QUALITY CONTROL FOR BATCH PROCESSES

USING MULTIVARIATE LATENT VARIABLE METHODS

By

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Abstract

The main goal in many processes is to obtain consistent and reproducible operation and end-quality properties. In this thesis the problem of product quality control in batch and semi-batch processes is addressed. Unlike from much of the published literature that uses first principles models, this thesis studies the end-quality feedback control problem using only empirical Partial Least Squares (PLS) models. Several simple, practical and effective regulatory control strategies are proposed.

The thesis consist of four main chapters: *i*) On-line control of a distributed end quality property (particle size distribution, PSD) using mid-course correction strategies (MCC), *ii*) an inferential-adaptive control approach that combines on-line and batch-to-batch control, *iii*) a novel reduced dimensional space control algorithm to obtain complete manipulated variable trajectories (MVT) consistent with past operation, and *iv*) incorporation of prior batch-to-batch information for batch analysis and monitoring.

In the first section, three on-line empirical MCC strategies are proposed for the control of bimodal PSDs in emulsion polymerization systems. The performance of the control strategies is evaluated using a detailed theoretical simulator. Control is applied only when the predicted properties falls outside a statistically defined "no-control" region. Each control strategy corresponds to a control objective: *i*) Control of second mode of the distribution, *ii*) control of the full bimodal PSDs and *iii*) control of relative distributions. Advantages and disadvantages of each one of the control strategies are discussed.

In the second part a combined on-line and batch-to-batch control strategy is presented. The approach extends MCC strategies used before to include multiple decision and correction points, batch-to-batch information to reject batch-wise correlated

disturbances, and an adaptive PLS approach to update the models from batch-to-batch to overcome model error, changing process conditions and unknown disturbances. The methodology is also illustrated with the control of PSD in emulsion polymerization. The problem of regulation about a fixed set-point PSD in the face of disturbances, and the problem of achieving new set-point PSDs are both illustrated.

In the third part a novel strategy for controlling end-product quality properties by solving on-line for complete MV trajectories, for the remainder of the batch, is presented. Control through the optimal solution for complete trajectories using empirical models is achieved by performing the model inversion and the MVT reconstruction in the reduce space of a latent variable model. The approach is illustrated with a condensation polymerization example for the production of nylon and with data gathered from an industrial emulsion polymerization process.

In the last section an extension of the multi-block multiway Principal Component Analysis (MPCA) and MPLS approaches is introduced to explicitly incorporate batch-to-batch trajectory information. It is shown that the advantage of using information on prior batches for analysis and monitoring is often small. However it can be useful for detecting problems when monitoring new batches in the early stages of their operation. The approach is illustrated using condensation polymerization and emulsion polymerization systems as examples.

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Chapter 1

Introduction

Batch and semi-batch processes are the main manufacturing process in many of the specialty industries such as polymers, pharmaceuticals and bio-chemicals. In these processes, it is often necessary to achieve tight final quality specifications. However, batch process control is difficult because unmeasured disturbances can have a large effect on the end-quality properties, on-line sensors for quality variables (or related quality variables) are rarely available, batch processes are time varying, have finite duration, and the ability to control the final product quality usually decreases as the reaction proceeds.

Disturbances affecting batch processes arise mainly from variations in raw material properties, from impurities and from initialization errors. Large errors in initialization and sequencing can be minimized through a high degree of automation, plant operation by well-trained personnel and feedback control of easily measured variables such as temperature, level and pressure. However, raw material variations and process condition changes may still affect the process variable trajectories and the final product qualities. In this situation, adjustments to the nominal trajectories have to be performed to achieve the desired product qualities.

Several control strategies to obtain these adjustments based on detailed fundamental models have been presented. Although these approaches are shown to be successful for quality control (in simulation and in a few laboratory studies), they have not found large acceptance in industry. This unacceptability comes from the fact that the developing of detailed theoretical models requires accurate description of the physicochemical, kinetic and heat transfer events that takes place during the process

(usually a time consuming and difficult task) and to the fact that to apply feed-back control, frequent measurements on the controlled quality variables are usually required. Moreover, there are situations, in which even if these theoretical models are available they may not be accurate enough to model the effect of subtle (but significant) impurities on the end-qualities due to model and parameter uncertainty and the possible absence of measurements on the disturbances and on the current state of the process.

The main goal of this research is to develop control strategies that use less demanding models and sensors, and yet still allow one to effectively monitor and control final quality properties. In particular, practical control strategies based on easily identifiable empirical models are proposed.

Empirical models obtained from process data (historical data with a few complementary experiments) represent a large advantage when compared to theoretical models because most of the data needed for their identification is usually available or easily obtained. Moreover, to built and maintain an empirical model is relatively easy.

Among the available methods for empirical model identification, latent variable methods such as Principal Component Analysis (PCA) and Partial Least Squares (PLS) are considered. Compared to other empirical methods such as artificial neural networks (ANNs) or multivariable linear regression (MLR), these multivariate statistical methods can effectively handle correlated data, replace missing data, and require only small training sets. These advantages make the proposed control strategies more robust and suitable for implementation.

The outline of the thesis is as follows: in Chapter 2 the important characteristics of batch processes are summarized, and a literature overview of some of the significant contributions made in the batch control area is given. Basic knowledge of empirical PLS model building is also introduced.

In Chapter 3 is shown how with the use of simple within-batch mid-course correction strategies based on easily identifiable empirical PLS models the otherwise

extremely difficult control of a high dimensional end quality property can be achieved. The control strategies are illustrated for the regulatory control of particle size distribution (PSD) in emulsion polymerization using a detailed theoretical simulator.

In Chapter 4 a combined batch-to-batch and on-line control strategy based also on simple empirical models is proposed. The control strategy incorporates information of previous batches on the controllers by exploiting the repetitive nature of the batches and the batch-to-batch correlation of the disturbances. Model parameter updating (batch-to-batch adaptation) is also performed to relax the initial model building requirements, and to overcome model error, changing process conditions and unknown disturbances. The approach is illustrated with the PSD regulation about a fixed set-point in the face of disturbances, and with the problem of achieving new set-point PSDs.

Chapter 5 introduces a novel methodology that allows control of the final product quality by on-line adjusting of complete manipulated variable trajectories (MVT) without increasing the complexity and number of experiments needed to build a model. The method perform the control computation (model inversion and MVT reconstruction) in the reduced latent variable space of an empirical PLS model. The methodology is illustrated in a condensation polymerization system for the control of final average molecular weight and amine concentration. Preliminary results are also shown for an industrial emulsion polymerization process.

Multi-way principal component analysis (MPCA) and multi-way partial least squares (MPLS) are well-established methods in batch analysis and monitoring. However, they often do not explicitly exploit the information available from prior batches. Chapter 6 introduces an extension of these methods to explicitly incorporate batch-to-batch information into batch monitoring. The approach is illustrated using condensation polymerization and emulsion polymerization systems as examples.

Chapter 2

Characteristics, Modeling and Control in Batch Processes

This Chapter presents some of the particular features of batch processes that are important for control and reviews some of the literature contributions to the batch control area. It also presents some of the reasons for using empirical modeling and introduces the basic ideas of Partial Least Squares (PLS).

2.1 Characteristics of batch process

Considerable knowledge and theory have been accumulated in designing, operating and controlling continuous processes. However, such knowledge cannot readily be used in batch/semi-batch operation because such processes have distinctive characteristics, which make that the achievement of end-quality properties and their adequate operation a challenging task. Among these characteristics we have (Bonvin [1998]):

1. Integrating Process. In a batch/semi-batch process, unlike continuous system there is no fixed operating point and therefore there is no static gain or time constant. The chemical reactions proceed from an initial point towards a final stage in such a way that any upset (disturbance) in the initial or in an intermediate condition would definitely affect the subsequent operation. This is because the influence of disturbances is generally cumulative until the end of the process. For example, small impurities in raw materials

can have a large impact on the end product properties. Once a disturbance that affects the final product quality occurs, it is often difficult to introduce remedial corrections.

2. Time specific control action. In batch process, the ability to influence the reaction typically decreases with time, therefore the upsets and disturbances have to be detected early and corrective adjustments have also to be taken at early (or at most at intermediate) stages of the processes in order for the compensation to be adequate. Moreover, in certain type of reactions it is generally required to perform the corrective adjustments at very specific time instants. For example, in order to control particle size distribution in emulsion polymerization the corrective actions need to be applied at the time when the nucleation take place (or before the nucleation is over).

3. Infrequent quality related measurements. Although process variables (such as temperature and pressure) can be easily measured in most reactor systems, on-line sensors for quality variable monitoring (especially those related to distributions and chemical composition) can only be obtained on-line with the aid of expensive instrumentation in a few research laboratories and are generally unavailable in industrial settings. For example, distributed qualities are, in the best of the cases, determined by taking a sample and analyzing it off-line using invasive and destructive methods. In this situation a long delay time is often expected, which may lead to inadequate monitoring and control schemes.

In addition to the above characteristics, all of which cause difficulties for the monitoring and control of batch/semi-batch processes, it worth pointing out that there are some features of batch processes that may aid in their monitoring and control.

4. Trajectory independent. In many situations, the main aim in batch operation is to achieve certain quality specifications of the end product. Therefore, a batch process is often considered satisfactory if the end-quality properties are achieved no matter what has been the pathway followed by the process trajectories (as long as these do not violate any constraints).

5. Repetitive behavior. Batch operation is repetitive. Therefore, it is expected that the information gained from past batch runs can be used in improving the subsequent ones, such as to design better trajectories or to reject correlated disturbances.

All of the above characteristics should be taken into account in identifying process models and in designing control strategies.

2.2 Literature review on product quality control for batch process

The research work related to quality control of batch process can be conveniently broken into three classes depending upon the objectives of control. If the objective is to search for unknown optimal operating trajectories for the manipulated variables that will optimise some economic or final product quality objective, then this can be referred to as optimisation. If the problem is to use an existing operating policy (with given nominal manipulated variable trajectories) and to control the product quality about a given set-point in the face of disturbances, then this can be referred to as regulatory control. If the goal is to check whether the nominal trajectories are followed and that the reaction variables are within the acceptable range, then this can be referred to as monitoring.

Each class can be further divided into three sub-classes according to the type of information (measurements) used: within batch control, batch-to-batch control and the combination of both. Within batch control, also referred as on-line control, is defined here as the one of utilizing all measurements available from the start of the batch to observe and assess the progress of the current batch (monitoring) or to adjust the manipulated variables for the remainder of the batch to achieve the desired performance for the current batch (regulation and optimisation). Batch-to-batch control, on the other hand can be understood as the use of data collected on previously completed batches to detect the disturbance behaviour in the system (monitoring) and/or to alter the operations of the next batch so as to bring its final quality closer to a desired target and/or to optimise some economic objective (regulation and optimisation).

2.2.1 Open-loop optimal control or one time optimization

Considerable amount of literature has been devoted to the so-called open-loop optimal control policies or one time optimization. Open-loop optimal control involves basically the *design* of new operative conditions and manipulated variables trajectories to achieve some optimal objective such as minimization of reaction time or cost using (generally) a detailed mechanistic model. As the objective is design, the computation of such process conditions is performed only one time off-line and no prediction or feed-back is employed. In spite of such a large amount of work devoted to this approach, it has not had a large impact in industry because it usually requires detailed theoretical models, is computationally intensive and the results are highly dependent on the accuracy of the models. Ponnuswamy et al. [1986] used temperature and initiator profiles to obtain desired average molecular weight and conversion in minimum time (results were corroborated experimentally) for the MMA polymerization. However, their optimal trajectories lead to similar results to those obtained by isothermal operation. Chen and co-workers [1986a, 1986b] and Wu et al. [1982] design optimal temperature (and initiator) policies to obtain a desired conversion and average molecular weight in minimum time, while Thomas and Kiparissides [1984] aim to obtain high conversion and desired number and weight average molecular weights in minimum time for the MMA polymerization (Experimental results are shown). Louie and Soong [1985a, 1985b] use a detailed theoretical model to obtain narrow molecular weight distribution polymers by designing optimal trajectories for the temperature, monomer and solvent injections. Their approach was also evaluated experimentally. Similar approaches were taken by Hsu and Chen [1987], Jang and Yang [1989], Vaid and Gupta [1991] among others. Open loop control of particle size distributions has been addressed by Crowley et al. [2000] and Immanuel et al. [2002] using sequential quadratic programming and genetic algorithms respectively. Optimal trajectories taking into account process constraints were obtained by Chen and Lee [1985, 1987], Choi and Butala [1991], Butala et al. [1992], and Secchi et al. [1990], among others. In some of these studies, experimental verification was also carried on.

2.2.2 Within batch control

On-line monitoring

Approaches to monitoring batch processes have focused on the use of three types of models: fundamental mathematical models, artificial intelligence models and statistical models.

The approaches using fundamental models are usually based on state estimation methods, which combine a fundamental model of the process with on-line measurements to provide on-line, estimates of the states of the system (Iserman [1984], Schuler and De Haas [1986]). The preferred method that has been employed for a long time (since early 60's, Seinfield [1970], Jazwinski [1970], MacGregor et al. [1986]) to perform such estimation is the use of observers or Kalman filters. Kalman filters are employed because they can reconstruct unobservable states and outputs of the system from process measurements. After the filter is designed, the most likely status of the process can then be evaluated using, for example, generalized likelihood ratio tests (Basseville [1988]). King [1986] presented a use of this approach to detect hazardous batch reactor conditions. In Kozub and MacGregor [1992b], stochastic disturbances arising from raw material impurities and parameter variations were incorporated as extended states into the model and estimator, and a scheme to monitor latex properties in the SBR emulsion copolymerization was set-up (similar approaches has been taken by Gagnon and MacGregor [1991], though in a continuous polymerization system). The main difficulty of utilizing the fundamental model-based approach is that the developing of such detailed model is time-consuming and difficult, and the filter may require considerable tuning (Alvarez et al. [1990]).

In artificial intelligence methods, those based on rule-based expert systems represent the process model by a set of qualitative and quantitative descriptions based on the knowledge about the process. Quantitative analysis can be brought to these rule-based expert systems through the use of probability theory or fuzzy logic (Petti et al. [1990],

Rojas and Kramer [1992]). Though these approaches do not require detailed models, formulation of such rules may be just as difficult and time consuming. The use of neural networks appeared in several publications for monitoring and fault detection (Himmelblau [1992], Bakshi and Stephanopoulos [1993]). However, the main drawback of this approach is that a training set with abundant faults must be available.

Fundamental model-based approaches and artificial intelligence approaches are *directional* in that they build into their models or rules the possible faults or reasons for deviations from normal behavior. Therefore, the detection and diagnostic abilities of these approaches depend on prior knowledge and disturbances that may occur, since these must be explicitly built into the estimator, or included as descriptions, or contained in the training set. Events or disturbances that are not considered in the model may lead to biased estimates and faulty diagnosis if they occur (Nomikos and MacGregor [1994]).

In the statistical approach, the use of multivariate statistical process control methods based on multi-way Principal Component Analysis (MPCA) and multi-way Partial Least Squares (MPLS) and their associated monitoring statistics (Nomikos and MacGregor [1992,1994,1995a, 1995b], Kourti et al. [1995,1996]) have been commonly used with industrial data for both the analysis of completed batches and for the on-line monitoring of new batches (Kourti and MacGregor [1995], MacGregor and Kourti [1995], Nomikos and MacGregor [1995a]). Nomikos and MacGregor [1994, 1995a, 1995b] illustrated the detection of abnormal batches using several criteria such as the Q-statistic (also known as square prediction error (SPE) or distance to the model in the X space (DMODX)), the instantaneous SPE (for on-line monitoring) and the PCA or PLS score plots or alternatively Hotelling's T^2. For on-line monitoring they proposed several alternatives for filling in the future missing measurements in order to be able to analyze the current batch in real time. Kourti et al. [1995, 1996] used multi-block methods (MBPCA/MBPLS) to incorporate different initial conditions, modes of operation, and prior processing conditions into the analysis and monitoring of batch processes. Kourti and MacGregor [1995] give an overview of conventional monitoring charts (cumulative

sum (CUSUM), exponentially weighted moving average (EWMA), Shewart, etc.) as well as MPCA. The authors give applications on the monitoring and diagnosis of a continuous and an industrial batch processes. Miller et al. [1993] and Kourti and MacGregor [1996] suggested methods to detect variables that most contribute to an out of control signal in multivariable charts (called contribution plots). The methods are illustrated on a simulated process of a high-pressure low-density polyethylene reactor and several batch industrial processes. Several other applications of MPCA and MPLS to batch processes have also been reported (MacGregor et al. [1994], Nelson et al. [1996], Westerhuis et al. [1999], Neogi and Schlags [1998], and Rännar et al. [1998] among others).

The advantage of all these methods is largely due to the fact that the normal operating data necessary for building the required model is always available, and to the fact that the statistical control charts used for analysis and monitoring are easily developed from these data. One of the main characteristics of such methods is that by projecting the evolving within-batch measurement trajectories (on-line and possibly off-line samples) into reduced dimensional spaces, the relevant information is kept and the monitoring charts built on such reduced spaces are easy to visually inspect and to interpret.

Within-batch regulatory control

Within-batch regulatory control strategies may be grouped in two different classes: control strategies based on the theory of differential geometry and control strategies based on non-linear model predictive control (NMPC).

The essential idea of differential geometry methods is to find an inverse (generally an analytical one) of a nonlinear process. This is performed by means of transformations to the states or control variables in such a way that the transformed process is linear and linear control theory can be applied. Kravaris et al. [1989, 1990] uses globally linearizing control (GLC) for the regulation of copolymer composition of SAN and for the control of copolymer composition and average molecular weight in a

VA/MMA solution reactor. Kozub and MacGregor [1992a] studied the control of many end-quality properties (instantaneous copolymer composition, conversion and molecular weight distribution) for styrene-butadiene (SBR) latex produced in a semi-batch emulsion polymerization process; the non-linear control was based on a feedback linearization scheme and the use of non-linear Kalman Filters. These methods are largely dependent on the process model and conditions that must be satisfied, therefore differential geometry control techniques may be difficult to implement on real processes.

In model predictive control, model predictions over a certain time horizon (prediction horizon) are used to generate optimal control policies over another time horizon (control horizon). The model used can be either detailed fundamental model or empirical model. Garcia [1984] applies dynamic matrix control (DMC) and quadratic DMC to a batch reactor. The non-linear model is linearized and updated as the states of the process change and it is used to obtain the step response coefficients. The non-linear model is used for prediction. Valappil and Georgakis [2001] used nonlinear model predictive control in emulsion polymerization reactors for controlling the final particle diameter, tensile strength and melt index. Crowley and Choi [1998] studied the on-line control of the molecular weight distribution (MWD) and conversion on the free radical polymerization of methyl methacrylate. Corrective control was obtained by solving a sequential quadratic programming (SQP) problem. Reaction calorimetry has been used as a non-invasive technique for estate estimation: Vicente et al. (2001) use a non-linear controller to compute the feed rates of monomer and chain-transfer agent to be added to the reactor to achieve a desired MWD.

Empirical model-based predictive control strategies have been based largely around either artificial neural networks (ANN), regression models obtained through latent variable methods (such as principal component regression (PCR) and partial least squares (PLS)) or subspace models. An ANN application can be found in Tsen et al. [1996] for the control of dispersity and MWD in batch emulsion polymerization. However, here theoretical models are still used for generation of an extended data set, which is necessary

to train the neural networks. Peterson et al. [1992] control the temperature and average molecular weight in a batch solution MMA reactor using a nonlinear model predictive control (NMPC) and a dynamic matrix controller (DMC). An example of using empirical state-space models is given by Russell et al. [1998a] for the control of amine end group concentration and number average molecular weight in batch polycondensation using the trajectories of reactor and jacket pressure as manipulated variables. Contributions based on latent variable modelling can be found in Kesavan, et al. [2000] for the control of batch digesters, in Yabuki, et al. [2000] for the control of average particle diameter in an industrial semi-batch process, and in Yabuki and MacGregor [1997] for the control of weight-average molecular weight and degree of crosslinking in the SBR emulsion process.

On-line optimisation

On-line optimisation can be seen as a general case of model predictive control. Therefore, there is only a subtle (and sometimes no clear) distinction between optimization and NMPC. In on-line optimization, instead of using the cost function reflecting the quality of controller performance, which typically is in quadratic form, the cost function is formulated to reflect also an economic objective, such as maximize the yield of a product, minimize time for a given productivity, etc. Ruppen et al. [1997] uses a theoretical model to perform an on-line time minimization and conversion control in an experimental set-up using SQP at several time intervals. Their approach consists of model identification (on-line parameter estimation) followed by the computation of the optimal profiles on the basis of the identified model (estimation-optimisation task), while Krothapally and Palanki [1997] calculate on-line optimal operating trajectories for the batch polymerization of styrene and methyl methacrylate using ANN. Compared to within batch regulatory control, few studies have been done in this area. This is largely because to obtain reliable results from optimisation more detailed model and/or a huge amount of computer work is required.

2.2.3 Batch-to-batch control

There are essentially two ways to utilize the information from the previous batches: either to use the information to update the parameter estimates of a model in order to account for slow process changes and to overcome model error, or to directly use the information for control if the batch-to-batch disturbances are reasonable highly batch-to-batch correlated.

Batch-to-batch regulatory control

In many batch processes, information gained from the operation of immediately prior batches may be useful in predicting the performance of the next batch. This implies that variations in the disturbances (e.g. raw materials) are reasonably highly auto-correlated from batch-to-batch and hence information gained from prior batches can be used to predict at least some of the performance for the next batch. Industrial examples of employing purely batch-to-batch regulation have been reported for the control of final polymer quality using adjustments in the initial catalyst formulations (Box and Jenkins (1970), Vander Wiel et al. [1992]). Clarke-Pringle and MacGregor (1998) use batch-to-batch corrections of the manipulated variable nominal conditions trajectories for the control of the molecular weight distribution (MWD) in linear polymers. The method uses qualitative fundamental process knowledge and errors between the measured and desired MWD at the end of the batch to update the manipulated variable trajectories for the next batch. Iterative learning control (ILC) is a technique that has been used extensively in the control of mechanical systems (Moore [1998]) and is especially suited for repetitive processes since it uses previous tracking error signals to adjust the manipulated variable trajectories and/or initial conditions for the upcoming batch run. In the microelectronics manufacturing, batch-to-batch control is known as run-to-run control. Edgar et al. [2000] give a comprehensive survey on the applications, problems and algorithms for process control in the semiconductor processing, while Bode et al. [2002] and Toprac et al. [2002] use run-to-run linear model predictive control (LMPC) in the manufacture of semiconductors.

Batch-to-batch optimisation

There is a large literature on batch-to-batch optimisation. Most of the approaches use the previous batch information to update model parameters and then again perform optimisation. The approaches using the concept of tendency models (Filippi-Bossy et al. [1989], Rastogi et al. [1992]) use simplified reaction mechanism models, update the estimates of the model parameters at the end of each batch, and then re-optimize the trajectories for the next batch. Empirical (PLS-ANN) models were used by Dong and McAvoy [1996] and Dong et al. [1996] to obtain input profiles that would achieve a target conversion and molecular weight in minimum time by solving a SQP problem. Model error was overcome using batch-to-batch adaptation. Crowley et al. [2001] use batch-to-batch optimisation to achieve a new desired PSD target in an emulsion polymerization system. The prediction is performed using a theoretical model but an updated PLS model is used to correct the prediction by relating the manipulated variables to the error from the theoretical model prediction and the measured distribution. An illustration of combining empirical information and theoretical modeling is given by Echavarria et al. (1995), who use experience and experimentation to estimate kinetic constants and based on them use an off-line optimization algorithm to compute optimum addition profiles in a emulsion copolymerisation process. Then this trajectory is implemented, new data is collected, the parameters up-dated and a new input trajectory (monomer flow rate) is again implemented.

Combined batch-to-batch, on-line control and optimization

Strategies which combine information about errors from past batches with information from the current batch in order to adjust the MV trajectories to regulate quality or to optimize some quality or economic objective function effectively combine the goals of batch-to-batch, within-batch (on-line) control and optimization.

Lee and coworkers [1996, 1999, 2000, 2001] combine the advantages of ILC and MPC into a single framework. Information from past error tracking signals is used along

with information from the current batch to control the process in real time. Contributions in this area include Lee et al. [2001] for minimization of reaction time, Lee et al. [1996] for the heat-up phase of a batch polymerization reactor, Chae et al. [2000] for the quality tracking control of poly-methyl methacrylate, and Bonne and Jørgensen [2001] for the trajectory tracking of a fed-batch fermentation reactor. Bonvin et al. [2002] and Srinivasan et al. [2003a, 2003b] have recognized that it is unrealistic to use detailed theoretical models for the control and *on-line optimization* of batch processes. They introduce a novel strategy in which the optimal structure of the parameterized inputs is determined using, for example an approximated model and then measurements (off-line and/or off-line) are employed to refine (update) them. Optimality is achieved by working close to the constraints that are active.

2.2.4 Modeling for monitoring, control and optimization

Appropriate mathematical system representations are the key to perform prediction and to compute manipulated variable adjustments in model-based monitoring, control and optimisation techniques. Usually the models used can be divided into three different types: fundamental models, empirical models and hybrid models.

Fundamental models

Fundamental models are also called first principle models, theoretical models or white-box models. These models are developed based on conservation (mass, moment and heat) theories and kinetic and thermodynamic principles. They are generally state space models, described by a set of highly coupled nonlinear algebraic, partial and partial differential equations, which require advanced numerical techniques for proper solutions to be obtained in a considerable amount of time. The main advantage of this type of the models is that they give a good understanding about the process and are well suited for a wide range of process operations. Therefore they have been considered as the preferred approaches in many studies. However, fundamental models are usually difficult and time consuming to build for real industrial systems. Generally for a model to be realistic, only

the part of the process that is most relevant to the control (and optimization) objectives is needed to be modeled and a few physical and kinetic parameters then estimated based on the process data.

For control purposes, a theoretical model is usually combined with Kalman filter to estimate model parameters, states and outputs based on process measurements (Jazwinski [1970], and MacGregor et al. [1986]). One of the advantages of Kalman filter is that it gives the possibility to adequately represent process and operational disturbances that are otherwise difficult to model and to measure (such as impurities) through extended nonstationary stochastic states (MacGregor et al. [1986], Kozub and MacGregor, [1992b], and Gagnon and MacGregor [1991]). If the model is good, then noisy measurements can be efficiently filtered out. On the other hand, if the model is poor, information-rich and accurate measurements are required to obtain acceptable estimates.

Empirical models

Empirical models are also called databased models or black-box models. These models describe the relation between input and output through an empirical relationship identified from a database, obtained from designed experiments and/or historical database. The models can be developed using neural networks or multivariate statistical regression methods. Compared to fundamental models, empirical models require little prior knowledge and are easy to build. However, there are several common arguments:

1. Empirical models lack extrapolative power. Because the information of empirical models comes mainly from the data used, they often can have good interpolative capabilities; however they rarely can predict the process behavior outside the range spanned by the data collected for model building. Obviously, such limitation restricts the use of empirical model in solving the process optimisation problem, in which new manipulated profiles may need to be determined. Although optimisation is a very important problem, in many real industrial settings nominal trajectories for producing a certain product are already fixed, and the main operational objectives are to monitor

and/or control the product quality about a given set-point in the face of disturbances (regulatory operation). In this situation, the operation region is always around the nominal conditions and extrapolative ability is not a critical problem.

2. Bonvin [1998] has pointed out that empirical models can only represent the relationship between variables that are manipulated or measured and have no prediction power for the variables not observed such as the heat of reaction. However, in many situations there is no need to estimate such unmeasured variables. If the purpose is to monitor the progress of the reactor, as long as the measurements reflect the influence of the disturbances, the abnormal operation can be easily detected. In the case of regulatory control of final product properties, the key is the relationship between manipulated variables, disturbances (the information often contained in the measured variables) and end product qualities. Any empirical model that adequately describes this relationship can achieve good control performance.

Hybrid models

Hybrid models are also called grey-box models. These models combine the two cases listed above. One type of hybrid models uses a simple model structure that is based on some qualitative knowledge of the process or a simplified model but retains the physical understanding of the system. The model parameters often are much easier to identify compared to detailed fundamental models. Tendency modeling (Filippi-Bossy et al. [1989], Rastogi et al. [1992]) can be classified in this type. Another type of hybrid models uses a detailed fundamental model to generate training data for developing the empirical model. An example can be found in Tsen et al. [1996], and in Krothapally and Palanki [1997].

From fundamental models to databased models, we can see the conflict between academic research and real industrial implementation. The research on fundamental modeling and using it for process monitoring, control and optimisation has been carried out since 1960s. In contrast to the large number of papers published, few have been

reported on the real industrial implementation of control algorithms. This is largely because: 1) to use a detailed fundamental model to solve an optimization problem is computationally intensive and unlikely can be performed on-line; 2) no model can perfectly describe the process and there always exist model error and uncertainty which might influence the reliability of the model; 3) to develop appropriate theoretical models is time consuming, and 4) to maintain the performance of the model generally requires in-house expertise. In many chemical and pharmaceutical companies the profit is made from the products they sell, and not directly from the engineering features they have implemented in their processing plants. Any time and money spent at improving the process has to pay off economically in the form of producing more or better products, improved safety operation and/or better usage of resources (Friedrich and Perne [1995]). Therefore, because of the high cost and uncertain profit of implementing a control system based on fundamental model, the implementation of such control strategies is a risky decision for many industries.

Empirical models and hybrid models use much simpler model structure and most of the information comes directly from process measurements. Obviously these models have more probabilities to be implemented in real industrial settings. The criticism mainly is whether using the simple model structure is sufficient. This question evidently is problem dependent. However, as mentioned before, in the case of monitoring and regulatory control, databased models are often adequate. Moreover, there are certain situations in which empirical models may be even better suited than the theoretical ones. One classic example of these situations is the control of particle concentration and particle size distribution (PSD) in emulsion polymerization [Kiparissides et al. [1981], Crowley et al. [2000, 2001]), which is studied in Chapters 3 and 4. In emulsion polymerization systems small variations in the impurity levels and in the surface chemistry of the emulsifier can exhibit a large influence on the particle nucleation (Penlidis et al. [1985], Huo et al. [1987]). Data on these impurities and surfactant variations are never available, and even if they were, the theoretical models are still insufficient to model their subtle effects on nucleation (Kiparissides et al. [1981], Penlidis

et al. [1989]). Therefore, information on their effects must still come from measurements made in the process, and the main requirement of any model for regulatory control is to be easy to built and be flexible enough to use the available measurements to infer the final quality. It will be shown in Chapter 3 and 4 that PLS methods are very suitable in these situations because their excellent interpolation power and because their efficient and flexible use of the information contained in process measurements.

2.3 Empirical modeling using latent variable methods

This thesis focuses on developing practical on-line monitoring and regulatory control schemes for batch/semi-batch processes. Batch-to-batch information is also effectively used. Latent variable methods, such as principal component analysis (PCA) and partial least squares (PLS), are used for model building. Compared to other empirical methods (i.e. neural networks or multivariate regression), the advantages of these methods are: 1) they can effectively handle highly correlated data; 2) they are fast and easy to build and update; 3) they do not require large training dataset; 4) they provide a model for both the X and Y spaces; 5) they can easily detect outliers and handle missing data; 6) they provide simple tools (e.g. Hotelling T^2 and square prediction error (SPE)) to check the validity of the new coming data; and 7) they provide effective interpretation tools, such as score plot, loading plot and contribution plot to help understand the process. These advantages give the possibilities to implement these methods to real industrial settings.

In industry, on-line process measurements (e.g. temperature, pressure, etc.) and off-line quality measurements (usually at the end of the process) are routinely collected and stored. Since the number of factors (movements in manipulated variables and disturbances) that drive the system is usually much smaller than the number of measured variables, there exist high correlation among the measured variables. It is well know that this high correlation among variables causes ill-conditioning of the data matrices and leads to poor parameter estimation when use ordinary linear regression. Qin (1997) showed that a regular backpropagation training can result in large prediction error under

correlated inputs and suggested using latent variable methods as a preprocessing step. In the same paper, he also pointed out that neural networks lack capabilities on several practical issues including detecting outliers, replacing missing data and checking the validity of the new data and it would be beneficial to combine ANN models with multivariate statistical models. Another drawback of neural networks is that they have large number of model parameters and therefore generally need a lot of training observations to avoid the overfit problem.

PCA and PLS are designed to handle multivariate data by projecting the information contained in a set of highly correlated process variables onto low-dimensional spaces defined by a few variables, known as principal components or latent variables.

The mathematical representation of PCA is:

$$\mathbf{X} = \mathbf{\Gamma V}^T + \mathbf{G} = \sum_{a=1}^{A} \boldsymbol{\tau}_a \mathbf{v}_a^T + \mathbf{G}$$

where $\mathbf{X}$ is the input matrix ($K \times M$); $\mathbf{\Gamma}$ is the score matrix ($K \times A$), which is the new coordinates in the lower dimensional space; $\mathbf{V}$ is the loading matrix ($M \times A$), which is orthonormal and is the basis to span the low dimensional space; and $\mathbf{G}$ is the residual matrix ($K \times M$). K is the number of observations, M is the number of variables of the $\mathbf{X}$ space and A is the number of principal components. $\boldsymbol{\tau}_a$ and $\mathbf{v}_a$, a-th column vectors of $\mathbf{\Gamma}$ and $\mathbf{V}$, are the score vector and loading vector of the a-th principal component respectively. The score matrix $\mathbf{\Gamma}$ can be computed from $\mathbf{\Gamma} = \mathbf{XV}$. Each score vector $\boldsymbol{\tau}_a$ is a linear combination of original variables and $\mathbf{v}_a$ is the corresponding combination weights. The principal components are ordered in a way such that the amount of variance described by each principal component decreases as the number of principal components increases. The squared prediction error (SPE) for the k-th row of $\mathbf{X}$ is given by $SPE_k = \sum_{m=1}^{M} g_{km}^2$.

A PLS model can be generally expressed as:

$$\mathbf{X}=\mathbf{T}\mathbf{P}^{\mathrm{T}}+\mathbf{F}$$

$$\mathbf{Y}=\mathbf{T}\mathbf{Q}^{\mathrm{T}}+\mathbf{E}$$

$$\mathbf{T}=\mathbf{X}\mathbf{W}$$

where $\mathbf{W}$ is a $(M \times A)$ weight matrix that maximizes the covariance between $\mathbf{T}$ $(K \times A)$ and $\mathbf{X}$ $(K \times M$, predictors), $\mathbf{Q}$ is a $(R \times A)$ matrix of regression coefficients (loadings) for $\mathbf{T}$ forming a linear model for the response variables ($\mathbf{Y}$, $K \times R$), while $\mathbf{P}$ is a $(M \times A)$ loading matrix that forms a model for $\mathbf{X}$. $\mathbf{E}$ and $\mathbf{F}$ are the error (noise) terms. R is the number of response variables and A is the number of latent variables. Several algorithms can be used to compute the matrices $\mathbf{W}$, $\mathbf{Q}$ and $\mathbf{P}$, among which NIPALS algorithm is the most commonly used. For further details please refer to (Wold [1978], Krzanowski [1987], Geladi and Kowalski [1986], Jollife [1986], Wold et al. [1987], Höskuldsson [1988], Geladi [1988], Geladi [1989], Jackson, [1991], Kourti and MacGregor [1995]). Notice that it is the model for the X-space that allows PLS models to handle missing data, check for outliers, monitor the process variables, and invert the model to yield new $\mathbf{x}$ values that are consistent with past operating conditions.

PLS model can also be expressed in the conventional linear regression form:

$$\mathbf{Y} = \mathbf{T}\mathbf{Q}^{\mathrm{T}} = \mathbf{X}\mathbf{W}\mathbf{Q}^{\mathrm{T}} = \mathbf{X}\hat{\mathbf{B}}$$

where $\hat{\mathbf{B}} = \mathbf{W}\mathbf{Q}^{\mathrm{T}}$. $\hat{\mathbf{B}}$ is an $(M \times R)$ regression coefficient matrix.

Batch process data consists of a three-dimensional data array containing the different process variables (1,… ,N) across both the batch (1,…,K) and time dimensions (1,…, L) as shown in Figure 2.1. Denote this data array as $\underline{\mathbf{X}}(K \times N \times L)$. To deal with this three-dimensional data structure, multiway PCA/PLS is applied. Multiway PCA/PLS collapses the "within-batch" dimensions (N and L) and creates a large unfolded data matrix $\mathbf{X}(K \times NL)$. Then normal PCA and PLS is performed on $\mathbf{X}$. In this PCA/PLS model,

each batch can be represented by a point in the score space spanned by the latent variables. Since the dimension of the score space is much smaller than the original data matrix, observing the batch behaviour in the score space is much easier.

For monitoring the batch process, generally a multiway PCA/PLS (MPCA/MPLS) model is built using historical data collected when only common cause variation was present and only good quality product has been obtained. For a new batch, its behaviour can be compared with this model of good behaviour. This involves plotting the scores and the SPE of new observations, and testing whether or not they are consistent with past good behaviour. This is accomplished by establishing control limits for the monitoring charts using the statistical properties of the training data. For on-line monitoring, a missing data algorithm can be used at first to predict the future measurements and then project the data onto the model defined by PCA/PLS. Many industrial applications for monitoring have been reported using MPCA/MPLS approach.

