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Application of data mining techniques of batch profiles for process understanding and improvement

Mark Johnson

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To the Graduate Council:

I am submitting herewith a dissertation written by Mark Johnson entitled "Application of data mining techniques of batch profiles for process understanding and improvement." I have examined the final electronic copy of this dissertation for form and content and recommend that it be accepted in partial fulfillment of the requirements for the degree of Doctor of Philosophy, with a major in Chemical Engineering.

Charles F. Moore, Major Professor

We have read this dissertation and recommend its acceptance:

John Collier, Belle Upadhyaya, J. Weslet Hines, Duane Bruns

Accepted for the Council:

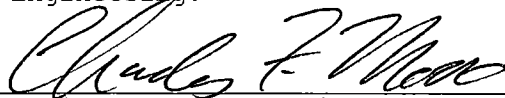
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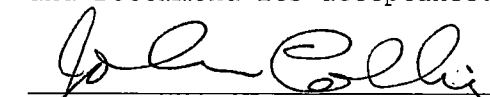
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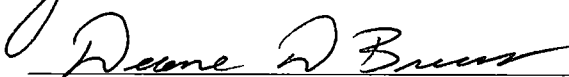
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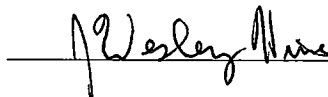

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Accepted for the Council:


Interim Vice Provost and
Dean of The Graduate School

**Application of Data Mining Techniques to Batch Profiles for
Process Understanding and Improvement**

A Dissertation Presented for
the Doctor of Philosophy Degree
The University of Tennessee, Knoxville

Mark Johnson

August 2001

Dedication

This work is dedicated first to my Lord and Savior, Jesus Christ, who has given me life. I would also like to dedicate this work to my beautiful wife Kimberly Anne, to our blessing of a son, Isaac Thomas, to my parents, Elmer Miller Johnson and Sue Bowlin Johnson, to my uncle, James E. Bowlin, to my aunt and uncle, Novella and Walmer Gardner, and to the rest of my extended family and friends who have been so supportive of me throughout my life.

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Soli Deo Gloria

Abstract

Batch processes are widely used in the chemical industry. Recently, much attention has been given to the monitoring and analysis of batch measurement data, or profiles, with an emphasis on the detection of problems. Similarly, methods to improve the final product quality in batch processes have multiplied in the literature. However, an area that is virtually unexplored is the utilization of the data mining techniques for monitoring and analysis of batch profiles for better understanding batch processes, rather than identifying problems in batches, in order to improve the process. The thrust of this work is to apply a systematic method to increase batch process understanding by sifting through the existing historical database of past batches, to discern directions for process improvement from the increased understanding, and to subsequently demonstrate better quality control through the use of online recipe adjustments.

A database of past batches is generated from a simulated nylon-6,6 process, with the main quality variable of interest being the number average molecular weight. The time and measurement variability in raw batch measurement profiles is characterized through scale parameters. These scale parameters are subjected to a standard principal component analysis (PCA) to understand the principal sources of variation present in a historical database of past batches. Directions for process improvement are discovered from the data mining study and appropriate manipulated variables to implement recipe adjustments are identified. Online predictions of the molecular weight are demonstrated which indicate off-target quality batches well before the end of the batch. A split-range linear molecular weight-based controller is developed that is able to reduce the variability in the quality around the target.

Further process improvement is accomplished by reducing the cycle time in addition to tightly controlling the final quality.

The approach for systematically analyzing batch process data is general and can be applied to any batch system, including non-reactive systems.

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Chapter 1

Scope of Work

Introduction

One of the primary objectives of batch manufacturing in the chemical industry is the consistent production of on-target quality batches. This is due to the premium on the quality of the value-added chemicals manufactured in batch processes. Online quality measurements are seldom available and the batch operation is typically based on a fixed recipe, with variables considered important, such as reactor temperature, being controlled to a pre-specified trajectory. In the absence of online quality measurements, a completed batch is characterized as either on-spec or poor quality based on laborious analytical measurements on a sample of the product.

Common cause variation, such as variability in the raw material quality, changes in equipment characteristics, etc., can have a significant impact on the final product quality and is not compensated for in the recipe-based operation. The principal sources of common cause variation can usually be inferred from the various online measurements, such as temperatures, pressures, flows, etc. Existing historical databases of the measurement trajectories (profiles) are a rich source of information on the sources of common cause variation affecting the process. The database can be systematically studied to understand these sources of variability and also the correlation of the measurement profiles with the final product quality. Historical data mining is thus a powerful tool that can be used to enhance process

understanding. The increased understanding can lead to process improvement through appropriate modifications for improved quality control. Process improvement is defined as making modifications to the process such that the quality more consistently reaches the target with lower variability in less time. In particular, strategies for online recipe adjustments can be proposed when good quality / disturbance predictions from the measurement profiles are obtained much before the completion of the batch. These adjustments compensate for the effect of the disturbances on the product quality so that tighter quality control is achieved. This is referred to as within batch control.

