoring and fault diagnosis of a continuous polymerization process and for the on-line monitoring of an industrial batch polymerization reactor.

Keywords: Batch processes, Control charts, Fault diagnosis, Multivariable processes, Principal component analysis, Process monitoring, Statistical process control.

1. INTRODUCTION

Statistical Process Control (SPC) concepts and methods have become very important in the manufacturing and process industries. Their objective is to monitor the performance of a process over time in order to verify that the process is remaining in a "state of statistical control". Such a state of control is said to exist if certain process or product variables remain close to their desired values and the only source of variation is "common-cause" variation, that is, variation which affects the process all the time and is essentially unavoidable within the current process. SPC charts such as Shewhart, CUSUM and EWMA charts are used to monitor key product variables in order to detect the occurrence of any event having a "special" or "assignable" cause. By finding assignable causes, long-term improvements in the process and in product quality can be achieved by eliminating the causes or improving the process or its operating procedures.

It is important to note that both the concepts and methods of SPC are totally different from those of automatic feedback process control. In general the two approaches are totally complementary. Automatic feedback control should be applied wherever possible to reduce variability in important process and product variables. Feedback controllers compensate for the predictable component of disturbances in important variables by adjusting other process variables and thereby transferring the variability into these less important manipulated variables (Downs and Doss, 1991). SPC monitoring methods should be applied on top of the process and its automatic control system in order to detect process behaviour that indicates the occurrence of a special event. By diagnosing causes for the event and removing them (rather than simply continuing to compensate for them), the process is improved.

Unfortunately, most SPC methods are based on charting only a small number of variables, usually the final product quality variables (Y), and examining them one at a time. These approaches are totally inadequate for most modern process industries. They ignore the fact that with computers hooked up to nearly every industrial process, massive amounts of data are being collected continually on perhaps hundreds of process variables (X). Measurements on variables such as temperatures, pressure, flowrates, etc., are available every second. Final product quality variables (Y), such as polymer molecular weights or melt index, cut points in distillations, etc., are available on a much less frequent basis. All such data should be used to extract information in any effective scheme for monitoring and diagnosing operating performance. However, all these variables are not independent of one another. Only a few underlying events are driving a process at any time, and all these measurements are simply different reflections of these same underlying events. Therefore, examining them one variable at a time, as though they were independent, makes interpretation and diagnosis extremely difficult. Such methods only look at the magnitude of the deviation in each variable independently of all others. Only multivariate methods that treat all the data simultaneously can also extract information on the directionality of the process variations, that is on how all the variables are behaving relative to one another. Furthermore, when important events occur in processes they are often difficult to detect because the signal to noise ratio is very low in each variable. But multivariate methods can extract confirming information from observations on many variables and can reduce the noise levels through averaging.

This paper presents an overview on both traditional and new multivariate SPC methods for monitoring and diagnosing process operating performance. Traditional methods for multivariate quality control (based only on the product quality measurements) are reviewed in Section 2. Some multivariate statistical projection methods (Principal Components Analysis (PCA) and Partial Least Squares (PLS)) which form the basis of new approaches to multivariate SPC (which use all the process data X as well as the quality data Y) are summarized and their similarities and differences with the traditional methods are discussed in Section 3. The use of these multivariate statistical projection methods for analyzing and interpreting historical plant operating records available in computer databases is also discussed. New multivariate SPC methods for the on-line monitoring and diagnosis of process operating performance in both continuous and batch processes are presented and illustrated in Section 4. Section 5 discusses some practical issues concerning the correct application of these methods.

2. MULTIVARIATE CHARTS FOR STATISTICAL QUALITY CONTROL

In most industries, traditional univariate control charts like Shewhart (Shewhart, 1931), CUSUM (Woodward and Goldsmith, 1964) and EWMA (Roberts, 1959; Hunter, 1986) are used for separately monitoring key measurements on the final product which in some way define the quality of that product. The difficulty with this approach is that these quality variables are not independent of one another, nor does any one of them adequately define product quality by itself. Product quality is only defined by the correct simultaneous values of all the measured properties, that is, it is a multivariate property.

The difficulty with using independent univariate control charts can be illustrated by reference to Figure 1. Here only two quality variables (y_1, y_2) are considered for ease of illustration. Suppose that, when the process is in a state of statistical control where only common cause variation is present, y_1 and y_2 follow a multivariate Normal distribution and are correlated ($\rho_{y_1y_2} = -0.94$) as illustrated in the joint plot of y_1 vs y_2 in Figure 1. The ellipse represents a contour for the in-control process, with 99 % confidence limits, and the asterisks represent a set of observations from this distribution. The same observations are also plotted in Figure 1 as individual Shewhart charts on y_1 and y_2 versus sample number (time) with their corresponding upper (UCL) and lower (LCL) control limits (99% confidence limits). Note that by inspection of each of the individual Shewhart charts the process appears to be clearly in a state of statistical control, and none of the individual observations gives any indication of a problem. The only indication of any difficulty is that a customer has complained about the performance of the product corresponding to the ⊗ in Figure 1. If only univariate charts were used, one would clearly be confused. The same customer apparently liked all the other lots of product sent to him, many of them with values of y_1 and y_2 much further from target. The true situation is only revealed in the multivariate y_1 vs y_2 plot where it is seen that the lot of product indicated by the ⊗ is clearly outside the joint confidence region, and is clearly different from the normal "in - control" population of product.

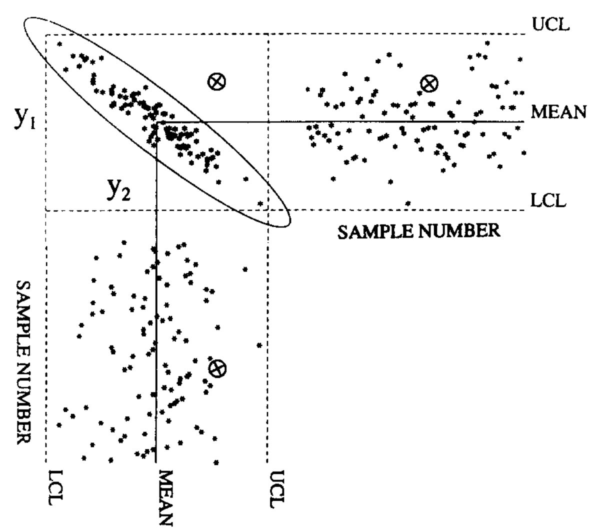

Figure 1. Quality control of two variables illustrating the misleading nature of univariate charts.

In spite of the misleading nature of univariate quality control charts they continue to be almost the only form of monitoring used by industry. However, several multivariate extensions of the Shewhart, CUSUM and EWMA based on Hotelling's T^2 statistic have been proposed in the literature (see review articles by Wierda (1994) and Sparks (1992)).

2.1 Multivariate Shewhart Charts

In situations where one observes a vector of q variables y_{qx1} at each time period multivariate χ^2 and T^2 charts are used. The T^2 chart has its origins in the work of Hotelling (1947), and several references discuss the charts in more detail (Alt, 1977, 1985; Alt and Smith, 1988; Ryan, 1989; and Jackson, 1991).

Given a (q×1) vector of measurements **y** on q, normally distributed variables with an in-control covariance matrix Σ one can test whether the vector μ of the means of these variables is at its desired target τ by computing the statistic :

$$\chi^2 = (\mathbf{y}-\tau)^T\Sigma^{-1}(\mathbf{y}-\tau) \qquad (1)$$

This statistic will be distributed as a central Chi-squared distribution with q degrees of freedom if $\mu = \tau$. A multivariate Chi-squared control chart can be constructed by plotting χ^2 versus time with an upper control limit (UCL) given by $\chi^2_\alpha(q)$ where α is an appropriate level of significance for performing the test (e.g. α = 0.01).

Note that this multivariate test overcomes the difficulty illustrated in the example of Figure 1, where univariate charts were incapable of detecting the special event denoted by ⊗. The χ^2 statistic in equation (1) represents the directed or weighted distance (Mahalanobis distance) of any point from the target τ. All points lying on the ellipse in Figure 1 would have the same value of χ^2. Hence, a χ^2 chart would detect as a special event any point lying outside of the ellipse. (Notice that the ellipse is the solution to equation (1) for $\chi^2 = \chi^2_\alpha(q)$, for two variables).