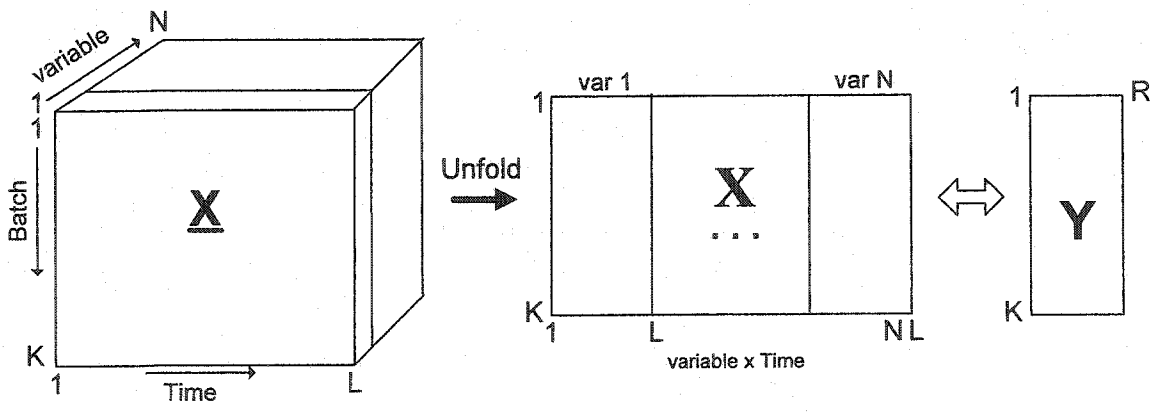


Figure 2.1 Unfolding of batch process data

A few attempts at using MPLS for the control of batch process are reported in literature (Yabuki and MacGregor [1997], Kesavan et al. [2000]). The main criticism on using PLS model for control is that its linear model structure may not be good enough to describe the non-liner behaviour frequently found in batch processes. However, it should be pointed out that in the case of regulatory control, since the main objective is to reject

disturbances about a nominal and fixed operating plant (usual situation in most industrial practice), the process trajectories should not deviate too far from their nominal conditions. Since batch PLS models are based on deviations from the mean (or nominal) trajectories, and since the model provides a local linear model about every point along that trajectory, it has rarely found that nonlinearities are a problem. If necessary, proper transformations can be used to overcome slight nonlinearities or the use of a nonlinear PLS regressor method employed. In this thesis, the use of empirical PLS models for regulatory control of batch processes is further studied and extended. It is shown that even when some processes are intrinsically highly non-linear (e.g. emulsion polymerisation), good regulatory control performance can be achieved by using simple PLS models.

Chapter 3

Control of PSDs using Mid-Course Correction Policies

The manufacture of emulsion polymers with consistent broad and bimodal particle size distribution (PSD) through in situ particle nucleation in semi-batch reactors is difficult due to the sensitivity of the particle nucleation phenomena to variations in reactor conditions, impurities, and surfactant and initiator properties. This Chapter presents several control strategies based on the use of readily available on-line and off-line measurements. Partial least squares (PLS) models are used to extract the necessary information from different sets of measurements to predict the final PSD. Using a simulated styrene emulsion polymerization process as example, these control strategies are shown to be effective, practical and with potential for industrial implementation.

3.1 Introduction

In emulsion polymerization, it is well known that one of the variables that determine the final latex properties (stability, film forming ability, covering capacity, "brushability", viscosity, opacity and texture among others) is the PSD. Control of the PSD can be very difficult since it is generated by the particle nucleation process, which is highly non-linear, of short duration, and highly sensitivity to even the smallest change in impurity concentrations, surfactant properties, temperature, etc.

One of the first attempts to control a PSD in batch processes is the work done by Liotta et al. [1997]. These authors show how a bi-disperse distribution can be created and

how it can be controlled to a limited extent through "competitive growth". "Competitive growth" exploits the fact that the weight fraction of polymer inside particles affects the relative narrowing (standard deviation divided by the mean) of a bidisperse PSD. They point out that the key to manipulate the growth is to change the ratio of average number of radicals for each particle population. To illustrate their approach, they built a semi batch reactor facility for the emulsion polymerization of styrene and they tried to control the relative broadening of distributions using the monomer feed rate as manipulated variable (MV). However, no full matching of a predetermined target shape was attempted.

A theoretical study on PSD controllability in continuous systems done by Semino and Ray [1995a, 1995b] showed that with an unbounded feed concentration of surfactant, initiator or inhibitor and knowledge of reactor states, one can guarantee the controllability of the PSD. Furthermore, they show how to extend the controllability region by using two manipulated variables with opposite effects (i.e., inhibitor and initiator). They specifically addressed the control of an unstable continuous stirred tank reactor for the emulsion polymerization of methylmethacrylate using single-input single-output (SISO) controllers.

Crowley et al. [2000] addressed the computation of surfactant feed profiles for matching a PSD to a pre-specified target in the batch emulsion polymerization of styrene. The manipulated variable was the surfactant feed profile in a 30 min span covering the particle nucleation period. In effect the free surfactant concentration profiles were being adjusted. They solved the optimization problem using sequential quadratic programming, and compared the performance of different objective function norms. In their work, the polymerization was limited to short reaction times (Interval I), in which with few movements in the manipulated variable, the desired target can be achieved. It is important to mention that at least in batch/semi-batch emulsion polymerization processes, this is the first successful simulation attempt at matching a full PSD to a target distribution. Recently, Immanuel et al. [2002] perform open-loop PSD control for an emulsion copolymerization process using genetic algorithms. In these studies, however, a full population balance model is needed to compute control actions, which may limit its

applicability in real processes. Moreover, no on-line adjustment of manipulated variables for disturbance rejection is performed.

In this Chapter a different approach to perform on-line control of the final PSDs is taken. Recognizing that to control the highly sensitivity nucleation phenomena during its short duration is extremely difficult using distributed parameter models, it is considered on-line control strategies based on simple empirical models which use less demanding on-line sensing to estimate the end result of the first nucleation stage and then make mid-course corrections to influence the subsequent nucleation stages so that the final PSD is controlled to its target. It is shown that various on-line and off-line sensors ranging from very simple to more sophisticated combinations can effectively estimate the state of the system between nucleations, and that the empirical models are easily developed to compute mid-course corrections. All these characteristics represent considerable advantages over alternative approaches.

3.2 Emulsion polymerization system

3.2.1 Base cases

An extensive model to simulate the styrene batch emulsion polymerization process was developed by Crowley et al. [2000] and is used in this work for data generation and model performance evaluation. This model, based also on the research of Cohen et al. [1998] computes the PSD, particle number (N_p), particle size, and it incorporates detailed kinetic and physical-chemistry mechanisms such as aqueous phase initiation, desorption, radical balances inside particles, and particle formation by micellar and homogeneous nucleation. The complete description of the reaction mechanisms and model parameters can be found in the original publications.

In the present study, two different target bimodal PSDs were generated using different initial recipes and then promoting secondary generations of particles using shots

of emulsifier at 150 min. The reaction was then carried out to 92% conversion. The nominal recipes for the two PSD targets are shown in Table 3.1.

Table 3.1 Recipes used in the generation of bimodal PSD.

Species	Recipe 1 (mol/l)	Recipe 2 (mol/l)
Water	23.611	23.460
Styrene	4.807	4.807
Initiator[a]	1.39×10^{-2}	1.39×10^{-2}
Emulsifier[b]	11.0×10^{-3}	20.0×10^{-3}
Emulsifier[c]	0.032	0.045

[a]potassium persulfate, [b]sodium dodecyl sulfate, [c]injection at 150min (mol).

3.2.2 Disturbances affecting the PSD

In emulsion polymerization reactions, the main process variations arise from different start-up conditions and changes in the quality of raw materials. The first type of process variations can be controlled, to a certain extent, by a high degree of automation (Yabuki and MacGregor [1997]). However, changes in the final quality due to raw materials variations cannot be controlled using this policy. The main raw material variations that affect the particle size distribution are those related to the characteristics of the emulsifier, the initiator and impurities present in the monomer. However, since impurities affect the polymerization in the same way as the initiator (Penlidis et al. [1985], Huo et al. [1987]), the effect of impurities will be lumped into the effect of the initiator in this study. This was also necessitated by the fact that the theoretical model used for simulation did not consider impurity effects.

In emulsion polymerization processes, emulsifier activity is mainly controlled by the particle surface coverage a_s (ability of an emulsifier molecule to stabilize a polymeric particle), while the initiator is controlled by its efficiency (f) or capacity to generate radicals in aqueous phase, k_{daq} (water soluble initiators). Variations in the emulsifier coverage capacity and the number of radicals generated from the initiator decomposition can arise from natural raw quality changes or from small contaminations due to inappropriate storage, long storage (aging) or improper equipment cleaning. The principal water-soluble impurity is dissolved oxygen, whose effect is an induction period at the

start of the polymerization with little impact on the end-quality properties (Huo et al. [1987]). On the other hand, organic impurities such as monomer inhibitor and trace organic compounds change the number of radicals. Even small changes in the number of polymer radicals or the emulsifier surface coverage (a_s) will have a great effect on the nucleation rate of particles and hence on the final PSD and particle concentration (N_p). Therefore, when the polymeric particles are nucleated in situ (a procedure that is common in the paint industry) control of PSD becomes essential.

In this simulation study, the major disturbances affecting the target PSD are introduced through random batch-to-batch variations in the particle size coverage (a_s) and a lumped initiator decomposition efficiency (f^*). This efficiency would represent variations in the initial initiator charge, impurity variations, and parameter mismatch arising from the aqueous phase decomposition constant (k_{daq}), etc. Considering normal variations in raw material qualities, the standard deviations of a_s and f^* are selected as 2% and 4%. These normal operation standard deviations do not include the effect of raw material variations due to contamination or different start-up conditions (i.e. due to special causes). A control policy that overcomes these special cause disturbances is developed in the following sections.

3.3 Manipulated variables

In the specific case of emulsion polymerization, potential manipulated variables for the control of PSD are those related to free emulsifier concentration, the number of radicals in the aqueous phase and monomer concentration inside the particles. The possible set of manipulated variables includes the emulsifier, initiator, inhibitor and monomer feed rates. In this research, we restrict control actions to shots of emulsifier ($u_{c,1}, u_{c,2}$) because they have an almost instantaneous effect on the free emulsifier concentration and on the number of particles nucleated. Shots of initiator/inhibitor could be alternative control actions. However, process measurements are needed earlier because their dynamics over the PSD are slower than those of emulsifier shots. In order to be able to both increase or decrease the number of particles nucleated (and hence affect the PSD

in a negative or positive direction), the nominal emulsifier shot ($u_{c,2}$) injected at 150min will be adjusted slightly in a positive or negative direction.

3.4 Sensor selection and model building for PSD prediction

For the Case studies presented in this Chapter and in Chapter 4, partial least square (PLS) models (linear and quadratic models) were employed to predict and then to control the final PSD. This is adequate due to the fact that, in the range under study, the effects of the manipulated variables (emulsifier adjustments at different times) on the controlled variables (PSDs) are almost linear as shown in Appendix A. The slight nonlinearities of the system, in such range, were properly taken into account by using quadratic and interaction terms (when needed) in the manipulated variables as well as simple transformations on the PSDs.

The data sets for model building are generated from the theoretical model of Crowley et al. [2000] by introducing random variations in a_s, f^* and changes in the manipulated variables (emulsifier shots at 30 and 150min) that resemble empirical control actions taken by operators when the quality properties are not on target. The measurements studied were various combinations of on-line discrete reactor and jacket temperature profiles (x_{on}) and off-line measurements on particle diameter (D_p), free emulsifier concentration (S_f), and PSD (x_{off}). The measurements considered and their sampling schedules are given in Table 3.2. Temperature values at every 10 min were used for convenience, but if continuous measurements of Tj were available, they could be incorporated in straightforward manner by augmenting the predictor matrix. In this work, except in the Case study II, we assume that on-line PSD measurements are not available, and that only one off-line (x_{off}) PSD measurement is made using a sample taken at 30 min. and whose results are available at 150min. If more PSD measurements were available, perhaps using on-line dynamic light scattering, etc., the methods proposed in this chapter could easily use this additional information, and would provide improved results. In order to simulate process conditions, white noise was added to all lumped process measurements. As the measurements are discrete, it is assumed that filtering may be

performed to reduce the effect of noise. As can be seen in Table 3.2, the noise level for the jacket temperature (Tj) measurements is σ=0.1% (approximately $\pm$ 323 $\times$ 0.001 $\approx$0.3°K); for Dp is (Yabuki and MacGregor [1997]) σ=0.576% ($\pm$ 60nm $\times$ 0.0057 $\approx$0.35 nm), and for S_f is σ=1% ($\pm$ 5 mmol/l $\times$ 0.01$\approx$.05mmol/l). In the case of the PSD measurement (x_{off}), the noise has a correlation that depends on the instrument providing the measurements. Therefore, noise structure identification was performed through principal component analysis (PCA), as described in Clarke-Pringle [1999], using actual repeated PSD measurements on a styrene latex sample by a dynamic light scattering instrument. From the identified structure, correlated noise (with standard deviation (σ)=1.0) was generated and added to all intermediate PSD measurements (see Appendix A).

Based on the five data sets shown in Table 3.2, each using a different combination of the process measurements, five PLS models were built to predict the final PSD. These PLS models were validated using the test set shown in Table 3.3. Predictions on the final PSD (at 92% conversion, 1000min) were made at a batch time of 150min (14% conversion) using all the indicated on-line and off-line measurements available at that time. As an illustration of the predictive performance of the models, some results are presented in Figures 3.1, 3.2 and 3.3 for Recipe 1 (Table 3.1). It was observed that all models that include either the intermediate PSD or D_p measurement have high predictive power for the final PSD. For example model 1, which uses an intermediate off-line measurement (x_{off}) of the PSD and on-line (x_{on}) jacket temperature measurements taken between 10 and 150 min has a predictive power of 97.1%. Figure 3.1 shows observed (-) and predicted (--) final PSDs for this model, using test batches 1 to 5 of Table 3.3.

In order to determine if the main process disturbances can be detected using only the most inexpensive measurement (T_j) model 5 was built. However its predictive power is only 47.9% indicating that only using on-line T_j measurements provides a poor prediction of the final PSD. Figure 3.2 shows the predicted (-) vs. observed (--) PSD for this model.

Table 3.2 Measurements for model building

Model	Measurements/(σ%)	Noise level[c]	Sampling times (min)
M1	PSD(*); Tj(0.1)	*; ± 0.3°K	30; 10,20…150
M2	Dp[a](0.576); Tj	± 0.35nm; 0.3°K	110; 10,20…150
M3	Dp; PSD; Tj	± 0.35nm; *; 0.3°K	110; 30; 10,20…150
M4	Dp; S_f[b](1); Tj	± 0.35nm; .0005mol/l; 0.3°K	110; 20; 10,20…150
M5	Tj	± 0.3°K	10,20…150

* correlated noise, [a]Yabuki and MacGregor [1997], [b]assumed, [c]using T_j=323°K, Dp=60nm and S_f=5mmol/l as base values.

Table 3.3 Test sets for prediction and control.

Test set	Multiplicative factor for a_s (dm^2)	Multiplicative factor for f*	Emulsifier shot at 150min (mol)
Base case	1	1	0.032
0	0.902	0.88	0.032
1	1.080	1.12	0.032
2	0.910	0.90	0.032
3	0.932	1.10	0.032
4	1.110	1.14	0.032
5	1.200	1.20	0.032
6	0.899	1.00	0.032

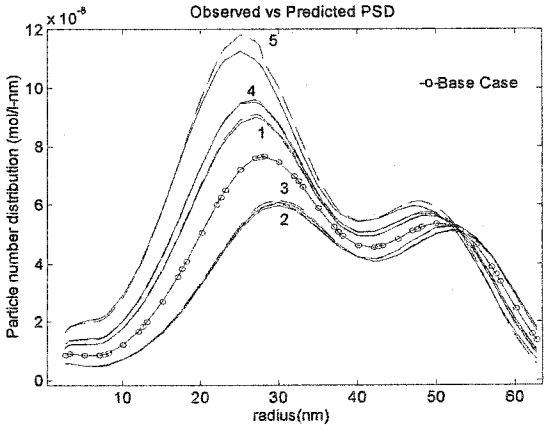


Figure 3.1 Predicted (– –) vs. observed (–) PSD for model 1, Recipe 1. The number indicates the test set in Table 3.3.

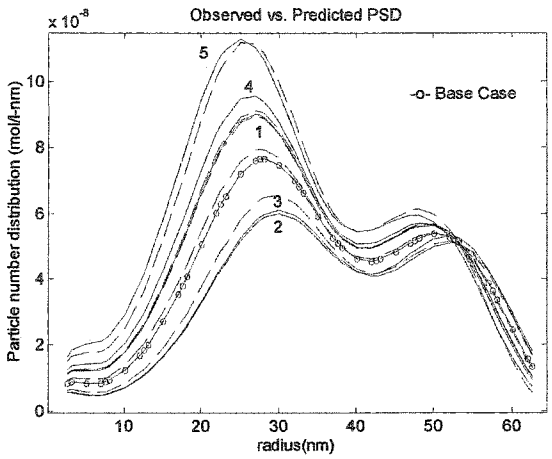


Figure 3.2 Predicted (– –) vs. observed (–) PSD for model 5, Recipe 1. The number indicates the test set in Table 3.3.

In order to measure model predictive performance, sum of squared prediction errors (PRESS) was taken over the set of m test batches:

$$PRESS = \sum_{k=1}^{m} \sum_{i=1}^{n} (y_{ik} - \hat{y}_{ik})^2 \tag{3.1}$$

where n is the number of variables obtained from the segmented PSD (60 in this case), and $\hat{y}$ the predicted PSD.

Figure 3.3 compares the PRESS of models 1 to 5 (using the different sensor sets shown in Table 3.2) for Recipe 1 and 2 of Table 3.1. The worst performance is from model 5, which uses only jacket temperature measurements (x_{on}). As seen before, models that use a single off-line measurement of PSD or D_p have good performance. As expected, the best models are those that include the most complete set of measurements. Figure 3.3 also shows that the performance of the model is relatively independent of the recipe employed and mainly relies on the measurement set used in model building.

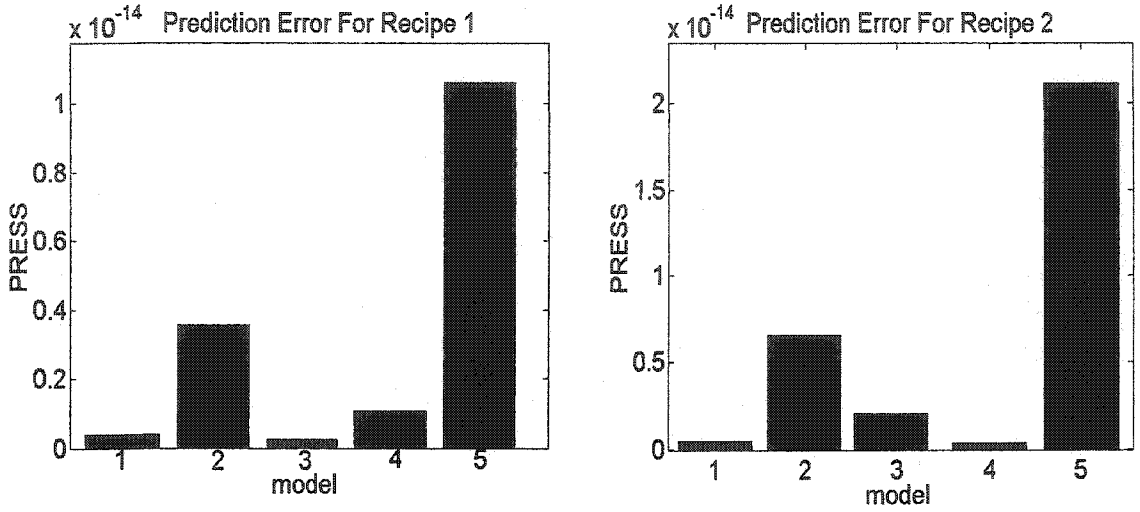


Figure 3.3 PRESS of Model 1-5 for Recipes 1 and 2

3.5 Control studies

3.5.1 Control methodology.

In this study, the methodology employed to control the PSD is an extension of the mid-course correction strategy for the control of high dimensional end-quality properties. In this methodology, introduced by Yabuki and MacGregor [1997], Yabuki et al. [2000] and Tsen et al. [1996], measurements are collected at early to mid-stages of the process and an empirical model is employed to predict the end-quality properties. The methodology is illustrated in Figure 3.4, where it can be seen that when a new batch is being produced, on-line (x_{on}) (jacket temperature) and off-line (x_{off}) (for example PSD) measurements are collected until a decision time (θ_l, l=1,2,..), where a prediction of the final product quality ($\hat{\mathbf{y}}_l$) (i.e. PSD) is then made on the basis that no future control action will be taken (i.e. the manipulated variable will be equal to their nominal values ($u_{n,1}$, $u_{n,2}$)).

If the prediction $\hat{\mathbf{y}}_l$ at time θ_l is outside a pre-determined no-control region (section 3.5.2), then the empirical model is inverted to obtain the control correction

adjustments (u_1, u_2) (shots of emulsifier) required to force the end-quality properties back to their target (section 3.5.3).

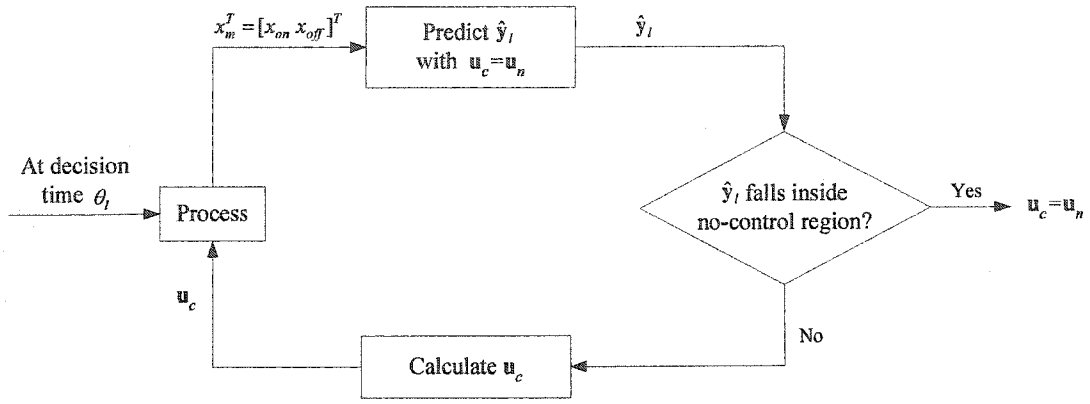


Figure 3.4 Mid-Course Correction (MCC) Strategy

Notice that at the mid-course control point (θ_1) almost any form of MV modification can be used (not only discrete control actions such as emulsifier shots). For example the profile of the base case addition rate of any component such as monomer, water, initiator, etc. could be modified at the control point by manipulating a parameter such as the slope, level or some other combination of parameters that will determine the shape of the profile of the MV for the remainder of the batch. The modifications would not alter the basic approach shown in Figure 3.4.

3.5.2 No-Control Region

With current automated batch sequencing and control systems, most batches seem proceed satisfactorily with no additional corrections necessary (Yabuki and MacGregor [1997]). Only if unusual disturbances occur is corrective action necessary to achieve the final quality targets. Therefore, operating personnel usually prefer that corrections only be made when they appear to be necessary. For this reason "no-control" regions were introduced to define limits for normal behaviour within which any variation cannot be distinguished from acceptable "common cause variation".

Because of the high collinearity among the quality measurements (PSD), the no-control region will be defined in the low dimensional principal component space $(t_1,\cdots,t_A)$ of the quality variables. To obtain this no-control region, principal component analysis (PCA) is performed on all the "good" or "in-control" quality data ($\mathbf{Y_g}$) from the historical data set.

$$\mathbf{Y}_g = \mathbf{TP}^T + \mathbf{E}' = \sum_{a=1}^{A} \mathbf{t}_a \mathbf{p}_a^T + \mathbf{E}' \tag{3.2}$$

where **T**, **P**, and **E'** are score, loading and residual matrices respectively, t_a and p_a are score and loading vectors for the a-th principal component, and A is the number of principal components.

Assuming that the principal components $\hat{t}_1,\cdots,\hat{t}_A$ are approximately normally distributed (which is reasonable by the Central Limit Theorem (Montgomery [1994], Draper [1997])), relating to linear combinations of variables), then the boundaries of the region can be defined in the latent variable space by Hotelling's T^2 statistic:

$$T^2 = \sum_{a=1}^{A} \frac{t_a^2}{s_a^2} = \frac{A(N^2-1)}{N(N-A)} F_\alpha(A, N-A) \tag{3.3}$$

where N= number of observations in the model training set; F_α = Critical value of the F distribution with A and N-A degree of freedom at the α level of significance, and s_a^2= variance of the score a.

For a predicted $\hat{\mathbf{y}}_I$ obtained from the model, its projection in the principal component subspace ($\hat{\mathbf{t}}_I^T = [t_{1I}, t_{2I}, \ldots t_{AI}]$) can be computed as:

$$\hat{\mathbf{t}}_I^T = \hat{\mathbf{y}}_I^T \mathbf{P} \tag{3.4}$$

If $\sum_{a=1}^{A} \frac{\hat{t}_{a,l}^2}{s_a^2} > \frac{A(N^2-1)}{N(N-A)} F_\alpha(A, N-A)$ the predicted quality is outside the (1-α)*100 no-control region, and hence a control action would be justified (α=0.05 in this study).

In the following examples, a normal operation region was determined as described above when the process was subject to only normally occurring raw material variations. Only 2 principal components, defining an ellipsoid, were required to summarize the final bimodal PSD space (according to cross-validation tests). If the predicted PSD falls outside this ellipse, then a statistically significant disturbance is deemed to have affected the process and a corrective control action adjustment is called for. In this case, the inferential PLS models built in the prediction section will be inverted to calculate the mid course corrections required to bring the PSD back to the target. The test data sets generated to evaluate the model prediction (Table 3.3) will also be used to evaluate the control performance. In this test set, batches with large upsets in a_s and f^* were included to provide a good test for the control methodology.

3.5.3 Control strategies

Three controls strategies that use different control actions are presented in this Chapter and outlined in Table 3.4. In Case I, the objective is to control only the secondary particle generation for Recipes 1 and 2 (Table 3.1) by adjusting the magnitude of the emulsifier shot ($u_{c,2}$) made at 150min. Case II addressed the control of the full bimodal PSD for Recipe 1 using two emulsifier shots ($u_{c,1}, u_{c,2}$) at 30min and 150min respectively to affect each of the two nucleation periods. Finally, in Case III control of relative distributions is attempted by adjusting the shot of emulsifier ($u_{c,2}$) at 150 min and the overall reaction time ($u_{c,2}^*$). Table 3.4 also shows the models and the test batch numbers that will be used to illustrate the prediction and control for each case. Detailed discussions of the control strategies are presented in the following sections.

Table 3.4 Control studies performed

	Case I	Case II		Case III
Recipe	1 and 2	1		1
Control objectives	Control of secondary particle generation	Control of bimodal distribution		Control of relative distributions
Control actions	Emulsifier shot ($u_{c,2}$) at 150min	Emulsifier shot ($u_{c,1}$) at 30 and emulsifier shot ($u_{c,2}$) at 150min		Emulsifier shot ($u_{c,2}$) at 150min and total reaction time ($u^*_{c,2}$)
Conversion(%)	92	92		50
Total Reaction Time (min)	1000	1000		380
Model	M1…M5	M1	M2	M1, M2
Measurements	Table 3.2	PSD; Tj	Dp; Tj	Table 3.2
Sampling Time (min)	Table 3.2	20;10,20	20;10,20	Table 3.2
Test set for control (Table 3.3)	1…5	0,2,3,6		1…5

3.5.4 Case I: Control of the secondary distribution.

In general, there is little that one can do to control the distribution of the first generation of particles nucleated other than automating the charging and initial conditions of the reactor as well as possible. However, once the first generation of particles has been nucleated, one of the sensor sets and the models proposed in the last section can be used to predict what the final PSD will be if no control action is taken. If this is outside the no-control region defining normal or common cause variations, then the emulsifier shot at 150min can be adjusted so that the second mode of the final PSD can be controlled to the target.

Control Algorithm:

The calculation steps for obtaining the amount of emulsifier shot needed are:

1) For a new batch, collect available on-line (x_{on}) and off-line (x_{off}) process measurements such as PSD, T_j, D_p, and S_f up to time 150 minutes.

2) Denote these observations by x_j, j=1,…,b in the PLS regression model:

$$\hat{y}_i(u_{c,2}) = \hat{\beta}_{i0} + \hat{\beta}_{i1}u_{c,2} + \sum_{j=1}^{b}\hat{\beta}_{ij+1}x_j \quad i=1,2,\ldots,n \tag{3.5}$$

where n is the number of points measured in the final PSD (60 in this case).

3) Solve for control action $u_{c,2}$ (emulsifier injection at time 150min) by minimizing the deviation of the final $\hat{y}_i$ from their target values $y_{sp,i}$ in a least squares sense if the predicted quality falls outside the acceptable control region in the score space:

$$\underbrace{\min}_{u_{c,2}} \sum_{i=1}^{n}(y_{sp,i} - \hat{y}_i(u_{c,2}))^2 \tag{3.6}$$

The solution is given by:

$$u_{c,2} = \frac{\sum_{i=1}^{n}\hat{\beta}_{i1}d_i}{\sum_{i=1}^{n}\hat{\beta}_{i1}^2} \tag{3.7}$$

where $d_i = y_{sp,i} - \hat{\beta}_{i0} - \sum_{j=1}^{b}\hat{\beta}_{ij+1}x_j$

Control results

Once the prediction for the final PSD is made and if it is determined that it falls outside the no-control region, a mid-course correction is taken (In this example, the no-control region is determined using only the secondary distribution to be controlled). Table 3.5 shows the results of the control actions for the set of test batches (Table 3.3) using the 5 different models and measurement sets defined in Table 3.2 and 3.4. As an illustration of the control performance, Figures 3.5a and 3.6a show the score plots for the controlled mode of the final PSD for model 1 and 5 (models 2, 3, and 4 have similar performance to that of model 1). In each Figure, the five points (*) indicate what would have happened if no control action has been taken, and the five points (o) indicate the

mid-course control results. It is interesting to notice that the reduced latent variable space is one-dimensional because the disturbances (a_s, f^*) affect the quality PSD in basically the same way. It is also important to notice that the control action acts in the same direction as both disturbances. This is the main reason why only one manipulated variable is used: From the theoretical point of view this makes sense. The performance of the control corrections can also be illustrated in the space of the PSD as shown in Figures 3.5b and 3.6b. Three distributions are presented: the target distribution (-), the distribution that would have occurred with no control action (···), and the distribution that resulted from the mid-course correction (--)

We can see from these Figures that mid-course corrections using model 1 yield excellent results since it controls the final PSD close to its desired target for all the test sets. This is also true for models 2 to 4. In general, a model that uses PSD or D_p together with on-line T_j measurements can both predict and control bad batches with excellent results. However, as seen in Fig. 3.6a, model 5, which uses only information obtained from on-line T_j measurements cannot predict that observation 3 is not on target and the resulting control action drives the PSD further from target. In practical situations, when a prediction falls inside the no-control region (observation 3), no mid-course correction should be taken, although for illustration purposes, we have shown what would have happened if action had been inappropriately taken. The correction for test batch 5 is also poor.

To compare the control performance with different models, the sum of squared error (SSE) was computed on the tracked PSD (y) with respect to the PSD target (y_{sp}) in the full space:

$$SSE = \sum_{j=1}^{m} \sum_{i=1}^{n} \left(y_{ij} - y_{sp,ij} \right)^2 \tag{3.8}$$

where m is the number of test batches (5). In Figure 3.7, the performance of five models is compared. We can see from this plot that models that use information directly related

with particle generation have low SSE. The high SSE of model 5 shows that only using jacket temperature cannot achieve good control.

Figures 3.8 and 3.9 illustrate the results obtained when this control strategy is applied to the PSD obtained with Recipe 2. These figures indicate that the control results are similar to those obtained with Recipe 1, and hence independent of the recipe employed. Figure 3.8a shows the reduced spaced control region for model 3 (no-control region determined using only the secondary distribution to be controlled), while Figure 3.8b the control in the full space, and Figure 3.9 the SSE.

From the results obtained, it can be noticed that in this approach, the secondary distribution is easily controlled since only one adjustment of the emulsifier shot ($u_{c,2}$) is needed to move the distribution back towards the target. Because an emulsifier shot is always performed, this approach has the advantage of not taking extra controller actions and only the amount of emulsifier ($u_{c,2}$) at the normal injection time (150min) needs to be corrected to compensate for the effect of the disturbances. Therefore, the algorithm is especially suitable when the second particle generation is greater or more important than the first one (for example Figure 3.8b) and when the PSD intermediate measurements can only be obtained with significant delay. However, in the case that the first generation is of great concern as well, then control of both modes of the full distribution should be attempted. This approach is presented in the next section.

Table 3.5 Moles of emulsifier to be injected at 150min for Case I, Recipe 1.

Test set	Model 1	Model 2	Model 3	Model 4	Model 5
1	0.0301	0.0304	0.0301	0.0303	0.0294
2	0.0343	0.0346	0.0344	0.0344	0.0341
3	0.0344	0.0341	0.0343	0.0343	0.0314
4	0.0294	0.0303	0.0294	0.0298	0.0296
5	0.0272	0.0287	0.0272	0.0282	0.0260

Base Case: Emulsifier moles injected=0.032

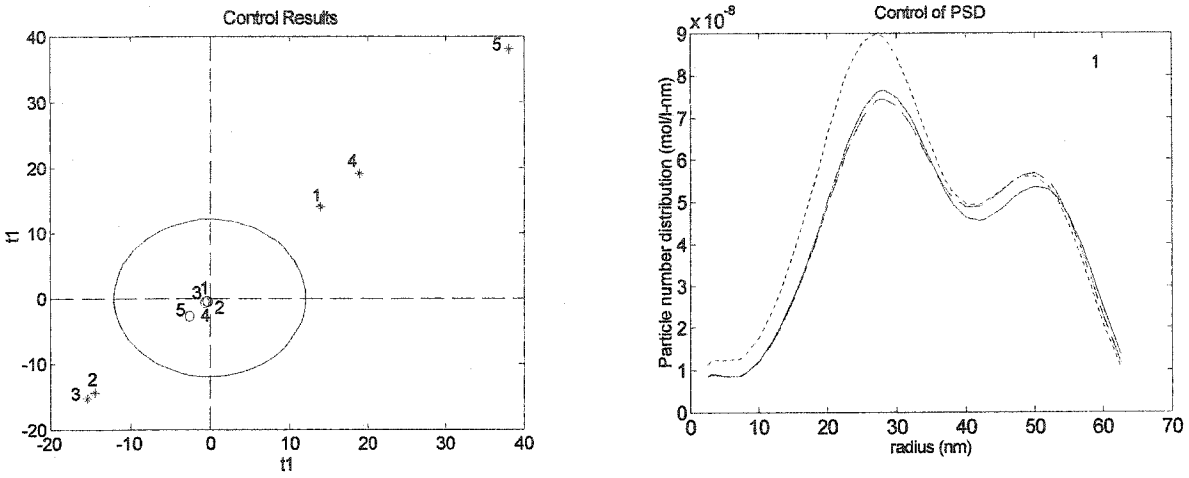


Figure 3.5 Control results for Case I, Recipe 1 using model 1. a) Score plot for the 5 test batches. Observation without control (*) and with control (o), and b) Final PSDs for test batch 1, Recipe 1: without control (---), with control based on model 1 (– –), and target distribution (–).

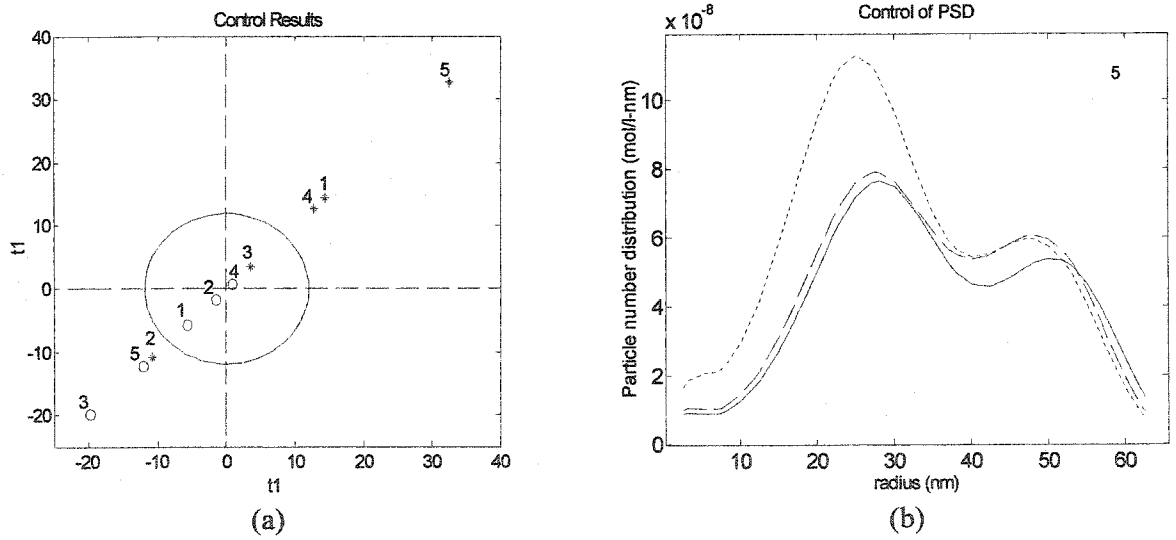


Figure 3.6 Control results for Case I, Recipe 1 using model 5 a) score plot for the 5 test batches. Observation without control (*) and with control (o), and b) Final PSDs for test batch 5, Recipe 1: without control (---), with control based on model 5 (– –), and target distribution (–).

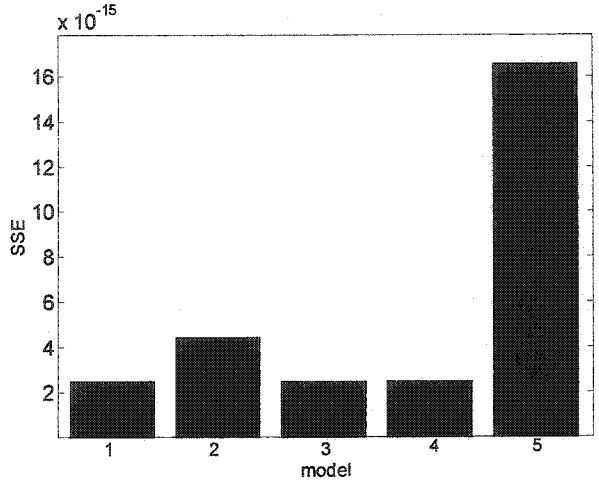
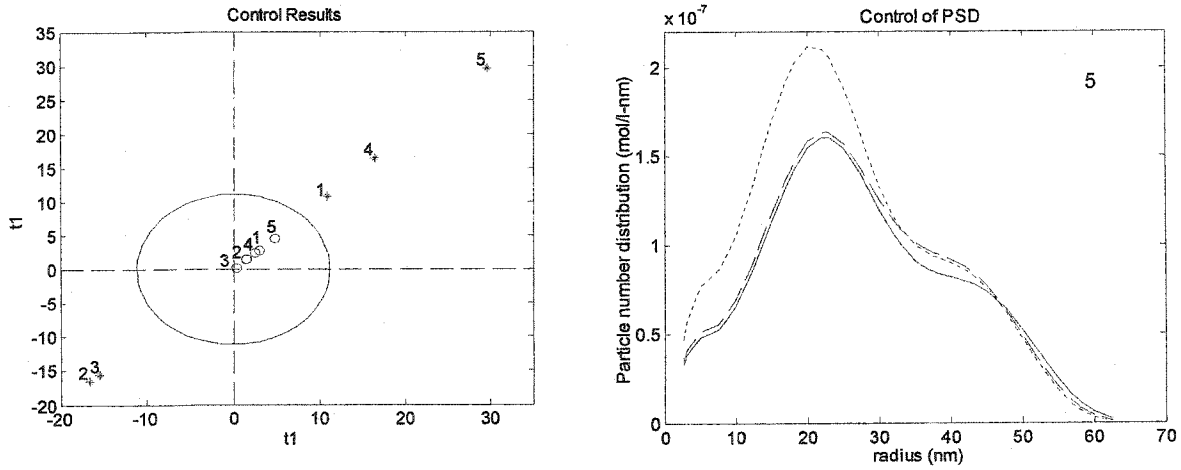


Figure 3.7 Control performance of models 1-5 for Case I, Recipe 1.