Objectives

The objectives of this work are as follows: 1. To utilize data mining tools in order to understand both the primary disturbances affecting the process and their effect(s) on the product quality, 2. To develop effective within batch control schemes to compensate for the primary disturbances and resulting in on-target quality batches with reduced quality variability, and 3. To reduce the batch production time in addition to tightly controlling the product quality. A batch polymerization reactor simulation is used to demonstrate these objectives.

Contributions

The contributions of this work are as follows: 1. Development of a methodology to systematically mine the historical database for increased understanding of the principal sources of common cause variability in batch processes, 2. Development of a systematic method for online predictions of a final quality variable, 3. Demonstration of

each methodology on the example batch process. For the example process, non-linear online predictions of the output quality and primary disturbances are demonstrated well before the end of the batch. An online recipe adjustment strategy is developed from the results of the data mining studies that improves the process by reducing the output variability in the quality. Reduced cycle time for the process with good quality control is also demonstrated.

Dissertation Layout

The dissertation is organized as follows. Chapter 2 gives a brief introduction to batch processes. Chapter 3 provides a survey of general batch processing literature, as well as giving an extensive review of batch monitoring and control literature. Chapter 4 describes the nylon-6,6 process simulation that is used to demonstrate process improvement through data mining. The historical profile database for fixed recipe operation is generated and described. The dynamic material and energy balances, batch operation recipe, available measurements and batch quality parameters are presented in the appendix.

In chapter 5, a systematic method is developed to examine the database of past batches for greater process understanding. In particular, existing multivariate methods are used to study and compare the correlation of the measurement profiles with the final product quality and to infer the primary disturbances affecting the process. Emphasis is placed on understanding the relation of the empirical parameters characterizing the profiles in a batch to the physics of the process. Offline predictions of the quality and primary disturbances are obtained using the methodology proposed by Kaistha [72] and incorporating non-linear terms in the regression.

In chapter 6, a methodology is developed for online predictions of the final quality. It is shown that good online predictions of the quality and primary disturbances are obtained much before the completion of the batch. Non-linear terms are used in the regression to develop the best predictions. These good online predictions open the possibility of within batch control. A within batch quality control strategy is proposed that is based on the final quality predictions. Simple proportional and non-linear algorithms for recipe adjustments are studied. The improvement in quality control over the recipe-based operation is quantified.

In addition to tight product quality control, a secondary objective in batch manufacturing is reduction in the batch production time representing increased yields. Chapter 7 broadens the scope of process improvement for the nylon-6,6 process to reducing the cycle time in addition to quality control. Appropriate within batch recipe adjustments are proposed for achieving both on-target product quality and reduced cycle time. The novel feature of the study is the manipulation of the batch end time in addition to the adjustments used in chapter 6. Results show that the cycle time can be significantly reduced while achieving tight product quality control.

The dissertation ends in chapter 8 with a brief summary of the work and the conclusions that can be drawn. Tentative suggestions for further research are also given.

Chapter 2

Batch Processes

Introduction

Properties of Batch Processes

In recent years, batch processing has received increased attention in the chemical industry. Batch processes are used for the manufacture of finite quantities of material, and are commonly used in the production of high value added chemicals, such as biochemicals, food products, pharmaceuticals, polymers, and specialty chemicals. Batch processes are also popular because of their flexibility in manufacturing. The competition from increased use of batch processes has generated a great deal of interest in improving batch operations in order to achieve consistent product quality, maximize production, increase the reliability of batch operations, and speed up process and product development. Improving batch operations depends on knowledge of current and past process operations. Current operations are monitored through field instruments that collect data about process conditions through the batch cycle. Monitoring these data is important for the purposes of within batch control, batch-to-batch control, problem detection, and problem diagnosis. Current process measurements are typically archived. These collected measurements form a rich historical database that can be mined for valuable insight for batch control and operation, problem detection and problem diagnosis, and process design and improvement.

Measurements and Process Profiles

Process measurements are important to batch processing for monitoring initial conditions (e.g., weights, volumes, raw material purities, etc.), process conditions (e.g., temperatures, pressures, flows, agitator speeds, etc.), and batch outcomes (e.g., compositions, yields, viscosities, densities, etc.). Unfortunately, product quality measurements are usually determined by laboratory analysis only at the end of the batch. Often, these analyses take time and are not available before a new batch starts. Although indirect, other batch data, if analyzed properly, can provide insight into the progress of a batch. Measurements during the batch (batch profiles) are an indication of variability, which affects batch operation. Variability arises from sources such as material balances, energy balances, kinetics, measurement uncertainty, process noise, equipment problems, etc. Some variability is to be expected and does not necessarily lead to poor operations, however, some sources of variability can affect quality, throughput, yield, etc. Analyzing and interpreting the time-related variability of profiles can provide significant insight. One of the problems in analyzing variability in a batch is separating variability which is inherent to the batch from variability which reflects unusual, or abnormal, behavior. Batch monitoring and analysis methods are useful for determining which profiles exhibit the inherent, or consistent, variability and which profiles exhibit abnormal, or inconsistent, variability.