When the in-control covariance matrix Σ is not known, it must be estimated from a sample of n past multivariate observations as

$$S = (n-1)^{-1} \sum_{i=1}^{n} (y_i - \bar{y})(y_i - \bar{y})^T \quad (2)$$

When new multivariate observations (y) are obtained, then Hotelling's T^2 statistic given by

$$T^2 = (\mathbf{y} - \tau)^T \mathbf{S}^{-1} (\mathbf{y} - \tau) \quad (3)$$

can be plotted against time. An upper control limit on this chart is given by:

$$T^2_{UCL} = \frac{(n-1)(n+1)q}{n(n-q)} F_\alpha(q, n-q) \quad (4)$$

where $F_\alpha(q, n - q)$ is the upper 100α% critical point of the F-distribution with q and n - q degrees of freedom (Tracy et al., 1992).

The above charts are for a single new multivariate observation vector at each time. If an average of m new multivariate observations are to be used at each time or if the estimate of the variance S is based on pooling estimates from rational subgroups, then the above definitions of the χ^2 and T^2 charts and their UCL's must be correspondingly redefined (Wierda, 1994). Furthermore, if the charts are utilized to examine past data that are also used in computing S, then the distributional properties of T^2 are different from the above (Tracy et al., 1992; Wierda, 1994).

Once an out of control signal is detected the challenge is to determine which variables are responsible for it (i.e., identify the variables whose means have shifted). Several approaches have been suggested for this task (Wierda, 1994; Kourti and MacGregor, 1994; Fuchs and Benjamini, 1994).

2.2 Multivariate CUSUM Charts

Cumulative Sum (CUSUM) charts are based on using a sequence of sequential probability ratio tests. Consider a sequence of random variables $\mathbf{y}_1, \mathbf{y}_2$ ... distributed independently as $N_p(\mu, \Sigma)$, and it is desired to test H_0 : $\mu = \tau$ versus $H_1 : \mu = \mu_1$. Let $d^2 = (\mu_1 - \tau)^T \Sigma^{-1} (\mu_1 - \tau)$ define the squared length of the shift in mean, then the sequential probability ratio test rejects the null hypothesis whenever

$$\sum_{i=1}^{n} \{d^{-1}(\mu_1 - \tau)\Sigma^{-1}(\mathbf{y}_i - \tau) - d/2\} > -\frac{\log \alpha}{d} \quad (5)$$

where α is the level of significance chosen. Healy (1987) used this result to propose plotting the CUSUM

$$C_i = \text{Max}\{0, C_{i-1} + d^{-1}(\mu_1 - \tau)\Sigma^{-1}(\mathbf{y}_i - \tau) - d/2\} \quad (6)$$

to test for this specific shift in mean from τ to μ_1.

To generalize the test against all alternatives (H_1:$\mu \neq \tau$) several extensions of the univariate CUSUM have been proposed. Crosier (1988) proposed computing T^2 at each point and then computing the CUSUM of the scalar distance T^2 (or its square root) as

$$C_i = \text{Max}\{0, C_{i-1} + T_i - k\} \quad (7)$$

with initial condition $C_0 \geq 0$. This CUSUM scheme signals an out-of-control situation when $C_i > h$.

Crosier also proposed replacing the scalar quantities of the univariate CUSUM by their vector counterparts and computing the vector CUSUM

$$\begin{aligned} \mathbf{s}_i &= 0 && \textit{if} \quad C_i \leq k \\ &= (\mathbf{s}_{i-1} + \mathbf{y}_i - \tau)(1 - k/C_i) && \textit{if} \quad C_i > k \end{aligned} \quad (8)$$

where C_i is the weighted length $\{(\mathbf{s}_{i-1} + \mathbf{y}_i - \tau)^T \Sigma^{-1} (\mathbf{s}_{i-1} + \mathbf{y}_i - \tau)\}^{1/2}$.

The scheme signals an out-of-control situation whenever

$$Z_i = \max\{0, C_i - k\} > h. \quad (9)$$

A reference value $k = d^2/2$ is usually chosen. This choice minimizes the average run length or ARL (average number of samples required to detect a deviation for the first time) at deviation d for a given on-target ARL. The on-target ARL is determined by the choice of the control limit h. Pignatiello and Runger (1990) have proposed another variant of the multivariate CUSUM.

These CUSUM charts use all of the observations since the detection of the last special event rather than only the last observation vector as in the Shewhart type charts. Their advantage over the latter charts is that their average run lengths are smaller for small shifts in the process mean.

2.3 The Multivariate EWMA

Multivariate EWMA charts compute the exponentially weighted moving average of the vector process (Lowry et al., 1992)

$$\mathbf{z}_i = \mathbf{R}(\mathbf{y}_i - \tau) + (\mathbf{I} - \mathbf{R})\mathbf{z}_{i-1} \quad (10)$$

where $\mathbf{R}$ = diagonal $\{r_1, r_2, ..., r_p\}$ and $0 < r_j \leq 1$; j = 1, ..., p. Large values of r_j give more smoothing and better detection of small shifts. The MEWMA gives an out-of-control signal when

$$Q_i^2 = \mathbf{z}_i^T \Omega_i^{-1} \mathbf{z}_i > h \quad (11)$$

where the control limit h is chosen to achieve a specified in-control ARL. When all the r_j are equal (r_j = r, for j=1,2,...p) the covariance matrix is given by

$$\Omega_i = \frac{r[1-(1-r)^{2i}]}{2-r} \Sigma \quad (12)$$

where Σ = Cov (Y).

The properties of the MEWMA chart are quite similar to those of the multivariate CUSUMs.

3. MULTIVARIATE STATISTICAL PROJECTION METHODS

3.1 Principal Component Analysis

When the number of measured quality variables (q) is large one often finds that they are highly correlated with one another and their covariance matrix Σ is nearly singular. A common procedure for reducing the dimensionality of the quality variable space is Principal Component Analysis (PCA). (Wold et al., 1987; Mardia et al., 1989; Jackson, 1991). The first principal component (PC) of **y** is defined as that linear combination $t_1 = \mathbf{p}_1^T\mathbf{y}$ that has maximum variance subject to $|p_1| = 1$. The second PC is that linear combination defined by $t_2 = \mathbf{p}_2^T\mathbf{y}$ which has next greatest variance subject to $|p_2| = 1$, and subject to the condition that it be uncorrelated with (orthogonal to) the first PC (t_1). Additional PC's up to q are similarly defined. It is easily shown that the principal component loading vectors $\mathbf{p}_i$ are the eigenvectors of the covariance matrix (Σ) of **Y**, and the corresponding eigenvalues (λ_i) are the variances of the PC's (i.e., $\mathrm{Var}(t_i) = \lambda_i$). In practice Σ is not known and is estimated by $(n-1)^{-1}\mathbf{Y}^T\mathbf{Y}$ where **Y** is the (n×q) matrix of mean centered and scaled measurements. Hence the sample principal component loadings are computed as the eigenvectors of the (q × q) matrix ($\mathbf{Y}^T\mathbf{Y}$). The principal component scores are defined as the observed values of the principal components for each of the n observation vectors (i.e., $\mathbf{t}_i = \mathbf{Y}\,\mathbf{p}_i^T$, i = 1, 2, ..., q). In effect PCA decomposes the observation matrix **Y** as:

$$\mathbf{Y} = \mathbf{T}\mathbf{P}^T = \sum_{i=1}^{q} \mathbf{t}_i\mathbf{p}_i^T \tag{13}$$

PCA is scale-dependent, and so the **Y** matrix must be scaled in some meaningful way. The most usual form of scaling is to scale all variables to unit variance and then perform PCA on the correlation matrix. Alternatively, in quality-control situations, scaling the Y's inversely proportional to their specification limits or some other measure of relative importance is usually more meaningful.