(a) (b)

Figure 3.8 Control results for Case I, Recipe 2 using model 3. a) score plot for the 5 test batches. Observation without control (*) and with control (o), and b) Final PSDs for test batch 4, Recipe 2: without control (---), with control using model 3 (– –), and target distribution (–).

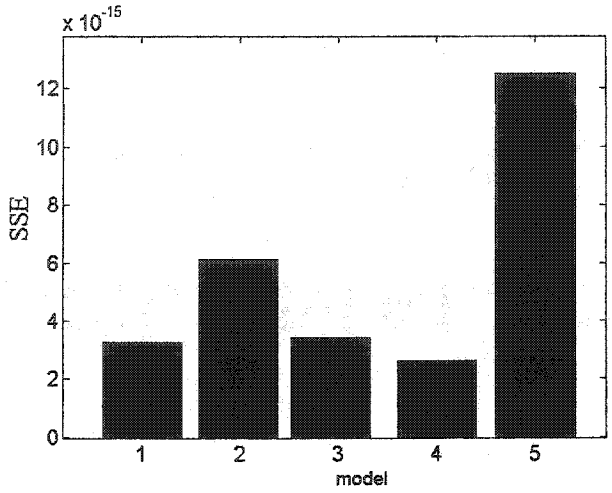


Figure 3.9 Control performance of model 1-5 for Case I, Recipe 2.

3.5.5 Case II: Control both modes of the bimodal distribution.

Control of the full distribution by regulating both the primary and secondary particle generations was also studied. This is possible because the particle generation in the emulsion polymerization of styrene takes approximately 25-35min (depending of the formulation and type of process operation) so some measurements can be taken before the first particle generation is over. The sampling time for the single measurements of the particle diameter or the particle size distribution is now performed at 20min instead of 30min as can be seen in Table 3.4. The mid-course control actions now consists of using the information available (x_{on}, x_{off}) up to 20 minutes and computing the emulsifier adjustments ($u_{c,1}, u_{c,2}$) to be performed at two time points, one ($u_{c,1}$) at 30 minutes to compensate for disturbances in the first particle generation, and the other ($u_{c,2}$) at 150 minutes to ensure that the secondary generation is compensated for both the effect of the disturbances detected and for the effect of the first adjustment ($u_{c,1}$) made at 30 minutes. Both corrective actions ($u_{c,1}, u_{c,2}$) are computed simultaneously at 30 min (i.e it is used only one decision point and therefore one model at 30 min.) Note that in order to

implement this policy one need to have a fast analyzer to obtain Dp or PSD in order to use the results before the first nucleation period is over.

The algorithm for computing the emulsifier shots needed at 30 min and 150min is similar to that of Case I. However, two control actions (instead of one) have to be obtained from the following objective function:

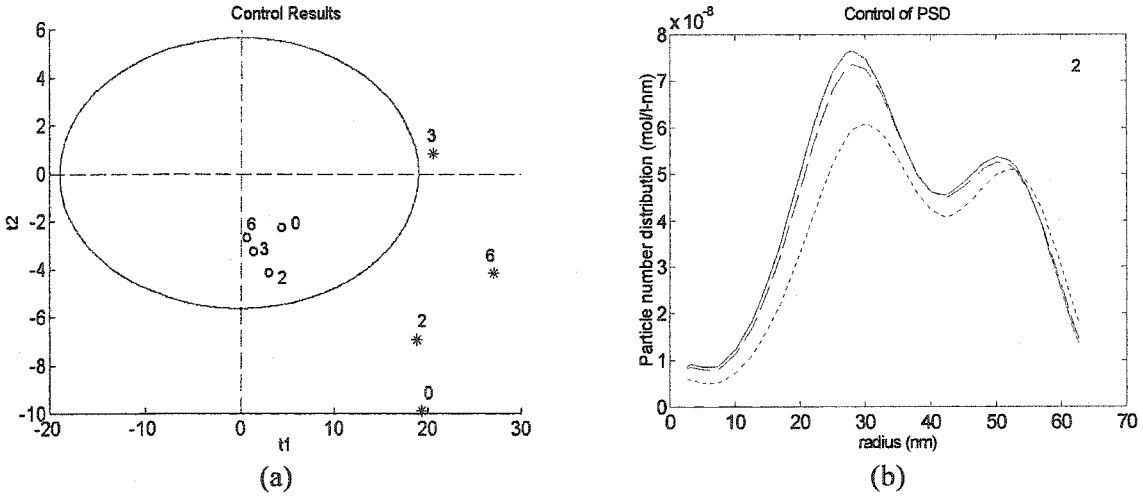
$$\begin{aligned} &\underbrace{\min}_{u_{c,1},u_{c,2}} \sum_{i=1}^{n}(y_{sp,i} - \hat{y}_i(u_{c,1},u_{c,2}))^2 \\ &s.t. \qquad u_{c,1} \geq 0 \end{aligned} \tag{3.9}$$

where $u_{c,1}$ and $u_{c,2}$ are the amount of emulsifier to be injected at 30 and 150min respectively.

As an illustration of the control obtained with this approach, the performance using model 1 is shown in Figure 3.10. Figure 3.10a shows the predicted PSDs if no control actions were taken (*) and those when control action was taken (o) in the principal component score space for test batches 0,2,3 and 6 which led to too few particles being nucleated in the first period. Only in this case will the control call for additional emulsifier to be injected at the 30 minute interval. In the other test batches, too many particles were nucleated and no compensation can be done to affect the first particle generation since emulsifier cannot be removed from the reactor. Hence in test batches 1, 4 and 5 the emulsifier shot ($u_{c,1}$) at 30 minutes is zero. The results of all mid-course corrections using this strategy are shown in Table 3.6. It is worth to mention that these results are similar to those obtained in Case I. In spite of the good results obtained with this approach (Case II), its main drawback lies in that samples have to be taken earlier in the process and the analyses have to be on-line and fast. To overcome this limitation, a more flexible approach to control the full PSD is described in the next section.

Table 3.6 Moles of emulsifier to be injected at 30 and 150min for Case II.

Test set	Model 1		Model 2	
Injection time	30min	150min	30min	150min
0	0.0031	0.0314	0.0037	0.0333
1	0	0.0304	0	0.0307
2	0.0029	0.0316	0.0037	0.0332
3	0.0025	0.0323	0.0036	0.0330
4	0	0.0300	0	0.0291
5	0	0.0284	0	0.0290
6	0.0031	0.0326	0.0037	0.0336



(a) (b)

Figure 3.10 Control results for Case II using model 1: Score plot for model 1. Batches without control (*) and with control (o). Ellipse denotes no-control region. Final PSDs for test batch 2: without control (-- -), control correction (– –), and target distribution (–).

3.5.6 Case III: Control of relative distributions.

In this Case, rather than just controlling the secondary distribution to a specified PSD regardless what happens to the first PSD generation (Case I), it would be easier to adjust the target for the second generation based on what was achieved in the first one, so that the desired shape of the overall distribution was kept the same. In the earlier studies (Case I and II) one was effectively trying to control the absolute PSD, i.e., both the shape of the distribution and the total number (or mol) of particles in each bin. Here we only focus on controlling the shape and let the total amount of polymer vary slightly. This type of control is recommended when the first particle generation is fast and there exists long

delays in the process measurements. Two correction variables are chosen for this control objective: an emulsifier shot ($u_{c,2}$) at 150min and the total reaction time ($u^*_{c,2}$). The shot of emulsifier controls the relative height of the two distribution peaks while the reaction time adjusts the location of the peaks to match the target distribution.

Control computation

The key idea in this approach is to solve for the control actions and unknown distribution simultaneously based on the information provided by the process measurements. This is necessary because the final distribution will be determined from the first nucleation. For example, if more particles are generated in the first generation, then more particles will have to be generated in the second one in order for the overall distribution shape to match that of the target. To perform this control the following steps are needed:

1) Denote the available observations at time 150min by x_j, $j=1, \ldots, b$ in the regression model:

$$\hat{y}_i = \hat{\beta}_{i0} + \hat{\beta}_{i1} u_{c,2} + \hat{\beta}_{i2} u^*_{c,2} + \sum_{j=1}^{b} \hat{\beta}_{ij+2} x_j \qquad (3.10)$$

2) Solve for control action, $u_{c,2}$ (emulsifier shot at 150min), and $u^*_{c,2}$ (time of reaction) which will minimize the deviation $\hat{y}_i$ from the target values $y_{sp,i}$ using constrained linear least-squares:

$$\underbrace{\min}_{\mathbf{z}} \frac{1}{2}\left\|\mathbf{Cz}-\mathbf{d}\right\|_2^2 \qquad \text{such that}$$

$$y_{sp,\max 1} \geq y_{sp,i} \quad i=1\ldots p$$

$$y_{sp,\max 2} \geq y_{sp,i} \quad i=p+1\ldots n; \tag{3.11}$$

$$y_{sp,\max 1} = r y_{sp,\max 2}; \quad \text{and} \quad \mathbf{z} \geq \mathbf{0}$$

where

$$\mathbf{C} = \underbrace{\begin{bmatrix} \hat{\beta}_{11} & \hat{\beta}_{12} & -1 & 0 & \cdots & 0 \\ \hat{\beta}_{21} & \hat{\beta}_{22} & 0 & -1 & \cdots & 0 \\ \vdots & \vdots & \vdots & \vdots & \ddots & \vdots \\ \hat{\beta}_{n,1} & \hat{\beta}_{n,2} & 0 & 0 & \cdots & -1 \end{bmatrix}}_{(n\mathrm{x}(n+2))} \quad \mathbf{z} = \underbrace{\begin{bmatrix} u_{c,2} \\ u^*_{c,2} \\ y_{sp,1} \\ \vdots \\ y_{sp,n} \end{bmatrix}}_{((n+2)\mathrm{x}1)} \quad \mathbf{d} = \underbrace{\begin{bmatrix} \hat{\beta}_{10} + \sum_{j=1}^{b} \hat{\beta}_{1j+2} x_j \\ \hat{\beta}_{20} + \sum_{j=1}^{b} \hat{\beta}_{2j+2} x_j \\ \vdots \\ \hat{\beta}_{n,0} + \sum_{j=1}^{b} \hat{\beta}_{nj+2} x_j \end{bmatrix}}_{(n\mathrm{x}1)}$$

$\mathbf{z}$ is the vector containing the unknown distribution ($y_1 \ldots y_n$) and control actions $u_{c,1}, u_{c,2}$; $\mathbf{d}$ is a vector obtained from process measurements; r is the ratio of the desired peak maxima of the first and second particle distributions, p is the interval at which the second generation begins (in this case p=40), and $y_{sp,max1}$ and $y_{sp,max2}$ are the maxima of the two peaks in the target distribution.

Control results

Once it is determined that the prediction falls outside the no-control region, a mid-course correction has to be taken. Table 3.7 shows the control actions implemented for the test batches 1 to 5. Figure 3.11a shows the control region in the reduced latent variable space for model 1, while Figure 3.11b shows uncontrolled and controlled PSD in the full space. For comparison purposes, the final PSDs of Figures 3.11a and 3.11b has been normalized according to:

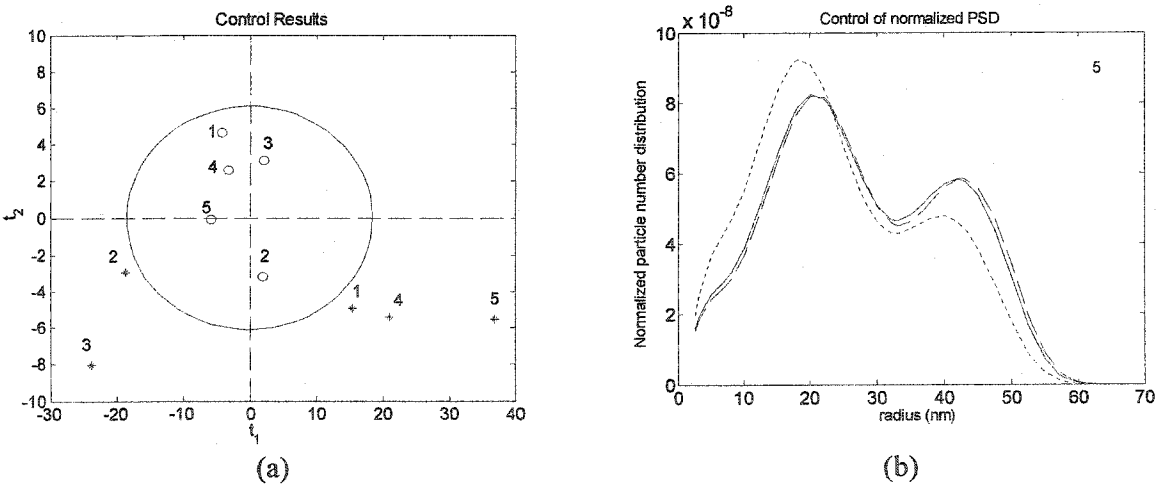
$$y_i^* = y_i \left(\frac{a_0}{a_k} \right) \tag{3.12}$$

where y_i^* represents the normalized fraction of particles at radius i (i=1,…,n, n=60) of the PSDs, y_i the original fraction of particles, a_0 the area of the desired PSDs set-point and a_k the area of the achieved PSD for the k-th test batch. The area can be computed as: $a_k = \int_0^n y(i)di$.

The results of using this approach show that the shape of the full PSD can be controlled without the disadvantages of the approach in Case II.

Table 3.7 Moles of emulsifier to be injected at 150 and end reaction time for Case III.

	Model 1		Model 2	
Test Set	Shot at 150min	End Time (min)	Shot at 150min	End Time (min)
1	0.0309	412	0.0310	412
2	0.0335	384	0.0327	393
3	0.0341	382	0.0339	384
4	0.0304	416	0.0307	414
5	0.0288	425	0.0284	422



(a) (b)

Figure 3.11 Control results for Case III using model 1. a) Normalized score plot for model 1. Observation without control (*) and with control (o). Ellipse denotes no-control region, and b) Normalized final PSD for test batch 2: without control (---), with control (– –), and target distribution (–).

3.6 Fault detection

Even with only one off-line process measurement (for example the PSD obtained from a sample taken at 30 min), it is possible to control the final PSD using mid-course corrections in all the proposed strategies. However, with a single sample measurement the control approach will be sensitive to any sensor fault. A bad measurement could easily lead to the predicted final PSD lying outside the no-control region and result in an inappropriate mid-course correction even when the batch is actually a good batch. This can be avoided by using sets of measurements from different sensors as given in Table 3.2 (e.g. off-line PSD and on-line T_j). One can then always test for measurement inconsistencies among different sensors.

PLS provides a powerful way of accomplishing this because, unlike normal regression and neural network methods it provides a model for the regressor (i.e. sensor response) space as well as giving a prediction of the final PSD (Burnham et al. [1999]). Therefore, prior to using any measured data for control of a new batch, the square prediction error (SPE) of the new vector of observations can be computed. This SPE provides a measure of any inconsistency between the vector of measurements for the new batch and the behavior of the set of measurements used to develop the PLS model (Kourti and MacGregor [1996] and Nomikos and MacGregor [1994]).

This monitoring capability of the PLS model is illustrated in Figure 3.12 using model 1 in Table 3.3. Batch 48 represents a good batch in which the measurements were good and so its SPE in the upper plot in Figure 3.12 small and below statistical 95% test limit. Batch 49 is a repeat of batch 48 but with a bias in the PSD measurement, while batch 50 is also a repeat of batch 48 but with a bias in the T_j measurements. In both cases the SPE statistic clearly detects a measurement problem. An alternatively approach could be to build a PCA model for each sensor separately. In this approach, each sensor would be tested separately by computing the SPE of each specific sensor group from its own PCA model. This is illustrated in the lower plots of figure 3.12. On the left is the SPE plot for the PSD measurements while in the right is that from the Tj measurements. In all

cases the faulty sensor was detected and the control would be suspended until the sensor is fixed.

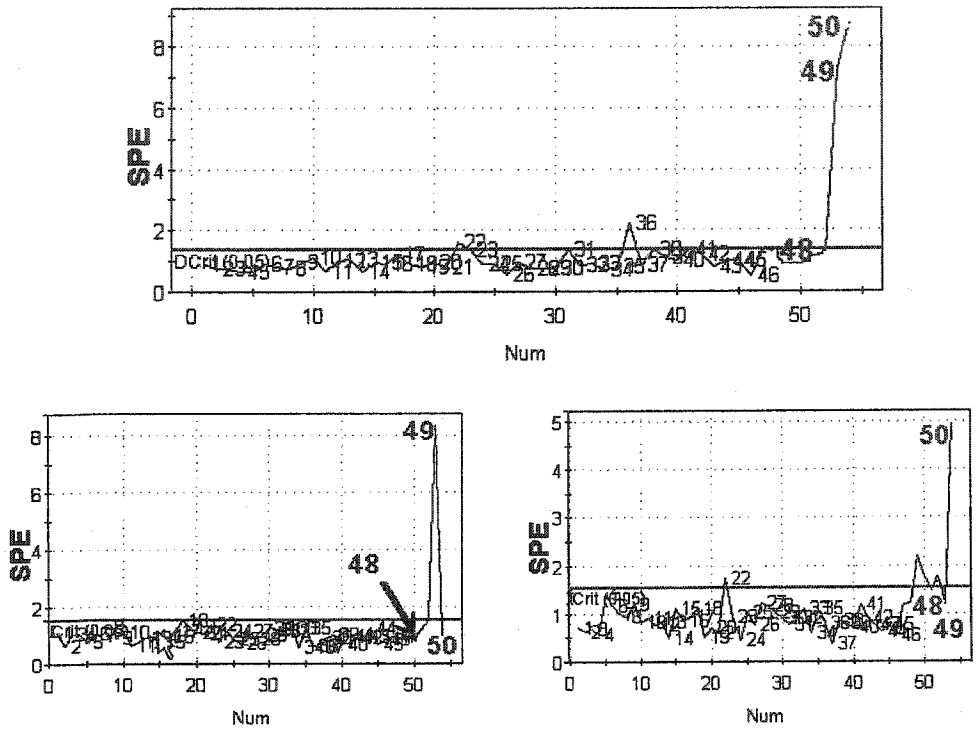


Figure 3.12 SPE for a good batch (48) in which exists analysis fault in process measurements PSD (49) and T_j (50)

3.7 Extensions to multi-modal distributions and model error.

PLS Models for the prediction of Multi-modal distributions can be obtained in the same way as those described in section 3.5.5 for the bimodal case. However, in the case of control, the number of emulsifier shots may potentially increase to the number of dominant distribution modes (one emulsifier shot for every particle generation). Therefore, information regarding the effect of every emulsifier shot (or other(s) potentially manipulated variable(s)) on the particle generations has to be present in the dataset used for model building. The computation of the control actions can basically follow two different paths: 1) Obtain all control actions simultaneously in the same form as that described in Case II (assuming that no disturbances other than those originally present since the start of the reaction will affect the subsequent particle nucleations and

that the information obtained from the available measurements can predict adequately such incoming distributions), or 2) Obtain every control action from an independent control computation (as will be described in Chapter 4). In the latter approach, it would be necessary to build a model for every particle nucleation that would relate the control action to the corresponding particle generation. The advantage of the second approach is that it can account for new incoming disturbances as well as the effect of any previous incorrect control actions that were taken.

In the case that new or larger disturbances affect the current batch than those present in the dataset used for model building and control, it may be possible that the prediction, and therefore, the control computation is inadequate. In this situation one might be able to use the new incoming information (assuming that this incoming data has information about the new disturbances affecting the process) in a recursive batch-to-batch way. This issue is addressed in detail in Chapter 4.

3.8 Conclusions

Several approaches to the multivariate end quality property control in semi-batch emulsion polymerization using mid-course correction policies were presented. The feasibility of these approaches was demonstrated based on simulation studies on styrene emulsion polymerization for the control of broad and bimodal particle size distribution using different sets of on-line and off-line measurements.

The results show that off quality PSD due to raw material disturbances can be controlled to a target distribution using off-line measurements of particle diameter and free emulsifier concentration, and/or particle size distribution from a sample taken early in the reaction together with on-line jacket temperature measurements. The manipulated variables were emulsifier shots at some mid-points in the reaction and the total reaction time. Since the approach is based on the use of readily available measurements, easily developed empirical models, and only occasional control actions at a mid-point time, the

approach could easily be implemented in industry. It appears to offer excellent control of the final PSD with much less effort than alternative approaches.

Nomenclature

$\hat{\boldsymbol{\beta}}$ = vector of estimated regression coefficient

σ = standard deviation

α = significance level

A = number of principal components

a_0 = PSD area set-point

a_k = PSD area for a test batch k

$\mathbf{E}'$ = residual matrix from PCA

F_α = critical value of the F distribution

f^* = lumped effective initiator efficiency

N = number of observations in the training set

$\mathbf{P}$ = loading matrix from PCA

$\mathbf{p}$ = loading vector from PCA

r = ratio of peak maxima between the first and second distribution

s_a^2 = variance of the score a

$\mathbf{T}$ = score matrix from PCA

$\mathbf{t}$ = score vector from PCA

$\mathbf{u}$ = vector of control action adjustments.

$\mathbf{x}$ = vector of measurements and control actions

y = observed or tracked PSD

y^* = normalized PSD

$\mathbf{y}_{sp}$ = target PSD

$\hat{\mathbf{y}}$ = predicted PSD

$\mathbf{z}$ = vector of unknown particle size targets and control actions

index

b = number of process measurements

i = bin number from the distribution segmentation (at certain radius)

j = measurements

k = test batch

l = time index

m = number of test batches

n = total number of distribution segments

p = interval at which the second nucleation begins

superscript

^ = indicates that the variable is estimated

subscript

sp = set-points

Chapter 4

Within-Batch and Batch-to-Batch Inferential-Adaptive Control of Batch Reactors

The purpose of this chapter is to present a control strategy that combines within-batch information from process variable trajectories and information from prior batches. The approach extends mid-course correction (MCC) strategies presented in Chapter 3 by including multiple decision and correction points, batch-to-batch information in the controllers, and an adaptive Partial Least Squares (PLS) approach to update the models from batch-to-batch. As with the approaches presented in Chapter 3, the scheme retains the "no-control region" concept where control is taken at various stages during the batch only if the projected error in the final quality is deemed to be statistically significant. The methodology is applied to the control of particle size distribution (PSD) in emulsion polymerization. The problem of regulation about a fixed set-point PSD in the face of disturbances, and the problem of achieving new set-point PSDs are both illustrated.

4.1 Introduction

In spite of the success of data driven approaches, they are mainly dependent on the quality of the training data used for identification. In the approaches taken before (Yabuki and MacGregor [1997], Yabuki et al. [2000], Russell et al. [1988a], Kesavan et al. [2000], Chapter 3), it is assumed that a training data set contains sufficient input

movements and disturbance information to allow proper model identification. If the above assumption is not accomplished, model error arising from low quality datasets (as those arising from only historical data) or changing disturbances and process conditions may degrade model prediction and control. For this, an important part of the control strategies presented in this Chapter is adapting poor initial models using batch-to-batch information.

The approach taken in this Chapter combines batch-to-batch and on-line batch control. It avoids the use of theoretical models by making efficient use of the on-line process trajectory data from the current and *previous* batches, as well as quality control data from occasional samples collected during the batches. The data requirements to build the models for this control are generally much less than with other empirical approaches that have been used. The approach incorporates adaptive PLS model updating at the end of the batch to overcome initial modelling errors and to adapt the model to new conditions when new PSD targets are specified.

The system used to illustrate the control strategies is the same one used in Chapter 3 (styrene batch emulsion polymerization for the PSD control developed by Crowley et al. [2000]). This theoretical simulator is used for data generation and control performance evaluation.

4.2 Disturbances, manipulated and controlled variables

As mentioned in Chapter 3, in nucleated emulsion polymerization systems the major disturbances affecting the PSD are those derived from raw material and/or reactive impurity variations. Therefore, in the simulations, variations are introduced into the emulsifier surface coverage potential (a_s) and the aqueous phase initiator dissociation constant (k_{daq}). Any water-soluble impurities present would have the effect of reducing the apparent value of k_{daq}. From the set of potential manipulated variables, $\mathbf{u}_c$ (injections of initiator, inhibitor, monomer and emulsifier; reactor temperature), injections of

emulsifier at different times were selected because they have the greatest and fastest effect on the particle nucleation rate.

4.3 Control methodology

The following description extends the mid-course correction strategies presented in Chapter 3 to include more than one decision point, model updating and batch-to-batch control. In this approach, one or more decision times (θ_l, $l = 1,2,...$) are specified at which control actions, involving a vector of manipulated variables ($\mathbf{u}_{c,l}$ $l=1,2,...$), can be taken. All the available information up to these decision times can be used in PLS models to predict the final product qualities at the end of the batch. If the projection of these predicted quality variables into a reduced dimensional latent variable space ($t_1,\cdots,t_A$) for the quality falls outside an acceptable region, then control action ($\mathbf{u}_c$) is taken. This is repeated at each decision time θ_l.

4.3.1 Prediction

Consider a PLS model for the batch process using the mean centred and scaled data available at certain decision times θ_l ($l = 1,2,...$) (For simplicity, only linear modelling is considered, although non-linear models and control can also be used):

$$\begin{aligned} \mathbf{X}_l &= \mathbf{\Gamma}_l \mathbf{V}_l^T + \mathbf{E}_l \\ \mathbf{Y} &= \mathbf{\Gamma}_l \mathbf{Q}_l^T + \mathbf{F}_l \end{aligned} \tag{4.1}$$

where the matrix $\mathbf{X}_l$ denotes the data on all the variables available at the decision time θ_l to be used for prediction, $\mathbf{Y}$ denotes the multivariate quality space, $\mathbf{\Gamma}_l$ contains vectors of new latent variables that captures most of the data variability (Höskuldsson [1988], Wold et al. [1989], Wold [1992], and Geladi and Kowalski [1986]), $\mathbf{V}_l$ and $\mathbf{Q}_l$ are matrices of coefficients and $\mathbf{E}_l$ and $\mathbf{F}_l$ error matrices.

The PLS model for any time θ_l may include, in the matrix of regressor variables ($\mathbf{X}_l$), data on the following variables from all the training batches: (*i*) on-line measurements on process variables such as temperatures, flows, pressures, etc. ($\mathbf{x}_{on,\,l}$) that are available up to time θ_l; (*ii*) any measurement made off-line in a quality control lab or by an infrequent analyser ($\mathbf{x}_{off,\,l}$) (e.g. an average particle size or a particle size distribution) that is available by time θ_l; (*iii*) any control action changes $\mathbf{u}^T_{c-prev,l} = [\mathbf{u}^T_{c,1}, \cdots \mathbf{u}^T_{c,l-1}]$ made at previous decision times θ_1, $\theta_2 \ldots \theta_{l-1}$, control action changes $\mathbf{u}_{c,l}$ made at the current decision time θ_l and under certain conditions future control actions $\mathbf{u}_{c-future,l}$ (θ_{l+1}, $\theta_{l+2} \ldots \theta_{end}$) for the current batch; (*iv*) any relevant information from immediately previous batches ($\mathbf{x}_{prior}$) that is useful for predicting the behaviour of the current batch (e.g. deviations of the final quality variables from their set-points at the last batch ($\mathbf{y}^{(k-1)}$), deviations in $\mathbf{x}^{(k-1)}_{on}$ and $\mathbf{x}^{(k-1)}_{off}$ from the last batch, or any control actions, $\mathbf{u}^{(k-1)}_{c}$, taken during the last batch). Which of these sets of variables is used, in any particular case, will be discussed later in the control studies. The final product quality matrix, $\mathbf{Y}$, contains, as rows, the observations on each quality variable for each batch ($\mathbf{y}^T$) in the training set.

The prediction from the PLS model (4.1) at each decision time θ_l for the final vector of quality variables for the current batch can be rearranged and expressed in linear regression form as:

$$\hat{\mathbf{y}}^T(\theta_l) = \mathbf{x}_l^T \hat{\boldsymbol{\beta}}_l$$
$$\text{where } \mathbf{x}_l^T = [\mathbf{x}^T_{on,l}, \mathbf{x}^T_{off,l}, \mathbf{x}^T_{prior}, \mathbf{u}^T_{c-prev,l}, \mathbf{u}^T_{c,l}, \mathbf{u}^T_{c-future,l}] \tag{4.2}$$

Again, which of these sets of variables is used, in any particular case, will be discussed in the control studies section.

4.3.2 Control computation

If it is determined that a control correction action at time θ_l is needed (according to the no-control region concept described in Chapter 3), this can be obtained using a Linear Quadratic Regulator (LQR):

$$\underbrace{\min}_{\mathbf{u}_c}(\mathbf{y}_{sp} - \hat{\mathbf{y}}(\theta_l))^T \mathbf{Q}_1 (\mathbf{y}_{sp} - \hat{\mathbf{y}}(\theta_l)) + \mathbf{u}_c^T \mathbf{Q}_2 \mathbf{u}_c \tag{4.3}$$

where $\hat{\mathbf{y}}(\theta_l)$ is given by eq. (4.2), and $\mathbf{u}_c$ is a vector of manipulated variables that will be obtained by solving (4.3). Depending on the set of variables used for model building (eq. 4.2), $\mathbf{u}_c$ may be composed of $\mathbf{u}_c = \mathbf{u}_{c,l}$ or $\mathbf{u}_c^T = [\mathbf{u}_{c,l}^T \ \mathbf{u}_{c-future,l}^T]$. Hard constraints can be introduced in the manipulated variables (MV) $\mathbf{u}_{c\min} \le \mathbf{u}_c \le \mathbf{u}_{c\max}$ if needed. The solution to the LQR problem in (4.3) is easily obtained using optimisation. Alternatively, a Minimum Variance Controller (MVC) can be used:

$$\underbrace{\min}_{\mathbf{u}_c}(\mathbf{y}_{sp} - \hat{\mathbf{y}}(\theta_l))^T \mathbf{Q}_1 (\mathbf{y}_{sp} - \hat{\mathbf{y}}(\theta_l)) \tag{4.4}$$

together with a simple minimum variance detuning factor

$$\mathbf{u} = \boldsymbol{\delta} \mathbf{u}_c \tag{4.5}$$

where $\mathbf{u}_c$ is the computed and $\mathbf{u}$ is the implemented vector of control actions, and $\mathbf{0} \le \boldsymbol{\delta} \le \mathbf{1}$ is a detuning factor.

4.4 Adaptive model

If the model obtained from eq. (4.1) is based on a poor data set, it is possible that model error lead to poor control actions and then to poor product quality. To overcome this limitation, batch-to-batch model parameter adaptation is introduced.

Using the training data set $\mathbf{Y}^{(0)} = \mathbf{Y}_{tr}$ and $\mathbf{X}_l^{(0)} = \mathbf{X}_{tr,l}$ the initial (nominal) model is given by equation (4.2).

Updating of such model with new batch data (k) is:

$$\text{for a batch } k \quad (k = 1,2...)$$

$$\mathbf{Y}^{(k)} = \begin{bmatrix} \mathbf{Y}^{(k-1)} \\ \mathbf{y}^{(k)T} \end{bmatrix}; \quad \mathbf{X}_l^{(k)} = \begin{bmatrix} \mathbf{X}_l^{(k-1)} \\ \mathbf{x}_l^{(k)T} \end{bmatrix} \tag{4.6}$$

$$\left[\mathbf{Y}^{(k)} \middle| \mathbf{X}_l^{(k)}\right] \xrightarrow{PLS} \hat{\boldsymbol{\beta}}_l^{(k)}$$

The updated model is:

$$\hat{\mathbf{y}}^{(k)T}(\theta_l) = \mathbf{x}_l^{(k)T} \hat{\boldsymbol{\beta}}_l^{(k)}$$

The model updating can be achieved by simply augmenting the $\mathbf{X}_l$ and $\mathbf{Y}$ matrices with the new data at the end of each batch and refitting the PLS model, or by using a recursive exponentially weighted adaptive PLS algorithm (Dayal and MacGregor [1997a,b]).

4.5 Control studies

The inherent flexibility of empirical models allows having several alternatives for model building (different sets of variables used as regressors in eq. (4.2)). Selection of the regressor vector depends on *i*) available measurements and *ii*) nature of the disturbances affecting the system. To illustrate such flexibility and the effectiveness of the methodology, within-batch and batch-to-batch control for an emulsion polymerization process is presented. Case study I involves within-batch control of the PSD. It is shown how an initially very poor model is improved using batch-to-batch adaptation while rejecting different types of disturbances using only within-batch control. In case study II both batch-to-batch and within-batch information is used for the PSD control. Batch-wise constant disturbances are rejected while improving the quality of the models using batch-

to-batch updating. Furthermore, a set-point change in the shape of the particle size distribution is shown to be achieved within a few batches when starting with information limited to a region around a completely different PSD set-point. These control case studies are summarized in Table 4.1.

Table 4.1 PSD Case studies and control strategies*

Case	Control Strategy	Disturbance type
I	Within-batch only	Batch-wise constant and batch-wise uncorrelated
II	Batch-to-batch + within-batch	Batch-wise constant and set-point change

*In both Case studies, batch-to-batch adaptation is performed.

4.6 Case Study I: On-line inferential control using batch-to batch adaptation.

In the examples that follow, one off-line measurements, $\mathbf{x}_{off}$ (the average particle diameter, Dp or the full PSD sampled at 20 min with a 10 min analysis time delay) together with on-line jacket temperatures (T_j), $\mathbf{x}_{on}$ are used to predict a final bimodal PSD. As in Case II of Chapter 3, two emulsifier shot adjustments (u_{c1}, u_{c2}) at θ_1=30 and θ_2=150 min (total reaction time is now 380 min) are performed; each one of these emulsifier shots will be used to control one of the particle generations: a 30 min shot adjustment, $u_{c,1}$ for the first generation while an adjustment to the nominal shot at 150 min, $u_{c,2}$ will be made to control the second generation distribution. In this example it is assumed that a fast analysis (10 minutes) can be performed or that an on-line PSD or Dp analyser is available; in the case that it is not possible to obtain such rapid measurement at the 30 min interval, batch-to-batch control can be applied using emulsifier adjustments at the beginning of the batch (θ_0=0min, as will be described in section 4.7). In this example we first consider that the PSD measurement is error free and that the control corrective actions computed from the algorithm (eq. 4.4) can be perfectly implemented (using, for example, a precision metering pump). Measurements on T_j have a normal random error with standard deviation (σ) of 0.1% (approximately $\pm 323 \times .001 \approx 0.3^{\circ}K$), while the noise for Dp is σ=1% (30nm $\times 0.01 \approx .3$nm). The effect that measurement errors on the

intermediate (grab sample) PSD measurements will have on control performance is addressed in section 4.7.3. The disturbances considered are batch-to-batch variations in emulsifier surface coverage (a_s) and the aqueous phase initiator dissociation rate constant (k_{daq}) with standard deviation equal to 2 and 3% respectively.

As shown in Case II of Chapter 3, the effect that the raw material disturbances have on the end-quality properties (PSD) can be adequately predicted using on-line process temperature measurements ($\mathbf{x}_{on,l}$) and an intermediate off-line measurement $\mathbf{x}_{off,l}$ (either Dp or PSD) at a point early in the batch (i.e. a single decision point at θ_l=30 min is used). To improve the prediction, the PSD space was linearized using a square root transformation and the PLS model (with 5 LV) was extended with a quadratic and an interaction term in the manipulated variables in order to handle some nonlinearities in the effects of the manipulated variables.

$$\text{At } \theta_l=30 \text{ min} \quad \hat{\mathbf{y}}^T = [u_{c,1}\ u_{c,2}\ \mathbf{x}_{on,1}^T\ \mathbf{x}_{off,1}^T\ (u_{c,1}^2)\ (u_{c,1}u_{c,2})]\hat{\boldsymbol{\beta}} \tag{4.7}$$

By using eq. (4.8) both emulsifier control actions, $u_{c,1}$ and $u_{c,2}$, are determined simultaneously at time θ_l=30 min using the MV objective function:

$$\min_{\mathbf{u}_{c,1},\mathbf{u}_{c,2}} (\mathbf{y}_{sp} - \hat{\mathbf{y}}(\theta_1))^T \mathbf{Q}_1 (\mathbf{y}_{sp} - \hat{\mathbf{y}}(\theta_1)) \tag{4.8}$$

together with a detuning factor (δ).

It should be mentioned that if disturbances were to enter into the system at different times (other than at time θ=0) during the batch or if more quality related measurements were available at future times (off-line grab samples for PSD or Dp), then a multi-decision point (θ_l, l=1,2...) control scheme approach may be preferred. Such a multi-decision point approach is illustrated in Case study II (section 4.7).

In order to evaluate the robustness of the methodology, a Monte Carlo study was performed in which 40 different data sets were generated and used as a training set to

obtain 40 slightly different nominal PLS models, each of which was used as the starting point for the adaptive algorithm (eq. 4.6). The training data set is deliberately chosen to be rather poor in information content. It consists of observations on 22 batches: 19 subject only to normal random variations in a_s and k_{daq}, and *only* 3 batches in which some mid-course correction was performed. The PSDs from a typical training data set of 22 batches used for model building is shown in Figure 4.1a while the corresponding projections of these PSDs in the two dimensional PCA latent variable space (section 3.5.2, Chapter 3) for this **Y** data are shown in Figure 4.1b. Each point in the score plot (Figure 4.1b) corresponds to one of the distributions shown in Figure 4.1a, and summarizes its important deviation from the average PSD (given by t_1=t_2=0 in Fig. 4.1b).