Process Improvement

The various batch monitoring and analysis techniques have been very useful for determining when process problems occur in batches. However, very little or no research has been done to utilize those tools

to mine the historical database for the purpose of improving batch processes. The focus of this research is to utilize the batch monitoring and analysis techniques in order to extract cause and effect relationships leading to process improvement. Chapter 3 gives an extensive literature survey on general batch processing, batch monitoring and analysis tools, and batch process control research.

Chapter 3

Literature Review

Introduction

The literature review is divided into six broad areas: 1. Batch processing, 2. Engineering process control (EPC) and statistical process control (SPC), 3. projection methods, 4. Batch monitoring and control, 5. Time-alignment issues, and 6. Batch polymerization reactor simulation.

Batch Processing

Levels of Batch Control

This review emphasizes batch control and related topics. Batch control can be divided into roughly three levels. At the highest level of control is production planning and scheduling, which involves assignment of particular plants to meet production needs, as well as allocating plant resources to a particular batch unit. The next level of control is recipe management and implementation. The appropriate instructions regarding the ingredients, sequence of operations, and process conditions are implemented at the process. The bottom level is within batch and batch-to-batch control. Literature regarding these three levels of batch control mentioned is covered, as well as some other issues related to batch processing.

Batch Processes

A definition for a batch process can be found in Fisher [1, p.1], where Shaw is quoted as saying that "a process is considered to be batch in nature if, due to physical structuring of the process equipment or due to other factors, the process consists of a sequence of one or more steps (or phases) that must be performed in a defined order. [The] completion of this sequence of steps creates a finite quantity of finished product." Fisher gives an exhaustive treatment of the issues in batch control. Burton [2] gives historical perspective on the development of batch control since the early 1960's. Dean [3] summarizes the fundamental requirements of a batch control system as follows: 1. It "must be able to perform sequence and continuous control," 2. "It must provide a workstation interface for interaction with process designers, process and commissioning engineers, production managers, production operators, maintenance personnel, and plant managers, 3. "It must provide records of production, material usage, and a level of traceability of the production process of each batch for use by all the plant and production personnel, quality assurance department, and the accounting system." In the mid 1990's, batch processing standards were introduced by the Instrument Society of America's SP88 Batch Control Systems standards committee. Fisher [1, p.xiv] states that the purpose of the standards committee, which began working in October 1988, was to "provide standards and recommended practices, as appropriate, for the design and specification of batch control systems as used in the process industries." Bunch et al [4] summarize the major aspects of the SP88.01 standard and identify several important issues. Recommendations for better implementing the standard are given by Nelson et al in [5]. Various aspects of batch automation illuminated by the standard are given in [6-8]. Owen et al [9] discuss benefits of

implementing the standard in the bulk pharmaceutical industry. Schaefer [10] looks at the how the standard has impacted the design and implementation of batch control projects. The SP88.01 standard has been supplemented by two additional standards, the SP88.02 and SP95.01. Nowicki et al [11] note that the SP88.01 standard "focused on the models and terminology of batch control," but that the new SP88.02's purpose is "to standardize the electronic exchange of batch information." Furthermore, the SP95.01 standard was introduced for the purpose of integrating not just batch manufacturing, but also continuous processes into the larger business system. Nowicki gives a summary of the SP88.02 standard as well as reviewing the key concepts of SP95.01. Lau [12] discusses the advantages of integrating batch manufacturing into a systems approach to business.

Batch Scheduling

Reklaitis [13] gives a summary of the major issues involved in the scheduling control problem in batch processing. Reklaitis defines scheduling as "the decision procedures and processes under which resources are allocated to activities so as to achieve desired outcomes in a timely and / or cost effective fashion." Approaches to the scheduling problem can be grouped into five categories: 1. Rule based methods, 2. Search methods, 3. Artificial intelligence related methods, 4. Simulation methods, and 5. Model-based optimization. Reklaitis suggests that "batch process scheduling problems are most appropriately solved using model based optimization methodology, supplemented with simulation as a schedule disaggregation and validation tool." Several scheduling methods are covered in [14-18]. A scheduling method that takes into account the equipment failure uncertainty is given by Sanmarti et al in [19].

Batch Recipe Management

Wang et al [20] note that recipe management has not been given much attention and then proceed to give detailed treatment to the development of a recipe management system. Årpen et al [21] present how object-oriented sequential function charts can be used to implement a batch recipe management system. In recipe implementation, the reactor charging issues of target setting and alarm generation are given detailed treatment by Tsai et al in [22]. Another recipe implementation issue, cleaning of batch equipment in-place is covered in Sakmar et al [23].