In practice, one rarely needs to compute all the q eigenvectors, since most of the variability in the data are captured in the first few PC's; 2 or 3 PC's are often sufficient to explain most of the predictable variations in the product. The NIPALS algorithm, (Geladi and Kowalski, 1986) is ideal for computing the principal components in a sequential manner when the number of variables is large. The number of PC's that provide an adequate description of the data can be assessed using a number of methods (Jackson, 1991) with cross-validation (Wold, 1978) being perhaps the most reliable. By retaining only the first A PC's the **Y** matrix is approximated by:

$$\hat{\mathbf{Y}} = \sum_{i=1}^{A} \mathbf{t}_i\mathbf{p}_i^T. \tag{14}$$

3.2 Quality Control Charts Based on Principal Components

Having established a PCA model based on historical data collected when only common cause variation was present, future behaviour can be referenced against this "in-control" model. New multivariate observations can be projected onto the plane defined by the PCA loading vectors to obtain their scores ($t_{i,new} = \mathbf{p}_i^T\mathbf{y}_{new}$), and the residuals $\mathbf{e}_{new} = \mathbf{y}_{new} - \hat{\mathbf{y}}_{new}$, where $\hat{\mathbf{y}}_{new} = \mathbf{P}_A\mathbf{t}_{A,new}$, and $\mathbf{t}_{A,new}$ is the (A × 1) vector of scores from the model and $\mathbf{P}_A$ is the (q × A) matrix of loadings. Multivariate control charts based on Hotelling's T^2 can be plotted based on the first A PC's, where

$$T_A^2 = \sum_{i=1}^{A} \frac{t_i^2}{s_{t_i}^2} \tag{15}$$

and $s_{t_i}^2 (= \lambda_i)$ is the estimated variance of t_i. If A = 2, a joint t_1 vs t_2 plot can be used.

Note that the traditional Hotelling's T^2 in equation (3) is equivalent (Mardia, Kent and Bibby, 1989; Kourti and MacGregor, 1994) to:

$$T^2 = \sum_{i=1}^{q} \frac{t_i^2}{\lambda_i} = \sum_{i=1}^{q} \frac{t_i^2}{s_{t_i}^2} = \sum_{i=1}^{A} \frac{t_i^2}{s_{t_i}^2} + \sum_{i=A+1}^{q} \frac{t_i^2}{s_{t_i}^2} \tag{16}$$

By scaling each t_i^2 by the reciprocal of its variance each PC term plays an equal role in the computation of T^2 irrespective of the amount of variance it explains in the **Y** matrix. This illustrates some of the problems with using T^2 when the variables are highly correlated and Σ is very ill-conditioned. When the number of variables (q) is large, Σ is often singular and cannot be inverted, nor can all the eigenvectors be obtained. Even if it can, the last PC's (i = A + 1, ..., q) in equation (16) explain very little of the variance of **Y** and generally represent random noise. By dividing these t_i's by their very small variances, slight deviations in these t_i's which have almost no effect on **Y** will lead to an out-of-control signal in T^2. Therefore, T_A^2 based on the first A (cross-validated) PC's provides a test for deviations in the product quality variables that are of greatest importance to the variance of Y.

However, monitoring product quality via T_A^2 based on the first A PC's is not sufficient. This will only detect whether or not the variation in the quality variables in the plane of the first A PC's is greater than can be explained by common cause. If a totally new type of special event occurs which was not present in the reference data used to develop the in-control PCA model, then new PC's will appear and the new observations $\mathbf{y}_{new}$ will move off the plane. Such new events can be detected by computing the squared prediction error (SPE_y) of the residuals of new observations (Kresta et al., 1991).

$$SPE_y = \sum_{i=1}^{q} (y_{new,i} - \hat{y}_{new,i})^2 \tag{17}$$

This statistic is also referred to as the Q-statistic (Jackson, 1991), or distance to the model. It represents the squared perpendicular distance of a new multivariate observation from the plane. When the process is "in-control", this value of SPE_y or Q should be small. Upper control limits for this statistic can be computed, from historical data, using approximate results for the distribution of quadratic forms (Jackson, 1991; Nomikos and MacGregor, 1995). In the modelling stage and when the process is "in control", SPE_y represents unstructured fluctuations (noise) that cannot be accounted for by the model. When an unusual event occurs that results in a change in the covariance structure of Y, it will be detected by a high value of SPE_y; a high value of SPE_y means that the projection model is not valid for that observation. A very effective set of multivariate control charts is therefore a T^2 chart on the A dominant orthogonal PC's ($t_1, ..., t_A$) plus a SPE_y chart.

3.3 PLS - Partial Least Squares

Given two matrices, an (n × m) process variable data matrix **X**, and an (n × q) matrix of corresponding product quality data **Y**, one would like to extract latent variables that not only explain the variation in the process data (**X**), but that variation in **X** which is most predictive of the product quality data (**Y**). PLS is a method (or really a class of methods) which accomplish this by working on the sample covariance matrix $(\mathbf{X}^T\mathbf{Y})(\mathbf{Y}^T\mathbf{X})$. In the most common version of PLS (Höskuldsson, 1988), the first PLS latent variable $t_1 = \mathbf{w}_1^T\mathbf{x}$ is that linear combination of the x-variables that maximizes the covariance between it and the **Y** space. The first PLS loading vector $\mathbf{w}_1$ is the first eigenvector of the sample covariance matrix $\mathbf{X}^T\mathbf{Y}\mathbf{Y}^T\mathbf{X}$. Once the scores $\mathbf{t}_1 = \mathbf{X}\mathbf{w}_1$ for the first component have been computed the columns of **X** are regressed on $\mathbf{t}_1$ to give a regression vector $\mathbf{p}_1 = \mathbf{X}\mathbf{t}_1/\mathbf{t}_1^T\mathbf{t}_1$ and the **X** matrix is deflated to give residuals $\mathbf{X}_2 = \mathbf{X} - \mathbf{t}_1\mathbf{p}_1^T$. The second latent variable is then computed as $t_2 = \mathbf{w}_2^T\mathbf{x}$ where $\mathbf{w}_2$ is the first eigenvector of $\mathbf{X}_2^T\mathbf{Y}\mathbf{Y}^T\mathbf{X}_2$ and so on. As in PCA the new latent vectors or scores ($\mathbf{t}_1, \mathbf{t}_2, ...$) and the loading vectors ($\mathbf{w}_1, \mathbf{w}_2, ...$) are orthogonal. For large ill-conditioned data sets, it is usually convenient to calculate the PLS latent variables sequentially via the NIPALS algorithm (Geladi and Kowalski, 1986) and to stop based on cross-validation criteria.

3.4 Analysis of Historical Process Data Sets

Although massive amounts of process data are being collected and stored in databases for most industrial processes, very little analysis and interpretation of these data is being performed. This is because of the overwhelming size of the databases and the very ill-conditioned nature of the routine operating data being collected. Furthermore, the signal-to-noise ratio is often poor in these data, and there are often significant amounts of missing data. However, all these problems are well addressed by the multivariate statistical projection methods of PCA and PLS. By examining the behaviour of the process data in the projection spaces defined by the small number of latent variables ($t_1, t_2, ..., t_A$), and interpreting process movements in this reduced space by examining the corresponding space defined by the loading vectors ($\mathbf{p}_1, \mathbf{p}_2, ..., \mathbf{p}_A$) or ($\mathbf{w}_1, \mathbf{w}_2, ..., \mathbf{w}_A$) in the case of PLS, it is often possible to extract very useful information from these databases, and to use this information to improve the process.

There are several interesting examples of using these methods to analyze process data. Denney et al. (1985) applied the methods to the analysis of an industrial sulphur recovery unit. Moteki and Arai (1986) used them to analyse a low-density polyethylene process and to find new operating conditions. Wise et al. (1991) applied PCA to analyze and diagnose systematic variations in the behaviour of a slurry-fed ceramic melter process. Slama (1991) used PCA and PLS to analyze data on more than 300 process variables and 11 product grades from the fluidized bed catalytic cracking and fractionation section of a refinery. Skagerberg et al. (1992) applied PLS to predict polymer properties from measured temperature profiles in a tubular low-density polyethylene process, and to interpret the behaviour of this process. Dayal et al. (1994) used PLS to model the dynamic behaviour of a continuous Kamyr digester in a pulp mill, and diagnosed the reasons for poor control of Kappa number by examining the loading plots ($\mathbf{w}_1$, $\mathbf{w}_2$).