The models obtained from these data sets are rather poor since most of the batches (19) contain little information other than on the correlation structure that exists during the production of good batches subject to small disturbances. This data on the 19 batches is used to define the "no-control" region. Only 3 batches (squares in Figure 4.1b) contain any effects of the MVs.

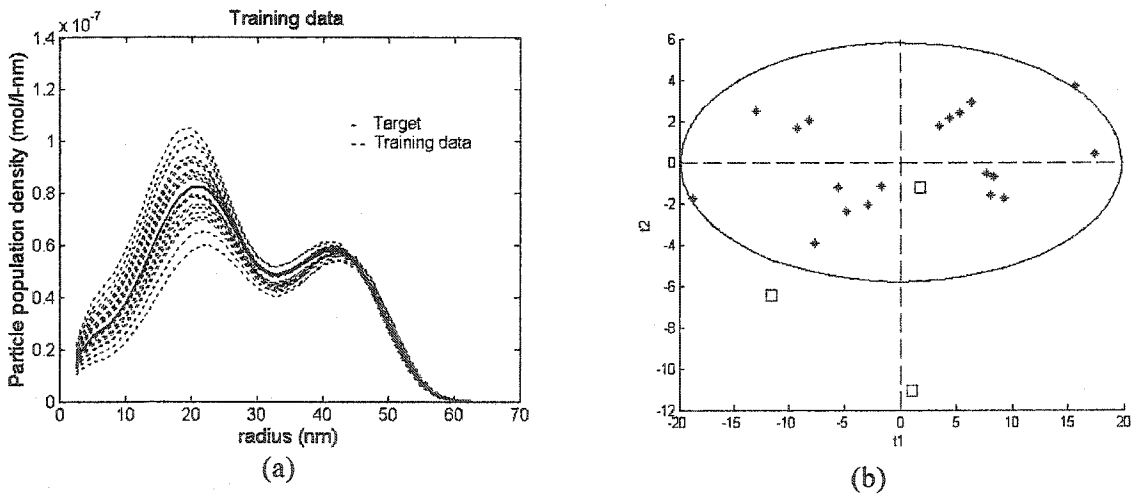


Figure 4.1 (a) PSDs of the training data set, solid curve is the target. (b) PSDs projected into the PCA score space. Ellipse denotes no-control region, *'s results from nominal operation conditions, and □'s results with a movement in the MVs.

To illustrate the control performance of the adaptive scheme (equations 4.6, 4.7 and 4.8) some results are presented in Figures 4.2 and 4.3. The averaged integral absolute error (IAE) is shown in Fig. 4.2 for all the different training data sets when the system is affected by a constant batch-wise disturbance (bias) in a_s (-28%) and k_{daq} (+20%). The IAE is computed as

$$IAE = \frac{\sum_{j=1}^{m}\sum_{i=1}^{r}\left|y_{ij} - y_{sp,ij}\right|}{m} \tag{4.9}$$

where m= number of Monte Carlo data sets and r= number of variables arising from the PSD segmentation (60 in this study). In Figure 4.2, (*) and (o) indicate the controller IAE performance when a single intermediate off-line Dp or a single PSD measurement, respectively is used as $\mathbf{x}_{off}$, along with the jacket temperature measurements up to θ_I=30 min ($\mathbf{x}_{on}$) in equation (4.7). It can be observed that better control is obtained when it is possible to have a full PSD measurement. However, even a simple Dp measurement can achieve reasonable performance, but at the expense of slower convergence. As an illustration of the control of the final PSD for one of the training data sets (this result is typical of the others) the progress of the PSD is shown in Figure 4.3a, while the progress of control in the reduced principal component score space is shown in Figure 4.3b (each PSD in Fig. 4.3a is summarized by a point in the score space of Fig. 4.3b). In Figures 4.3a,b it can be seen that control using the poor initial model (denoted by batch run number 1) only performs slightly better than when no control action is taken (indicated as batch zero (□)). However, by using the adaptive control algorithm (equations 4.6, 4.7 and 4.8), after only a few batches (4-10) the control is almost perfect (In this example, minimum variance tuning factor was kept δ=1 to show how even with a very large model error, the control algorithm is successful. However, in practice is recommended to use δ<1 to achieve some robustness to model error. Alternatively the LQC in equation (4.3) could be used.) Convergence of the parameters estimates ($\hat{\boldsymbol{\beta}}$) in the model (eq. 4.7) corresponding to the linear effects of the two manipulated variables ($u_{c,1}$ and $u_{c,2}$) on the

PSD at the 60 different radii for the PLS model is shown in Figure 4.4 (similar convergence for all other parameters is also achieved). The emulsifier shots values ($u_{c,1}$ and $u_{c,2}$) are shown in Figure 4.5 for the first 10 batches.

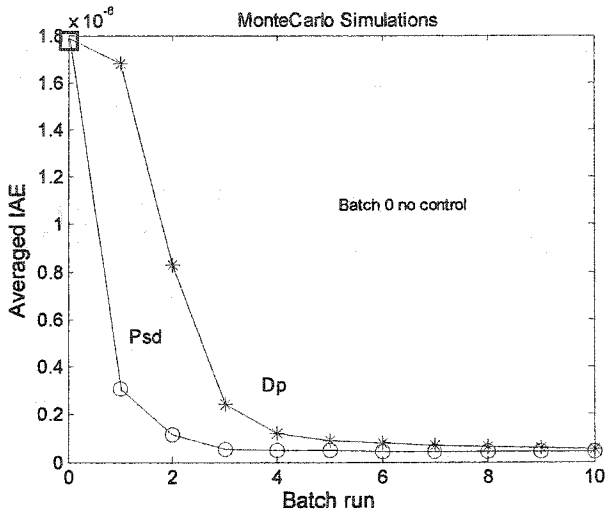
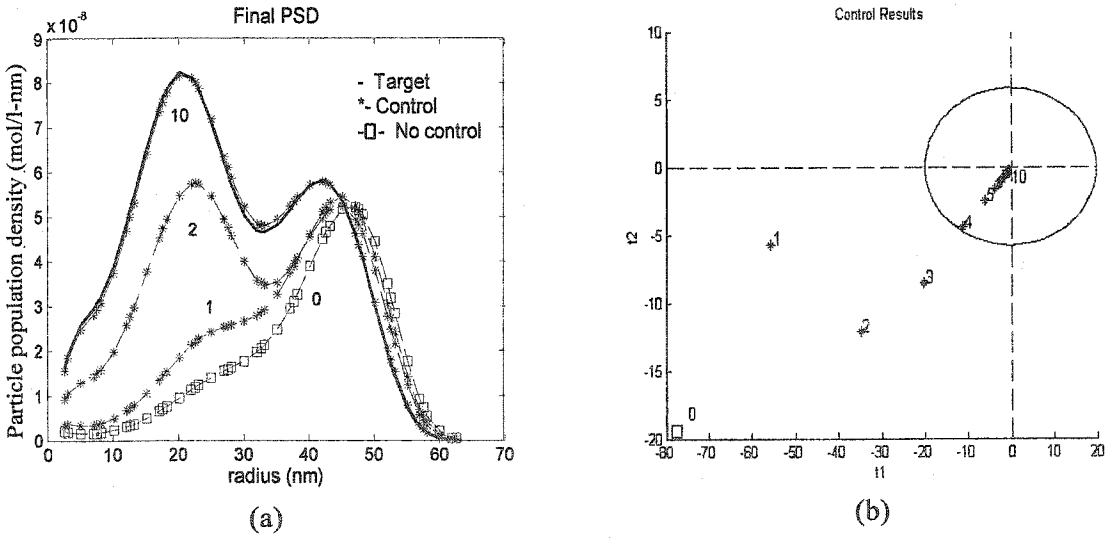


Figure 4.2 IAE of the PSD control using adaptive algorithm. Constant disturbance in a_s=0.72a^*_s, k_{daq}=1.2 k^*_{daq}. (□) no control, (o) control achieved using PSD and (*) Dp measurements.



(a) (b)

Figure 4.3 Adaptive PSD control with $x_{off,1}$=Dp subject to constant disturbance. (a) Control performance of the PSDs, and (b) Control in the reduced dimension PCA score space where ellipse denotes the no-control region, (*) achieved control and (□) no control. The number indicates the batch run.

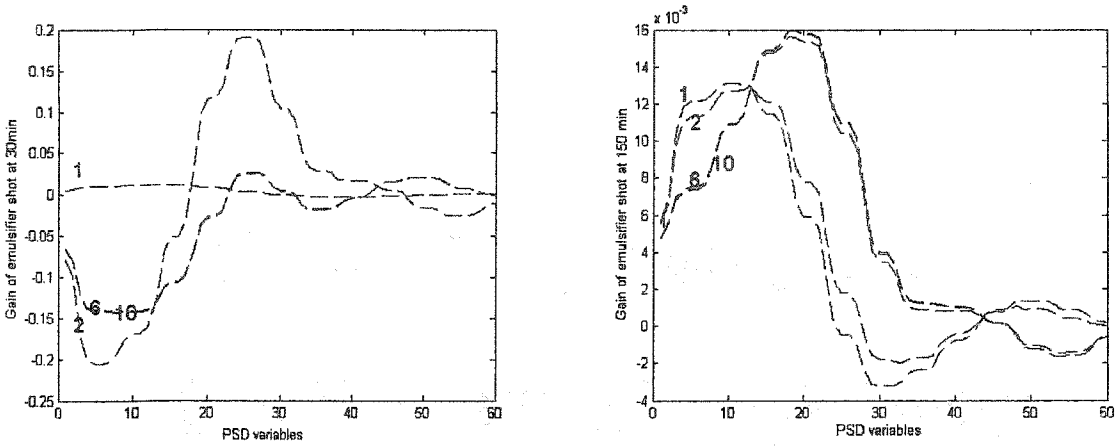


Figure 4.4 Adaptive estimates of the gains ($\hat{\beta}$) in the PLS model (equation 4.7) relating the MVs at 30 and 150 min (u_{c1}, u_{c2}) to the PSD (at 60 different sizes). The number indicates the batch run.

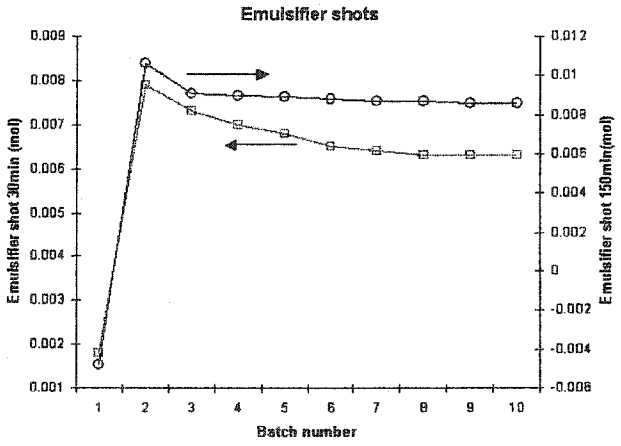


Figure 4.5 Manipulated variable adjustments (emulsifier shots). Control actions: u_{c1} at 30 min (□) and u_{c2} at 150 min (o).

The strategy proposed above is purely an on-line scheme (the adaptation is performed off-line, but the control action uses only within batch information) and therefore can reject both batch-wise uncorrelated as well as correlated disturbances. In Figure 4.6, the PSD control, in the reduced dimensional space, is shown for a series of batches suffering from frequent irregular changes (biases of different magnitudes) in the raw materials affecting the emulsifier surface coverage potential a_s. A single PSD (o) measurement, taken at 20 min ($x_{off,1}$), together with jacket temperatures ($x_{on,1}$) are used as

predictors. In this Figure, it can be seen that the final PSD is controlled into the desired target region for most of the batches. As the model improves, due to adaptation between batches, the control is seen to improve. For example, only batches 3,5, 6 and 7 are outside the control limits, but following the control scheme achieves acceptable control (inside the "common cause variation" region) for all subsequent batches.

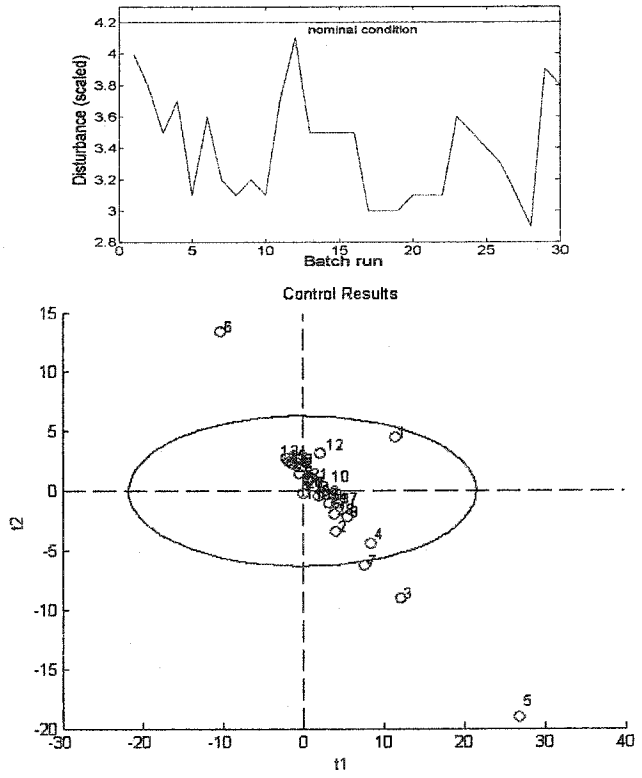


Figure 4.6 Adaptive PSD control for frequent changes in a_s (k_{daq}=1.2 k^*_{daq}) using an intermediate PSD measurement. The ellipse denotes the target region.

To illustrate that the control methodology is independent of the number and type of disturbances affecting the system, as long as these disturbances are detected by either on-line or off-line measurements, an example involving multiple (batch wise repetitive) disturbances and model mismatch in several parameters no present in the initial training data set, (Table 4.2) is shown in Figure 4.7. In this Figure it can be seen that after a few batches the control is satisfactory and that the model has been adapted to account for different disturbances as well as for the parameter mismatches.

Table 4.2 Disturbance magnitudes

Disturbance or parameter mismatch	Factor*
Critical micelle concentration (mol/*l*)	0.9
Propagation rate constant (*l*/mol-s)	0.95
Particle surface coverage potential (a_s, dm^2)	0.8
Monomer partitioning aqueous/droplet	0.9
Chain transfer rate constant (*l*/mol-s)	1.05
Diffusivity of monomer radical (cm^2/s)	0.95
Aqueous termination (*l*/mol-s)	0.95
Buffer concentration (mol/*l*)	0.9
Aqueous phase initiator dissociation constant (k_{daq}, s^{-1})	1.2

*Multiplicative factor in nominal conditions.

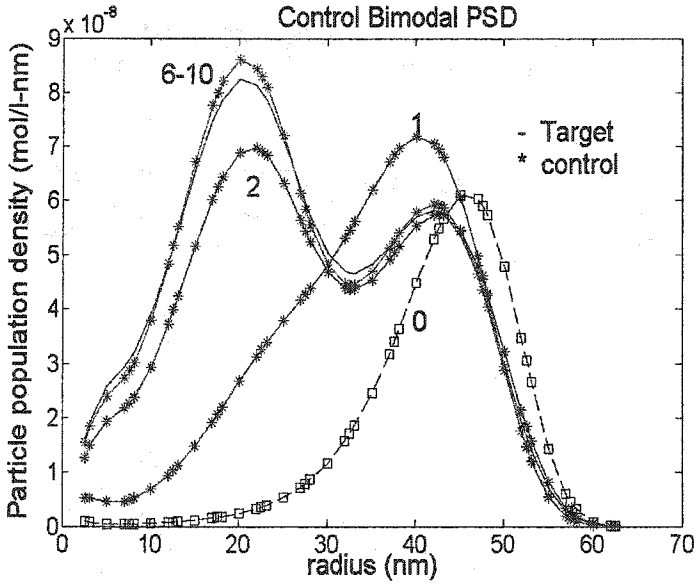


Figure 4.7 Adaptive PSD control with $x_{off,i}$=PSD. The number indicates the batch run (Batch 0 is no control (-□-)).

Although on-line (within-batch) control is always desirable, when on-line sensors are not available or the dynamics of the process are fast (for example the particle nucleation period in vinyl acetate emulsion polymerization lasts only about 2-3 min), then only batch-to-batch control is possible for the first particle generation. Therefore, in the next section a combination of batch-to-batch and inferential on-line control is proposed for this situation. Batch-to-batch information is used to control the first particle

generation, while on-line control is applied for the secondary distribution. Since the initial models are poor, batch-to-batch adaptation is also performed.

4.7 Case study II: Batch-to-batch and on-line inferential control with batch-to batch adaptation.

In Case study I, two emulsifier adjustments ($u_{c,1}$ and $u_{c,2}$) were simultaneously calculated at time θ_1=30 min, and implemented at θ_1=30 min and θ_2=150 min respectively to control the overall final PSD. In case study II, the adjustment to the shot of emulsifier at 150 min ($u_{c,2}$) will be used to control the second generation of particles. However, for the control of the first generation, the initial emulsifier concentration ($u_{c,0}$ at θ_0=0 min) will now be adjusted based on information from prior batches. This approach has the advantage that do not need fast analysis measurements in the PSD or Dp.

For this Case study, several alternatives for PLS models are possible. These alternatives differ from one another in the regressors used in the models. In all alternatives two PLS models are developed, one to control the first particle generation and one for the second. For both models, the PSD space was linearized using a square root transformation.

The first model to be applied at θ_0=0 min to predict the final PSD related to the first generation of particles ($\mathbf{y}_{\mathrm{I}}$) can be based on the initial charge of emulsifier ($u_{c,0}$, the deviation from the nominal recipe) and on information from previous batches such as the first generation of the final PSD of the immediately previous batch ($\mathbf{y}_{\mathrm{I}}^{(k-1)}$):

$$\hat{\mathbf{y}}_{\mathrm{I}}^{T} = [u_{c,0} \; u_{c,0}^{2} \; \mathbf{y}_{\mathrm{I}}^{(k-1)T}]\hat{\gamma} \tag{4.10}$$

However, if there exists long time delay in measuring the final PSD measurement from the previous batch ($\mathbf{y}^{(k-1)}$), eq. (4.10) cannot be used, but can be modified to include

on-line ($\mathbf{x}_{on}^{(k-1)}$) and off-line ($\mathbf{x}_{off}^{(k-1)}$) measurements and control action $u_{c,0}^{(k-1)}$ from the previous batch (a quadratic term was also included to account for slight nonlinearities):

$$\hat{\mathbf{y}}_{\mathrm{I}}^{T} = [u_{c,0}\ u_{c,0}^{(k-1)}\ \mathbf{x}_{on}^{(k-1)T}\ \mathbf{x}_{off}^{(k-1)T}\ u_{c,0}^{2}\ (u_{c,0}^{(k-1)})^{2}]\hat{\boldsymbol{\varphi}} \tag{4.11}$$

Both of these equations should contain information on disturbances occurring in the preceding batch. Before a new batch begins, an adjustment to the nominal initial emulsifier concentration ($u_{c,0}$) is calculated by optimising a LQR or MV objective function such as:

$$\underbrace{\min}_{u_{c,0}} (\mathbf{y}_{\mathrm{I},sp} - \hat{\mathbf{y}}_{\mathrm{I}}(u_{c,0}, u_{c,0}^{(k-1)}, \mathbf{x}_{on}^{(k-1)}, \mathbf{x}_{off}^{(k-1)}))^{T} \mathbf{Q}_{1} (\mathbf{y}_{\mathrm{I},sp} - \hat{\mathbf{y}}_{\mathrm{I}}(u_{c,0}, u_{c,0}^{(k-1)}, \mathbf{x}_{on}^{(k-1)}, \mathbf{x}_{off}^{(k-1)})) \tag{4.12}$$

A second PLS model is built to relate the emulsifier shot at 150 min ($u_{c,2}$), the on-line and off-line measurements taken during the current batch (k) and any initial emulsifier adjustment ($u_{c,0}$) made in the current batch, with the second generation of the final PSD ($\mathbf{y}_{\mathrm{II}}$):

$$\hat{\mathbf{y}}_{\mathrm{II}}^{T} = [u_{c,0}\ u_{c,2}\ \mathbf{x}_{on}^{T}\ \mathbf{x}_{off}^{T}\ u_{c,0}^{2}]\hat{\boldsymbol{\beta}} \tag{4.13}$$

The adjustment of the second emulsifier shot ($u_{c,2}$) is computed by using the MV objective function:

$$\underbrace{\min}_{u_{c,2}} (\mathbf{y}_{\mathrm{II},sp} - \hat{\mathbf{y}}_{\mathrm{II}}(u_{c,0}, u_{c,2}, \mathbf{x}_{on}, \mathbf{x}_{off}))^{T} \mathbf{Q}_{1} (\mathbf{y}_{\mathrm{II},sp} - \hat{\mathbf{y}}_{\mathrm{II}}(u_{c,0}, u_{c,2}, \mathbf{x}_{on}, \mathbf{x}_{off})) \tag{4.14}$$

Equations (4.12) and (4.14) can be used in conjunction with the minimum variance detuning factor (δ) shown in eq. (4.5). Alternatively, a LQ control objective as in equation (4.3) could be used.

Both of these models are updated from batch-to-batch as in equation (4.6), and control actions are calculated only if the predicted PSDs fall outside their no-control

region in the PCA score space. Clearly any control action (emulsifier adjustment) taken at θ_0=0 min is based only on batch-to-batch information, using deviations in the first PSD or process variable trajectories from the previous batch. At the second decision time (θ_2=150 min), only information on the current batch, including the first control action $u_{c,0}$, the on-line trajectory data ($\mathbf{x}_{on}$) and off-line PSD measurements ($\mathbf{x}_{off}$), is sufficient to summarize the effect of disturbances coming from both prior batches and from the current batch, and so control based only on within-batch information is performed.

In the following examples, it is assumed that an error free off-line PSD measurement is taken at 40 min ($\mathbf{x}_{off}$) and on-line Tj measurements ($\mathbf{x}_{on}$) (every 10 min up to 150 min with normal distributed random error σ=0.1%, approximately ±323 ×.001 ≈0.3°K) are available. (A study on the effect of noise in PSD measurement is addressed at the end of this section). To test the sensitivity of the models, 20 slightly different data sets were generated and used as training sets to obtain different nominal PLS models (eq. 4.11 and 4.13). Each training data set consists of 22 batches: 15 subject only to normal random variations in a_s and k_{daq}, and only 7 in which some mid-course corrections or initial condition change was performed as well.

4.7.1 Control for constant batch-wise disturbances

To illustrate the performance of the adaptive control scheme (eq. 4.6, 4.12 and 4.14), together with the effect of the minimum variance de-tuning factor (eq. 4.5) some examples are shown. (In the examples that follow the minimum variance tuning factor is applied only to the control action arising from equation 4.14, $u_{c,2}$). However, if needed, it can also be applied to the initial emulsifier concentration adjustment or constraints in the manipulated variables can be imposed. In this example, the system is affected by a constant batch-wise disturbance in a_s (-28%) and k_{daq} (+20%). The averaged integral absolute error (IAE) computed for all the different training data sets is shown in Fig. 4.8a for two values of the de-tuning parameter (δ=1 and δ=0.85) as a function of batch number.

The control results on the final PSD is shown in Fig. 4.8b for one typical sequence of batch control runs. In these figures, (x) represents the control performance achieved when minimum variance control (MVC) action is taken (δ=1); (o) when the control action is detuned (δ=0.85) and (-□-) what would happen if no control action is taken (indicated as batch zero). The average magnitude of the control actions (detuned case, δ=0.85) for the 20 datasets is shown in Figure 4.9.

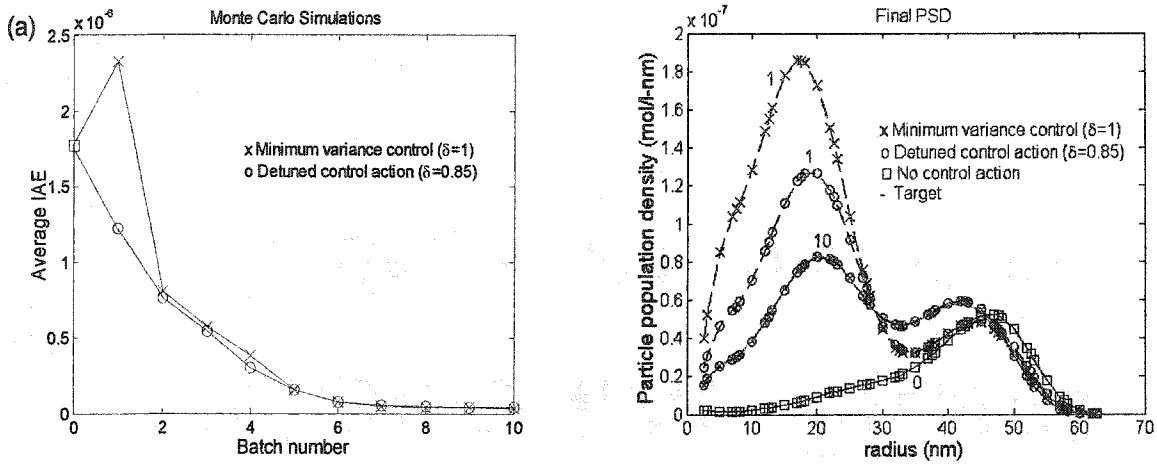


Figure 4.8 Adaptive PSD control for a batch-wise constant disturbance in a_s=0.72a^*_s, k_{daq}=1.2 k^*_{daq}. (a) IAE and (b) PSD progress versus batch number.

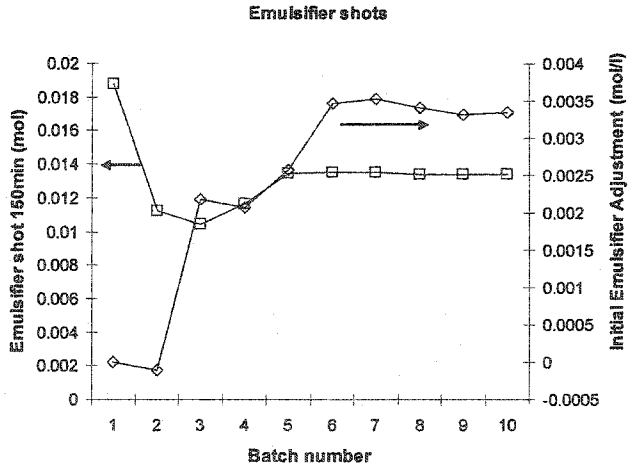


Figure 4.9 Manipulated variable adjustments (u_{co}, u_{c2}) versus batch number.

4.7.2 Control for PSD set-point change

So far the control performance has been illustrated for the case when the system is affected by model error and disturbances around one target PSD (i.e. regulatory control). Now we evaluate the performance of the adaptive schemes (eq. 4.6, 4.12 and 4.14) when it is desired to achieve a completely new bimodal PSD using a model built from operating data obtained around a very different initial PSD target. Figure 4.10 illustrates the adaptive scheme for an extreme case. In this Figure the new target PSD is shown together with the PSDs obtained from applying the minimum variance adaptive algorithm (for batches 1,2,3 and 10). Also is shown the original target PSD (--) around which the initial (nominal) model was developed. As can be seen in this Figure, the adaptive MVC scheme (δ=0.6) is able to achieve the desired target within a few batches.

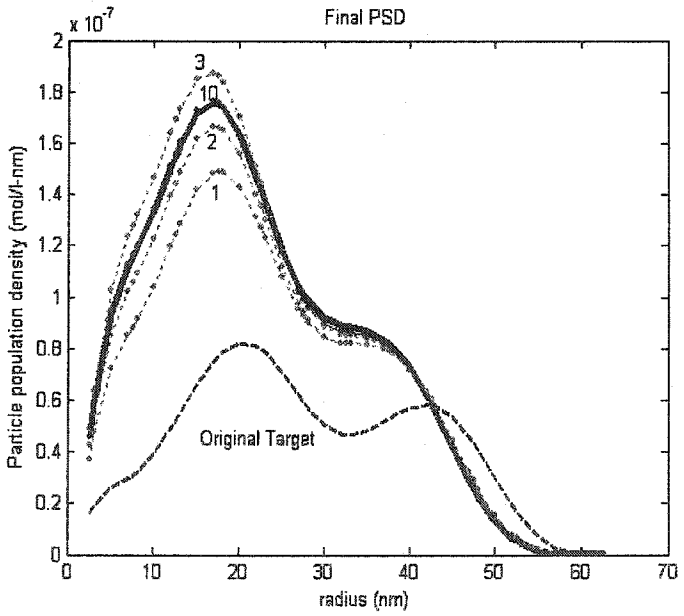
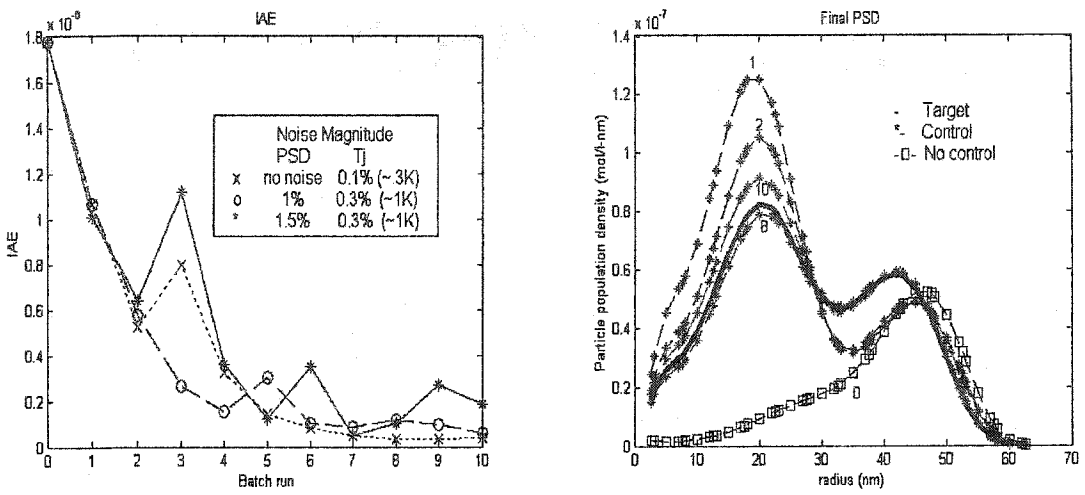


Figure 4.10 Performance of adaptive algorithm for a new PSD target. (--) Original target, (•-) achieved PSD and (–) desired target. The number indicates the batch run.

4.7.3 PSD measurement noise

In practice, the PSD measurement is also affected by measurement noise. This noise has a correlated structure that depends on the instrument providing the measurements. Therefore, noise structure identification was performed through PCA as described in Clarke-Pringle and MacGregor [1998], on repeated laboratory PSD measurements on one styrene emulsion latex sample. From the identified structure, new correlated noise was generated and added to the intermediate PSD sampled at 40 min (Appendix A). The IAE obtained from different magnitudes of noise in the PSD (structural noise) and Tj (random noise) are shown in Fig. 4.11a for one data set of the Case study II (batch-wise constant disturbance in a_s and k_{daq}, and δ=0.85). In Figure 11b the resulting PSDs are shown for the case in which σ=1.5% for the PSD noise and σ=0.3% for Tj (approximately $\pm 323 \times .003 \approx 1^{\circ}K$). It is clear that the performance trends are similar in all cases, but, as the measurement noises increase, the IAE from batch-to-batch is more variable and settles around a higher value, and the PSD continues to bounce around the target. The use of a no-control region that reflects the impact of these measurement noises would prevent continued control actions from being implemented based solely on noise once the PSD has attained the final no-control region.



(a) (b)

Figure 4.11 Effect of noise on the performance of the adaptive PSD control. (a) IAE with different levels of noise. Batch zero is no control. (b) Control results for the final PSD when the noise in the grab sample PSD, x_{off}, is σ=1.5%, and Tj σ=0.3% (±1°K using as base T_j=323°K). The number indicates the batch run.

4.8 Conclusions

An inferential-adaptive methodology for the control of multivariate quality properties in semi-batch processes is presented. The methodology can utilize information both from previous batches and from the current batch to make adjustments to the nominal manipulated variable values at several time intervals throughout the batch. PLS latent variable models are able to easily incorporate the highly correlated process measurements into the model, and to achieve adequate prediction and control of the high dimensional product quality space. The data requirements for such a strategy are modest and the models are easily built, making the approach suitable for industrial processes. The adaptive algorithm was tested for the control of the full particle size distribution of the final product in an emulsion polymerization process subject to batch-wise constant and random disturbances, as well as for tracking changes in the PSD set-point. The disturbances were shown to be easily rejected, and optimal process variable adjustments necessary to achieve completely new PSD set-points were obtained.

Nomenclature

A = number of principal components

$\mathbf{E}$ = residual matrix from PLS

$\mathbf{E}'$ = residual matrix from PCA

$\mathbf{F}$ = residual matrix from PLS

m = number of data-sets in Monte Carlo simulations

$\mathbf{Q}_1$ = weighting matrix in the controlled variables

$\mathbf{Q}_2$ = suppression movement matrix

$\mathbf{Q}$ = loading matrix for $\mathbf{Y}$ from PLS

$\mathbf{t}$ = score vector from PCA

$\mathbf{u}$ = vector of implemented manipulated variables

$\mathbf{u}_c$ = vector of manipulated variables

$\mathbf{u}_{c\text{-prev}}$ = vector of past manipulated variables

$\mathbf{u}_{c\text{-future}}$ = vector of future unknown manipulated variables

$\mathbf{V}$= loading matrix from PLS

$\mathbf{X}$ = regressor matrix

$\mathbf{x}$ = regressor vector that includes on-line and off-line measurements, and control actions

$\mathbf{x}_{off}$ = vector of off-line measurements

$\mathbf{x}_{on}$ = vector of on-line trajectory measurements

$\mathbf{x}_{prior}$ = vector containing previous batch information

$\mathbf{Y}$ = quality matrix

$\mathbf{y}$ = quality variables

$\hat{\mathbf{y}}$ = estimated quality variables

Greek symbols

$\hat{\gamma}$ = matrix of regression coefficients

$\hat{\varphi}$ = matrix of regression coefficients

θ = decision times

δ = de-tuning factor

Γ = score matrix from PLS

$\hat{\beta}$ = matrix of regression coefficients

Index

a = latent variable index

l = time index

k = batch index

superscript

^ = indicates that the variable is estimated

* = indicates nominal conditions

subscript

sp = set-points

Chapter 5

Trajectory Control in Batch Reactors using Latent Variable Models

In this Chapter, a novel inferential strategy for controlling end-product quality properties by adjusting the complete trajectories of the manipulated variables is presented. Control through complete trajectory manipulation using empirical models is possible by controlling the process in the reduce space (scores) of a latent variable model rather than in the real space of the manipulated variables. Model inversion and trajectory reconstruction is achieved by exploiting the correlation structure in the manipulated variable trajectories captured by a Partial Least Squares (PLS) model. The approach is illustrated with a condensation polymerization example for the production of nylon and with data gathered from an industrial emulsion polymerization process. The data requirements for building the model are shown to be modest.

5.1 Introduction

Control of product quality usually requires the on-line adjustment of several manipulated variable trajectories (MVTs) such as the pressure and temperature trajectories. Several approaches based on detailed theoretical models have been presented before (see Chapter 2 for a review). However, many of these strategies are difficult to implement because they are computationally intensive and/or require substantial model knowledge.

Empirical modelling, on the other hand, has the advantage of ease in model building. Yabuki and MacGregor [1997] and the approaches presented in Chapter 3 and 4 used empirical models for the control of product quality properties, but in these approaches the control action has been restricted to only a few movements in the manipulated variables (injection of additional reactants) because in these cases these few adjustments were enough to reject the disturbances and to achieve the desired end-qualities. However, if the operation calls for adjustments to MVTs through most of the duration of the process, another approach needs to be taken. The approach often used in these cases is to segment the MVTs into a small number of intervals (e.g. 5-10) and force the behaviour of the MVTs over the duration of each interval to follow a zero or first order hold. Control is then accomplished by manipulating the slope or the level (stair-case parameterisation) at the start of each interval (decision points). Studies involving this type of parameterisation can be found in Chin et al. [2000] and Russell et al. [1998a] among others. However, in many batch processes such a stair-case parameterization of the MVTs, just for convenience of the control engineers, may not be acceptable. The operation of the batch may require, or historically be based on, smooth MVTs, and converting them to stair-case approximations might represent a radical departure from normal practice, with the implication that control schemes based on them will never be implemented. Moreover, model inversion in the control algorithm would be usually difficult with this approach (stair case parameterization) because a large number of highly correlated control actions need to be determined at every decision point.

A solution to this problem comes from recognizing that within the range of normal process operation all the process variable trajectories (both MVTs and measured variables) are very highly correlated with one another, both contemporaneously (i.e. at the same time period) and temporally (over the time history of the batch). This implies that their behaviour can be represented in a much lower dimensional space using latent variable models based on PCA or PLS. In this Chapter is shown that by projecting all the process variable trajectory data into low dimensional latent variable spaces, all control decisions can be performed on the latent variables, and the entire MVTs for the

remainder of the batch then reconstructed from the latent variable models. In this reduced dimensional space, the data requirements for modelling and for model parameter estimation are much less demanding, the control computation is easier, and the computed MVTs are smooth and consistent with past operation of the process.

In spite of these inherent advantages in controlling the MVTs of batch processes in a latent variable space, no literature has yet addressed this issue. Statistical controllers for continuous processes (a binary distillation column simulator and the Tennessee Eastman process) based on Principal Component Analysis (PCA) have been proposed (Chen and McAvoy [1996], Chen et al. [1998], McAvoy [2002]) which express the control objective in the score space of a PCA model, but the dimension of the manipulated variable space is still small since no trajectories need to be computed.

The purpose of this Chapter is to introduce an inferential control strategy that allows a much finer characterization and smoother reconstruction of optimal manipulated variable trajectories than those obtained using staircase parameterisation, without increasing the complexity and number of identification experiments needed for model building. The outline of the Chapter is as follows: in section 5.2 the methodology is introduced; in section 5.3, the control approach is illustrated with a condensation polymerisation case study for the production of nylon and preliminary results are shown for an industrial emulsion polymerization process.