Miscellaneous Issues

In other miscellaneous issues, Read [24] offers several suggestions to improve batch operations. Also in operational improvement, Srinivasan et al [25] cover an automated approach to perform a batch process Hazard and Operability (HAZOP) study. Allgor et al [26] offer a "systematic process development methodology" for performing optimal batch process development. Soroush et al [27-28] present a theoretical framework for the design and operation of batch reactors and demonstrate the framework in a case study. Troy et al [29] present a development environment for the development of software for programmable logic controllers (PLCs) in batch process control. Methodologies for batch process development, which takes into consideration the impact on the environment, are given in [30-31]. A new method for the batch process design, which considers heat integration, utility system design, and water minimization along with constraints on time, is given in [32]. The design of energy storage systems for batch processes is covered in [33]. At the within batch and

batch-to-batch control level, the literature of batch monitoring and control, which is a primary thrust of this work, will be covered in a subsequent section.

Engineering Process Control and Statistical Process Control

Engineering Process Control

The two bodies of literature that exist in process control can be sorted into the areas of either statistics or engineering systems. The term engineering process control (EPC) is used to distinguish the engineering systems literature from the statistics based methods. Marlin [34] provides an introduction to engineering process control. Marlin [34, p. 6] defines control as follows: "To maintain desired conditions in a physical system by adjusting selected variables in the system." An important type of control is feedback control, which Marlin [34, p. 6] defines as follows: "Feedback control makes use of an output of a system to influence an input to the same system." The inputs that are influenced by feedback are the manipulated variables (MV). A manipulated variable is used by an automatic control system to transfer variability from one part of the process to another. Ogunnaike and Ray [35] provide an advanced treatment of engineering process control.

Statistical Process Control

Oakland [36] provides an introduction to the concepts and applications of statistical process control (SPC). SPC is a process monitoring technique, which considers the statistics of measurements in order to determine if a process exhibits normal variation. Each product manufactured by a system shows variability. Oakland [36, p. 52] defines 'common' cause variability as the "complex interaction of 'random' ...

causes, each of which is slight, [and which cannot] be traced to a single cause." Excessive variation is referred to as 'special' cause variability. A process showing only common cause variability is said to be 'in control'. Oakland [36, p. 70] states that "To control a process using variable data, it is necessary to keep a check on the current state of the accuracy (central tendency) and precision (spread) of the distribution." Control charts are used to monitor the current state of the process. SPC that is applied to more than one variable at a time for the same system is called multivariate analysis. Tatsuoka [37] gives an exhaustive exposition of the methods of multivariate statistical analysis, which also incorporates methods of linear algebra that are covered later. Furthermore, SPC has the capability of giving process insight leading to process improvement.

SPC Literature

Vander Wiel et al [38] detail a new methodology, algorithmic statistical process control (ASPC), which is "an integrated approach to quality improvement ... that realizes quality gains through appropriate process adjustment (i.e., process control) and through elimination of root causes of variability signaled by statistical process monitors." Guh et al [39] apply a neural network model for the recognition of abnormal SPC charts. Saraiva et al [40] develop a methodology with the goal "not to detect and diagnose 'abnormal' situations, but rather to uncover improvement opportunities from the large amounts of 'normal' operation data that are collected from the process but often end up not being explored at all." Saraiva cites estimates that only 6-20 % of production problems arise from special causes with the remaining bulk of 80-94 % being due to common causes, which are addressed through process improvement. Reducing the common cause variability to improvement the

process is a main thrust of this work. Finally, Marsh and Tucker [41] have investigated the application of SPC techniques to batch units. However, as shown later, the authors make a major assumption about the time dependence that typically does not hold for batch processes.

Projection-based Methods

Linear Algebra Background

Strang [42] gives a treatment of the subject that covers not only the basics of linear algebra, but also provides a geometric interpretation of linear operations. The fundamental problem of linear algebra is the solution of the matrix equation,

$$\mathbf{Y} = \mathbf{X}\beta \quad (3.1)$$

Where: β = an m (rows) \times n (columns) transformation matrix
 \mathbf{X} = an $n \times p$ input matrix
 \mathbf{Y} = an $m \times p$ output matrix

This system of equations has an exact solution, β , only if \mathbf{Y} can be expressed as a combination of the columns of \mathbf{X} , given that the columns of \mathbf{X} are linearly independent. The columns of \mathbf{X} are linearly independent if no column can be expressed as a combination of the others. In this case, \mathbf{X} is said to be full rank. If \mathbf{X} is not full rank, then two other solution possibilities exist: 1. an underdetermined case in which infinitely many solutions exist, or 2. the overdetermined, or inconsistent, case in which no solution exists. The second case can effectively be dealt with using projection methods. When a system of equations is inconsistent, a "best" solution may still be found by minimizing an error criterion; e.g., a least squares regression of the data. In regression, the columns of the original \mathbf{Y} are projected onto the column space, or set of all possible exact

solutions, of the original \mathbf{X} . The solution closest to the original \mathbf{Y} in the column space of \mathbf{X} is the projected matrix $\underline{\mathbf{Y}}$. The appropriate $\underline{\beta}$ that finds $\underline{\mathbf{Y}}$ is expressed by the following equation:

$$\underline{\beta} = (\mathbf{X}^T \mathbf{X})^{-1} \mathbf{X}^T \mathbf{Y} \quad (3.2)$$