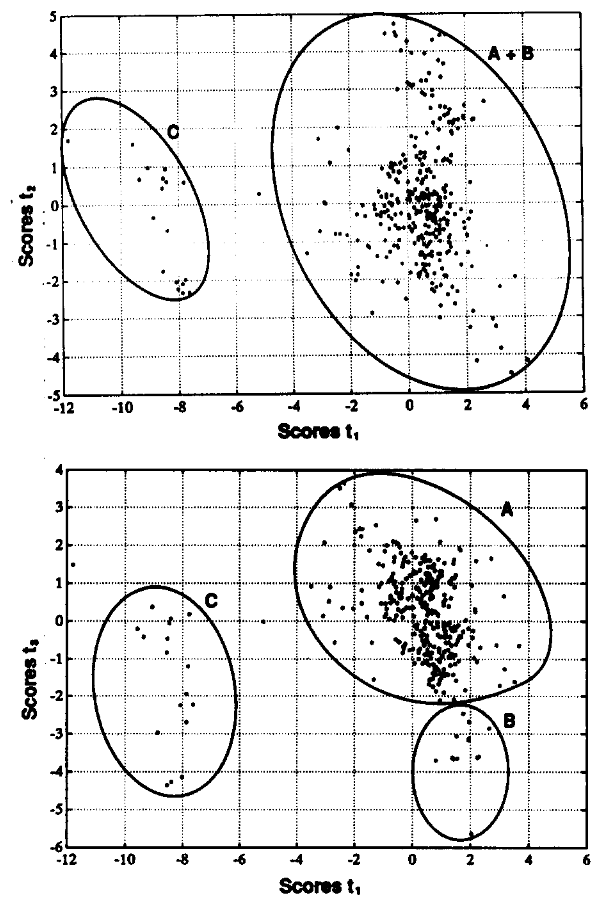

Figure 2. PLS scores plots for several hundred hours of operation of the rougher scavenger unit of a mineral flotation process. t_1 vs t_2, and t_1 vs t_3. *(Hodouin et al. (1993); With permission from CIM Bulletin).*

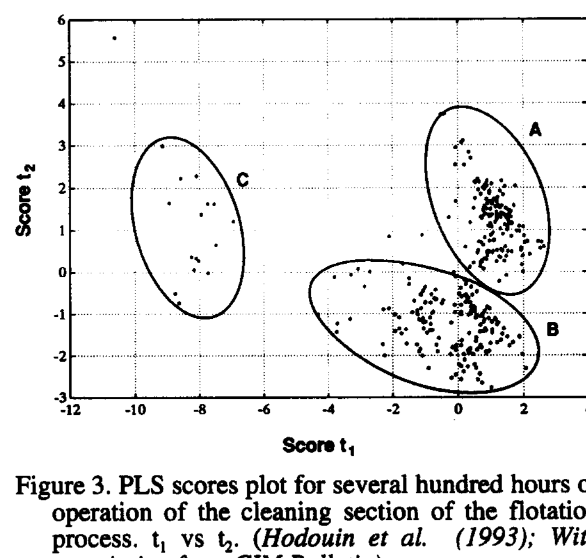

Figure 3. PLS scores plot for several hundred hours of operation of the cleaning section of the flotation process. t_1 vs t_2. *(Hodouin et al. (1993); With permission from CIM Bulletin).*

Hodouin et al. (1993) used PCA and PLS to analyze and interpret the behaviour of mineral flotation and grinding circuits in a large mineral processing plant. Historical data from 350 hours of operation were retrieved from computer files. The difficulty with such massive data sets is first to find out where in the data there is useful information. The projections of hourly average data (after gross outliers were removed) into the plane defined by the first three latent variables are shown in Figure 2 for the rougher scavenger (RS) unit and in Figure 3 for the cleaner regrinding (CR) unit. The data points appear to cluster into 3 distinct regions which corresponded to different operating conditions. Simply examining the individual plots of the process variables would be confusing and would not reveal such information. By focusing attention on the transitions between the regions one can learn most of what there is to know about the 350 hours of operation. To help diagnose the reasons for these shifts in process operation, one can interrogate the underlying multivariate model (as discussed below in Section 4.2) and display the process variable contributions to these shifts. Cluster C for example corresponds to low copper in the feed, low RS concentrate flowrate, low lead recovery, high frother and low promoter feedrate.

4. MULTIVARIATE STATISTICAL PROCESS CONTROL

The main approach of statistical quality control (SQC) methods developed throughout the statistical literature has been to monitor only product quality data (**Y**). However, in these approaches, all of the data on the process variables (**X**) are being ignored. If one truly wants to do Statistical Process Control (SPC), one must look at all of these process data as well. There are often hundreds of process variables, and they are measured much more frequently (and usually more accurately) than the product quality data (**Y**). Furthermore, any special events which occur will also have their fingerprints in these process data (**X**). Sometimes product quality is only determined by the performance of the product later, in another process (e.g. catalyst conditioning- performance of catalyst is assessed later in polymer production). It would be useful to know if the product is good before using it; monitoring the process would help in the early detection of poor-quality product.

There are several other reasons why monitoring the process is advantageous. Sometimes, only a few properties of the product are measured, but these are not sufficient to define entirely the product quality. For example, if only rheological properties of a polymer are measured, any variation in end-use application that arise due to variation in chemical structure (branching, composition, end-group concentration) will not be captured by following only product properties. In these cases the process data may contain more information about events with special causes that may affect the product quality (product performance).

Finally, even if product quality measurements are frequently available, monitoring the process may help in diagnosing assignable causes for an event. When monitoring product quality, even if it is determined which quality variable caused the multivariate chart to go out of limits, it may still be difficult to determine what went wrong in the process. Several combinations of process conditions may cause the same product property to change. Monitoring the process would help identify one combination of process variables and therefore determine the underlying cause more easily.

Certainly, one could apply the previously discussed SQC charting methods directly to the x variables as well (Kourti and MacGregor, 1994). However, as discussed previously, with large numbers of highly correlated variables, these methods are impractical. Furthermore, they offer no way of relating the **X** and **Y** data, and least-squares regression analysis is also impractical in this situation. Another problem is that these methods cannot handle missing data arising from sensor failure, etc.. The most practical approaches to multivariate SPC appear to be those based on multivariate statistical projection methods such as PCA and PLS. The methods are ideal for handling the large number of highly correlated and noisy process variable measurements that are being collected by process computers on a routine basis; these methods can also handle missing data.

4.1 Monitoring Continuous Processes

An essential part of SPC is to establish multivariate control charts to detect special events as they occur, and to diagnose possible causes for them while the information is fresh. The philosophy applied in developing multivariate SPC procedures based on projection methods, is the same as that used for the univariate or multivariate Shewhart charts. An appropriate reference set is chosen which defines the normal operating conditions for a particular process. In other words, a PCA or PLS model is built based on data collected from various periods of plant operation when performance was good. Any periods containing variations arising from special events that one would like to detect in the future are omitted at this stage. The choice of this reference set is critical to the successful application of the procedure, as discussed by Kresta et al. (1991).

The multivariate control chart is now a T^2-chart on the first A latent variables (equation (15)). Added to this is a chart on SPE_x where:

$$SPE_x = \sum_{i=1}^{m} (x_{new,i} - \hat{x}_{new,i})^2 \qquad (18)$$

where $\hat{x}_{new}$ is computed from the reference PLS or PCA model. This latter plot will detect the occurrence of any new events which cause the process to move away from the hyperplane defined by the reference model. Control limits for the T^2 charts are chosen in the same manner as previously discussed, and the UCL on SPE_x is based on the Chi-squared approximation (Q-statistic (Jackson, 1991); Nomikos and MacGregor, 1995).

The main concepts behind the development and use of these multivariate SPC charts for monitoring continuous processes were laid out by Kresta et al. (1991), Wise et al. (1991), Wise and Ricker (1991), and MacGregor et al. (1991a, b). Several illustrations of the methods were also presented in those papers, along with the algorithms and details on estimating control limits.

The basic approach is illustrated here with the monitoring of a simulated multi-section high-pressure tubular reactor process for the manufacture of low-density polyethylene (LDPE) (MacGregor et al., 1994a). Simulated data were generated for this study using the models described in (Kiparissides et al., 1993). Measurements are available on a frequent basis on all process variables (X) - reactor temperature profiles in each section, feedrates on all component streams, cooling system flows and temperatures, and pressures in each reactor section. Every hour, measurements are available on product quality and productivity (Y) - polymer molecular weights and branching properties, and conversion of monomer to polymer. Using data collected (**X, Y**) when the process was operating well, and no special events were present, a PLS model using only three latent variables (A = 3) was able to explain 90.0 % of the variation in the Y data.