5.2 Control methodology

5.2.1 Model building

The proposed methodology uses historical data bases and a few complementary identification experiments for model building. The empirical model is obtained using Partial Least Squares (PLS). However, other projection methods such as principal component regression may also be applied.

The database from which the PLS model is identified is shown in Figure 5.1. It consists of a ($K \times M$) response matrix $\mathbf{Y}$ and an originally three-dimensional array $\underline{\mathbf{X}}$, which after unfolding (Nomikos and MacGregor [1995a], Kourti et al. [1995]) would yield a ($K \times N$) regressor matrix $\mathbf{X}$ where K is the number of batches. Each row vector of $\mathbf{Y}$, denoted as $\mathbf{y}^T$, contains M quality properties measured at the end of each batch. Each row vector of $\mathbf{X}$, denoted as $\mathbf{x}^T$, is composed of:

$$\mathbf{x}^T = [\mathbf{x}_{on}^T \; \mathbf{x}_{off}^T \; \mathbf{u}_c^T]$$

where $\mathbf{x}_{\mathbf{on}}^T = [\mathbf{x}_{\mathbf{on},1}^T \; \mathbf{x}_{\mathbf{on},2}^T \cdots \mathbf{x}_{\mathbf{on},l}^T]$ is a vector of the trajectories of l on-line process variables such as temperature and pressure obtained from on-line sensors; $\mathbf{x}_{\mathbf{off}}^T = [\mathbf{x}_{\mathbf{off},1}^T \; \mathbf{x}_{\mathbf{off},2}^T \cdots \mathbf{x}_{\mathbf{off},r}^T]$ is the set of any off-line measurements collected occasionally on r variables during the batch, and $\mathbf{u}_{\mathbf{c}}^{\mathbf{T}} = [\mathbf{u}_{\mathbf{c},1}^{\mathbf{T}} \; \mathbf{u}_{\mathbf{c},2}^{\mathbf{T}} \cdots \mathbf{u}_{\mathbf{c},n}^{\mathbf{T}}]$ is a vector of the trajectories of n manipulated variables. As can be seen in Figure 5.1, $\mathbf{x}_{\mathbf{on},j}^T = [x_{on,1}, \cdots, x_{on,f}]_j$ and $\mathbf{x}_{\mathbf{off},s}^T = [x_{off,1}, \cdots, x_{off,g}]_s$ denotes, respectively, the row vector of observations obtained from on-line measurements on the j-th variable, and from off-line measurements on the s-th variable over the course of the batch, while $\mathbf{u}_{\mathbf{c},m}^{\mathbf{T}} = [u_{c,1}, \cdots u_{c,w}]_m$ denotes the trajectory of the m-th MV. Here, f, g and w are, respectively, the number of on-line measurements, off-line analysis and MV segments for the corresponding variable in each category. In the following text $\mathbf{x}_{\mathbf{on}}^T$ and $\mathbf{x}_{\mathbf{off}}^T$ are combined into a single row vector $\mathbf{x}_m^T = [\mathbf{x}_{on}^T \; \mathbf{x}_{off}^T]$, and then $\mathbf{x}^T = [\mathbf{x}_{\mathbf{m}}^T \; \mathbf{u}_{\mathbf{c}}^T]$.

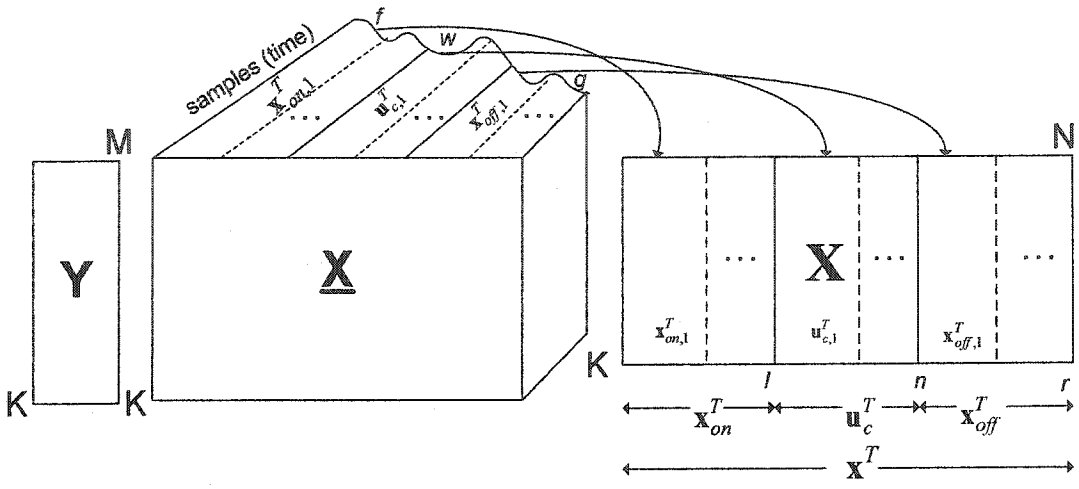


Figure 5.1 Unfolding of database for model building.

Full MVTs are obtained through trajectory segmentation as illustrated in Figure 5.2. The MVTs are segmented into a (possibly) large number of intervals (w) and control decision points (θ_i, i=1,2,...) are selected. At each decision point (θ_i), final properties ($\mathbf{y}$) are predicted and the adjustments to the remaining MVTs (after this decision point) are computed if the predicted final properties ($\hat{\mathbf{y}}$) are not within desired specifications. Notice that the segment size is not necessarily uniform and that decisions points may be chosen arbitrarily but are assumed to be the same for each batch. (The decision points will usually be selected using prior process knowledge.) In the limit, control action can be taken at every segment (i.e. every segment would represent a decision point), but this is almost never necessary, as a small number is usually adequate.

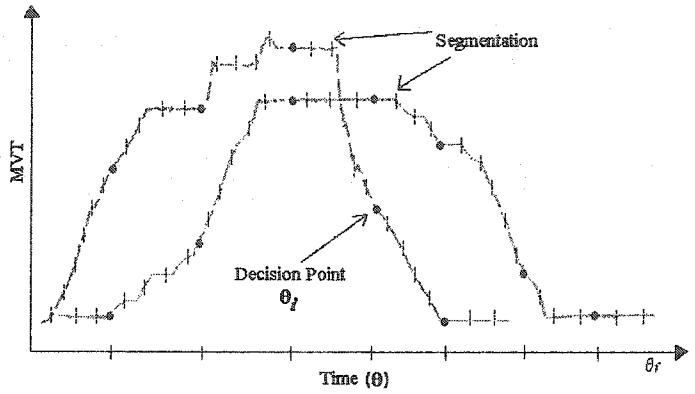


Figure 5.2 Fine segmentation of MVTs and decision points.

The data-set used for model building consists of normal operating data as well as data in which some changes in the manipulated variable trajectories (at each decision point) has been performed. Linear PLS regression is then performed by projecting the scaled (unit variance) data (expressed as deviations from their nominal conditions) onto lower dimensional subspaces:

$$\begin{aligned} \mathbf{X} &= \mathbf{T}\mathbf{P}^T + \mathbf{E} \\ \mathbf{Y} &= \mathbf{T}\mathbf{Q}^T + \mathbf{F} \end{aligned} \tag{5.1}$$

where the columns of $\mathbf{T}$ are values of new latent variables ($\mathbf{T} = \mathbf{X}\mathbf{W}$) that capture most of the variability in the data, $\mathbf{P}$ and $\mathbf{Q}$ are the loading matrices for $\mathbf{X}$ and $\mathbf{Y}$ respectively, and $\mathbf{E}$ and $\mathbf{F}$ are residual matrices. Non-linear PLS regression can also be used as will be shown at the end of section 5.3.1 and in Appendix B. However, for simplicity, in the following discussion linear models are assumed.

The control methodology used in this work consists of two stages: at predetermined decision times (θ_i, i=1,2,…) an inferential end-quality prediction using on-line and possible off-line process measurements ($\mathbf{x}_m$) and MVTs ($\mathbf{u}_c$) available up to that time is performed to determine whether or not the controlled end-qualities ($\mathbf{y}$) fall outside a pre-determined "no-control" region, and then if needed, control action is computed in the latent variable space followed by model inversion to obtain the modified MVTs for

the remainder of the batch that will yield the desired final qualities. This two-stage procedure is repeated at every decision point (θ_i) using all available measurements on the process variable and manipulated variable trajectories available up to that time. The novelty of the proposed approach is that the control and the model inversion stage is performed in the reduced dimensional space (latent variable or score space) of a PLS model rather than in the real space of the MVTs. Due to the high correlation of measurements and control actions, the true dimensionality of the process, determined by the score variable space (t_a, a=1,2,...,A) of the PLS model, is generally much smaller than the number of manipulated variable points obtained from the MVT segmentation ($\mathbf{u_c}$). Therefore, the control computation performed in the reduced latent variable space ($\mathbf{t}$) is much simpler than the one performed in the real space. In the following, the control methodology is described for one control decision point (θ_i) during the batch. This is simply repeated at each future decision point. Notice that although the method is illustrated with an example in which the decision points are defined at fixed clock times (θ_i, i=1,2,...), these decision points could easily be based on measured variables other than time, such as specified values of conversion or energy production. This would be an advantage on batches that do not have the same duration (due to, for example, seasonal variations in cooling capacity and varying row material properties), since the process trajectories can then be aligned using such indicator variables (Kassidas et al. [1998], Nomikos and MacGregor [1995b], Kourti et al. [1996], Neogi and Schlags [1998]).

5.2.2 Prediction

For on-line end-quality estimation ($\hat{\mathbf{y}}$), when a new batch k is being processed, at every decision point (θ_i, i=1,2,...) $0 \leq \theta_i \leq \theta_f$, there exists a regressor row vector $\mathbf{x}^T$ composed of at least the following variables:

$$\mathbf{x}^T = [\mathbf{x}_m^T \quad \mathbf{u}_c^T] = [\mathbf{x}_{m,measured}^T{}_{,\theta_i} \quad \mathbf{x}_{m,future}^T \quad \mathbf{u}_{c,implemented}^T{}_{,\theta_i} \quad \mathbf{u}_{c,future}^T] \tag{5.2}$$

The regressor vector $\mathbf{x}$ consists of: all measured variables ($\mathbf{x}_{m,measured}$) available up to time θ_i ($0 \leq \theta < \theta_i$); unmeasured variables ($\mathbf{x}_{m,future}$) not available at θ_i, but that will be available in the future ($\theta_i \leq \theta \leq \theta_f$); implemented control actions $\mathbf{u}_{c,implemented}$ ($0 \leq \theta < \theta_i$); and future control actions $\mathbf{u}_{c,future}$, ($\theta_i \leq \theta \leq \theta_f$) which will be determined through the control algorithm. Note that at the model building stage, the $\mathbf{x}_{m,future}$ and $\mathbf{u}_{c,future}$ vectors are available for each batch.

To estimate whether or not the final quality properties for a new batch will lie within an acceptable region, the prediction is performed considering $\mathbf{u}_{c,future} = \mathbf{u}_{c,nominal}$ (i.e. assuming that the remaining MV trajectories will be kept at their nominal conditions) using the PLS model:

$$\hat{\mathbf{t}}^T_{present} = [\mathbf{x}^T_m \quad \mathbf{u}^T_c]\mathbf{W} = [\mathbf{x}^T_{m,measured,\theta_i} \quad \mathbf{x}^T_{m,future} \quad \mathbf{u}^T_{c,implemented,\theta_i} \quad \mathbf{u}^T_{c,nominal}]\mathbf{W} \tag{5.3}$$

$$\hat{\mathbf{y}}^T = \hat{\mathbf{t}}^T_{present}\mathbf{Q}^T \tag{5.4}$$

$\mathbf{W}$ and $\mathbf{Q}$ are projection matrices obtained from the PLS model building stage. The vector of scores, $\hat{\mathbf{t}}_{present}$, for the new batch is the projection of the $\mathbf{x}$ vector onto the reduced dimension space of the latent variable model at time θ_i, and $\hat{\mathbf{y}}$ is the vector of predicted end-quality properties. From the above equations, it can be noticed that changes in batch operation detected by measurements of the process variable trajectories ($\mathbf{x}_{m,measured,\theta_i}$) or produced by changes in the MVTs ($\mathbf{u}_{c,implemented,\theta_i}$) would produce changes in the scores ($\hat{\mathbf{t}}_{present}$) and therefore in the end-quality properties (i.e. changes in the end-qualities can be detected through changes in the scores). From equation (5.3), it can also be noticed that in order to compute $\hat{\mathbf{t}}_{present}$ and $\hat{\mathbf{y}}$, it is necessary to have an estimate of the unknown future measurements ($\mathbf{x}_{m,future}$) from ($\theta_i \leq \theta \leq \theta_f$). These can be imputed

from the PLS model using efficient missing data algorithms available in the literature (Nelson et al. [1996], Arteaga and Ferrer [2002]). Missing data imputation based on, for example, conditional expectation or expectation/maximization (EM) are statistically efficient estimators for the remaining portions of the batch trajectories, based on the PLS model developed from data on completed batches (García-Muñoz and MacGregor [2003]). Alternatively, a multi-model approach in which a different model is identified at every decision point can be used (Russell et al. [1998a]). The decision of one alternative over other depends on the number of decision points and/or performance of the missing data algorithm. In the example shown in this paper a *single* PLS model is used for prediction and control, and the estimation of unknown future measurements is performed using the PLS model and a missing data algorithm.

The "no-control region" can be determined in several ways, such as one that takes into account the uncertainty of the model for prediction (Yabuki and MacGregor [1997]), the product specifications, or quality data under normal ("in-control") operating conditions (Chapter 4). In this Chapter a simple control region based on product quality specifications will be used. The issue of whether or not to use a "no-control" region is at the discretion of the user, and is not essential to the control methodology presented here.

If the quality prediction is outside the "no-control" region, then a control action, and model inversion to obtain the MVTs for the remainder of the batch $\mathbf{u}^T_{c,\,future}$ is needed. Obtaining the full MVTs consist of two stages: 1) computation of the adjustments required in the latent variable scores $\Delta\mathbf{t}$, followed by 2) model inversion of the PLS model to obtain the real MVTs for the remainder of the batch. These two stages are explained in the following sections.

5.2.3 Score adjustment computation

At every decision point (θ_i), the change in the scores ($\Delta\mathbf{t}$) needed to track the end-qualities closer to their set-points ($\mathbf{y}_{sp}$) can be obtained by solving (5.5):

$$\begin{aligned} &\underbrace{\min}_{\Delta \mathbf{t}(\theta_i)} (\hat{\mathbf{y}} - \mathbf{y}_{\mathbf{sp}})^T \mathbf{Q}_1 (\hat{\mathbf{y}} - \mathbf{y}_{\mathbf{sp}}) + \Delta \mathbf{t}^T \mathbf{Q}_2 \Delta \mathbf{t} + \lambda T^2 \\ &s.t. \quad \hat{\mathbf{y}}^T = (\Delta \mathbf{t} + \hat{\mathbf{t}}_{\mathbf{present}})^T \mathbf{Q}^T \\ &T^2 = \sum_{a=1}^{A} \frac{(\Delta t + \hat{t}_{present})_a^2}{s_a^2} \\ &\Delta \mathbf{t}_{\mathbf{min}} \le \Delta \mathbf{t} \le \Delta \mathbf{t}_{\mathbf{max}} \end{aligned} \tag{5.5}$$

where $\Delta \mathbf{t}^T = \mathbf{t}^T - \hat{\mathbf{t}}^T_{\mathbf{present}}$, $\mathbf{Q_1}$ is a diagonal weighting matrix, $\mathbf{Q_2}$ is a movement suppression matrix, T^2 is the Hotelling's statistic, s_a^2 is the variance of the score t_a, and λ is a weighting factor. Hard constrains in the adjustment to the scores ($\Delta \mathbf{t}_{\mathbf{min}} \le \Delta \mathbf{t} \le \Delta \mathbf{t}_{\mathbf{max}}$) are problem dependent and may or not need to be included. Soft constraints on $\Delta \mathbf{t}$ are contained in the quadratic objective function. The soft constraint on the score magnitudes through, Hotelling's T^2 statistic, is intended to constrain the solution in the region where the model is valid.

Equation (5.5) is a quadratic programming problem that can be restated as:

$$\underbrace{\min}_{\Delta \mathbf{t}(\theta_i)} \frac{1}{2} \Delta \mathbf{t}^T \mathbf{H} \Delta \mathbf{t} + \mathbf{f}^T \Delta \mathbf{t} \tag{5.6}$$

where

$$\begin{aligned} &\mathbf{H} = \mathbf{Q}^T \mathbf{Q}_1 \mathbf{Q} + \mathbf{Q}_2 + \mathbf{Q}_3 \\ &\mathbf{f}^T = (\mathbf{Q}\hat{\mathbf{t}}_{\mathbf{present}} - \mathbf{y}_{\mathbf{sp}})^T \mathbf{Q}_1 \mathbf{Q} + \hat{\mathbf{t}}^T_{\mathbf{present}} \mathbf{Q}_3 \\ &\mathbf{Q}_3 = \mathrm{diag}\left[\lambda / s_a^2 \right] \\ &\Delta \mathbf{t}_{\mathbf{min}} \le \Delta \mathbf{t} \le \Delta \mathbf{t}_{\mathbf{max}} \end{aligned} \tag{5.7}$$

In the case of no hard constraints, the solution is easily obtained as:

$$\Delta \mathbf{t}^{\mathbf{T}} = -\mathbf{f}^{\mathbf{T}} \mathbf{H}^{-1} \tag{5.8}$$

The aim of eq. (5.8) is to obtain the change in the scores ($\Delta \mathbf{t}$) that would drive the final quality variables closer to their desired set-points ($\mathbf{y}_{sp}$). Due to the movement suppression matrix ($\mathbf{Q}_2$) and/or λ, the computed ($\Delta \mathbf{t}$) may not drive the process all the way to their set-points.

Choosing $\mathbf{Q}_2=\mathbf{0}$ and $\lambda=0$, gives the minimum variance controller, which, at each decision point would force the predicted qualities ($\hat{\mathbf{y}}$) to be equal to their set-points ($\hat{\mathbf{y}} = \mathbf{y}_{sp}$) at the end of the batch:

$$\begin{aligned} &\underbrace{\min}_{\Delta \mathbf{t}(\theta_i)} (\hat{\mathbf{y}} - \mathbf{y}_{sp})^T \mathbf{Q}_1 (\hat{\mathbf{y}} - \mathbf{y}_{sp}) \\ &st \quad \hat{\mathbf{y}}^T = (\Delta \mathbf{t} + \hat{\mathbf{t}}_{present})^T \mathbf{Q}^T \end{aligned} \tag{5.9}$$

Three situations arise (for the unconstrained case) in finding a solution to (5.9) depending on the statistical dimensions of $\mathbf{y}_{sp}$ and $\Delta \mathbf{t}$ ($\mathbf{Q}_2=\mathbf{I}$):

1. dim ($\Delta \mathbf{t}$) = dim ($\mathbf{y}_{sp}$)

In this situation a unique solution exists that can be directly obtained from (5.9):

$$\Delta \mathbf{t}^T = \mathbf{y}_{sp}^T (\mathbf{Q}^T)^{-1} - \mathbf{t}_{present}^T \tag{5.10}$$

2. dim ($\Delta \mathbf{t}$) < dim ($\mathbf{y}_{sp}$)

In this case a least square solution is needed:

$$\Delta \mathbf{t}^T = (\mathbf{y}_{sp}^T - \mathbf{t}_{present}^T \mathbf{Q}^T) \mathbf{Q} (\mathbf{Q}^T \mathbf{Q})^{-1} \tag{5.11}$$

3. dim ($\Delta \mathbf{t}$) > dim ($\mathbf{y}_{sp}$)

This case is a common situation. Although the number of variables to be used in the control algorithm has been reduced to A latent variables, a projection from a lower to

higher space is still required. In this situation eq. (5.9) has an infinite number of solutions. Therefore, a natural choice is to select the $\Delta t(\theta_i)$ having the minimum norm-2:

$$\begin{aligned} &\underbrace{\min}_{\Delta t(\theta_i)} \Delta t^T \Delta t \\ st \quad & y_{sp}{}^T = (\Delta t + \hat{t}_{present})^T Q^T \end{aligned} \tag{5.12}$$

and whose solution can be easily obtained as:

$$\Delta t^T = (y_{sp}{}^T - \hat{t}^T_{present} Q^T)(QQ^T)^{-1} Q \tag{5.13}$$

A detuning factor ($0 \leq \delta \leq 1$) may be included for this reduced space controller:

$$\Delta t^T = \delta(y_{sp}^T - \hat{t}^T_{present} Q^T)(QQ^T)^{-1} Q \tag{5.14}$$

A Δt vector is computed at every decision point (θ_i).

Equations 5.10, 5.11 and 5.13 are consistent with the PLS model inversion results found by Jaeckle and MacGregor [1998].

Notice that in this last situation (eq. (5.14)), the matrix QQ^T has dimension $m \times m$ (m being the number of quality properties). Therefore, in order to avoid ill-conditioned matrix inversion, the quality properties should not be highly correlated. This poses no problem since one can always perform a PCA on the Y quality matrix to obtain a set of orthogonal variables (τ) that can be used as new controlled variables. Alternatively, if it is decided to retain an independent set of physical y variables, selective PCA (Jaeckle and MacGregor [1998]) can be performed on the Y matrix to determine that subset of quality variables which best defines the Y space.

5.2.4 Inversion of PLS model to obtain the MVTs

Once the low dimensional ($A \times 1$) vector Δt is computed via one of the control algorithms described in the last section, it remains to reconstruct from $\mathbf{t}^T = \Delta \mathbf{t}^T + \hat{\mathbf{t}}^T_{\text{present}}$, estimates for the high dimensional trajectories for the future process variables ($\mathbf{x}_{m,\text{future}}$) and for the future manipulated variables ($\mathbf{u}_{c,\text{future}}$) over the remainder of the batch. These future trajectories can be computed from the PLS model (5.1) in such a way that their covariance structure is consistent with past operation. If there were no restrictions on the trajectories, such as might be the case for a control action at θ=0, then the model for the X-space can be used directly to compute the $\mathbf{x}$ vector trajectory ($\mathbf{x}^T = [\mathbf{x}_m^T \ \mathbf{u}_c^T]$) for the entire batch (Jaeckle and MacGregor [1998]) as:

$$\mathbf{x}^T = \mathbf{t}^T \mathbf{P}^T \tag{5.15}$$

However for control intervals at times $\theta_i > 0$ the $\mathbf{x}$ vector trajectory ($\mathbf{x}^T = [\mathbf{x}^T_{m,\text{measured}(0:\theta_i)} \ \mathbf{u}^T_{c,\text{implemented}(0:\theta_i)} \ \mathbf{x}^T_{m,\text{future}(\theta_i:\theta_f)} \ \mathbf{u}^T_{c,\text{future}(\theta_i:\theta_f)}]$) is composed of measured process variables ($\mathbf{x}^T_{m,\text{measured}(0:\theta_i)}$) for the interval $0 \le \theta < \theta_i$, and for the already implemented manipulated variables ($\mathbf{u}^T_{c,\text{implemented}(0:\theta_i)}$) that must be respected when computing the trajectories for the remainder of the batch ($\theta_i \le \theta < \theta_f$).

Denote $\mathbf{x}_1^T = [\mathbf{x}^T_{m,\text{measured}(0:\theta_i)} \ \mathbf{u}^T_{c,\text{implemented}(0:\theta_i)}]$ the known trajectories over the time interval ($0:\theta_i$) that must be respected, $\mathbf{x}_2^T = [\mathbf{x}^T_{m,\text{future}(\theta_i:\theta_f)} \ \mathbf{u}^T_{c,\text{future}(\theta_i:\theta_f)}]$ the remaining trajectories to be computed, and $\mathbf{P}_1^T$ and $\mathbf{P}_2^T$ their corresponding loading matrices.

At times $\theta_i > 0$, if $\mathbf{x}$ is directly reconstructed using (5.15) as $\mathbf{x}^T = \mathbf{t}^T \mathbf{P}^T$ then

$$[\mathbf{x}_1^T \ \mathbf{x}_2^T] = [\mathbf{t}^T \mathbf{P}_1^T \ \mathbf{t}^T \mathbf{P}_2^T] \tag{5.16}$$

However, the computed $\mathbf{t}^T\mathbf{P}_1^T$ will generally not be equal to the actually observed trajectories at time θ_i $\mathbf{x}_1^T = [\mathbf{x}^T_{\mathbf{m,measured(0:\theta_i)}}\ \mathbf{u}^T_{\mathbf{c,implemented(0:\theta_i)}}]$. Therefore, simply selecting $\mathbf{x}_2^T = \mathbf{t}^T\mathbf{P}_2^T$ would not be correct as it does not account for what has actually been observed for $\mathbf{x}_1^T$ in the first part of batch.

Therefore, assume that the remaining trajectories (future manipulated variables and measurements) are:

$$\mathbf{x}_2^T = (\mathbf{t}^T + \boldsymbol{\alpha}^T)\mathbf{P}_2^T \tag{5.17}$$

where $\boldsymbol{\alpha}^T\mathbf{P}_2^T$ is an adjustment to $\mathbf{x}_2^T$ that accounts for the effects of discrepancy between $\mathbf{t}^T\mathbf{P}_1^T$ and $\mathbf{x}_1^T$ during the first part of the batch. (Selection of such a relationship will also ensure that the correlation structure of the PLS model is kept.) However, we still wish to achieve the computed value in score space $\mathbf{t}$ that will satisfy the overall PLS model. Therefore, we must have:

$$\mathbf{t}^T = [\mathbf{x}_1^T\ \mathbf{x}_2^T]\begin{bmatrix}\mathbf{W}_1\\ \mathbf{W}_2\end{bmatrix} = \mathbf{x}_1^T\mathbf{W}_1 + \mathbf{x}_2^T\mathbf{W}_2 \tag{5.18}$$

then
$$\mathbf{x}_2^T\mathbf{W}_2 = \mathbf{t}^T - \mathbf{x}_1^T\mathbf{W}_1 \tag{5.19}$$

Substituting $\mathbf{x}_2^T = (\mathbf{t}^T + \boldsymbol{\alpha}^T)\mathbf{P}_2^T$ in (5.19):

$$(\mathbf{t}^T + \boldsymbol{\alpha}^T)\mathbf{P}_2^T\mathbf{W}_2 = \mathbf{t}^T - \mathbf{x}_1^T\mathbf{W}_1$$

Therefore

$$(\mathbf{t}^T + \boldsymbol{\alpha}^T) = (\mathbf{t}^T - \mathbf{x}_1^T\mathbf{W}_1)(\mathbf{P}_2^T\mathbf{W}_2)^{-1} \tag{5.20}$$

And by substituting (5.20) in (5.17) the remaining MVTs to be implemented are obtained ($\theta_i \leq \theta < \theta_f$):

$$\mathbf{x}_2^T = (\mathbf{t}^T - \mathbf{x}_1^T \mathbf{W}_1)(\mathbf{P}_2^T \mathbf{W}_2)^{-1} \mathbf{P}_2^T \tag{5.21}$$

It is easy shown that this equation reduces to the relationship in (5.15) when $\theta_i = 0$ where there are no existing trajectory measurements or manipulated variables. The $(A \times A)$ matrix $\mathbf{P}_2^T \mathbf{W}_2$ is nearly always well conditioned, and so there is no problem with performing the inversion (Appendix B). This inferential control algorithm is then repeated at every decision point (θ_i) until completion of the batch.

5.3 Case studies

5.3.1 Case study 1: Condensation polymerization

In the batch condensation polymerisation of nylon 6,6 the end product properties are mainly affected by disturbances in the water content of the feed. In plant operation, feed water content disturbances occur because a single evaporator usually feeds several reactors (Russell et al. [1998a]). The non-linear mechanistic model of nylon 6-6 batch polymerization used in this work for data generation and model performance evaluation was developed by Russell et al. [1998b]. The complete description of the model and model parameters can be found in the original publication.

This system was studied by Russell et al. [1998a,b] where several control strategies including conventional control (PID and gain scheduled PID), non-linear model based control and empirical control based on linear state-space models were evaluated. In the data-based approach (Russell et al. [1998a]) control of the system was achieved by reactor and jacket pressure manipulation. These two manipulated variables were segmented and characterized by slope and level (stair-case parameterization) leading to 10 control variables. A total of 7 intervals (decision points) were used. An empirical state

space model was identified from 69 batches arising from an experimental design in the 10 manipulated variables. Several differences between the control strategy proposed here and the one used by Russell et al. [1998a] can be noticed, the most important being: (*i*) the control is computed in the reduce latent variable space rather than in the real space of the MVTs, (ii) only 2 decision points are needed to achieve good control; thereby simplifying the implementation and decreasing the number of identification experiments needed to build a model, and (*iii*) a much finer MVT reconstruction is achieved.

Control objectives and trajectory segmentation.

The control objective is to maintain the end-amine concentration (NH_2) and the number average molecular weight (MWN) at their set-points to produce nylon 6,6 when the system is affected by changes in the initial water content (W). The MVTs used to control the end-qualities are the jacket and reactor pressure trajectories. These manipulated variable trajectories are finely segmented every 5 min. starting at 35 min. from the beginning of the reaction until 30 min. before the completion of the batch (total reaction time 200 min), giving trajectories defined at 40 discrete time points in the interval ($35 \leq \theta \leq 170$). The trajectories for the first 35min and the last 30min were fixed for all batches. Two control decision points at 35 and 75 min. were found to be sufficient for good control for the conditions used in this example. In order to predict NH_2 and MWN, on-line measurements of the reactor temperature (Tr) and venting (v) are considered available every two minutes.

Data generation

A PLS model with 5 latent variables (determined by cross-validation) was built from a data set consisting of 45 batches in which the initial water content (W) was randomly varied. In 30 of the batches some movement in the MVT (at the two decision points) was performed (some of these batches would normally be available from historical data). The effect that the number batches used for identification of the PLS

model has on control performance is discussed at the end of this section and in Appendix B.

Prediction

The first step is to evaluate the performance of the PLS model prediction with different missing data algorithms at each decision point. Several missing data algorithms were tested and as an illustration some results are shown in Figure 5.3 and in Figure 5.4. The predicted trajectories, made using the available data up to the first decision point (θ_i=35min), for venting (v) and reactor temperature (Tr) when the process is affected by a disturbance of –10% (mass) in the initial water content are shown in Figures 5.3. Each predicted trajectory is obtained using: (-x-) expectation-maximisation (EM), (-□-) iterative-imputation (IPM), (···) single component projection (SCP), and (-o-) projection to the plane using PLS (PTP) method (Arteaga and Ferrer [2002], García-Muñoz and MacGregor [2003]). As judged from this example and many similar simulations, all the missing data algorithms provide reasonable estimates of the trajectories, except perhaps the SCP method. In Figure 5.4 predictions of the final qualities made at the first decision point (θ_i=35min) using the IMP approach are shown when the initial water content randomly varies for 15 batches in the range of ±10% (mass). As can be seen in Figure 5.4, the predicted final quality properties (at θ=200min) (□) made using the PLS model at the first decision point (θ_i=35min) are in good agreement with the observed values (o).

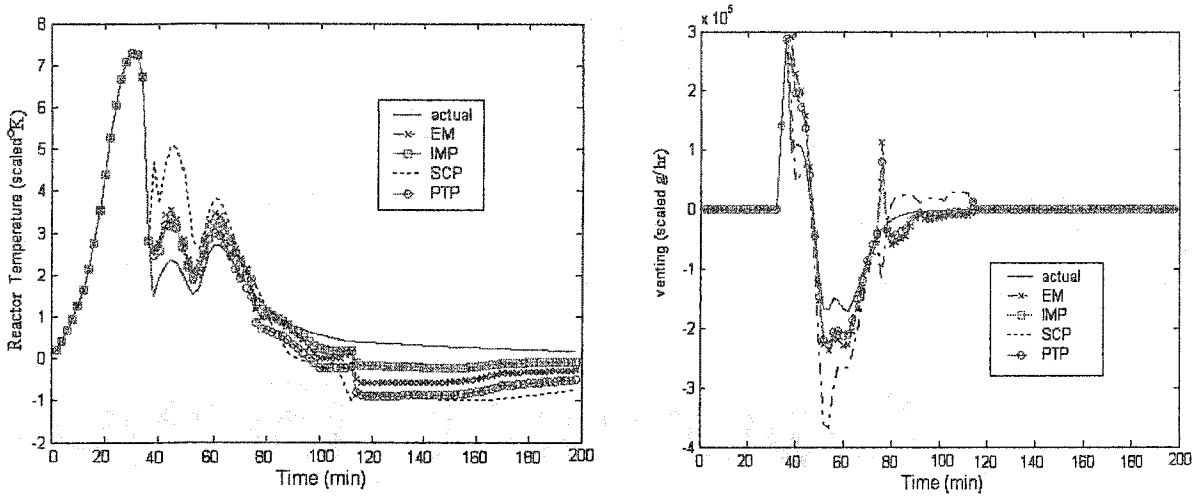


Figure 5.3 PLS model predictions of the MVTs made at the first decision point (35min) using different missing data imputation methods: (-x-) expectation-maximisation (EM), (-□-) iterative-imputation (IPM), (···) single component projection (SCP), (-o-) projection to the plane using PLS (PTP) and (—) actual value.

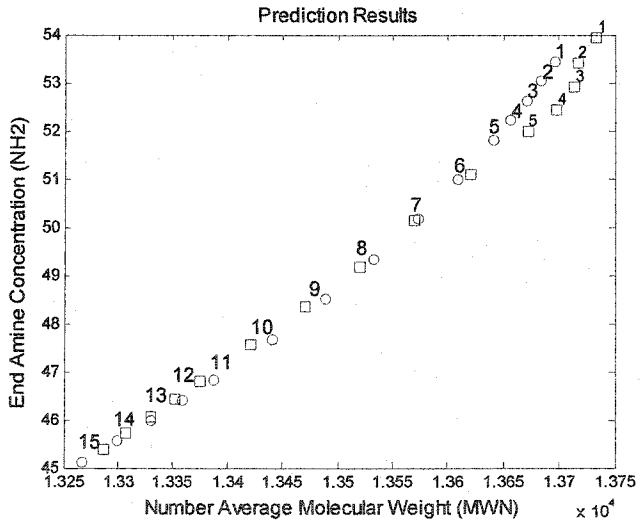


Figure 5.4 Observed (o) and predicted (□) end-quality properties using PLS model.

Slight improvement in the predictions at high MWN and NH_2 values can be obtained with a non-linear quadratic PLS model (Appendix B). However, the linear PLS model is very good in the target region (mid-values) and adequate in the extremes. Moreover, the control performance obtained using linear PLS model and that obtained

using a non-linear quadratic PLS model, for the conditions used in this example, were found to be quite similar (Appendix B).

Estimation and model prediction assessment

One of the advantages of using PLS models for control it is that it provides a powerful way to asses the validity of the PLS model for trajectory estimation of the missing measurements, and for quality prediction, and it enables one to detect sensor failures, etc. Therefore, prior to computing new control trajectories, the square prediction error (SPE) of the new vector of measurements should be computed at each decision point. This SPE provides a measure of any inconsistency between the measurements and imputed missing values for the new batch and the behavior of the set of measurements used to develop the PLS model (Nomikos and MacGregor [1995b]). If the SPE is larger that a statistically determined limit (Nomikos and MacGregor [1994]), the quality prediction and the control computation from the PLS model should be considered to be unreliable. In this situation, it might be preferable not to recompute the MVTs at the current decision point, but simply continue to apply those from the last decision point.

Regulatory Control

At each decision time (θ_i) a prediction of the final quality is made. If it is determined that control action is needed any of the control algorithms given by equations (5.5) through (5.14) can be used to compute a correction, $\Delta \mathbf{t}$, in the latent variable score space, and then the new manipulated variable trajectories for the remainder of the batch can be reconstructed from equation (5.21). The performance of the linear minimum variance controller algorithm (equation (5.14) and (5.21) with δ=**1.0**) is shown in Figure 5.5. The final quality properties of the 15 batches shown in Figure 5.4 that are affected by disturbances in the initial water concentration are shown with and without control. An "in-control" region (dotted lines) was defined considering that the final product is acceptable if their predicted values lie in the specified ranges $48 \leq NH_2 \leq 50.6$ and $13463 \leq MWN \leq 13590$. In Figure 5.5, the o's show what happens if no control action is

taken and the □'s show the end qualities obtained after control is performed. As can be seen in this Figure, the proposed control scheme corrects all batches and brings the final quality into the acceptable region. Figure 5.6 shows the jacket and reactor pressure MVTs, for runs 1 and 15 together with their nominal conditions. In this Figure, (—) represents the MVTs computed to reject a disturbance of –10% in the initial mass of water, and (- – -) those needed to reject a disturbance of +10%. Their nominal conditions of the MVTs are indicated with (---). Note that the controller computes new MVTs that are very smooth and consistent with their behavior during past operations. This consistency with past operation is, of course, forced to be true through use of the PLS model for MVT reconstruction (eq. 5.17 and 5.21).

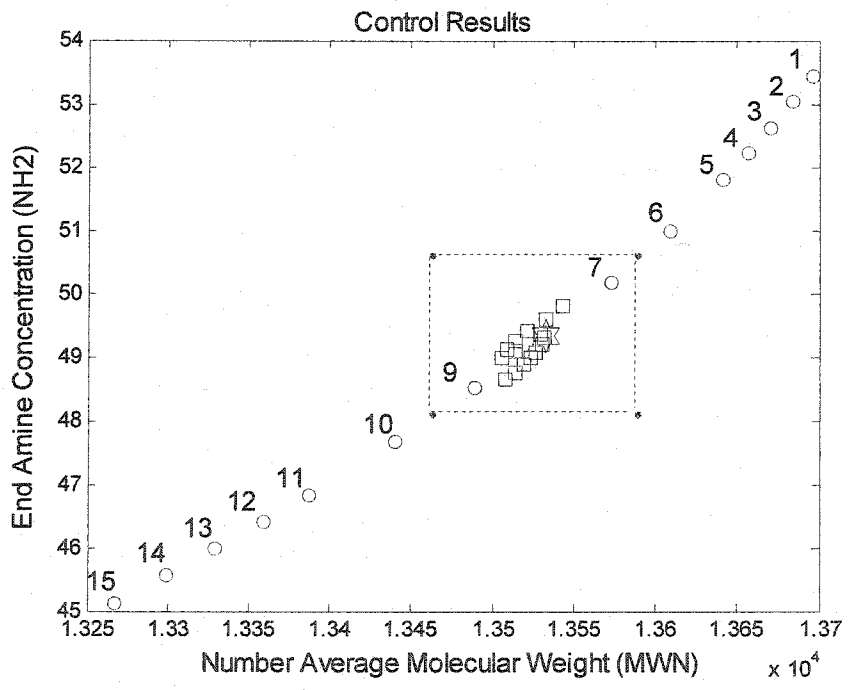


Figure 5.5 Control results. End-quality properties without control (o); after control is taken (□) and set-point (*).