Where: $\mathbf{X}^T = \mathbf{X}$ transposed
 $\mathbf{X}^T \mathbf{X}$ = a matrix product
 $()^{-1}$ = the matrix inverse

The projected $\underline{\mathbf{Y}}$ is given by:

$$\underline{\mathbf{Y}} = \mathbf{X} \underline{\beta} \quad (3.3)$$

The distance, Q , from the original \mathbf{Y} to $\underline{\mathbf{Y}}$ is the perpendicular distance, or error, which is orthogonal to the column space of \mathbf{X} . Note that, in the special case that the columns of \mathbf{X} are orthogonal, the product $(\mathbf{X}^T \mathbf{X})^{-1}$ becomes the identity matrix, \mathbf{I} , and the regression coefficients are found by:

$$\underline{\beta} = \mathbf{X}^T \mathbf{Y} \quad (3.4)$$

The above method is used in multiple linear regression (MLR). Higher order terms may augment the \mathbf{X} matrix, if a non-linear regression is desired. For example, if quadratic terms are included, the input augmented matrix, \mathbf{X}_{aug} , can be written as:

$$\mathbf{X}_{aug} = [\mathbf{X} \ \mathbf{X}^2] \quad (3.5)$$

Where: \mathbf{X}^2 = Squared terms from \mathbf{X} matrix.

Linear terms may also be excluded for an exclusively non-linear regression. For example, in a second-order case, the input matrix, \mathbf{X}_2 , is written as:

$$\mathbf{X}_2 = \mathbf{X}^2 \quad (3.6)$$

Furthermore, the method of least squares is only one of many types of error minimization. A more general discussion of optimization, which also applies to neural networks, can be found in Chong and Zak [43].

Principal Component Analysis

Wold [44] describes principal component analysis (PCA) as the decomposition of a data matrix as follows:

$$\mathbf{X} = \mathbf{TP}^T \quad (3.7)$$

Where: \mathbf{X} = an $m \times n$ data matrix of row vectors
 \mathbf{T} = an $m \times n$ scores matrix ($= \mathbf{XP}$)
 \mathbf{P}^T = an $n \times n$ loadings, or principal directions, matrix

The \mathbf{T} matrix is a set column vector scores, or projection magnitudes, arising from the projection of \mathbf{X} onto the loadings. The loadings in \mathbf{P}^T are row vectors that are also called principal components (PCs). The first PC is in the direction of maximum variance, the second is in the second highest direction of variance, and so on, until the last PC, which is in the direction of the least variance. The loadings and scores are orthogonal; $\mathbf{t}_i^T \mathbf{t}_j = 0$, $i \neq j$, and $\mathbf{p}_i^T \mathbf{p}_j = 0$, $i \neq j$. In essence, when the decomposition is performed, the frame of reference has been rotated such that \mathbf{X} is represented by decorrelated variables, i.e., the scores. PCA is effective when the primary information about the data can be expressed using the first few PCs, with the remaining PCs representing the noise in the data. Thus, equation (3.7) could be better expressed as follows:

$$\mathbf{X} = \mathbf{T}_r \mathbf{P}_r^T + \mathbf{E} \quad (3.8)$$

Where: \mathbf{r} = the retained PCs
 \mathbf{E} = an $m \times n$ residual matrix representing noise

Data typically need some pre-processing, such as mean-centering and scaling, before PCA is done. The NIPALS algorithm for finding the PCs and scores is given in Wold [44]. Linear PCA is sufficient for the purposes of this work.

Strang [42] discusses a different method of doing PCA that is very closely related to the above. The method of singular value

decomposition (SVD) as described is used for calculating the loadings and scores. The singular value decomposition is expressed as follows:

$$\mathbf{X} = \mathbf{U} \Sigma \mathbf{V}^T \quad (3.9)$$

Where: \mathbf{U} = Left singular vectors
 Σ = Singular values
 \mathbf{V}^T = Right singular vectors

The right singular vectors are the principal components ($= \mathbf{P}^T$ from (3.7)). The scores matrix in (3.7) can be found as follows:

$$\mathbf{T} = \mathbf{U} \Sigma \quad (3.10)$$

Where: \mathbf{U} = Left singular vectors
 Σ = Singular values
 \mathbf{T} = Scores

Alternatively, the scores may be found by:

$$\mathbf{T} = \mathbf{X} \mathbf{V} \quad (3.11)$$

Where: \mathbf{V} = Right singular vectors
 \mathbf{X} = Original data
 \mathbf{T} = Scores