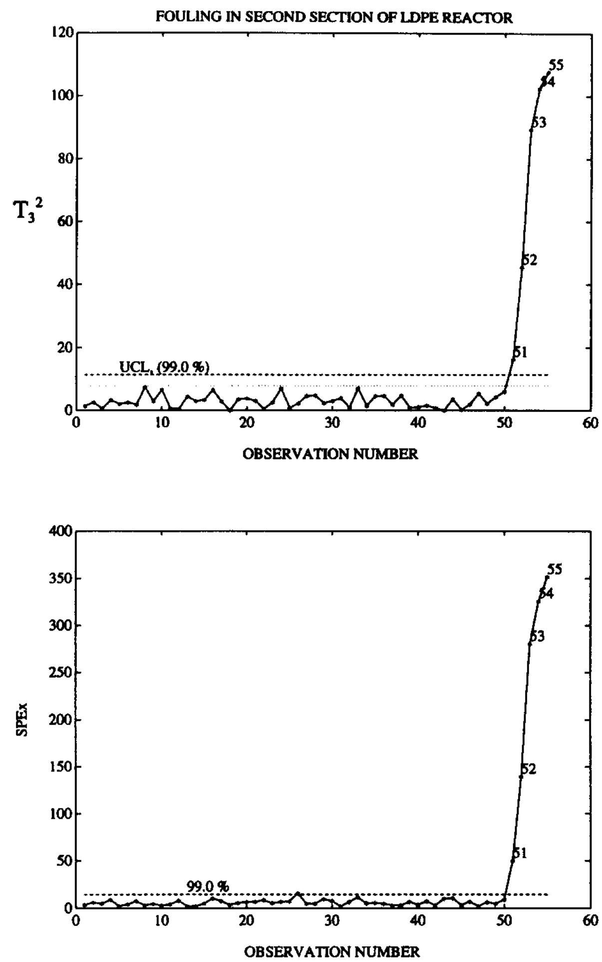

Figure 4. T^2 Chart on 3 scores and squared prediction error chart for monitoring a LDPE process. Points 51 - 55 denote a period where fouling gradually occurred in the second zone of the reactor.

Figure 4 illustrates the use of a T^2_A (A=3) chart and an SPE_x chart to monitor the behaviour of the reactor when there is an increasing level of fouling in the second section of the reactor. Unnumbered points indicate past conditions of normal operation. Fouling starts at point 51. Notice that both the T^2_3 and the squared prediction error plots quickly detected the onset of this special event and alarmed an out-of-control situation, on-line, before lab data on product quality became available.

4.2 Fault Diagnosis

Both univariate and multivariate SPC charts are based on statistical tests to detect any deviations from the in-control reference distribution upon which the models and charts have been built. In classical quality control approaches which chart only quality variables, once an out-of-control signal has been given, it is then left up to the process operators and engineers to try to diagnose an assignable cause using their process knowledge and a one-at-a-time inspection of process variables. However, multivariate charts based on PLS or PCA provide a much greater capability for diagnosing assignable causes. By interrogating the underlying PLS or PCA model at the point where an event has been detected, one can extract diagnostic or contribution plots which reveal the group of process variables making the greatest contributions to the deviations in the SPE_x and the scores (Miller et al., 1993; MacGregor et al., 1994a; Kourti and MacGregor, 1994; Wise and Ricker, 1991). Although these plots will not unequivocally diagnose the cause, they will provide much greater insight into possible causes and thereby greatly narrow the search.

Consider the out-of-control alarms shown in Figure 4 for the LDPE process. Diagnostic plots showing the contribution of the process variables to the SPE_x at point 51 are shown in Figure 5. These contribution plots point to the temperature of the reaction mixture at the exit from zone 2 and the temperature of the cooling agent into the jacket of the second zone as being the main process variables that are showing inconsistency (and contributing to the large values of SPE_x). This combination of variables would imply heat transfer problems and could lead the operator to suspect fouling.

Diagnostic plots can also be constructed for the variable contribution on the scores. If the SPE_x is within limits and T_A^2 out of limits, contribution plots on the scores (t_1, or t_2, ..., or t_A) would indicate combinations of process variables that contributed to out of normal values for the scores. (The detection of the score(s) responsible for the out of limits signal for T_A^2 is discussed in (Kourti and MacGregor, 1994)).

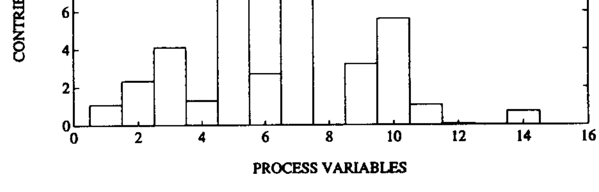

Figure 5. Contribution plot showing the process variable contributions to the SPEx for point 51.

4.3 Multi-block PLS

The monitoring and diagnostic charts discussed in the previous sections may be difficult to interpret when the number of variables included in the X space is very large. The combined use of multi-block PLS (MB-PLS) and contribution plots may facilitate this task. In the MB-PLS approach, large sets of process variables (X) are broken into meaningful blocks; usually each block corresponds to a process unit, or a section of a unit.

Multivariate monitoring charts for important subsections of the plant, as well as for the entire process, can be constructed. The principles behind multi-block data analysis methods and their algorithms can be found in (Wold, 1982) and (Wangen and Kowalski, 1988). Details on the application of multi-block PLS for process monitoring and the corresponding algorithm can be found in (MacGregor et al., 1994a) where MB-PLS is used for monitoring and diagnosing an LDPE reactor. Each block corresponds to one zone. Plots of t_1 vs t_2 and SPE, obtained for each block of the process, were utilized to detect an abnormal event in the zone it occurred in; then contribution plots were successfully used to assign causes for it.

Multiblock PLS is not simply a PLS between each X block and Y. The blocks are weighted in such a way that they are most predictive of Y. The monitoring space is determined by one model rather than separate models for each block. If there are time delays between process units the data can be time shifted to accommodate for the delays.

4.4 Data Compression

A major difficulty that often arises in computer databases is that univariate data compression methods are used to minimize the amount of data that needs to be stored. These methods usually destroy the essential multivariate nature of the process data, and eliminate much of the useful information. A much more useful method of data compression, and one that would retain the true multivariate nature of the data, would be to store the scores of the first A latent variables ($\mathbf{T}_A$) and the loading matrix ($\mathbf{P}_A$). From this, the original variables can always be reconstructed as $\hat{\mathbf{X}} = \mathbf{T}_A\mathbf{P}_A^T$ as long as no special event occurs that is not predicted by the model. In this latter case, the SPC detection scheme would alarm such an event, and the data during this period should not be compressed, but retained in its entirety for analysis and fault diagnosis. By employing multivariate SPC schemes based on PCA and PLS as discussed earlier, these latent vectors will be computed in real-time, and hence such a data-compression scheme is already included within the SPC scheme.

4.5 Multivariate SPC for Batch Processes

In much of the specialty chemical, pharmaceutical and other manufacturing industries, batch processes are used extensively. The use of the multivariate statistical projection methods has been extended to the analysis and the on-line monitoring and diagnosis of batch processes (MacGregor and Nomikos, 1992; Nomikos and MacGregor, 1994a, 1994b, 1995; MacGregor et al., 1994b). Typical data from batch processes include time-varying trajectories of all the measured process variables throughout the duration of each batch (X), product quality measurements (Y) at the end of each batch, and batch recipe and charge conditions (Z) at the start of each batch. If such data are available in a historical database on many past batches, multivariate PCA and PLS models can be developed for analyzing these historical batches and for establishing on-line SPC charts for monitoring the progress of each new batch. Since the process data (**X**) is now a three-dimensional array (batch run × variable × time), Nomikos and MacGregor used three-dimensional or multi-way PCA (MPCA) and PLS (MPLS) methods. Multi-way PCA and PLS methods have been discussed in a series of articles (Lohmöller and Wold, 1980; Wold et al., 1987; Geladi, 1989; Smilde and Doornbos, 1991). Nomikos and MacGregor proposed approaches for handling the fact that one dimension (time) is evolving during the progress of a new batch, and for establishing control limits on the multivariate SPE and score plots. In their approach the X matrix is manipulated in such a way, that the non-linear trajectory of each variable is subtracted upon meancentering, thus converting a non-linear problem to one that can be tackled with linear methods such as PCA and PLS.