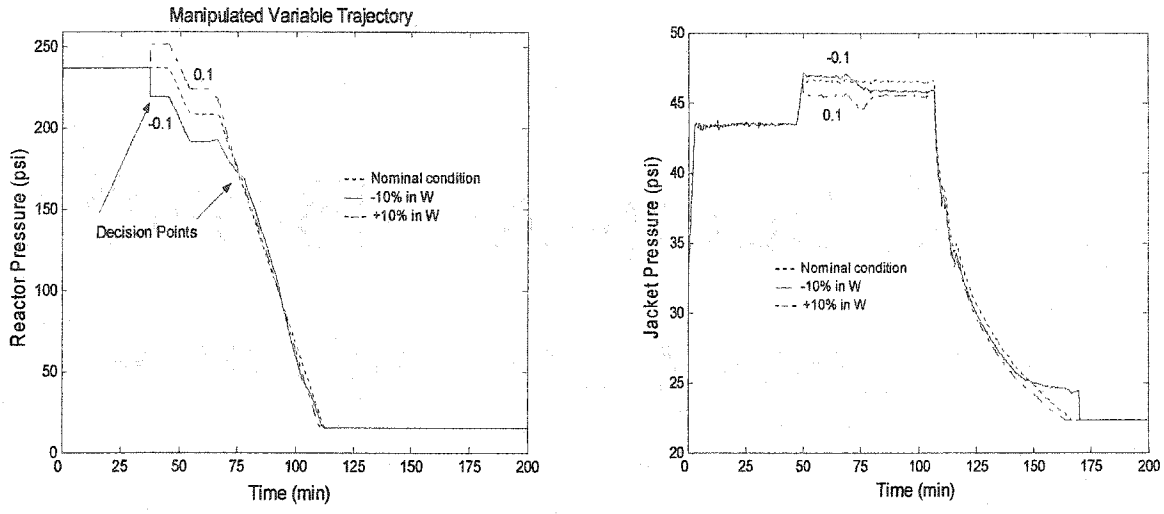


Figure 5.6 Manipulated Variable Trajectories. (- - -) nominal conditions, (—) when the disturbance is –10% mass in W, and (- – -) when disturbance is +10% in W.

Set-point Change or New Product Design

In this section the performance of the control algorithm is shown in the case that a set-point change (or new product design) is desired within the region of validity of the PLS model. No disturbances in W are included in this example, but, if present, the on-line control algorithm will easily reject them as illustrated before. The desired quality properties (o) and those obtained by using eq. (5.14) and (5.21) with δ=1 (□) are shown in Figure 5.7 for three different set-points. In Figure 5.8 the MVTs needed to achieve such set-points are shown. It can be seen that the performance of the algorithm in achieving the desired final quality set-points is very good (Fig. 5.7), and that the MVTs computed by the controller are smooth and very consistent with the shape of the trajectories from past operation.

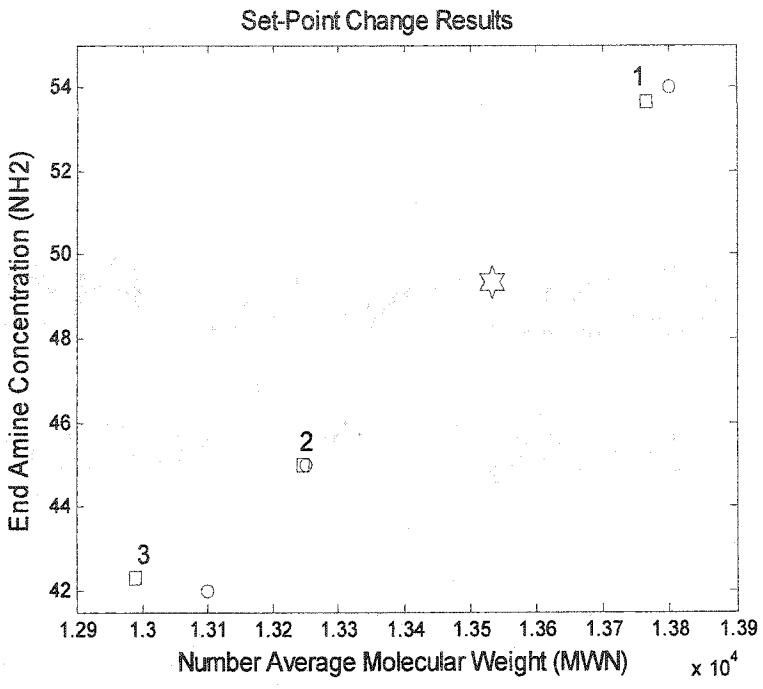


Figure 5.7 Set-point change. (o) Desired, (□) achived qualities using the control algorithm (eq. (5.14) and (5.21)) and (✳) Nominal operating point.

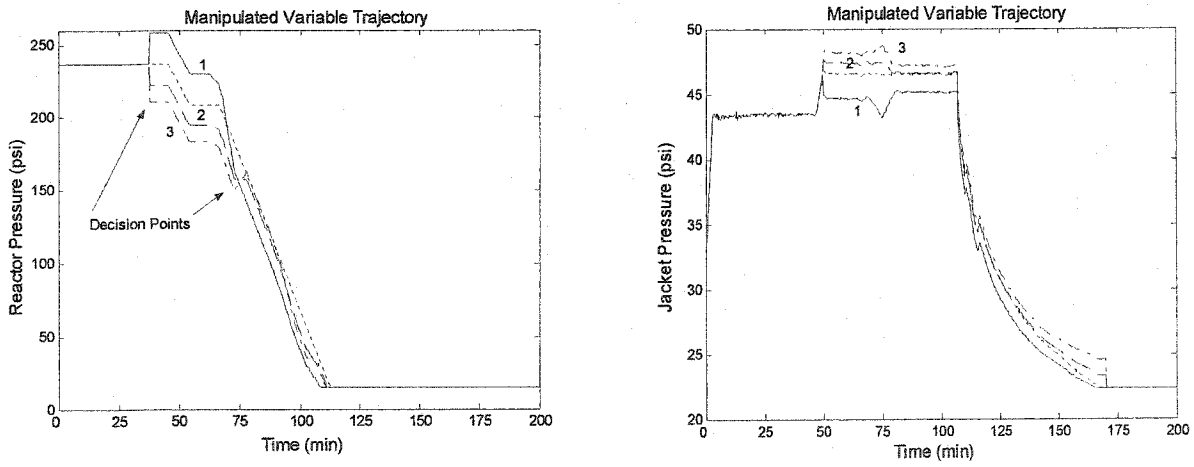


Figure 5.8 Manipulated variable trajectories for set point change. The number indicates the set-point change shown in Figure 5.7, and (- - -) the nominal MVT.

Discussion

Several practical issues may affect (to some extent) the performance of the proposed control algorithm. Some of them are briefly discussed here and more details are given in Appendix B.

The number of latent variables is generally decided by cross-validation methods at the model building stage. It was observed that too large a number of components (with respect to that obtained by cross-validation) may promote an ill-conditioned $\mathbf{P}_2\mathbf{W}_2$ inversion at the second decision point. This problem can be easily overcome by using a pseudo-inverse procedure based on singular value decomposition as shown in Appendix B. For the simulation system studied no significant degradation in performance is obtained by using a different number of PLS components.

The influence of using different missing data imputation algorithms was also studied. All the algorithms give adequate control performance. Those based on EM, IMP and PTP perform slightly better than the one in which SCP was used.

In the previous examples, a total of 45 batches (30 with a movement in the MVTs at the two decision points) were used for model identification. However, adequate control performance (all test batches falling inside the "in-control" region of Figure 5.5) was achieved using as few as 15 batches (10 in which some experiment in the MVTs was performed). This illustrates that the data requirements for PLS model building are modest. However, if the model has been identified using very limited or uninformative batch data-sets (as those arising from only historical data), batch-to-batch model parameter updating can be performed at the end of each new completed batch to improve the quality of the model parameter estimates, prediction and control for the upcoming batch (Flores-Cerrillo and MacGregor [2003]). Details are given in Appendix B.

To assess the impact of measurement noise on the performance of the algorithm, different levels of random noise were added to the on-line measurements of reactor temperature (T_r) and venting (v). It was found that adequate control performance (test batches falling inside the "in-control" region of Figure 5.5) is achieved with noise levels up to 35% in temperature and venting rate, respectively. The noise level here represents the percentage of the noise variance over the true variations of the temperature and venting rate changes observed in the training set. 35% noise level approximately represents variations in temperature of ±2°K and venting rate of ±42g/s (1 standard deviation, see Appendix B for details). For larger levels of noise (50%, for example) the control performance is slightly degraded because the random error added to the measurements becomes quite large when compared with the true variations in the MVs. A no-control region that reflects the impact of these measurement noises may be obtained by propagating such measurement errors with the PLS model as suggested in Yabuki and MacGregor [1997]. This would prevent control actions from being implemented based solely on the uncertainty arising from noise.

The control methodology outlined in section 5.2 can be easily extended to cases in which a non-linear PLS model and control is needed. This is achieved by simply modifying eq. (5.12) (case 3, dim ($\Delta\mathbf{t}$) > dim ($\mathbf{y_{sp}}$)) to take into account the non-linear nature of the PLS algorithm. For example, in the case of a quadratic PLS model, eq. (5.12) can be restated as:

$$\underbrace{\min}_{\Delta\mathbf{t}(\theta_i)} \Delta\mathbf{t}^T\Delta\mathbf{t} \qquad st \quad \mathbf{y_{sp}}^T = \mathbf{u}^T\mathbf{Q}^T \tag{5.22}$$

where $$\mathbf{u}^T = \beta_1 + \beta_2(\Delta\mathbf{t} + \hat{\mathbf{t}}_{present})^T + \beta_3(\Delta\mathbf{t}^T + \hat{\mathbf{t}}^T_{present})^2.$$

This equation can be easily solved for $\Delta\mathbf{t}$ using quadratic programming (or nonlinear least squares in the case of no constraints), and the MVTs can be reconstructed

in the same way as described in section 5.2.4. From the simulation study, the control performance of the quadratic PLS model is quite similar to that obtained using the linear PLS model (see Appendix B). This is not surprising because, in the region under study, the process is only slightly non-linear. However, if larger disturbances affect the process a non-linear PLS approach may be better suited.

Finally, a preliminary method to handle hard constraints in MVT is proposed in Appendix B.

5.3.2 Case study 2: Feasibility study on industrial data for an emulsion polymerisation process

Data

In this feasibility case study, industrial data for an emulsion polymerisation processes is used. The original data-set consists of 53 batches obtained from an experimental design in which the initial conditions and/or process variable trajectories were altered. No intermediate quality measurements were available during the reaction. However, final product physical properties (FP) and final product quality properties (FQ) are available at the end-of the process for most of the batches. Figure 5.9a shows the actual process variable trajectories that comprise the training data set (**X**), while Figure 5.9b shows the 6 quality properties (**Y** matrix), corresponding to these batches. In Figure 5.9a, it can be noticed that *i*) the process trajectories were aligned by using an indicator variable, the reaction extent (every interval represents a 0.5% increase in the reaction extent) since every batch had a different end reaction time, and that *ii*) some of the trajectories contain a noticeable level of noise. It was decided not to perform any pre-treatment such as filtering or smoothing on the process trajectories in order to test the performance of the prediction and control algorithm under this situation. It can also be seen in Fig 5.9b that FP-1 and FP-2 are highly correlated therefore, to avoid an ill condition matrix inversion in the control computation stage, FP-2 was removed and only

5 end quality properties controlled. Removing FP-2 poses no problem since by controlling FP-1 and the other quality variables we are controlling FP-2 indirectly. Alternatively, we can perform PCA on the quality property matrix (Y) and control the corresponding principal components instead of the actual properties. For poetry reasons no further details can be given regarding the nature of the process trajectories, initial conditions or product specifications.

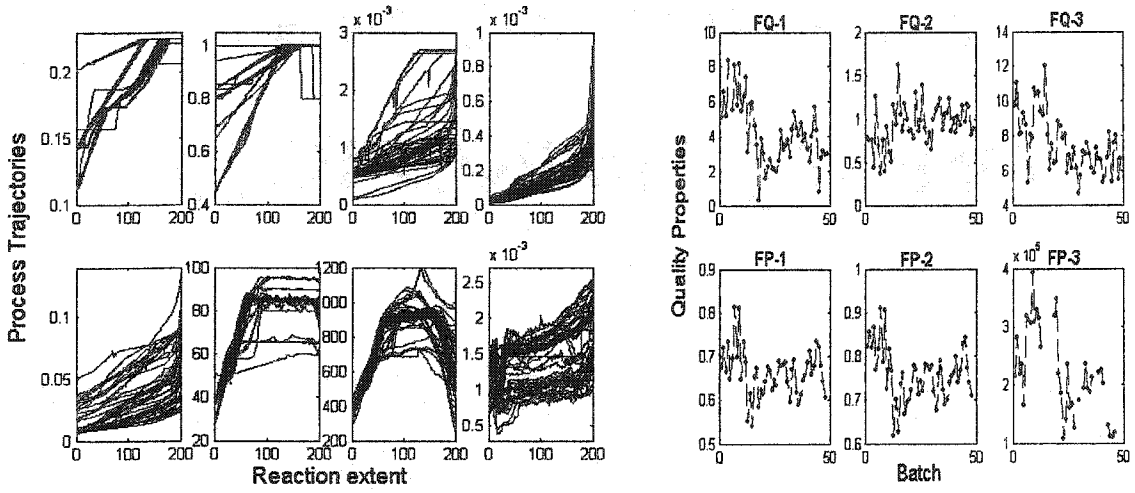


Figure 5.9 (a) Original process variable trajectories. Every interval represents 0.5% of reaction extent; (b) original quality properties

From the original data, 49 batches were used as a training data-set, while 4 batches were used as testing set. These four batches were selected to span different regions of the space far from the origin as can be seen in Figure 5.10. In this Figure the projection of all batches in the first two PLS dimensions (t_1-t_2) is shown. Batches 6,12,16 and 46 were removed from the dataset and used as test data. The 8 process variable trajectories are manipulated variables and each one of them is segmented in 200 intervals (every interval represents 0.5% of reaction extent). Therefore the data matrix used for model building consists of segmented MVTs [X] and initial conditions [Z] (regressor matrices), and the matrix of 5 physical and quality properties [Y]. The identified PLS model consists of 5 latent variables (obtained by cross-validation) that fits 76.8% of the X

space and 69.9% of the Y space. Based on cross-validation, 51.7% of the Y space can be predicted.

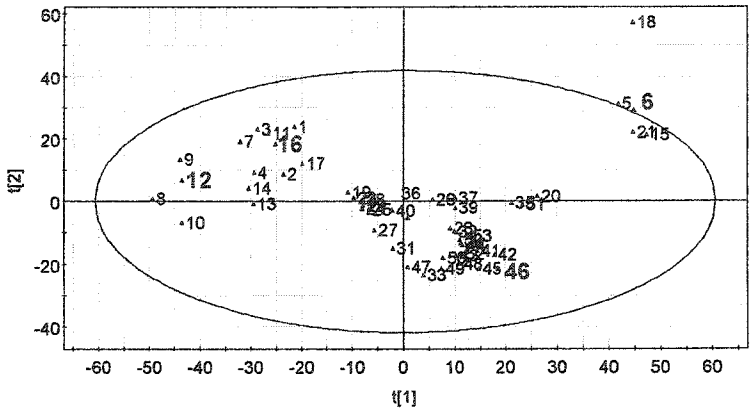


Figure 5.10 t_1- t_2 PLS space for the batches used in the training data-set. Batches 6,12,16, and 46 were removed from the original data set and used as test data.

Control objectives

The batch data in this study was the result of open-loop batch runs collected under different initial conditions and different MVTs. There was no possibility of implementing the resulting controller on the batches. Therefore, this data is simply used to test the feasibility of the prediction and control algorithms. One of the existing batch runs is taken as the nominal conditions and the final physical and quality variables (**y**) measured from it selected as the targets (set-points). Others batch runs with different initial conditions and different MVTs are then selected as initial disturbance conditions for a new batch. If no corrective action is taken to adjust the MVTs then the batch will follow the actual MVTs implemented throughout its duration, and the final quality (**y**) will be the measured values for that batch. Control is to be applied after a batch has reached 10% of completion (based on reaction extent).

Direct evaluation of the controller is not possible, but indirect validation can be obtained by comparing how close the recomputed MVTs follow the nominal MVTs from 10% of reaction extent until the end of the batch. Since the first 10% of the history of the

new batch is different from the nominal MVTs, then to achieve the desired final qualities (qualities of the nominal batch), one should not expect the recomputed MVTs to exactly follow those for the nominal batch, but they should be close to them. Notice that if the control algorithm is actually implemented, it would pose no problem to re-compute the MVTs at several decision points and not only at one as show here.

Prediction

To evaluate the performance of the PLS missing data algorithms, the total percent relative RMSE for all the qualities properties (5 in this study) is shown in Table 5.1 over the K=4 batches that compose the testing data set:

$$\%RMSE = \left(\frac{1}{5} \sum_{i=1}^{5} \frac{1}{K} \sqrt{\sum_{k=1}^{K} \left(\frac{y_{ik} - \hat{y}_{ik}}{y_{ik}} \right)^2} \right) \times 100$$

where y_{ik} is the i-th observed end-quality property for batch k and $\hat{y}_{ik}$ its predicted value.

As an illustration of the missing measurement reconstruction (at 10% of reaction extent using the EM approach), Figure 5.11 is shown for batch 12, where it can be noticed that the trajectory estimation is satisfactory in spite of the high level of noise.

Table 5.1 Performance of missing data algorithms for prediction: total percent relative RMSE for all 5 end quality properties.

Algorithm	EM	IMP	SCP	PTP-PLS
%RMSE	7.9	7.2	9.8	6.8

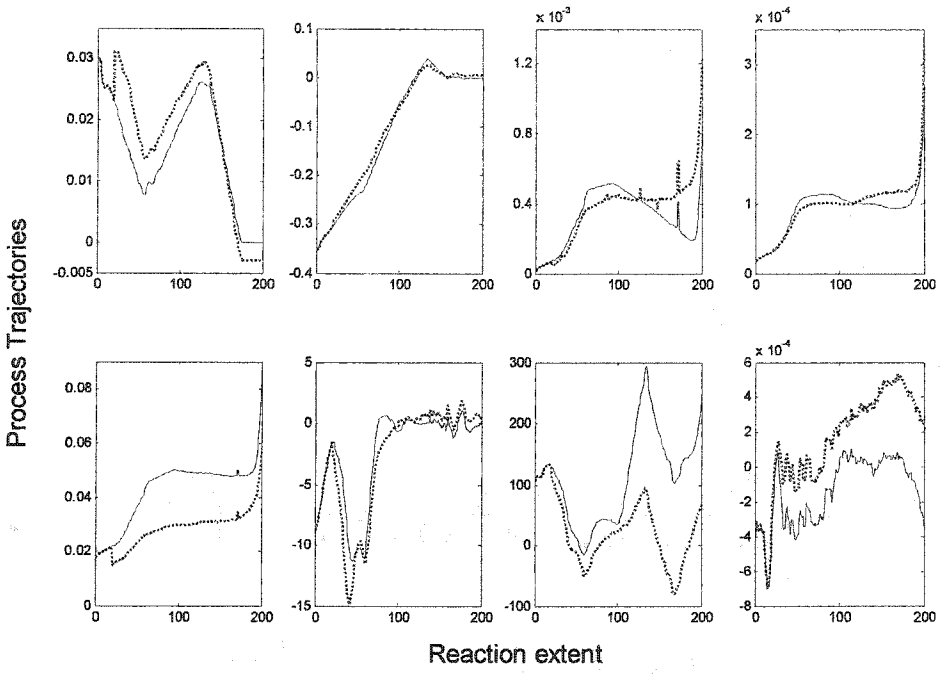


Figure 5.11 Performance of the missing data algorithm for reconstruction of process measurements. The prediction is performed at 10% of reaction extent (every interval represents 0.5% of reaction extent). (···) estimated trajectory using the EM algorithm and (—) observed trajectories (scaled units).

Control

As an illustration of the control performance using the proposed scheme (equation 5.10 and 5.21 with δ=1.0), results for one testing batch (batch 12) are shown. Figure 5.12 shows the measured final values of the y variables (o) for the batch when no control was taken, their predicted values at 10% of completion if no control were taken (*), the target values (□), and the *expected* quality properties obtained if control action *were* performed (✯). Since a minimum variance strategy was used (eq. 5.10 and 5.21), the values of the expected end quality properties resulting from the control algorithm will match their targets (□), (since these values were computed using simply the PLS model with the imputed MVT adjustments obtained from model inversion using the same PLS model.) A better way to evaluate the reasonableness of the control is to inspect the MVTs obtained

from the control algorithm. Figure 5.13 shows nominal trajectories (···), the current trajectories that would give "out-of-control" qualities (---) and the MVTs obtained from the control algorithm (—) (at 10% of reaction extent) that would drive the predicted physical and quality properties to the desired targets. In this Figure, notice that MVTs obtained from the control algorithm after 10% of completion are quite close to their nominal conditions and exhibit the desired shapes. It seems reasonable to assume that if these new trajectories were to be implemented, they would drive the process closer to the desired end-quality values, simply because the new MVTs are much closer to the nominal conditions than those when no control is performed. Note that they should not match the nominal trajectories exactly because they must also compensate for the first 10% of the batch being run at the wrong conditions. Furthermore, since the trajectories are highly correlated with one another, there are various trade-offs among the MVTs that might give quite similar final quality values. In summary, although the control could not actually be tested, these results indicate that the controller is behaving very much as one might expect and are providing the incentive for its implementation.

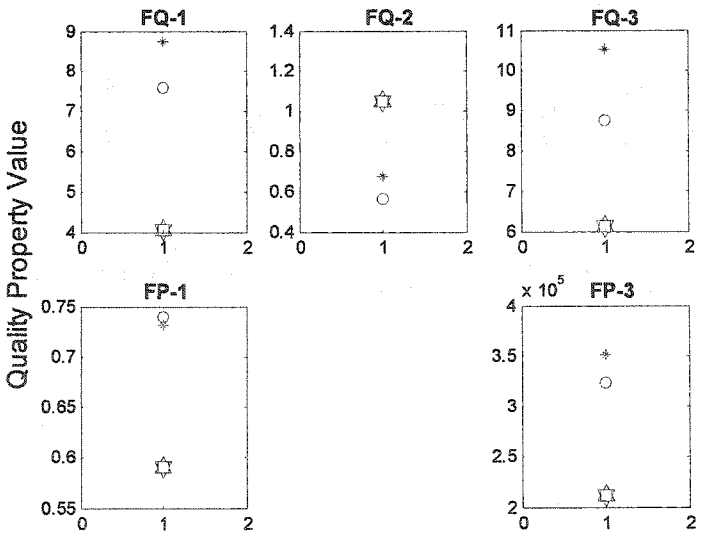


Figure 5.12 Control results (control action taken at 10% of completion of the batch). Target (□), predicted qualities (*), observed values if no control action is taken (o) and expected quality properties *if* control action *were* performed (✳).

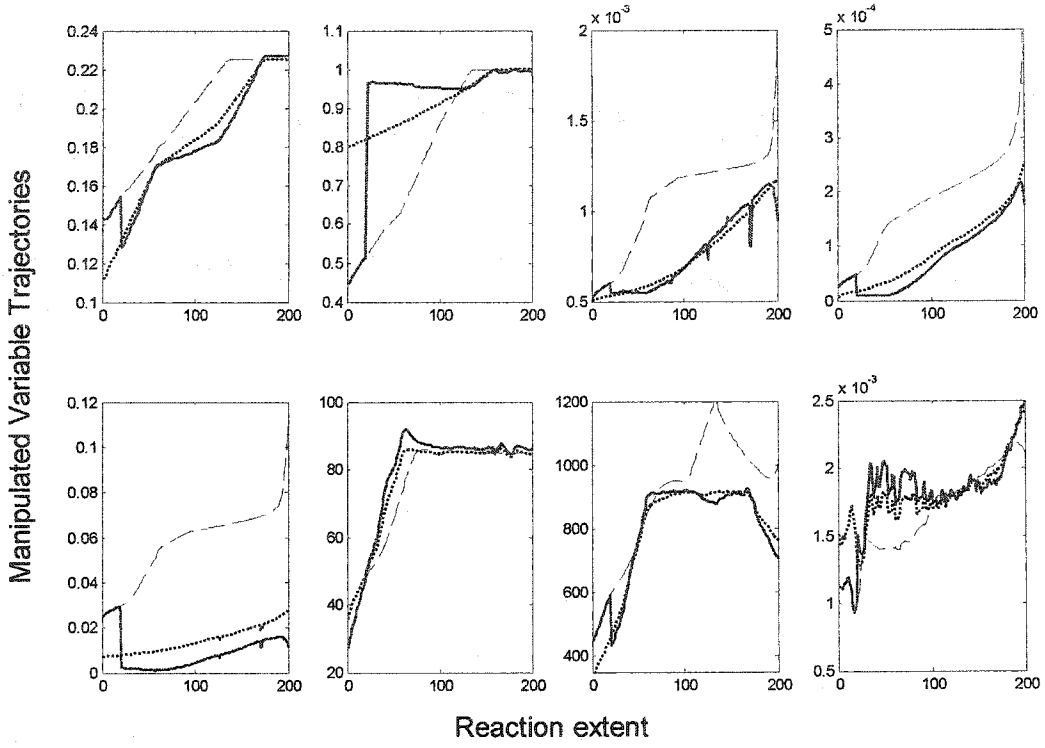


Figure 5.13 Manipulated variable trajectories (computed at 10% of reaction extent from the beginning of the process). (···) nominal conditions; (---) current trajectories that would give "out-of-control" qualities and (—) MVTs obtained from the control algorithm (equation 5.10 and 5.21, with δ=1.0).

5.4 Conclusions

A novel control strategy for final product quality control in batch and semi-batch processes is proposed that recomputes, on-line, the entire remaining trajectories for the MVs at several decision points. In spite of the fact that the resulting controller solves for the high dimensional manipulated variable trajectories (MVTs), the control algorithm involves solving for only a small number of latent variables in the reduced dimensional space of a PLS model. The high dimensional manipulated variable trajectories are then solved by inverting the PLS model. The only requirement of this approach (as with any other control algorithm that recomputes the MVTs) is that the lower level control scheme

can accept and track the computed modified trajectories. The strategy uses empirical PLS models identified from historical data and a few complementary experiments. The algorithm is illustrated using a simulated condensation polymerisation process and data obtained from an industrial emulsion polymerisation setting. Since smooth and continuous MV trajectories can be obtained, the approach seems well suited for use in processes and mechanical systems (robotics) where such smooth changes in the MVs are desirable.

Nomenclature

A = number of principal components

$\mathbf{E}$ = residual matrix

f = number of on-line measurements for the j-th variable

$\mathbf{F}$ = residual matrix

g = number of off-line analysis for the s-th variable

K= number of batches

l = number of trajectories for the on-line variables

M= number of quality properties

n = number of trajectories for the manipulated variables

$\mathbf{P}^T$ = loading matrix

$\mathbf{p}^T$ = loading vector

$\mathbf{Q}_1$ = weighting matrix in the controlled scores

$\mathbf{Q}_2$ = score suppression movement matrix

$\mathbf{Q}^T$ = projection matrix from PLS

r = number of the off-line variables

s^2 = variance of a score

$\mathbf{T}$ = score matrix

$\mathbf{t}$ = score vector

$\hat{\mathbf{t}}_{present}$ = vector of estimated scores

$\mathbf{u}_c$ = vector of manipulated variables trajectories

$\mathbf{u}_{c,\,future}$ = vector of future control actions ($\theta_i \leq \theta \leq \theta_f$)

$\mathbf{u}_{c,\,implemented}$ = vector of implemented control actions ($0 \leq \theta < \theta_i$)

$\mathbf{W}$ = projection matrix

$\mathbf{x}$ = regressor vector that includes on-line and off-line measurements, and control actions

$\mathbf{X}$ = unfolded regressor matrix of process trajectories (MVTs and measurements)

$\underline{\mathbf{X}}$ = three dimensional array

$\mathbf{x}_m$ = vector of total measurements (on-line and off-line)

$\mathbf{x}_{m,\,future}$ = vector of unmeasured variables at time θ_i ($\theta_i \leq \theta \leq \theta_f$)

$\mathbf{x}_{m,\,measured}$ = vector of measured variables at time θ_i ($0 \leq \theta < \theta_i$)

$\mathbf{x}_{off}$ = vector of off-line measurements

$\mathbf{x}_{on}$ = vector of on-line trajectory measurements

$\mathbf{Y}$ = matrix of quality properties

$\mathbf{y}$ = vector of quality variables

$\hat{\mathbf{y}}$ = vector of estimated quality variables

Greek symbols

λ = weighting factor

θ = time / decision time

δ = de-tuning factor

α = proportionality vector

β = coefficients for the PLS inner relation

Index

a = latent variable index

i = time index

j,s,m = variable index

k = batch index

f = final batch time

Chapter 6

Multivariate Analysis and Monitoring of Batch Processes using Batch-to-Batch Information

Multiway principal component analysis (MPCA) and multiway partial least squares (MPLS) are well-established methods for the analysis of historical data from batch processes, and for monitoring the progress of new batches. In this Chapter, an extension of the multi-block MPCA/MPLS approach is introduced to explicitly incorporate batch-to-batch trajectory information while keeping all the advantages and monitoring statistics of the traditional MPCA/MPLS. It is shown that the advantages of using information on prior batches for analysis and monitoring is often small, but it can be useful for detecting problems when monitoring new batches in the early stages of their operation. The approach is illustrated using condensation polymerization and emulsion polymerization systems as examples.

6.1 Introduction

Multi-way PCA and PLS for the analysis, monitoring and prediction of final product quality in batch processes were first introduced by Nomikos and MacGregor [1992, 1994, 1995a, 1995b]. They illustrated the detection of abnormal batches using several criteria such as the Q statistic (also known as square prediction error (SPE) or

distance to the model in the X space (DMODX)), the instantaneous SPE (for on-line monitoring), and the PCA or PLS score plots or equivalently Hotelling's T^2 statistic. Kourti et al. [1995, 1996] used multi-block methods (MBPCA/MBPLS) to incorporate different initial conditions, modes of operation, and prior processing conditions into the analysis and monitoring of batch processes. In this Chapter is proposed a variation of these multi-block methods to incorporate more efficiently previous batch-to-batch information.

Recently, Dorsey and Lee [2001] proposed a monitoring framework based on state-space models to take into account batch-to-batch variability more explicitly than the MPCA and MPLS methods. The approach first uses MPCA to extract the time and covariance structure of the data within batches, and then uses sub-space identification to obtain a state-space model for these principal components. However, the proposed methodology is useful mainly for detecting batch-to-batch variations only in the score space obtained from the MPCA model. By modeling only the score space (obtained by MPCA), the identified state-space model accounts only for the batch-to-batch variation in the score space of the normal (in-control) runs. Since the PCA score space is completely orthogonal to the SPE space of the MPCA model, then any charts based on the model states or innovations will not be able to detect faults that are detected primarily in the SPE space by the MSPC methods (e.g. faults that introduce totally new latent variables or PCs as opposed to those that simply induce larger variations in the existing PCs). Faults that affect both the score and SPE space of the MPCA model might be detected if their effect on the score space is strong enough. The shortcoming of their proposed monitoring methodology results from the fact that all information on the space orthogonal to the PCA score space (i.e. the SPE space) is lost. Furthermore, subspace methods for system identification usually require more training batches to build the state space models than is normally required to establish multivariate PCA/PLS models.

This Chapter introduces a modified MPCA/MPLS procedure that, besides retaining all the advantages of the MPCA/MPLS methods for batch analysis and on-line

monitoring also enables one to incorporate a summary of prior batch trajectories and performance. The approach and potential benefits to be gained from it are illustrated based on simulations of two batch polymerizations processes: the condensation polymerization of nylon and the emulsion polymerization of styrene.

6.2 MPCA and MPLS monitoring using batch-to-batch information

6.2.1 Preliminaries

An important question concerns "when data from prior batches would be useful for the analysis and monitoring of future batch processes?" Clearly, a minimum requirement is that they contain some information on effects that will have an influence on the performance of the future batches. This implies that there must exist some autocorrelation in important performance variables from batch-to-batch. Such a situation would arise if a common source of raw materials is being used for successive batches and the materials from this source have some characteristics (e.g. impurity concentrations, surface chemistry properties, etc.), which change slowly with time. Then one could expect the performance of future batches to be related to that of recent past batches. If no such batch-to-batch carry-over effect due to common disturbances is present, then the value of incorporating prior batch information into MPCA or MPLS analysis or monitoring schemes would be negligible.

6.2.2 Incorporation of batch-to-batch information into MPCA/MPLS

To capture information in prior batches, one could directly use the final quality measurement matrix ($\mathbf{Y}$) taken on the product from previous batches. These data can be easily incorporated into a matrix $\mathbf{Z}$ and then existing multi-block approaches used for the analysis and monitoring of batch processes (Kourti et al. [1995]). However, this may not always be possible. The product quality data for the last batch ($\mathbf{y}^{(k-1)}$) may not be available from the quality control laboratory before the next batch is started. Furthermore, not all the product quality data are measured for every batch, and even those that are

measured may have considerable measurement error and may not be sufficient to capture all the relevant information from past batches. Therefore, in this Chapter is proposed to use the final PCA or PLS scores values ($t_1, t_2 .. t_a$) from prior batches to summarize the histories of prior batches. The idea is then to include these summarizing scores from prior batches into a matrix **Z**. A multi-block MPCA or MPLS (MBPCA/MBPLS) is then used to combine this batch-to-batch information (**Z** matrix) with the trajectory data (**X**) for the current batches (k). The structure of the resulting data matrices is illustrated in Figure 6.1. Each row, $\mathbf{x}_k^T$, of the **X** matrix consists of measurements on all the process variables at all time intervals for the k^{th} batch, and the corresponding row $\mathbf{z}_k^T$ of the **Z** matrix contains score values for each of the past r batches, that is

$$\mathbf{z}_k^T = [t_1^{(k-1)}, t_1^{(k-2)} \cdots, t_1^{(k-r)}; t_2^{(k-1)}, t_2^{(k-2)} \cdots, t_2^{(k-r)}; \cdots; t_a^{(k-1)}, t_a^{(k-2)} \cdots, t_a^{(k-r)}] \quad (6.1)$$

Once the multi-block model has been built, this lagging of the prior batch scores in the **Z** matrix poses no problem, and the analysis and monitoring is easily accomplished with existing multi-block MPCA/MPLS approaches (Kourti et al. [1995]). However, a problem arises at the model building stage, when, in order to build the MBPCA or MBPLS model, one needs complete data for both the **Z** and **X** matrices for all batches in the training data. But the score values from prior batches needed to form the **Z** matrix will not be available until after the model has been built. Faced with this dilemma, the following iterative training approach to build the model is proposed.

1) An initial MPCA or MPLS is carried out on the **X** matrix. The scores for each batch from this model are then used to provide initial estimates of the scores for prior batches to be included in the **Z** matrix, as illustrated in Figure 6.1.

2) The **Z** matrix is then weighted, if desired, relative to the **X** matrix (e.g. block scaling) and an augmented [**Z X**] matrix is used to perform another PCA or PLS to obtain a complete model incorporating both the prior batch information and the current batch data. (If one does not wish to discard the first r batches, then a missing data algorithm

(e.g. Nelson et al. [1996]) can be used to account for the unknown prior batch information arising from the lagging of the first r batches.)

3) Repeat (2) until convergence of the scores (T) is achieved: $(\mathbf{Z}_i - \mathbf{Z}_{(i-1)}) = \boldsymbol{\Omega}; \|\boldsymbol{\Omega}\| \leq \varepsilon$, where i is the iteration number. In the examples considered in this work, convergence of the scores was achieved using direct-substitution in a few iterations.

4) At convergence, the final models that can be used for monitoring, analysis and prediction are given by:

For PCA:

$$[\mathbf{Z}\ \mathbf{X}] = \boldsymbol{\Gamma}\mathbf{V}^{\mathrm{T}} + \mathbf{E}^{*} \tag{6.2}$$

For linear PLS:

$$\begin{aligned} [\mathbf{Z}\ \mathbf{X}] &= \mathbf{T}\mathbf{P}^{\mathrm{T}} + \mathbf{E} \\ \mathbf{Y} &= \mathbf{T}\mathbf{Q}^{\mathrm{T}} + \mathbf{F} \end{aligned} \tag{6.3}$$

Each low dimensional ($1 \times a$) row of the ($k \times a$) score matrices $\boldsymbol{\Gamma}$ or $\mathbf{T}$ ($\boldsymbol{\Gamma} = [\mathbf{Z}\ \mathbf{X}]\mathbf{V}$ and $\mathbf{T} = [\mathbf{Z}\ \mathbf{X}]\mathbf{W}$) provides all the statistically significant information on the relationships among the prior batch histories (rows of $\mathbf{Z}$) and the time histories of all the variables in the current batch (rows of $\mathbf{X}$). Separate loadings and scores for the $\mathbf{Z}$ and $\mathbf{X}$ matrices in a multi-block scheme then can be obtained directly from the above MPCA or MPLS models (Westerhuis et al. [1998]).