As in (3.8), PCA is effective when the data are highly collinear and only a few PCs need to be retained. This is expressed as follows:

$$\mathbf{X} = \mathbf{U}_r \Sigma_r \mathbf{V}_r^T + \mathbf{E} \quad (3.12)$$

Where: \mathbf{r} = the retained PCs
 \mathbf{E} = an $m \times n$ residual matrix representing noise

The singular value decomposition is related to the eigendecomposition [42]. The singular values are simply the square roots of the corresponding eigenvalues [42]. The singular values are arranged in descending order by most software and measure the variance explained by each respective loading. The first loading, or principal component (PC), describes the largest direction of variation and has the largest singular value, the second PC describes the second largest direction of variation and has the second largest singular value, etc. The scree plot is commonly used to visually express the amount of variance

explained by the singular values. The scree plot is shown in two plots as a cumulative sum of the squared singular values expressed as a percentage of the total variance and as the percentage variance explained for a given PC.

Further information on PCA is given in Jackson [45-47]. Wise et al [48] explore a theoretical basis for using PCA to monitor multivariate processes. Wachs et al [49] propose an improved PCA method for monitoring time-dependent relationships by recursively summing the last s PCA scores. Dong et al [50] extend PCA for non-linear analyses.

Partial Least Squares

While MLR explains the variance in the output data and PCA explains the variance in the input data, a third method, partial least squares (PLS), is used to explain the covariance between the input and output data. Essentially, the covariance between the input data matrix, \mathbf{X} , and the output data matrix, \mathbf{Y} , is maximized. A set of latent vectors, similar to principal components, are then available for predictions. The \mathbf{X} and \mathbf{Y} matrices are decomposed in the following form:

$$\mathbf{X} = \mathbf{TP}^T + \mathbf{E} \quad (3.13)$$

$$\mathbf{Y} = \mathbf{UQ}^T + \mathbf{F} \quad (3.14)$$

with an inner relation consisting of a linear regression between \mathbf{U} and \mathbf{T} :

$$\mathbf{U} = \mathbf{T}\beta \quad (3.15)$$

Where: \mathbf{X} = input data matrix
 \mathbf{T} = score matrix
 \mathbf{P} = latent vectors from \mathbf{X}
 \mathbf{E} = residual matrix
 \mathbf{U} = score matrix
 \mathbf{Q} = latent vectors from \mathbf{Y}
 \mathbf{F} = residual matrix
 β = regression coefficients

For a detailed discussion of the algorithm for performing PLS, as well as other background information, see Höskuldsson [51]. Qin et al [52]

extend the framework of PLS by the embedding of neural networks. Phatak et al [53] provide insight into the geometric interpretation of two common algorithms for performing PLS. Westerhuis et al [54] compare multiblock and hierarchical PCA and PLS models. Simoglou et al [55] present a comparison of PLS with a conceptually similar technique, canonical variates analysis (CVA), that is receiving increased interest in linear modeling.

Overview Literature

Wise et al [56] provide a technical review of several popular techniques in the field of chemometrics. Brown [57] gives an excellent qualitative overview of the state of the field in information and data handling techniques from a chemometrics point of view.

Batch Monitoring and Control

The major focus of this section will be the literature of batch monitoring with special emphasis given to the works of Nomikos and MacGregor [58-59], Darnell [71], and Kaistha [72]. Subsequently, within batch modeling and within batch and batch-to-batch control applications are reviewed.

Batch Monitoring using Projection Methods

Method of Nomikos and MacGregor

Nomikos and MacGregor [58-59] show that multiway PCA (MPCA), a technique from the field of image analysis, is appropriate for analyzing batch profiles in a multivariate sense. The MPCA method is performed by taking a 3-D array of batch data, which consists of multiple batches having multivariate measurements across time, and unfolding it into a 2-

D matrix that is subsequently analyzed by PCA. The form of the 2-D matrix is that the rows correspond to the batches and the columns are the measurements grouped together by time sample. For example, 20 batches and 5 measurements across a time of 300 minutes, when unfolded to a 2-D matrix, would have dimensions 20 x 1500, with every fifth column being the same variable at each time sample. The data are first mean-centered and the columns are scaled to unit variance, to eliminate the effects of differing engineering units. Since batch data variables are highly correlated, MPCA serves to identify the correlation structure as well as allowing data reduction. Only the first few principal components are kept in order to capture the major sources of variability in the batch. Two statistical measures are used to identify abnormal operation: the standard prediction error and the Hotelling's t^2 -statistic. The SPE is the remaining sum-of-squared error (SSE) after a new batch's trajectory has been projected onto the retained principal components. It provides a measure of the perpendicular distance of the projection from the hyperplane and indicates the lack of fit. The Hotelling's t^2 -statistic is a measure within the principal plane of how far away the projection is from the origin. Abnormal t^2 indicate a deviation that is too great, but not a different model regime. To use the technique for problem detection, a database of normal, or 'good', batches must first be assembled. From these batches, a PCA model is built to characterize the nominal correlation in the profiles. Standard statistical control limits are constructed in order to identify abnormal performance of new batches. Normal control limits can be constructed on each PC profile, on the Hotelling's t^2 -statistic, and the SPE. Ellipsoidal limits can be constructed on the PC scores to identify abnormal batches. An on-line framework is proposed which assumes that future deviations in the new batch trajectories will remain at their

current values for the duration of the batch. The problem with the method of MPCA is that time-axis variability is not considered. Furthermore, as presented, the method is designed to detect special cause variation in batches, but does not address the detection of sources of common cause variation in batches for the purpose of process understanding.