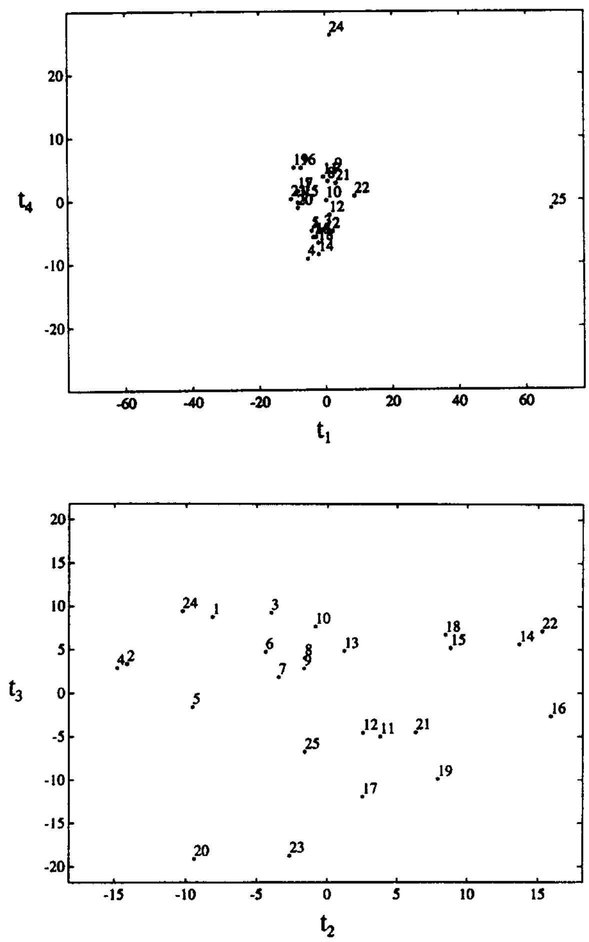

Figure 6. Plots of t_1 - t_4 and t_2 - t_3 for 25 batches. Batches #24 and #25 have produced products with unusual properties.

The application of MPCA to monitoring batch processes is illustrated here with an example. Data from 25 batches from an industrial polymerization reactor were provided. Four quality properties (y_1, y_2, y_3, y_4) are used to characterize the product and measurements on these properties become available several hours after the production of the polymer. For each batch, the quality measurements and the corresponding trajectories of 10 process variables for 90 time intervals were provided. (After proper alignment of the data 83 time intervals were used.) Two batches were characterized as "bad" by the producer. For one of the batch products the value of y_1 was lower than usual while for the other the values for y_2, y_3 and y_4 were unusually high. In a preliminary analysis, MPCA was performed on all the batches (i.e., on the three - way array **X** with dimensions 25×10×83), to test if the method would be able to discriminate between "good" and "bad" batches with the available process data; in other words to assess if the system was *observable*. Figure 6 shows the projections of the process conditions of these 25 batches on the score planes (t_1 - t_4) and (t_2 - t_3) defined by the 4 first principal components. It can be seen that in the (t_1 - t_4) plot, batches #24 (low y_1) and #25 (unusual y_2, y_3, y_4) are out of the main cluster (or, normal operating region) formed by the rest of the batches. It is interesting to see that the problems in these two batches appear in only two scores and that these are different scores, which means that different combinations of variables affect different properties (t_4 for y_1, and t_1 for y_2, y_3, y_4).

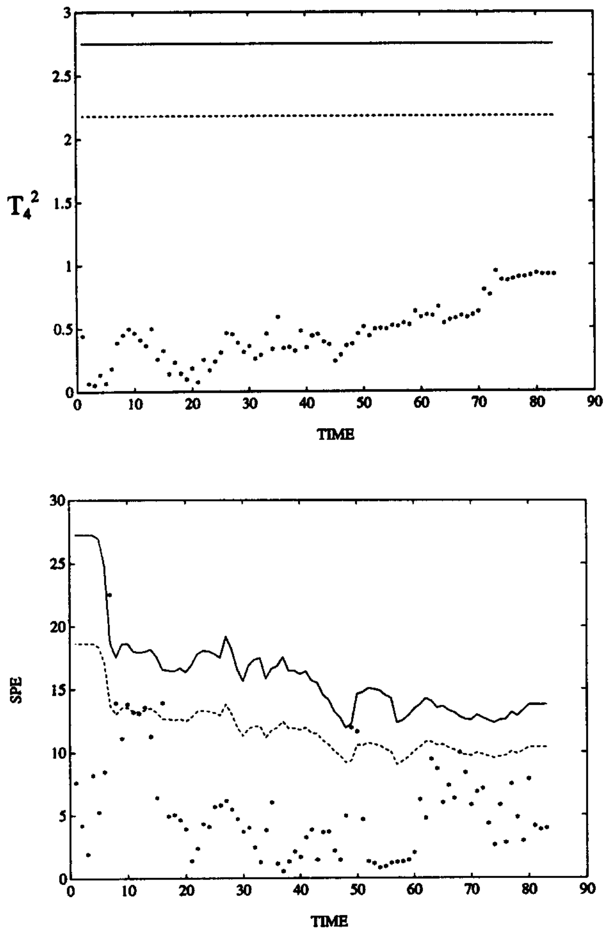

Figure 7. Monitoring a Good Batch. T_A^2 statistic and SPE_x.

Having established the observability of faults with the analysis of past data, a model was built to summarize the information contained in the 23 good batches about the normal operating region of the process. This model was then used as statistical reference to classify new batches as normal ("good") or abnormal ("bad"), in the way described in (Nomikos and MacGregor, 1994a, 1995). New batches are classified by monitoring the T_A^2 statistic calculated from the first A latent variables (scores) and by monitoring SPE at each time interval k; when both of these quantities stay within the limits of normal operation (specified by the model) then the batch is accepted as good.

Figure 7 shows the SPE response as a function of time, and the T_A^2 (A=4) statistic of a batch that was eventually classified as "good". Notice that both of these quantities remain well within the confidence intervals throughout the batch. (The solid line corresponds to a 99% limit and the dashed line to a 95% limit for the SPE plots; for T_A^2 the solid line corresponds to 95 % limit and the dashed to 90 % limit.)

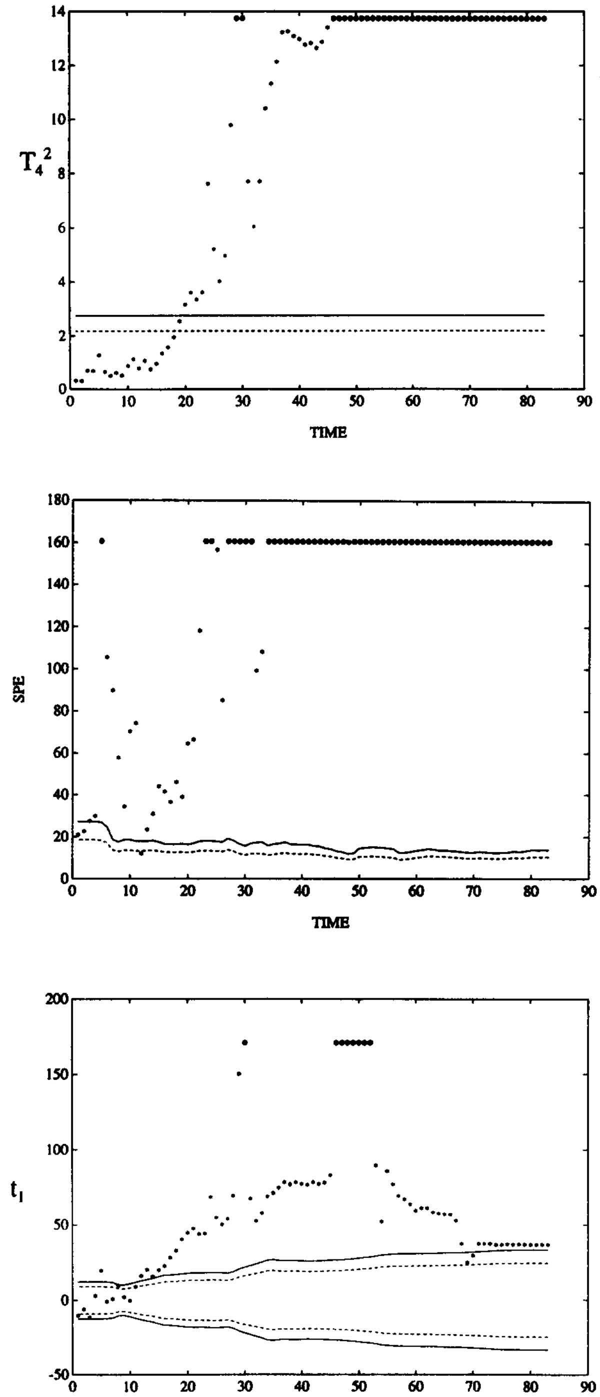

Figure 8. Monitoring Batch #25 (a bad batch). T_A^2 statistic, SPE_x and of t_1 for the duration of the batch.