Notice that the iterative scheme is only necessary for model building. Once the model is identified, the iterative scheme is not needed and monitoring, analysis and prediction can be performed in the same way as is done in normal MPCA and MPLS methods. For example, for each new batch, the history of prior batches (rows of $\mathbf{Z}$), and the process trajectory (a new row, $\mathbf{x}_{new}^{\mathrm{T}}$, of $\mathbf{X}$) are both available to calculate the score

values $t_1, t_2 ... t_a$. Therefore, the use of the proposed approach (Figure 6.1) will allow one to efficiently incorporate prior batch information into a single model for both the analysis of completed batches and for the on-line monitoring of new ones. Moreover, all the benefits and statistical analysis tools of the conventional MPCA and MPLS methods will be retained. The approach can also be easily extended in cases that a non-linear PLS method is needed (Wold et al. [1989], Frank [1990], Wold [1992], Berglund et al. [1997]).

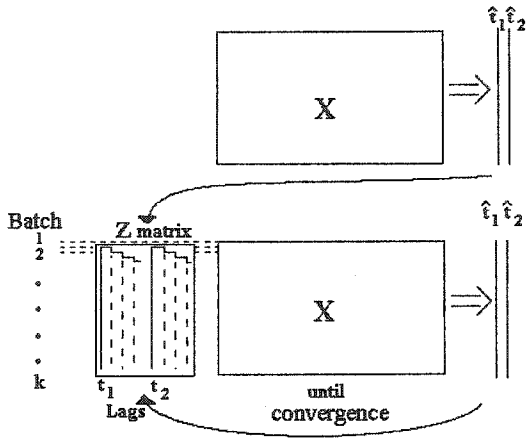


Figure 6.1 MPCA monitoring scheme using batch-to-batch information.

For analysis or monitoring, several different combinations of the matrices shown in Fig. 6.1 can be used. To monitor only batch-to-batch changes, the **Z** matrix contains all the necessary information. Multivariate control charts based on **Z** alone would be sufficient for detecting changes in the autocorrelation structure of the batch-to-batch behavior such as might arise from changing feedstocks of raw materials. Using the **X** matrix alone would allow one to analyze and monitor only the within batch changes. Using both the **Z** and **X** matrices allows for complete treatment of both batch-to-batch and within batch changes within the same MBPCA/MBPLS model.

6.2.3 Model Building: Selection of number of lags

After convergence of the lagged-scores (**Z**) has been achieved, the number of significant lags (number of prior batches, r, used in the **Z** matrix) can be determined by

inspecting the values of the loadings for the lagged scores in **Z** ($\mathbf{p}_z$). If the loadings on all the scores in **Z** for all batches beyond a certain lag are small, then these lags can be dropped and the model reiterated again until convergence. To illustrate this, an example is shown in Figure 6.2a and 6.2b (Case study 1, section 6.3.3, for the emulsion polymerization of styrene for large positive batch-to-batch correlation (ϕ=0.9) and for small batch-to-batch correlation (ϕ=0.2) respectively.) In Figure 6.2a, it can be seen that only the two loadings $p_{z,1}^{(k-1)}, p_{z,2}^{(k-1)}$ (from the first principal component, $\mathbf{t}_1$) associated with the immediately preceding batch (k-1) are large indicating that only one lag (and two scores (**T**), eq. (6.1)) need to be used in model building. Moreover, the method can also detect when the incorporation of previous batch information would be of little value, as shown in Figure 6.2b, where it can be seen that the values of $\mathbf{p}_z$ are small for the previous scores at all past lags (since the correlation from batch-to-batch is low, ϕ=0.2).

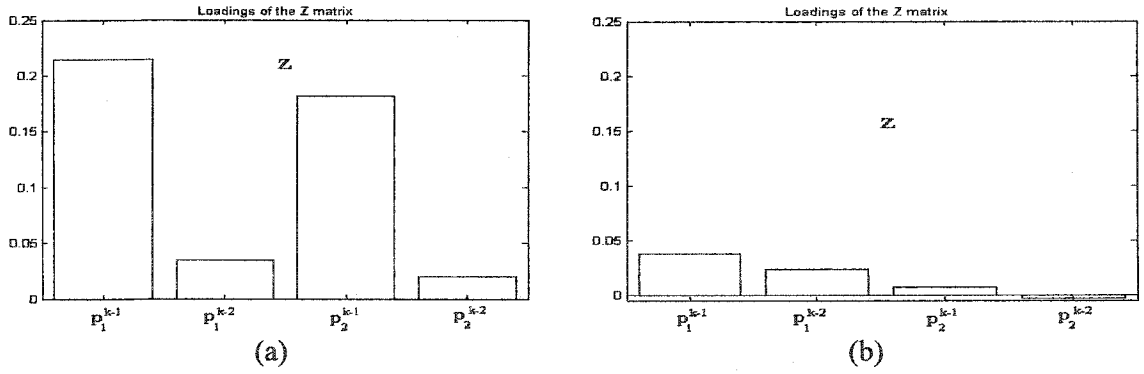


Figure 6.2 Selection of the number of significant lags for model building. (a) large (ϕ=0.9) and (b) small (ϕ=0.2) degree of batch-to-batch correlation (Case 1, section 6.3.3).

6.3 Off-line analysis and on-line monitoring studies

6.3.1 Systems

The approach is illustrated using two simulated polymerization systems, one is the condensation polymerization of nylon 6,6 and the other is the emulsion polymerization of styrene. It is important to notice that the usefulness of the previous batch information for

on-line monitoring will be greater in early stages of the process, since in later stages most of the information is contained in the on-line measurements taken from the current batch (k). For many industrial processes, as those used here for illustration purposes, the main sources of disturbances occur at the start of the process due to initial charge conditions or impurities property variations in the raw materials. Therefore a monitoring scheme containing batch-to-batch information may be useful in providing a more consistent and faster detection of early disturbances (as long as these have a degree of batch-to-batch correlation). In what follows a brief description of the systems and conditions on which the monitoring studies were performed is presented.

Condensation Polymerization

The first process considered here is the batch condensation polymerization of nylon 6,6. A detailed theoretical model for this process was developed by Russell et al. [1998b] and is used in this work. Details about the model and model parameters are described in the original publication. The focus of this study will be on the initial polymerization stage of the batch, from the initialization of the process to the opening of the vent valve (around 35 min). This is because the decision when to open the vent valve is key to control the achievable product quality (Russell et al. [1998b]). Besides, the results of this initial polymerization stage are indicative of the complete batch since the main source of disturbance is the fluctuation of the feed water (W) (a single evaporator usually serves several reactors (Russell et al. [1998a])).

For on-line monitoring, measurements include reactor pressure, steam jacket pressure, reactor temperature and vent rate. However, steam jacket and reactor pressure are considered here as manipulated variables that remain at their initial set points during the early phase and so are not included in **X** (If variations exist in these variables, their trajectories should be included in the **X** matrix). Moreover, at the initial heating stage the vent readings are zero because the vent valve is close. Therefore, the available on-line information comes from the reactor temperature readings, T_r (every 15s). The end quality variables are amine end groups (NH_2) on the polymer molecules and the number average

molecular weight (MW) at the end of the reaction (200min). The quality and process measurements are corrupted by normally distributed random error with magnitudes reported in Table 6.1. As can be seen in this Table, the noise level for the reactor temperature measurements (T_r) is σ=0.1% (approximately $\pm 450 \times 0.001 \approx 0.5^{o}K$); for NH_2 is σ=0.1% ($\pm 49.7\mu mol/Kg \times 0.001 \approx 0.05\mu mol/Kg$), and for MW is σ=0.3% ($\pm 13500 g/mol \times 0.003 \approx 40 g/mol$).

Emulsion Polymerization

A non-linear model, with simple kinetics, to simulate the styrene emulsion polymerization was developed by Lynch and Kiparissides [1981], and is used in this work. This model, originally developed for tubular reactors with full recycle, has been adapted for use in batch and semi-batch processes. For a complete description of the model and model parameters the reader is referred to the original publication. The focus of this study will be on the initial polymerization stage of the batch, from the initialization of the process up to 40 min. This is because the particle generation is of short duration and early detection of abnormal conditions of the batch would allow one to take faster corrective action. Moreover, the results of this initial polymerization stage are indicative of the complete batch because once the particle generation is over there is almost no further change in the number of particles. The main source of disturbance considered in this study is variation in the surface chemistry properties of the emulsifier. In particular, these surface chemistry variations affect the surface covering potential (a_s) of the emulsifier. This disturbance has a great effect on the number of micelles formed and hence on the number of polymer particles nucleated (Np), as well as on the resulting conversion. On-line reactor (Tr) and jacket temperature (Tj) measurements are considered to be available every minute. The end-quality variables are conversion (C) and number of particles (Np) at the end of the batch (480min). The quality and process measurements are corrupted by normally distributed random error with magnitudes reported in Table 6.2. As can be seen in this Table, the noise level for the reactor and jacket temperature measurements (T_r and T_j respectively) is σ=0.05% (approximately

$\pm 323 \times 0.0005 \approx 0.2^{\circ}K$); for Dp is (Yabuki and MacGregor [1997]) σ=0.5% ($\pm 400nm \times 0.005 \approx 2$ nm); for C is σ=0.1% ($\pm 92\% \times 0.001 \approx 0.1\%$), and for Np σ=0.1% ($\pm 5\times10^{19} \times 0.001 \approx 5\times10^{16}$particles/l). Notice that due to the low sampling rate of the on-line measurements some filtering can be performed.

Table 6.1 Measurement noise for condensation system

Measurements		σ%	Noise level[a]
X	Tr	0.1	± 0.5°K
Y	NH_2	0.1	± 0.05μmol/Kg
	MW	0.3	± 40g/mol

[a]using T_r=450°K, NH_2=50μmol/Kg, and MW=13500g/mol as base values

Table 6.2 Measurement noise for emulsion system

Measurements		σ%	Noise level[a]
X	Tr	0.05	± 0.2°K
	T_j	0.05	± 0.2°K
Y	C	0.1	± 0.1%
	N_p	0.1	± 5×10^{16}
	D_p	0.5	± 2nm

[a]using $T_r \approx T_j$=323°K, Dp=400nm, C=92% and Np=5×10^{19} as base values

Data History Generation

For the condensation polymerization system, 200 batches were used to generate the normal data history used as a training set, while in the case of emulsion polymerization 150 batches were generated. However, adequate MPCA/MPLS models can be built with many fewer batches (Nomikos and MacGregor [1995a], Kourti et al. [1996]). For both systems, the normal batch history was generated by assuming that the disturbances, d_k (W for condensation polymerization and a_s for emulsion polymerization), vary from batch-to-batch in a correlated manner according to the autoregressive model:

$$d_k = \phi d_{k-1} + \xi_k \tag{6.4}$$

where ξ represents normal distributed random error and ϕ the degree of batch-to-batch correlation. Two types of correlation are studied for each system: Strong (ϕ=0.8 for condensation and ϕ=0.9 for emulsion) and weak correlation (ϕ=0.2 for both systems). For each type of correlation, a training set was generated.

6.3.2 Case studies

Several case studies for the off-line analysis of completed batches and for the on-line monitoring of new batches were performed in both systems for low and high batch-to-batch correlation in the disturbances. Process faults or upsets were introduced including changes in the batch-to-batch correlation structure, slow drifts and short-lived up-sets. For the sake of brevity, only some of these case studies are presented as shown in Table 6.3 for the high correlation structure where the benefits of inclusion of prior batch information in the model are more evident. In the cases studies, normal MPCA/MPLS (using no information on prior batches) and the proposed approach (MBPLS/MBPCA with prior batch information, **Z** matrix) are compared.

Table 6.3 Monitoring case studies*

Case Studies	Off-line	On-line
1. Change in correlation	Emulsion	----
2. Small drift	Condensation	Condensation
3. Short-lived upset	----	Emulsion

*For strong (ϕ=0.9 and ϕ=0.8) and weak (ϕ=0.2) batch-to-batch correlation in disturbances.

6.3.3 Detection of changes in correlation from batch-to-batch

The purpose of this section is twofold: 1) to show that a proper SPC monitoring method based on MPCA/MPLS should not alarm for changes in batch-to-batch correlation if these changes are not important to product quality (final product quality only depends on the magnitude of the disturbances not on their time order or equivalently their batch-to-batch correlation structure) and 2) to show that, if needed, changes in correlation can be easily detected using previous batch information.

In this example, a change in the degree of correlation, of the testing data set, from ϕ=0.9 to ϕ=0.2 is performed (eq. 6.4) at batch 26 for the emulsion polymerization system (the variance of ξ was adjusted as described in Dorsey et al. [2001] to keep the total variance of the raw material qualities at the same level).

In Figure 6.3a, batch-to-batch evolution of the final quality properties (C, Np) is shown for this example using a Hotelling's T_y^2 chart (Tracy et al. [1992]). Figure 6.3b shows the corresponding batch-to-batch evolution of the SPE_x (Q_x statistic) obtained by performing normal MPLS on the batch data only (no inclusion of previous batch information) together with their 95 and 99% confidence limits (Nomikos and MacGregor [1994,1995a]). Note that the SPE_x does not detect the change in the degree of correlation. However, this is what should happen because, as can be seen in Figure 6.3a, the end quality properties are clearly still in a state of statistical control. Any method that alarms in this situation (Dorsey and Lee [2001]) is misleading as a monitoring method for the health of the batches. It is stressed this point because a recent publication (Dorsey and Lee [2001]) has used the lack of detection of this change in batch-to-batch correlation structure by MPCA/MPLS as a negative result for these methods. However, as shown above, it is clearly a positive feature if the objective is to monitor the health of the batches.

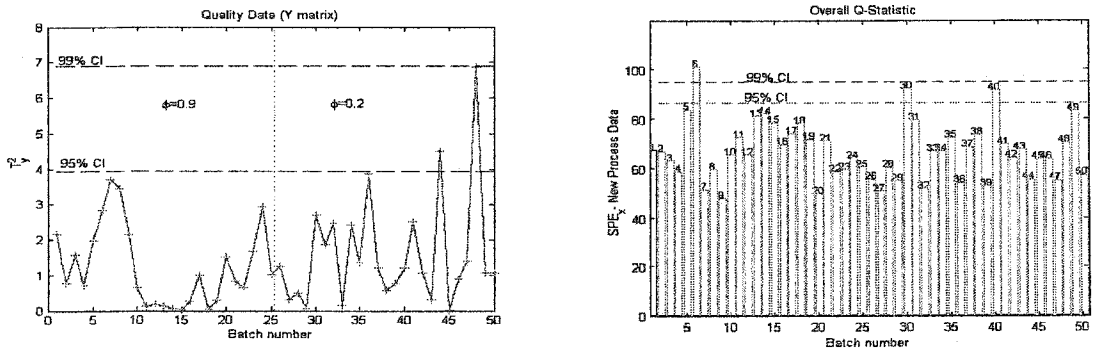


Figure 6.3 Off-line batch-to-batch monitoring of the emulsion polymerization system for a change in the batch-to-batch correlation structure of the disturbances at batch 26. a) Hotelling's T^2 for the final quality data (C, Np). b) SPE_x (Q_x) on X data using normal MPLS.

Changes in the disturbance correlation structure, although usually not important for product quality can be important, and should be monitored, if the disturbance correlation structure is being used somehow in a batch-to-batch control algorithm (Chin et al. [2000]). In that case the manipulated variables set-points are being changed for new

batches based on assuming a previously identified disturbance autocorrelation structure. If this autocorrelation structure were to change suddenly the batch-to-batch control scheme based on the assumed value should lead to poor results and degrade quality. In this situation, changes in the batch-to-batch correlation can be detected simply by performing PCA on the **Z** matrix of previous batch scores (Figure 6.1) and monitoring their SPE_Z (Q_Z statistic) and/or Hotelling's T_z^2. In this case, it may be necessary to include enough lags (r) to allow for adequate modeling of the batch-to-batch correlation structure. The detection of a change in the correlation structure of the emulsion process at batch 26 from ϕ=0.9 to ϕ=0.2 when the number of lags r=10, is shown in Figure 6.4.

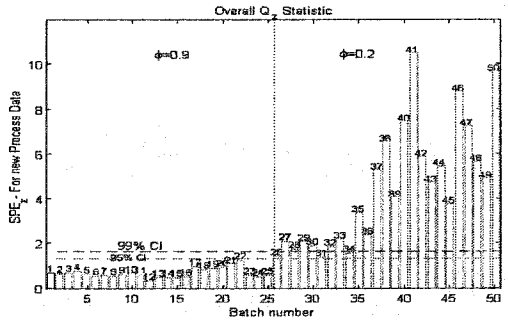


Figure 6.4 SPE_z for detection a change in correlation from ϕ=0.9 to ϕ=0.2 (change at batch 26) using r=10 lags.

Another simple alternative to detect changes in the degree of correlation is by fitting a time series model (for example AR(1), to the scores (**t**) obtained from the normal MPLS model on [**X Y**]. The estimated value of φ would then represent the degree of correlation. Change in the correlation structure of new incoming batches can be monitored using a simple Shewhart chart on the residuals (ξ) of such time series model

$$\xi^k = \mathbf{t}^k - \hat{\varphi}\,\mathbf{t}^{k-1} \qquad (6.5)$$

and confidence limits established based on the variance of the residuals. This is illustrated in Figure 6.5 for the case in which the first principal component (t_1), of the normal MPLS is modeled.

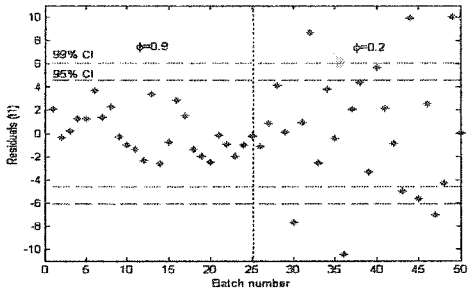


Figure 6.5 Monitoring of residuals (ξ) for t_1 from normal MPLS. Change in correlation from ϕ=0.9 to ϕ=0.2.

6.3.4 Detection of a slow drift in the water content of the salt (W) over many batches

Off-line Analysis

In this example, the objective is to detect a slow drift of the product quality for the condensation polymerization process when there is a slow drift in the initial water content (W) of the incoming batch, as well as a common cause batch-to-batch correlation of ϕ=0.8 in W. It is expected that, by using batch-to-batch information, faster detection of the drift will be achieved since, as can be seen in Figure 6.6a, W is progressively getting worse over many batches (drift begins at batch 26). In Figure 6.6b, a Hotelling's T^2 chart for monitoring the batch-to-batch evolution of the end-quality properties (NH_2, MW) is shown for the 50 batches, while in Figure 6.6c and 6.6d the t_1 score obtained from normal MPLS and MBPLS (using both **X** and **Z**) respectively is shown. In these Figures, a statistically significant out of control region (at α=0.01 significance level) is detected only at batch number 50. Note that including information (**Z**) on previous batches did not improve the time to detection (Figure 6.6d). This result shows that the use of such prior

batch information is of limited value once one has complete information on the current batch.

A faster detection of this type of events would be beneficial because would allow one to correct for the disturbance before the quality properties are off-specifications. The simplest way to detect small drifts, trends or mean shifts is by the use of a cumulative Hotelling's T^2 (ψ) obtained from either multi-block or normal MPLS. Two alternatives are shown: Crosier [1988] proposed computing Hotelling's T^2 at each observation and then computing the cumulative of the scalar distance T^2 as:

$$\psi_i = \max\{0, \psi_{i-1} + T_i - \eta\} \tag{6.6}$$

This multivariate CUSUM scheme signals an out-of control situation when $\psi_i > h$. The limit h and the parameter η where chosen as suggested by McNeese et al. [1991]: $\eta = \sigma/2$ and $h = 4.5\sigma$. Alternatively a finite-horizon cumulative Hotelling's T^2 (ψ_i) may be used (Dorsey and Lee [2001]):

$$\psi_i = \sum_{i=k-r+1}^{k} T_i^2 \tag{6.7}$$

where r is the number of previous batches over which the summation is taken. Figure 6.7a shows the CUSUM on Hotelling's T^2 (ψ_i) obtained from eq. (6.6) while in 6.7b ψ_i obtained from eq. (6.7) with r=10 for normal MPLS. It can be seen that, in both approaches, the small drift can be detected around batch 47, slightly earlier than with the direct use of Hotelling T^2 (Fig. 6.6c and 6.6d).

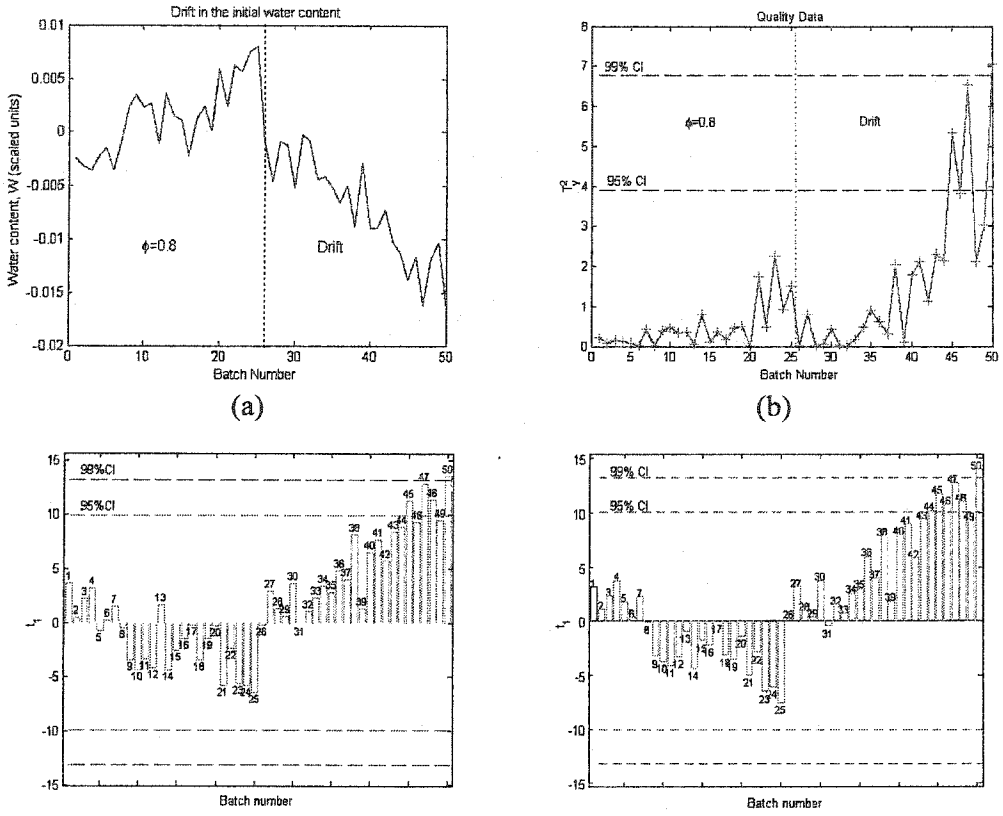


Figure 6.6 a) Disturbance trend for water content (W), b) Hotelling's T^2 on the batch final quality data (Y), c) t_1 score from MPLS, and d) t_1 score from MBPLS

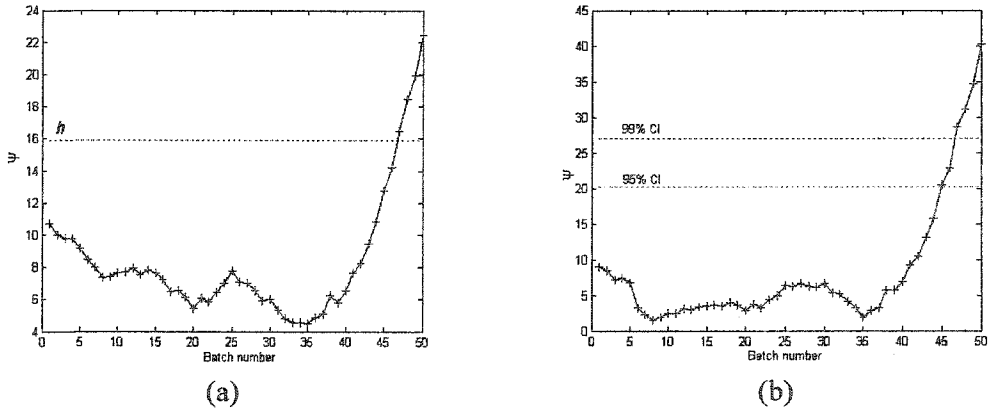


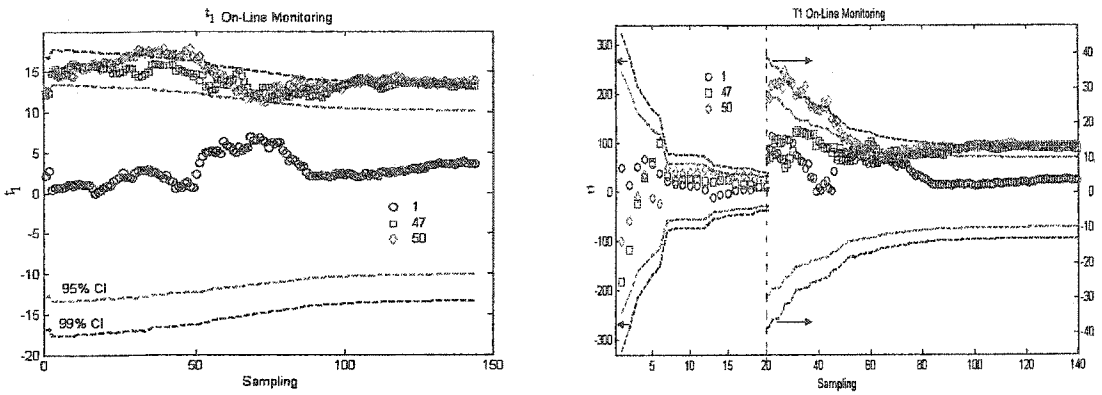
Figure 6.7 Detection of drift with cumulative Hotelling's T^2 (ψ). (a) Equation (6.6) and (b) Equation (6.7) with r=10 for normal MPLS.

In this section we have considered the use of previous batch information in the detection of changes in batch-to-batch correlation and for the detection of slow drifts for the case in which the batches have already been completed (off-line analysis). In the next section, the usefulness of prior batch information in the on-line monitoring of new batches is investigated.

On-line monitoring

In on-line monitoring, at any time during the batch, future unknown measurements need to be estimated. Nomikos and MacGregor [1994], [1995a] proposed three alternatives to estimate such measurements, and found that the approach that uses a missing data algorithm was generally superior and so is used here.

In this example, the slow batch-to-batch drift in the initial water content (W) together with a common-cause batch-to-batch correlation (ϕ=0.8), as shown in Figure 6.6 is also used to illustrate the effect that the inclusion of previous batch information ($\mathbf{Z}$ matrix, Figure 6.1) has on the on-line monitoring of new batches. The on-line $\mathbf{t}_1$ score plots from the MBPLS using prior batch information is shown in Fig. 6.8a for batches 1 (normal operation condition), 47 (out of 95% CI), and 50 (out of 99% CI). The on-line $\mathbf{t}_1$ score plot obtained from normal MPLS without use of prior batch information for the same batches is shown in Figure 6.8b (note the difference in scales from both figures).



(a) (b)

Figure 6.8 t_1 on-line monitoring for detection of drift with ϕ =0.8. (a) MBPLS with prior batch information. (b) normal MPLS.

From this Figure it is evident that with the incorporation of batch-to-batch information in the on-line monitoring scheme, smaller and more consistent confidence intervals are obtained. Detection of the abnormal situation caused by the slow drift in the inlet water concentration is also achieved faster. As shown in Figure 6.8a, by incorporating information on previous batches, the starting $\mathbf{t}_1$ score values for each successive batch slowly rises and by batch 47 to 50 it is evident that a fault is present almost from the first sample point. Without the information on prior batches to confirm this gradual batch-to-batch trend, the detection of the problem takes longer as seen in Figure 6.8b.

The importance of the previous batch information (**Z** matrix) can be scaled-up or scaled-down, allowing for increased sensitivity on batch-to-batch abnormalities. The effect of scaling-up the **Z** matrix is illustrated in Figure 6.9 for the same case as discussed in Figures 6.6, 6.7 and 6.8 (slow drift upset in the condensation polymerization system.) To allow for a direct comparison, normalization of the scores and confidence limits (CI) has been performed:

$$\mathbf{t}_1^* = \frac{\mathbf{t}_1}{99\%\,\mathrm{CI}}; \quad \mathrm{CI}^* = \frac{\mathrm{CI}}{99\%\,\mathrm{CI}}$$

Three cases are shown: (i) normal MPLS with no prior batch information (*); (ii) MBPLS including prior batch information, **Z**, (+) (unit variance scaling in the **Z** matrix, scaling factor (SF=1)) and (iii) MBPLS when the **Z** matrix is up-weighted by a factor of 2 (SF=2). Figure 6.9 also more clearly demonstrates the use of prior information (**Z**) in on-line monitoring. Note that, by the end of the batch, all methods end up giving approximately the same normalized score values and indicating an out of control situation. This is consistent with what was shown in the previous section on off-line analysis. However, with the use of prior batch information (**Z**) this slow batch-to-batch drift in the water content of the feed is detected much earlier in the batch. The prior batch information (**Z**) essentially gives the MBPLS a head start by giving initial score values close to the final score values of the previous batch, whereas, the regular MPLS approach has to start again from scratch and learn from the early data from each new batch.

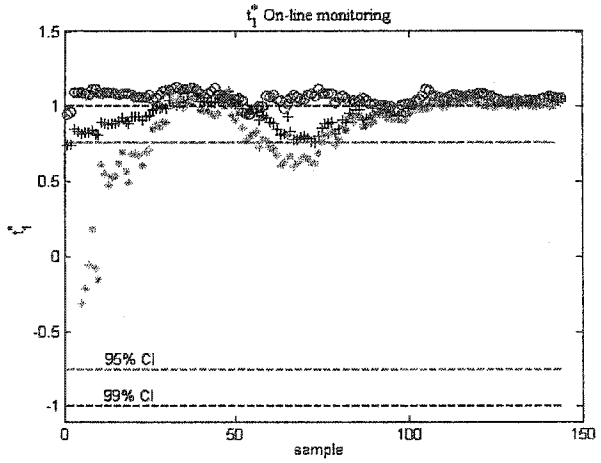


Figure 6.9 Normalized on-line score plots and the effect of scaling-up the Z matrix. Normalized t_1^* plot for batch 50 (ϕ=0.8). (*) normal MPLS, (+) MBPLS (unit variance scaling in the Z matrix, scaling factor (SF)=1), and (o) MPLS with SF=2 in the Z matrix.

Use of Contribution plots in assessing past batch information

In on-line monitoring of new batches, once an out of control signal is detected, contribution plots (MacGregor et al. [1994], Kourti and MacGregor [1995]) can be inspected to determine to what extent previous batches, ($t_1 \ldots t_r$ scores in the **Z** matrix) and

on-line measurements within the current batch (**x** data) are contributing to such a signal. The contribution plot for the t_1 score plot in Figure 6.9 at time interval 4 (99% control limit violated) for batch 50 (SF=2 in **Z** matrix) is shown in Figure 6.10. The contribution plot clearly shows that it is the variation in prior batch scores that are contributing to the alarm and not the temperature measurements (T_r) in the early part of the current batch.

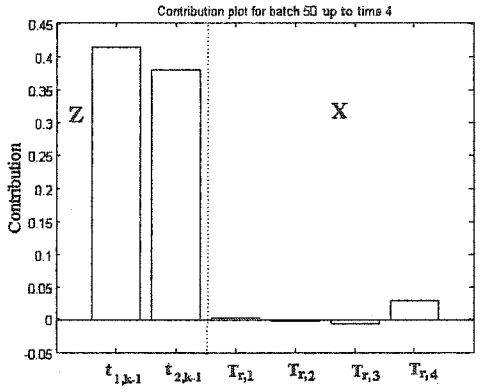


Figure 6.10 Contribution plot to component 1 up to time 4. Abnormal batch for slow drift case (batch 50).

6.3.5 Detection of a short-lived upset in a single batch

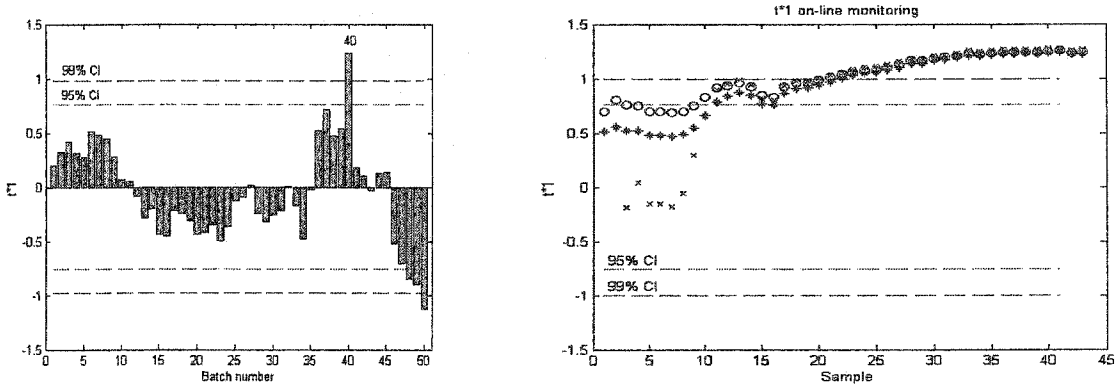
In this case, the objective is to show that for a short-lived upset in the current batch (affecting the end-quality properties of that batch), the use of batch-to-batch information will not add any benefit to an on-line monitoring scheme or off-line analysis. A Monte Carlo simulation study was performed, in which 50 different testing datasets with correlation ϕ=0.9 were generated for the emulsion polymerization system. In every new testing dataset a short-lived upset (arising from surfactant variations affecting a_s) was introduced at batch 40. Results from the Monte Carlo simulation are shown in Table 6.4. The average times to detection of the upset indicated by an out of control signal in the t_1 score (at α=0.05 and 0.01 probability limits) are seen to be even a little slower by including batch-to-batch information than when this is not included.

Table 6.4 Monte Carlo simulation results for short-lived upset.

(The numbers indicate the average time to detection of the fault.)

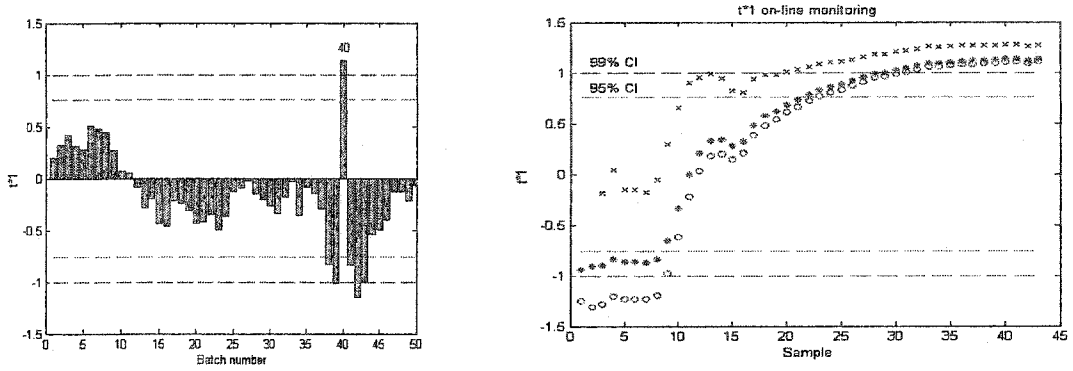
	MBPCA		MPCA	
CI	95%	99%	95%	99%
Average	13.8	23.24	9	18

From the Monte Carlo simulation study, two examples are shown to illustrate more clearly the effects of incorporation of previous batch information (**Z** matrix) into the model to detect this type of faults. In Figure 6.11a is shown the off-line $\mathbf{t}_1^*$ monitoring for a realization of the Monte Carlo study, while in Figure 6.11b is shown its normalized on-line $\mathbf{t}_1^*$ score for batch 40 from (x) normal MPCA, (*) MPCA with prior batch information (unit variance scaling, SF=1) and (o) when the **Z** matrix has been up-weighted (SF=2). In Figure 6.12 is shown the off-line $\mathbf{t}_1^*$ for *another* realization of the Monte Carlo study together with their normalized on-line $\mathbf{t}_1^*$ score for batch 40.



(a) (b)

Figure 6.11 Detection of short-lived upset with ϕ=0.9. (a) off-line $\mathbf{t}_1^*$ monitoring. (b) $\mathbf{t}_1^*$ on-line monitoring for batch 40. (x) MPCA, (*) multi-block MPCA (unit variance scaling in the Z matrix, scaling factor (SF)=1), and (o) MPCA with SF=2.



(a) (b)

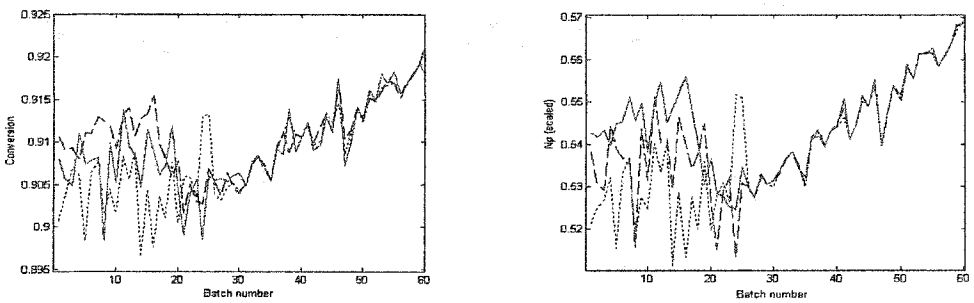
Figure 6.12 Detection of short-lived upset with ϕ=0.9. (a) off-line t_1^* monitoring. (b) t_1^* on-line monitoring for batch 40. (x) MPCA, (*) multi-block MPCA (unit variance scaling in the Z matrix, scaling factor (SF)=1), and (o) MPCA with SF=2.

From the above Figures, it can be seen that even in cases in which the disturbance, at batch 40, have the same direction as that of previous batches and these batches follows a trend, the benefit of incorporation of previous batch information is marginal (Figure 6.11). Moreover, if the disturbance breaks the trend, the detection will be much slower (Figure 6.12). Therefore, the use of batch-to-batch information will not add any benefit to an on-line monitoring scheme or off-line analysis for the detection of this type of upset.