Literature of Batch Monitoring

Nomikos [60] and Martin et al [61] give concise overviews of the MPCA technique. Martin et al [62] present an alternative, called the M^2 statistic, to the Hotelling's t -statistic to construct control limits whose form and shape are dictated by the data. Martin et al [63] review several issues and concerns in batch monitoring. An inverse PLS (IPLS) model is reviewed that generates a large number of nominal trajectories from a small number of complete batches such that a monitoring scheme can be adequately tested. Furthermore, a multi-group PCA model, which encompasses a range of product grades and recipes, is discussed. Dong et al [64] extend the MPCA technique by using non-linear PCA. Boque et al [65] explore using multiway covariates regression models to predict final product quality from process variable information. All these works utilize data mining tools to detect the special cause variation in batch processes, but say nothing about understanding the sources of common cause variation in those batches. Furthermore, none of the above consider variation along the time-axis.

Neogi et al [66] apply multivariate statistical methods to a real industrial process and use the reaction extent to align the batches in time. The difficulty with the time-scaling method used is that, for most industrial processes, the reaction extent is not available. The time-scaling information is not incorporated in the predictions of

product quality. Neogi et al to some extent utilize data mining tools to gain process insight, for improving the process under consideration. No details are given on how the process was improved. Gallagher et al [67] also provide an example of the application of MPCA to nuclear storage tank monitoring. Finally, several applications of multivariate statistical analysis are given in [68-70]. With the exception of Neogi, none of the applications address the time-scaling problem. Again it is emphasized that most of these works apply multivariate SPC only for the detection of special causes, not for determining the sources of common cause variation.

The following works by Darnell [71] and Kaistha [72] include insight into a problem that much of the research in the area of batch monitoring has in the past ignored: the importance of scaling the time axis first. The concept of time alignment is introduced and is further discussed in the time alignment section.

Method of Darnell

Darnell [71] develops a technique for analyzing batch profiles that quantifies the variability of batch profiles and allows an increased sensitivity SPC analysis. Darnell shows that batch profiles can be thought of in terms of a scaled reference profile. Three scaling parameters and the residual SSE are used to characterize the behavior of batch profiles. The initial condition (IC) and magnitude scale (MS), quantify measurement axis behavior, and are equivalent to the slope and intercept terms in a regression. The time scale (TS) measures the time axis behavior of the profile. The TS is extracted first in order to align batch profiles in time. The profiles in the study were linearly interpolated to a common length and the difference in the number of points gave a crude TS parameter. The IC and MS are subsequently

extracted by projecting the reference profile onto a time-scaled profile. The deviation between the reference and the projection is then calculated, squared, and summed to get the SSE. Statistical limits are then constructed for each of the parameters as well as for the residual error in order to detect process abnormalities.

SPC Framework of Kaistha

Kaistha [72] develops a general framework for characterizing batch profiles. The framework is also detailed in Moore et al [73]. The method was primarily developed from proprietary data from industry. Kaistha covers three important areas: 1. Understanding the nature of variability, 2. Time scaling, and 3. problem detection using projection methods.

Partitioning Variability

Kaistha provides a refinement in understanding the different types of variability that occur in batch profiles. Variability can be partitioned into two types: consistent and inconsistent. The consistent variability repeatedly occurs from batch-to-batch in a particular way. The inconsistent variability arises due to process noise and special causes. Furthermore, the consistent variability can be partitioned into two types, measurement-axis and time-axis.

Time Alignment Methods

Batch profiles exhibit time-axis variability that projection-based methods do not properly characterize. The batches must first be aligned in time before any subsequent projection-based method is applied. Kaistha considers three types of time scaling to perform the time alignment: 1. Linear interpolation, 2. Dynamic time warping, and 3.

Shapes and features. Linear interpolation is the simplest method, but sometimes creates spurious features as an after effect of scaling.