Figure 8 shows how batch #25 would have behaved, had the model been in use on-line, when the data for this batch were becoming available. (Points indicated by an asterisk in a circle, plot out of the co-ordinate limits of the graph). Notice that almost all the points are out of the 95% limit in the SPE, while the T_4^2 statistic goes out of limits after 15 time intervals into the batch run. (Similarly for batch #24 the two statistics moved out of their limits early in the run). A plot of t_1 versus time for batch #25 reveals that this latent variable goes out of limits after few time intervals into the run. Note that all of these charts quickly revealed that batches 24 and 25 were "unusual" very early in the runs. Upon diagnosis of the nature of the fault, corrective action might be taken. If such action to save the batch were not possible, at least the product from the batch could be isolated and not blended with good product.

This was a feasibility study for this on-going project. Usually more good batches (more than 50) are required in order to obtain a representative sample of sufficient size to correctly estimate the confidence limits for the normal operating region.
The proposed monitoring charts are in accordance with the SPC requirements in that they can be easily displayed and interpreted, and they can quickly detect a fault. Furthermore, it is also possible to provide the operators with diagnostic information by interrogating the underlying MPCA, MPLS or multi-way multiblock PLS model. Other industrial applications of these methods have been reported for the analysis of historical batch databases by Kosanovich et al. (1994), and for the monitoring of a different batch polymerization by Nomikos and MacGregor (1995).

5. PRACTICAL ISSUES

Several issues that arise when applying these methods to industrial data are briefly discussed here.

5.1 Reference Data Set for Modelling

When dealing with empirical modelling the data set upon which the model will be based should be carefully chosen to satisfy the needs of the intended application.

Inferential Modelling. When the objective is to infer the values of product quality (Y) from process data obtained at different operating regions, then the set of data that will be used for modelling should have representative values of the Y properties and the process variables over all the possible operating regions. (Values for Y should cover the whole range of specifications of the quality properties and data points should be evenly distributed.) Ideally, for inferential purposes, a designed set of experiments with data from different operating regions properly weighted, would be required.

Historical Analysis. When analyzing historical data, initially all the data should be used. If the projection indicates clusters with only a few points in them, or individual outliers, these data should not be discarded, but investigated. True outliers (measurement errors) should be discarded. If there are clusters of only a few points that are identified as reflecting some real, unusual event, then more data points are needed in this region to model the behaviour of these events.

Model For Monitoring Purposes. In monitoring, a specific operating region of interest is tackled. Only data corresponding to good / acceptable product, or acceptable conditions should be included; faults or disturbances are excluded from this model. The objective is to model good operating behaviour only, and to test for any future deviations from this model.

5.2 Linear vs Nonlinear Models

It has been argued that nonlinear models may be necessary to model batch or continuous processes. Again, the model depends on the application. If the model is used for monitoring, linear models are usually sufficient to describe process fluctuations around an operating point. Although the behaviour of the process variables is nonlinear in batch processes, the data are meancentered in the method discussed in Section 4.5 in such a way that the non-linear trajectory is subtracted and the deviations of the variables from the trajectory can be modelled with a linear model.

If the models are build for inferential purposes and cover a wide range of operating regions, then nonlinear models may be necessary. Nonlinear versions of PLS have been reported (Frank, 1990; Wold, 1992; Höskuldsson, 1992).

5.3 Dynamic Models

The projection methods discussed here are capable of dealing with dynamic situations. In batch monitoring dynamics has already been accounted for as illustrated in Section 4.5.

When modelling continuous dynamic processes, lagged variables of x and y variables can be included in the X matrix. Multivariate time series analysis is discussed for PCA by Jollife (1986) and Jackson (1991) and for PLS by Wold et al. (1984) and MacGregor et al. (1991c). Time delays are accounted for by time shifting. An industrial example where plant data was both time shifted, to account for time delays between X and Y, and lagged to account for autocorrelations in Y, is described in (Dayal et al., 1994).

5.4 Single or Multiple Models for Quality Properties

One of the advantages of PLS over linear regression is that all the quality properties (Y) can be modelled together and related to X in a single model. When the quality properties are not correlated, it is customary to built a model that relates X to each y variable separately. This approach is satisfactory in general if the model is just being used for calibration, inferential control or prediction. For monitoring purposes, however, since quality is a multivariate property, it is important to fit all the variables from the Y space in a single model in order to obtain a single low-dimensional monitoring space.

6. SUMMARY

This paper has provided justifications for the use of multivariate statistical process control, and has reviewed some of the traditional statistical control charting methods for monitoring product quality data (Y). However to truly perform multivariate statistical process control, one must utilize not just the final product data (Y) but all the data on process variables (X) that are being routinely collected by process computers. SPC approaches based on multivariate statistical projection methods (PCA and PLS) have been developed for this purpose. The ideas behind these new approaches, and the literature on them, are reviewed. Multivariate control charts in the projection spaces provide powerful methods for both detecting out-of-control situations, and diagnosing assignable causes, and they are applicable to both continuous and batch processes. The only requirement for applying these methods is the existence of a good database on past operations. For this reason, they have attracted wide interest, and are rapidly being applied in many industries.

REFERENCES

Alt, F.B. (1977). *Economic design of control charts for correlated, multivariate observations*. Ph.D. dissertation, Georgia Institute of Technology.

Alt, F.B. (1985). Multivariate quality control. In S. Kotz and N.L. Johnson (Eds.), *Encyclopedia of Statistical Sciences* Vol. 6, pp. 110-122. Wiley, New York.

Alt, F.B. and N.D. Smith (1988). Multivariate process control. In P.R. Krishnaiah and C.R. Rao (Eds.), *Handbook of Statistics,* Vol. 7, pp. 333-351. North-Holland, Amsterdam.

Crosier, R.B. (1988). Multivariate generalizations of cumulative sum quality-control schemes. *Technometrics,* **30,** 291-303.

Dayal, B., J.F. MacGregor, P.A. Taylor, R. Kildaw and S. Marcikic (1994). Application of feedforward neural networks and partial least squares regression for modelling Kappa number in a continuous Kamyr digester. *Pulp and Paper Canada*, **95**, No. 1, 26-32.

Denney, D.W., J. MacKay, T. MacHattie, C. Flora and E. Mastracci (1985). Application of pattern recognition techniques to process unit data. *CSChE Conference*, Sarnia, Ontario, Canada.

Downs, J.J. and J.E. Doss (1991). Present status and future needs - A view from North American industry. In Y. Arkun and H. Ray, (Eds.), *AIChE Symposium Proceedings of the Fourth International Conference on Chemical Process Control, Padre Island, TX, February 17-22*, AIChE Publication, No. P-67, New York.

Frank, I.E. (1990). A nonlinear PLS model. *Chemometrics and Intelligent Laboratory systems.* **8**, 109-119.

Fuchs, C. and Y. Benjamini (1994). Multivariate profile charts for statistical process control. *Technometrics*, **36**, 182-195.

Geladi, P. and B.R. Kowalski (1986). Partial least-squares regression: A tutorial. *Anal. Chim. Acta*, **185**, 1-17.

Geladi, P., (1989). Analysis of multi-way (multi-mode) data. *Chemometrics and Intelligent Laboratory Systems*, **7**, 11-30.

Healy, J.D. (1987). A note on multivariate CUSUM procedures. *Technometrics*, **29**, 409-412.

Hodouin, D., J.F. MacGregor, M. Hou and M. Franklin (1993). Multivariate statistical analysis of mineral processing plant data. *CIM Bulletin Mineral Processing*, **86** (975), 23 - 34.