6.3.6 Application of previous batch information in inferential prediction

In emulsion polymerization processes, on-line measurement of the quality properties (for example, Np, Dp, particle size distribution, MW, MWD, conversion) is frequently not available. Moreover, in certain situations (for example input saturation in cooling) the on-line measurements (T_j, T_r) may not be enough to capture the effect that disturbances may have on the end quality properties, limiting the prediction and control performance. In this section, it is shown how with the incorporation of previous batch information inferential prediction of end quality properties can be improved. (In this example a different recipe and conditions to those used in the examples presented before is employed. This is done to promote input saturation on T_j, which will lead to too few

information to infer the end-qualities from only on-line measurements). Three different historical data-bases, with different degree of batch-to-batch correlation, were generated: strong (ϕ=0.9), medium (ϕ=0.5) and weak (ϕ=0.2). Three testing data sets consisting of 60 batches were also generated: the first 25 batches keep the correlation from their corresponding training set, but at batch 26 a drift is induced as shown in Figure 6.13. Four different PLS models were built considering different availability of i) on-line T_j and T_r measurements, and ii) Dp and C on-line analysis made at 20min. These four models are shown in Table 6.5. In this Table is shown the SPE_y of the *end* quality properties for each one of the models and for each one of the correlations for all the batches composing the *testing* data set. It can be seen that the smallest SPE_y is obtained by models that use an on-line end quality related measurement (Dp, C). These models are model 1 (MPLS ([T_j T_r Dp C] **Y**)) and model 2 (multi-block MPLS ([**Z** T_j T_r Dp C] **Y**)). However, these models are frequently not possible to be obtained due to the requirement of the Dp and C on-line measurements. Model 3 (PLS ([T_r T_j] **Y**)) represents the most common situation in which only exits available temperature measurements. However, this model has the largest SPE_y indicating that an inferential control scheme based on such a model would be poor. Model 4 (proposed approach) only uses for the prediction of the end quality properties T_j and T_r measurements but with the inclusion of previous batch information ([**Z** T_j T_r] **Y**). This model obtains better prediction that the one in which previous batch information is discarded (model 3). In Table 6.5, it can also be seen that as the degree of batch correlation is smaller, the usefulness the **Z** matrix is lower. This corroborates the conclusions drawn in previous sections.



(a) (b)

Figure 6.13 Testing data set with drift. (a) Conversion, (b) Np. (-) ϕ =0.9, (--) ϕ =0.5 and (...) ϕ =0.2.

Table 6.5 Effect of the incorporation of previous batch information for different degrees of correlation

		Strong (ϕ =0.9)		Medium (ϕ =0.5)		Weak (ϕ =0.2)	
Models	Measurements	SPE C	SPE Np	SPE C	SPE Np	SPE C	SPE Np
1) MPLS	T_j,T_r,Dp,C	0.00163	0.00387	0.00225	0.00504	0.00211	0.00504
2) Multi-block MPLS	t_1-t_2,T_j, T_r,Dp,C	0.00142	0.00314	0.00215	0.00482	0.00207	0.00491
3) MPLS	T_j,T_r	0.00364	0.00919	0.00357	0.00825	0.00346	0.00824
4) Multi-block MPLS	t_1-t_2, T_j,T_r	0.002744	0.00675	0.00335	0.00772	0.00340	0.00802

6.4 Summary and Conclusions

The use of information from previous batches has often been of use in the optimization of batch processes. These batch-to-batch control and optimization methods utilize the repetitive nature of batch processes to learn about the effects of past optimization moves and hence to achieve better operating trajectories. In these problems prior batch information is essential. However, benefits of using information from previous batches to aid in the monitoring of existing batch processes that are being operated about a fixed set of manipulated variable trajectories is less certain. This latter problem was addressed in this Chapter.

An explicit procedure is presented for the incorporation of information from prior batches into multivariate statistical process control schemes based on multi-way PCA/PLS for monitoring batch processes. The approach involves incorporating the scores values, summarizing the operation of immediately preceding batches, into the MSPC scheme for the current batch. At the model building stage, this is shown to require an iterative scheme to develop the necessary PCA/PLS models. These models can then be use in the same straightforward (non-iterative) manner, as regular MSPC approaches to monitor new batches.

Simulations on two batch polymerization systems are used to demonstrate the method and to illustrate its potential. It is shown that, for off-line analysis or monitoring at the end of each batch, incorporating prior batches using this method provides little advantage over the usual MPCA/MPLS methods based on only the current batch data. In on-line monitoring, by incorporating prior-batch information, smaller and more consistent control limits and scores for the early stages of the process and faster upset detection of abnormal process conditions may be achieved. However, since the past batch information is more important at early stages of the batch and generally vanishes as more on-line measurements are available, only those process that have limited information at the beginning of the batch would benefit much from this methodology. Moreover, for processes that are only weakly batch-to-batch correlated or that suffer from random batch-to-batch disturbances, no benefit should be expected.

Nomenclature

A = number of principal components (scores or latent variables)

d = disturbance

$\mathbf{E}$ = residual matrix from PLS

$\mathbf{E}^*$ = residual matrix from PCA

$\mathbf{F}$ = residual matrix from PLS

h = adjustable parameter (eq. 6.6)

n = number of batches

$\mathbf{P}$ = loading matrix from PLS

$\mathbf{p}$ = loading vector from PLS

r = number of past batches (lags)

$\mathbf{T}$ = score matrix from PLS

$\mathbf{t}$ = score vector from PCA/PLS

$\mathbf{t}^*$ = normalized score

$\mathbf{V}$= loading matrix from PCA

$\mathbf{X}$ = regressor matrix

$\mathbf{Y}$ = quality matrix

$\mathbf{y}$ = quality variables

$\mathbf{z}$ = vector of previous scores

$\mathbf{Z}$ = matrix containing previous scores

Greek symbols

ψ_i = cumulative Hotelling's T^2

σ = sample standard deviation

η = adjustable parameter (eq. 6.6)

ϕ = degree of batch-to-batch correlation

ξ = random error

Γ = score matrix from PCA

α = significance level

Index

a = latent variable index

k = batch index or total number of batches

Chapter 7

Summary and Conclusions

Large amounts of process data are usually available in industry. This type of data has been exploited, for example, in the analysis and monitoring of batch processes. However, its use in batch control has been addressed to a less extent. The general objective of this thesis was to investigate the use of process data and latent variable methods for quality control in batch and semi-batch processes. In particular several practical control strategies were proposed based on PLS models identified from historical data and a few designed experiments.

The following problems were addressed: *i*) on-line inferential control of a distributed end quality property, namely particle size distribution (PSD), *ii*) development of an inferential strategy that combines batch-to-batch control, on-line information and model parameter updating, *iii*) reduced space control of batch processes using full manipulated variable trajectories consistent with past operation, and *iv*) incorporation of prior batch-to-batch information for analysis and monitoring. In as follows, the work done in each area is summarized, the contributions outlined, some conclusions are drawn, and some future work is proposed.

In Chapter 3 the problem of disturbance rejection to control broad and bimodal PSDs in emulsion polymerization using mid course corrections (MCC) was addressed. Three strategies were presented: *i*) control of second mode of the distribution, *ii*) control of the full bimodal PSDs, and *iii*) control of relative distributions. Each one of these alternatives has advantages and disadvantages and selection of one over the other mainly depends on the availability of intermediate quality related measurements. The

manipulated variables were emulsifier shots at some mid-points during the reaction and the total reaction time. These control actions were selected considering the fast dynamics that the emulsifier has on the particle nucleation. Adjustments in the amount of emulsifier are applied only when the predicted properties falls outside a statistically defined "no-control" region. This region delimits the variability present in normal operation to perform compensation only when a significant disturbance affects the process. The adequacy of these strategies was demonstrated based on simulation studies on a detailed theoretical simulator. These studies show that very good control is obtained. Since the approach is based on the use of readily available measurements, easily developed empirical models, and only occasional control actions at a mid-point time, the approach can be attractive for industry.

Chapter 3 has extended previous mid-course correction strategies to the control of high dimensional end-quality properties in a non-trivial problem. To the knowledge of the author, this is the first study in which full PSDs are successfully controlled on-line in polymerization processes. The approach can also be easily used in cases that many distributed quality properties such as molecular weight distribution and/or multimodal PSDs need to be controlled as well.

Chapter 4 proposes a control strategy that combines within-batch information from process variable trajectories and information from prior batches. Information from past batches is used in two forms: *i*) for model parameter updating to overcome model error, changing process conditions and unknown disturbances and *ii*) to feed-forward past measurements into the controller for the current batch to exploit the repetitive nature of the batch operation and to develop a batch-to-batch control algorithm. In the first part of that Chapter, the flexibility and easy maintenance of empirical models is illustrated for the within-batch control of bimodal PSDs in emulsion polymerization. It is shown how an initially very poor model is improved using batch-to-batch adaptation while rejecting different types of disturbances. Manipulated variables are emulsifier injections at two time intervals, each one to control a particle distribution. In the second part, both batch-

to-batch and within-batch information is used for the PSD control. Batch-wise constant disturbances are rejected while improving the quality of the models using batch-to-batch model updating. Batch-to-batch control is employed for controlling the first generation distribution using an initial condition (emulsifier concentration) as manipulated variable, while the second distribution is controlled with the within-batch control scheme using an emulsifier shot at a mid-point during the batch. A set-point change in the shape of the particle size distribution was also studied. The desired distribution is achieved within a few batches when starting with information limited to a region around a completely different PSD. In all approaches very good control performance is achieved in a few batches in spite of the initial large model error and the limited amount of measurements. Monte Carlo studies were also carried on to evaluate the robustness of the methodology. Several other practical aspects as measurement noise and off-line analysis delays were also consider. The data requirements for the strategy are modest and the models are easily built, making the approach suitable for industrial processes.

Chapter 4 has extended the mid-course correction strategies presented in Chapter 3 by including multiple decision and correction points, batch-to-batch information in the controllers, and an adaptive Partial Least Squares (PLS) approach. It is shown how batch-to-batch control can be easily performed and how new PSDs can be achieved. Some previous approaches in the literature have achieved new desired PSDs, but by using computationally intensive strategies based on theoretical distributed parameter models.

Chapter 5 presents a novel inferential control strategy that allows a much finer characterization and smoother reconstruction of the full manipulated variable trajectories (MVTs) than those obtained using staircase parameterisations, and that reduces the complexity and number of identification experiments needed for model building. This is possible by formulating the control strategy in the reduced dimensional space of a latent variable model, and then reconstructing the MVTs for the remainder of the batch using the structure of the model. The novelty of the approach relies precisely in that the control and the model inversion stage is performed in the reduced dimensional space of a PLS

model rather than in the real space of the MVTs. The strategy consists of three parts: *i*) prediction of the product quality using an estimate of unknown future measurements obtained with any of the missing data algorithms available in literature. (Here several missing data algorithms were evaluated and most of them show adequate performance); *ii*) a control adjustment in the latent variable score space where the statistical dimensions of the end qualities and scores have to be taken into account, and *iii*) reconstruction of the MVTs by inverting a PLS model while respecting the already implemented control actions and the observed process measurements. The control strategy was illustrated for disturbance rejection and set-point change (product design) for the control of number average molecular weight and amine end concentration in a condensation polymerization system. For robustness in implementation, several aspects were also studied, such as control performance using small data sets, large levels of measurement noise and different number of latent variables than those selected by cross-validation. In all cases the methodology obtained adequate results. Preliminary results are also shown for an industrial emulsion polymerization process. In this example, although the MVTs obtained from the control algorithm could not be implemented, the shape and magnitude of the computed MVTs seemed to be very reasonable and consistent with observed trajectories for the nominal batches.

This Chapter is probably the most innovative chapter of the thesis. It is the first empirical model based control methodology that is able to readjust the entire MVTs at each control point throughout the batch. Furthermore, it provides a smooth and almost continuous reconstruction of these trajectories that is consistent with past plant operation. This by itself represents a large advantage compared to other approaches that assume, just for computation convenience, that such trajectories can be represented by a stair-case parameterization. This characterization might represent a radical departure from normal practice, with the implication that control schemes based on them cannot be implemented. Furthermore, the control strategy do not need large data sets to build the models and do not suffer from numerical problems that may arise when determining simultaneously the large number of highly correlated control actions. Further work in this area includes an

explicitly handling of hard constraints. It would also be interesting to investigate the application of the methodology to other systems; especially in robotics were it seems that, for adequate operation, smooth MVTs need to be implemented.

Chapter 6 explored the use of prior batch data in the analysis and on-line monitoring of batch processes. A modified MPCA/MPLS procedure to explicitly incorporate batch-to-batch trajectory information while retaining all the advantages and monitoring statistics of the traditional MPCA/MPLSA was proposed. The method involves the use of the final PCA or PLS scores values ($t_1, t_2 ... t_a$) from prior batches to summarize all variables and time histories of prior batches. Then, these scores are combined with the trajectory data for the current batches by again performing MPCA/MPLS. At the model building stage, the method needs iterative training to obtain the scores values. However, once the model is identified, the iterative scheme is not needed and monitoring, analysis and prediction can be performed in the same way as is done in normal MPCA and MPLS methods.

Multivariate control charts based on prior scores alone were shown to be sufficient for detecting changes in the autocorrelation structure of the batch-to-batch behavior (such as might arise from changing feed-stocks of raw materials.) The process data matrix alone was shown to be sufficient to analyze and monitor only the within batch changes (conventional MPCA and MPLS). Using both allowed for complete treatment of both batch-to-batch and within batch changes within the same MBPCA/MBPLS model. The approach was illustrated using two simulated polymerization systems, the condensation polymerization of nylon 6,6 and the emulsion polymerization of styrene. Several studies for the off-line analysis of completed batches and for the on-line monitoring of new batches were performed in both systems and compared with MPCA/MPLS. The results show that incorporation on previous batch information for on-line monitoring does provide a more consistent and faster detection of early disturbances (as long as these have a degree of batch-to-batch correlation). However, the advantage of inclusion of such information is often small and it vanishes in

later stages since most of the information is already contained in the on-line measurements taken from the current batch.

In summary, the thesis has presented several novel control and monitoring strategies for batch processes based on empirical PLS models that are easily obtained from a combination of existing data and a modest amount of experimentation. The strategies should all be easy to implement and maintain in an industrial environment.

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Appendix A

Effect of MV on PSDs and PSD measurement noise

The objective of this Appendix is twofolds: 1) to show that the effects of the manipulated variables (MV) over the particle size distribution (PSD), in the range under study, are almost linear and, 2) to show the levels of noise added to the intermediate PSD measurement.

Effect of MV on PSDs

For all the case studies shown in Chapter 3 and 4, the slight non-linearities (in the range studied) of the system were taken into account by using quadratic and interaction terms (when needed) in the manipulated variables as well as the square root transformation on the final PSDs. As example, in Figure A.1 is shown the effects that the emulsifier adjustment $u_{c,2}$ (emulsifier adjustment at 150min, Chapter 3 and 4) has on the central bin (number of particles at certain radius) of the *second* mode of the final PSD (square root transformation has been applied in this PSD), while in Figure A.2, it is shown the effects of $u_{c,0}$ (initial emulsifier concentration adjustment, Chapter 3) on the central bin of the *first* mode of the final PSD (remember that each mode of the final PSD is generated by a corresponding emulsifier shot ($u_{c,2}$ or $u_{c,0}$)). As can be seen in such Figures, the dependency of the controlled variables on the manipulated ones is almost linear. The slight non-linearity shown in Figure A.2 (and corroborated by correlated residuals in Figure A.3, dotted line) can be further lessened by including into the model

the quadratic term of the manipulated variable ($u_{c,0}^2$). By adding such term, the regression coefficient is closer to unity (R=0.9993) and the residuals become uncorrelated as can be corroborated in Figure A.3.

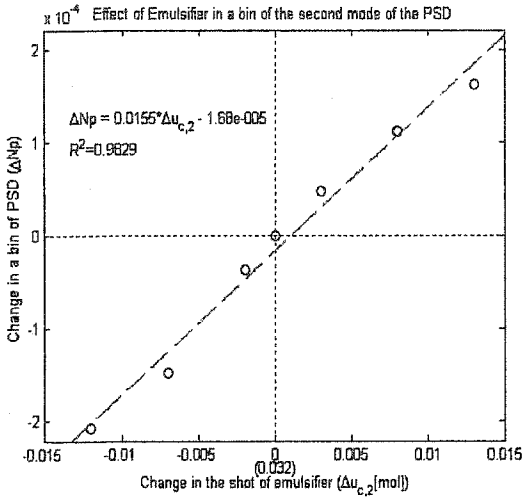

Figure A.1 Effects of $u_{c,2}$ in a bin (ΔNp) of the second mode of the PSD (square root transformation applied to the PSD).

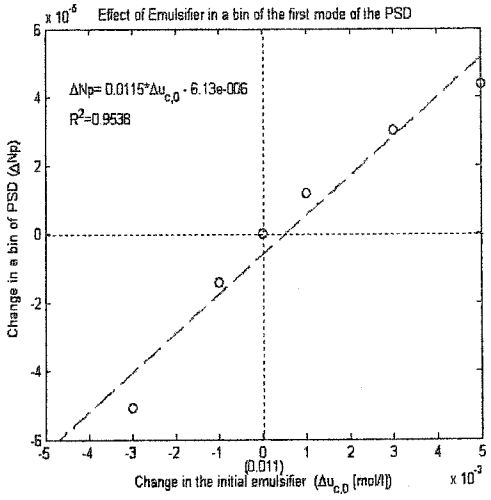

Figure A.2 Effects of $u_{c,0}$ in a bin (ΔNp) of the first mode of the PSD, (*no* quadratic term added in $u_{c,0}$ and square root transformation applied in the PSD)

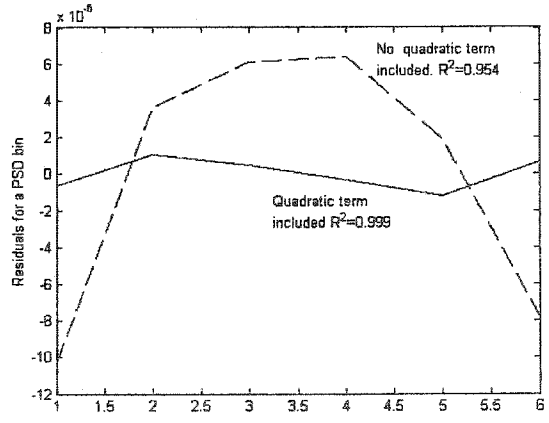

Figure A.3 Effect of the quadratic term ($u_{c,0}^2$) on the residuals for a PSD' bin. (--) no quadratic term added and (-) use of quadratic term ($u_{c,0}^2$).

PSD Measurement Noise

In practice, the PSD measurement is also affected by measurement noise. This noise generally has a correlated structure that depends on the instrument providing the measurements. Therefore, noise structure identification was performed through PCA as described in Clarke-Pringle and MacGregor [1998], on repeated laboratory PSD measurements on one styrene emulsion latex sample. From the identified structure, new correlated noise was generated and added to the intermediate PSD of the case studies shown in Chapter 3 and 4. Figure A.4 shows the intermediate PSD for a level of noise of σ=1% (Chapter 3 and 4) while in Figure A.5 σ=1.5% (Chapter 4).

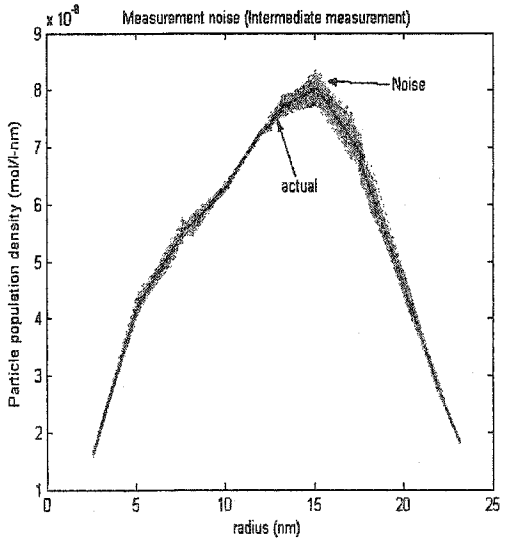

Figure A.4 Noise in the intermediate PSD (σ=1%). (—) actual measurement, (--) realization with noise.

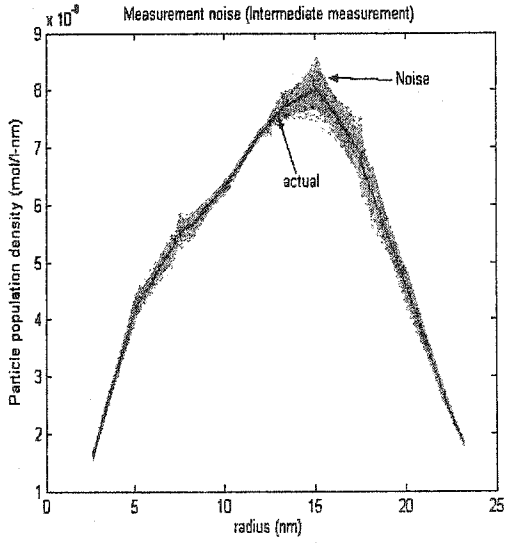

Figure A.5 Noise in the intermediate PSD (σ=1.5%). (—) actual measurement, (--) realization with noise.

Appendix B

Effect that Several Variables has on the Reduced Space Control Performance

The purpose of this Appendix is to detail the discussion given in Chapter 5 (section 5.3.1) on the effect that several variables has on the performance of the reduced space controller.

Effect of number of latent variables (LV)

To illustrate the effect that the number of LV has on control performance some results are presented in Table B.1. In this Table is shown the root mean squared error (RMSE, eq. (B.1)) for different number of components on the 15 batches (K) shown in Figure 5.4 (regulatory control). It can be observed that the RMSE is not largely increased by using a lower number of components to those obtained by cross-validation method (5 in this case). However, when using 6 and 7 components the error in MW increases considerably. This is because using a too large a number of components may promote an ill-conditioned $\mathbf{P}_2\mathbf{W}_2$ inversion at the second decision point. This problem can be easily overcome by using a pseudo-inverse procedure based on singular value decomposition (MATLAB function *pinv*). The results of using the pseudo-inverse are also shown in Table B.1, where it can be seen that by using this simple procedure the robustness of the algorithm is increased.

$$RMSE_i = \sqrt{\frac{\sum_{k=1}^{K}(y_{ik} - y_{sp,ik})^2}{K}} \qquad i = 1,2 \qquad \text{(B.1)}$$

where y_i are the qualities for the k-th batch obtained after control is performed.

Table B.1 Effect of number of LV in control performance: RMSE.

# Components	NH_2	MW	NH_2-pinv	MW-pinv
4	0.33	23.6	--	--
5*	0.34	15.6	--	--
6	0.49	141.1	0.45	17.6
7	0.43	60.9	0.43	30.0
8	0.64	9.2	0.40	23.7

*obtained by cross-validation

Effect of missing data algorithm

Different missing data algorithms were tested to see their effect on control performance. Table B.2 shows the RMSE for each one of the missing data algorithms in controlling the 15 batches shown in Figure 5.4 (regulatory control). From this Table, it can be noticed that all the algorithms give adequate control performance. Those based on EM, IMP and PTP perform slightly better than the one in which SCP was used.

Table B.2 Missing data algorithms on control performance: RMSE.

Algorithm	NH2	MW
EM	0.38	18.2
IMP	0.34	15.6
SCP	0.38	21.9
PTP-PLS	0.38	16.2

Effect of number of batches used in model building

In the previous examples a total of 45 observations were used as training set to built the PLS model used for prediction and control. In this section, it will be shown that adequate control performance (all test batches falling inside the in-control region shown in Figure 5.5) can be achieved using much less number of batches. To illustrate this, Table B.3 presents the RMSE for the 15 batches shown in Figure 5.5 (regulatory case study) when using a different number of batches to train the models. From this Table, it can be noticed that the control performance is very good using even only 15 batches. This illustrates that the data requirements for PLS model building are modest. However, if the model has been identified using very limited or uninformative batch data-sets (as those arising from only historical data), batch-to-batch model parameter updating can be performed at the end of each new completed batch to improve the quality of the model parameter estimates, prediction and control. An example is shown later in this Appendix.

Table B.3 Effect of number of batches on control performance: RMSE.

# of batches	NH2	MW
45	0.34	15.6
30	0.30	13.8
20	0.37	18.0
15	0.38	16.9

Effect of measurement noise

In order to determine the effect of noise on control performance, different levels of random noise were added to the on-line measurements of reactor temperature (Tr) and venting (v) as shown in Table B.4. It was found that adequate control performance (test batches falling inside the "in-control" region of Figure 5.5) is achieved with levels up to 35% in temperature and venting rate, respectively. The noise level here represents the percentage of the noise variance over the true variations of the temperature and venting

rate changes observed in the training set. 35% noise level approximately represents variations in temperature of ±2°K and venting rate of ±42g/s (1 standard deviation, Table B.4). For larger levels of noise (50%, for example) the control performance is slightly degraded because the random error added to the measurements becomes quite large when compared with the true variations in the MVs. This Table shows that even large levels of noise have little effect on the performance of the controller. To better illustrate the random noise level added to the temperature and venting measurements, Figures B.1a and B.1b are shown (50% of noise has been added). In these Figures (···) represents the measurements (temperature and venting respectively) used for model building and (—) the level of noise.

Table B.4 Effect of number of measurement noise on control performance: RMSE.

% noise level*	T_r (σ%)	Noise level T_r (°K)**	v (σ%)	Noise level v (g/s)**	NH_2 RMSE	MW RMSE
15	0.25	±1.3	8.3	±28.8	0.80	26.4
25	0.32	±1.6	10	±34.7	0.80	37.3
35	0.37	±1.9	12	±41.7	0.73	28.3
50	0.45	±2.3	15	±52.1	0.89	38.1

*respect to the variation in training set. **T_r=500°K and v=350 g/s used as base values.

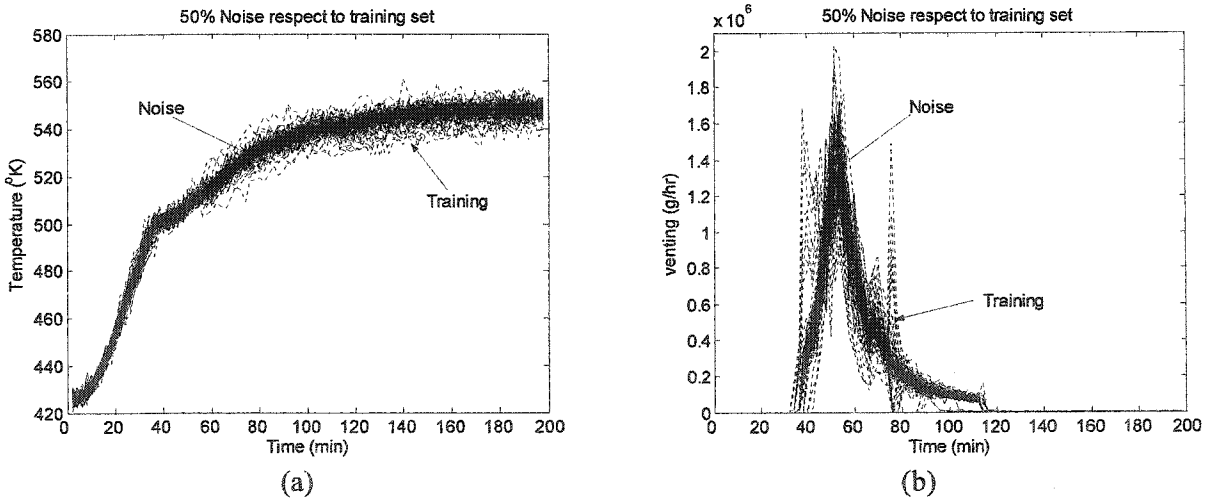

Figure B.1a-b Example of noise level added to process measurements (35% respect to training set). (a) temperature and (b) venting. (···) represents noise measurements used for model building and (—) the level of noise.

Extension to non-linear PLS models

The control methodology outlined in section 5.2 can be easily extended to cases in which a non-linear PLS model and control is needed. This is achieved by simply modify, for example, eq. (5.12) (case 3, dim ($\Delta \mathbf{t}$) > dim ($\mathbf{y}_{sp}$)) to take into account the non-linear nature of the PLS algorithm. In the case of a quadratic PLS model, eq. (5.12) can be restated as:

$$\begin{aligned} &\underbrace{\min}_{\Delta \mathbf{t}(\theta_i)} \Delta \mathbf{t}^T \Delta \mathbf{t} \\ st \quad &\mathbf{y}_{sp}{}^T = \mathbf{u}^T \mathbf{Q}^T \end{aligned} \tag{B.2}$$

where $\mathbf{u}^T = \beta_1 + \beta_2 (\Delta \mathbf{t} + \hat{\mathbf{t}}_{present})^T + \beta_3 (\Delta \mathbf{t}^T + \hat{\mathbf{t}}^T_{present})^2$. Once obtained the values of $\Delta \mathbf{t}$, the reconstruction of the MVT can be performed as described in section 5.2.4. As an illustration, some results using quadratic PLS are shown in Table B.5. In this Table it can be seen that the control performance of the quadratic PLS model (measured by RMSE on the 15 batches shown in Fig. 5.4) is similar to that obtained using the linear PLS model. This is not surprising because, in the region under study, the process is only slightly non-linear. However, if larger disturbances affect the process a non-linear PLS approach may be better suited.

Table B.5 Non-linear PLS modelling

	Linear PLS		Quadratic PLS	
	NH_2	MW	NH_2	MW
RMSE	0.34	15.6	0.36	14.9

Model Adaptation

As it is not always possible to identify a model from a data-set in which there exits enough information to determine the effect of disturbances and MVT on the end-

qualities, it is necessary to have a mechanism to improve gradually a poor model as more batches are being produced. One of the simplest ways to do this is by continuously update the model parameters at the end of each batch as shown in Chapter 4:

$$\mathbf{Y}^{(k-1)} = \mathbf{Y}_{tr}; \quad \mathbf{X}^{(k-1)} = \mathbf{X}_{tr}$$

for a new batch *k*:

$$\mathbf{Y}^{(k)} = \begin{bmatrix} \mathbf{Y}^{(k-1)} \\ \mathbf{y}^{(k)} \end{bmatrix}; \quad \mathbf{X}^{(k)} = \begin{bmatrix} \mathbf{X}^{(k-1)} \\ \mathbf{x}^{(k)} \end{bmatrix} \qquad \text{(B.3)}$$

$$\left[\mathbf{Y}^{(k)} \middle| \mathbf{X}^{(k)}\right] \xrightarrow{PLS} \mathbf{P}^{(k)}, \mathbf{W}^{(k)}, \mathbf{Q}^{(k)}$$

the updated model is:

$$\begin{aligned} \mathbf{X}^{(k)} &= \mathbf{T}\mathbf{P}^{(k)T} + \mathbf{E}^{(k)} \\ \mathbf{Y}^{(k)} &= \mathbf{T}\mathbf{Q}^{(k)T} + \mathbf{F}^{(k)} \end{aligned} \qquad \text{(B.4)}$$

where (tr) indicates the immediately previous training data set.

To show the performance of the adaptive controller algorithm, a subset of the data-set used in the previous example consisting of only 10 observations was used to train an initial model. In Figure B.2, it is shown *preliminary* results for the performance of the controller (eq. 5.21, δ=1, control action taken at 35 and 75min) when the system is affected by a constant batch-to-batch disturbance in the initial water content (W). In this Figure is shown (*) what happen if control action is not taken, (★) achieved qualities if no adaptation is performed (indicated with 1) and the end-qualities using the adaptive scheme (□). In Figures B.3a and B3.b, it is shown the effect of adaptation on the MVTs for batches 1 (no adaptation, original training data), 3 and 10.

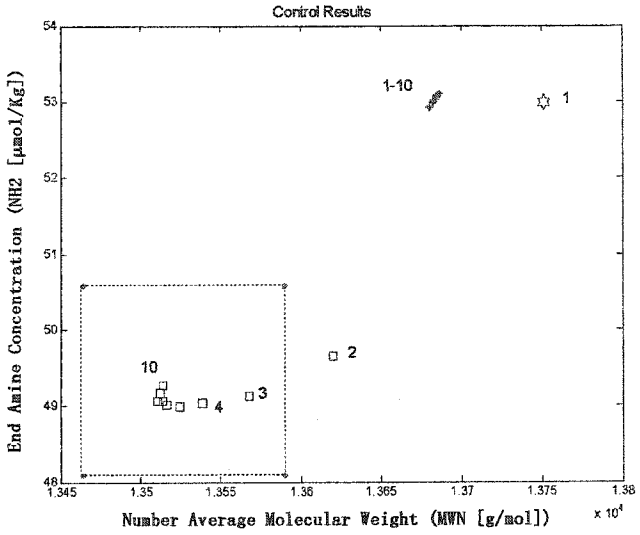

Figure B.2 Control results. (*) End-quality properties without control, (★) control without adaptation and (□) control with adaptation (equation 5.21).

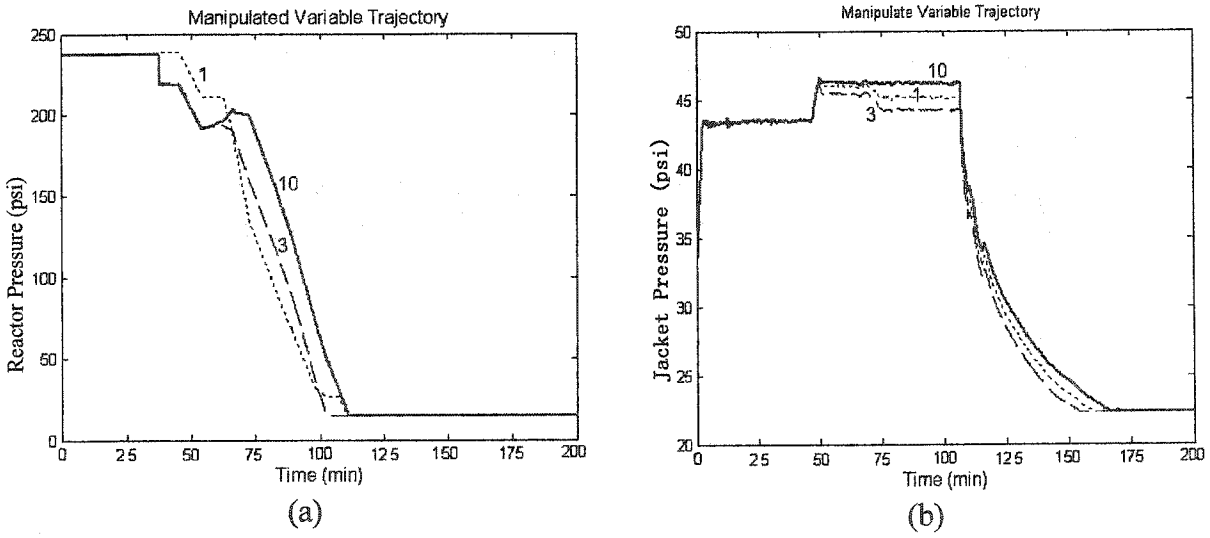

Figure B.3a-b Manipulated variable trajectories for batch 1, 3 and 10 when the process is affected by a batch-to-batch repetitive disturbance.

Constraint handling

In spite of that MV constraints are implicitly taken into account by the structure of the PLS model, it would be undoubtedly beneficial if the algorithm can explicitly take

them into account. In this last section, it is proposed a preliminary algorithm to explicitly handle hard constraints in the inputs. This algorithm still needs to be evaluated. Alternatively, the full optimization approach may be taken.

From eq. (5.21) the optimal adjustments to the manipulated variable trajectories (MVT) are obtained ($\mathbf{x}_2^T$). In the case of input saturation, $\mathbf{x}_2$ will be composed of $\mathbf{x}_2^T = [\mathbf{x}_2^* \ \mathbf{C}]^T$, where $\mathbf{x}_2^*$ are the part of the trajectories, computed from (5.21), that can be implemented, while $\mathbf{C}$ are those that would hit a constraint. In spite of hitting a constraint, we still wish to achieve the computed value in the score space $\mathbf{t}$ that will satisfy the overall PLS model. Therefore:

$$\mathbf{t}^{\mathrm{T}} = [\mathbf{x}_1 \ \mathbf{x}_2^* \ \mathbf{C}]^T \begin{bmatrix} \mathbf{W}_1 \\ \mathbf{W}_2^* \\ \mathbf{W}_c \end{bmatrix} = \mathbf{x}_1^{\mathrm{T}}\mathbf{W}_1 + \mathbf{x}_2^{*\mathrm{T}}\mathbf{W}_2^* + \mathbf{C}^{\mathrm{T}}\mathbf{W}_c \qquad \text{(B.5)}$$

then $$\mathbf{x}_2^{*\mathrm{T}}\mathbf{W}_2^* = \mathbf{t}^{\mathrm{T}} - \mathbf{x}_1^{\mathrm{T}}\mathbf{W}_1 - \mathbf{C}^{\mathrm{T}}\mathbf{W}_c \qquad \text{(B.6)}$$

Also we want to maintain the correlation in the PLS model:

$$\mathbf{x}_2^{*\mathrm{T}} = (\mathbf{t}^{\mathrm{T}} + \boldsymbol{\alpha}^{\mathrm{T}})\mathbf{P}_2^{*T} \qquad \text{(B.7)}$$

Substituting (B.7) in (B.6):

$$(\mathbf{t}^{\mathrm{T}} + \boldsymbol{\alpha}^{\mathrm{T}})\mathbf{P}_2^{*T}\mathbf{W}_2^* = \mathbf{t}^{\mathrm{T}} - \mathbf{x}_1^{\mathrm{T}}\mathbf{W}_1 - \mathbf{C}^{\mathrm{T}}\mathbf{W}_c^*$$

Therefore $$(\mathbf{t}^{\mathrm{T}} + \boldsymbol{\alpha}^{\mathrm{T}}) = (\mathbf{t}^{\mathrm{T}} - \mathbf{x}_1^{\mathrm{T}}\mathbf{W}_1 - \mathbf{C}^{\mathrm{T}}\mathbf{W}_c^*)(\mathbf{P}_2^{*\mathrm{T}}\mathbf{W}_2^*)^{-1} \qquad \text{(B.8)}$$

And by substituting (B.8) in (B.7) the new MVTs that can be implemented are obtained (i.e. those trajectories that try to compensate for the effect of the constraints):

$$\mathbf{x}_2^{*\mathrm{T}} = (\mathbf{t}^{\mathrm{T}} - \mathbf{x}_1^{\mathrm{T}}\mathbf{W}_1 - \mathbf{C}^{\mathrm{T}}\mathbf{W}_c^*)(\mathbf{P}_2^{*\mathrm{T}}\mathbf{W}_2^*)^{-1}\mathbf{P}_2^{*T} \qquad \text{(B.9)}$$