Höskuldsson, A. (1988). PLS regression methods. *J. Chemometrics*, **2**, 211-228.

Höskuldsson, A. (1992). Quadratic PLS regression. *J. Chemometrics*, **6**, 307-334.

Hotelling, H. (1947). Multivariate quality control, illustrated by the air testing of sample bombsights. In C. Eisenhart, M.W. Hastay, and W.A. Wallis (Eds.), *Techniques of Statistical Analysis*, pp. 113-184. McGraw-Hill, New York.

Hunter, J.S. (1986). Exponentially weighted moving average. *J. Qual. Technol.*, **18**, 203-210.

Jackson, J.E. (1991). *A User's Guide to Principal Components*, John Wiley and Sons, Inc., New York.

Jollife, I.T. (1986). *Principal Component Analysis,* Springer Series in Statistics, Springer Verlag, New York.

Kiparissides, C., G. Veros, G. Kalfas, M. Koutoudi and C. Kantzia. (1993). A comprehensive mathematical model of a multizone tubular high pressure LDPE reactor. *Chem. Eng. Communications*

Kosanovich, K.A., M.J. Piovoso, K.S. Dahl, J.F. MacGregor and P. Nomikos (1994). Multi- way PCA applied to an industrial batch process. *Proceedings, American Control Conference 1994, Baltimore MD.* Volume 2, pp. 1294 - 1298.

Kourti, T. and J.F. MacGregor (1994). Recent developments in multivariate SPC methods for monitoring and diagnosing process and product performance. *Submitted to the Journal of Quality Technology.*

Kresta, J., J.F. MacGregor and T.E. Marlin (1991). Multivariate statistical monitoring of process operating performance. *Can. J. Chem. Eng.*, **69**, 35-47.

Lohmöller, J. and Wold, H. (1980). Presented at the *European Meeting of Psycometrics Society*, Gronigen, Netherlands.

Lowry, C.A., W.H. Woodall, C.W. Champ, and S.E. Rigdon (1992). A multivariate exponentially weighted moving average control chart. *Technometrics*, **34**, 46-53.

MacGregor, J.F., T.E. Marlin, J. Kresta and B. Skagerberg (1991a). Multivariate statistical methods in process analysis and control. In Y. Arkun and H. Ray, (Eds.), *AIChE Symposium Proceedings of the Fourth International Conference on Chemical Process Control, Padre Island, TX, February 17-22*, AIChE Publication, No. P-67, pp. 17-22, New York.

MacGregor, J.F., B. Skagerberg and C. Kiparissides (1991b). Multivariate statistical process control and property inference applied to low density polyethylene reactors. *IFAC Symp. ADCHEM'91, Toulouse, France, Oct. 1991.*, pp. 131-135, Pergamon Press.

MacGregor, J.F., T. Kourti and J. Kresta (1991c). Multivariate Identification: A study of several methods. *IFAC International Symposium, ADCHEM'91, (Toulouse, France) Proceedings,* Pergamon Press, pp. 369-375.

MacGregor, J.F. and P. Nomikos (1992). Monitoring batch processes. In Reklaitis, Rippin, Hortacsu and Sunol (Eds.), *Proceedings NATO Advanced Study Institute (ASI) for Batch Processing Systems Eng., May 29-June 7, 1992, Antalya, Turkey,* Springer-Verlag, New York.

MacGregor, J.F., C. Jaeckle, C. Kiparissides and M. Koutoudi (1994a). Monitoring and diagnosis of process operating performance by multi-block PLS methods with an application to low density polyethylene production, *AIChE* J., **40**, 826-838.

MacGregor, J.F., P. Nomikos and T. Kourti (1994b). Multivariate statistical process control of batch processes using PCA and PLS. In *Preprints IFAC, ADCHEM 1994, Kyoto, Japan*. pp. 525-530.

Mardia, K.V., J.T. Kent and J.M. Bibby (1989). *Multivariate Analysis.* Academic Press, London.

Miller, P., R.E. Swanson and C.F. Heckler (1993). Contribution plots: The missing link in multivariate quality control. Presented at the *37th Annual Fall Conf. ASQC, Rochester, NY.*

Moteki, Y. and Y. Arai (1986). Operation planning and quality design of a polymer process. *Proc. IFAC Symp., DYCORD-86, Bournemouth, UK*, pp. 159-166.

Nomikos, P. and J.F. MacGregor (1994a). Monitoring of batch processes using multi-way principal component analysis. *Amer. Inst. Chem. Eng.* J.,**40**, 1361-1375.

Nomikos, P. and J.F. MacGregor (1994b). Multi-way partial least squares in monitoring batch processes. *First International Chemometrics Internet Conference*. September 1994.

Nomikos, P. and J.F. MacGregor (1995). Multivariate SPC charts for monitoring batch processes. *Technometrics*, **37**(1).

Pignatiello, J.J., Jr. and G.C. Runger (1990). Comparisons of multivariate CUSUM charts. *Journal of Quality Technology*, **22**, 173-186.

Ryan, T.P. (1989). *Statistical Methods for Quality Improvement,* Wiley, New York.

Roberts, S. W. (1959) Control chart tests based on geometric moving averages. *Technometrics*, **1**, 239-250.

Shewhart, W.A. (1931). *Economic Control of Quality of Manufactured Product.* Van Nostrand, Princeton, N.J.

Skagerberg, B., J.F. MacGregor and C. Kiparissides (1992). Multivariate data analysis applied to low-density polyethylene reactors. *Chemometrics and Int. Lab. Syst.*, **14**, 341-356.

Slama, C.F. (1991). *Multivariate statistical analysis of data obtained from an industrial fluidized catalytic process using PCA and PLS.* M.Eng. Thesis, Department of Chemical Engineering, McMaster University, Hamilton, Ontario, Canada.

Smilde, A.K., and D.A. Doornbos. (1991). Three-way methods for the calibration of chromatographic systems: comparing PARAFAC and three-way PLS. *Journal of Chemometrics,* **5**, 345-360.

Sparks, R.S. (1992). Quality control with multivariate data. *Austral. J. Statist.*, **34**, 375-390.

Tracy, N.D., J.C. Young, and R.L. Mason (1992). Multivariate control charts for individual observations. *Journal of Quality Technology*, **24**, 88-95.

Wangen, L.E. and Kowalski, B. (1988). A multiblock partial least squares algorithm for investigating complex chemical systems. *J. Chemometrics,* **3**, 3-20.

Wierda, S. J. (1994). Multivariate statistical process control - recent results and directions for future research. *Statistica Neerlandica*, **48 (2)** 147-168.

Wise, B.M., D.J. Veltkamp, N.L. Ricker, B.R. Kowalski, S. Barnes and V. Arakali (1991). Application of multivariate statistical process control (MSPC) to the West Valley slurry-red ceramic melter process. *Waste Management '91 Proc., Tucson, Arizona.*

Wise, B.M. and N.L. Ricker (1991). Recent advances in multivariate statistical process control improving robustness and sensitivity. *IFAC Symp. ADCHEM'91, Toulouse, France, Oct. 1991.*, pp. 125-130, Pergamon Press.

Wold, S. (1978). Cross-validatory estimation of the number of components in factor and principal components model. *Technometrics,* **20** (4), 397-405.

Wold, H. (1982). Soft Modelling. The basic design and some extensions. In K.G. Joreskog and H. Wold (Eds)., *Systems under Indirect Observation, 2.*, Chapter 1, North Holland, Amsterdam.

Wold, S., C. Albano, W.J. Dunn III, U. Edlund, K. Esbensen, P. Geladi, S. Hellberg, E. Johansson, W. Lindberg and M. Sjöström (1984). Multivariate Data Analysis in Chemistry. In: *Chemometrics. Mathematics and Statistics in Chemistry* (B. Kowalski, Ed.), NATO - ASI, D. Reidel Publishing Company, pp. 17-95.

Wold, S., P. Geladi, K. Esbensen, and J. Ohman (1987). Multi-way principal components and PLS analysis. *Journal of Chemometrics,* **1**, 41-56.

Wold, S. (1992). Nonlinear partial least squares modelling. II Spline inner relation. *Chemometrics and Intelligent Laboratory Systems*, **14,** 71-84.

Woodward, R.H., and P.L. Goldsmith (1964). *Cumulative Sum Techniques.* Oliver and Boyd. London.