Dynamic time warping (DTW) is a technique from speech recognition that is investigated as a possible technique for aligning batch profiles. Essentially, DTW calculates a non-linear mapping, or warping, of either one profile onto another or two profiles to a fixed time interval. In the mapping the optimum path is calculated subject to local and global constraints on the path. At present, the computation time to do DTW is excessively lengthy, but should improve with increasing processor speeds and parallel computing methods. Another drawback to the method is the introduction of yet another profile, the warping function, to analyze. The warping function is a historical record of the steps taken to minimize the distance between two profiles. These steps are subject to certain constraints in the DTW algorithm. DTW works best whenever an indicator variable, e.g., reaction conversion, is available, but such variables are rare in industry. DTW does show promise, though, in that it does a good job of mapping a profile onto another profile.

Kaistha develops the third technique, shapes and features, to automatically time scale batches by extracting the consistently occurring features in a profile. Each shape is translated and projected and a normalized minimum SSE is calculated. After each shape is tried, the minimum SSE for all shapes is used to locate the event time of the feature. This method has a computational advantage over DTW.

Problem Detection

Kaistha covers the area of problem detection using projection-based methods. Kaistha notes that PCA is a scaling dependent technique. The correlation structure in batch data changes from phase to phase, and so PCA is unable to explain the variability across an entire batch. Furthermore, the resultant PCA loadings are difficult to interpret for physical insight into the batch. Kaistha develops the method of moving window PCA (MWPCA). MWPCA explains the process variability via PCA loadings of time dependent regions of the data. These loadings, or factors, develop according to the amount of variance explained by the first principal loading within an extending time window. Whenever the variability explained by the current factor decreases below a certain threshold in the window, a new source of variability is identified. The old factor is stored, and a new factor is calculated. Because the factors explain zones of variability, they can be interpreted more easily; e.g., each factor may correspond to a particular physical phenomenon in a phase of a batch.

Within Batch and Batch-to-batch Control

Within Batch Modeling and Control

Kresta et al [74] develop a method for inferential process models based on PLS for continuous processes. Although not for batch processes, the principles in this paper can be extended to inferential monitoring in batches. Care must be taken to consider the possibility of time-axis variability. Shen et al [75] use a first order model to analyze industrial batch data in order to see the change in the dynamics both within a batch and from batch-to-batch. The goal of the study is to improve the process by understanding the process dynamics. The process

is modeled piecewise for the various phases, but no consideration to time-variability within a particular phase is given. Also, conducting the various step tests of manipulated variables for understanding the process is not typically acceptable in most industrial settings. Espuña et al [76] use neural networks and genetic algorithms on snapshot data to model the performance of an industrial process. The difficulty with the method is that no process understanding can be developed from the black box model in order to further improve the process.

Palanki et al [77] apply optimal control principles and neural networks to optimize batch manipulated variables on-line. Ni et al [78] use a fuzzy logic supervised neural network combined with a simple PI controller to perform temperature control on an exothermic batch reactor simulation. Martinez et al [79] introduce the concept of a performance function that includes in its definition end-product quality constraints and operational preferences. In each of these approaches, future process improvement cannot be done due to the black box nature of the neural network. None of the methods increases process understanding for the purpose of process improvement. Soroush et al [80] use a global linearizing controller (GLC) to control a non-linear polymerization reactor. The main difficulty with the method is that it is dependent on a detailed first principles model, which is not available for most industrial processes.

Robertson et al [81] use historical profiles to supply the setpoints for profile tracking controllers. However, this method is found to be "insufficient to guarantee consistent product when the process conditions change." Russell et al [82] implement a Kalman filter with principal component regression (PCR) and PLS to predict quality outcomes on-line and use this information as feedback to a model predictive controller. The difficulty with the practical implementation

of the method is that numerous control moves were simulated along with typical disturbances in order to develop the model. No consideration is given to time-axis variability in the process. In [83], Russell et al develop a scheduled proportional-integral-derivative (SPID) technique which combines model-based inferential monitoring and control with parameterization of normal setpoint trajectories. The problem with the method is that the parameterization of the normal setpoint profiles is not typically allowed in an industrial setting. Again, time-scaling is not considered. Process improvement deriving from better process understanding is not emphasized.

Joseph and Hanratty [84] develop a shrinking horizon model predictive control for control of an autoclave process using a neural network approach. The model is applied to a process with no time-axis variability, and so does not consider such changes in general. Process measurement trajectories are also ignored due to the nature of the process under consideration. Process improvement possibilities through improved process understanding are limited due to the black box nature of the neural net model. The model does incorporate batch-to-batch learning. Joseph, Tsen, et al [85] modify the approach in [84] by generating training data from actual experimental measurements through weighted estimates of unknown experimental points. The augmented dataset is then used to train the neural network. A mid-course correction policy is proposed for recipe adjustment. The method has the potential to become complicated very quickly as only one primary disturbance and one control action is considered. Furthermore, process improvement through increased process understanding is discouraged due to the nature of the neural net.

Kozub et al [86] develop an inferential feedback control scheme for the control of a semi-batch free radical polymerization process.

The approach depends on the availability of a detailed first-principles model. Time-axis variability is not considered. Yabuki et al [87] consider practical approaches for final quality control in an industrial process. The first approach is to highly automate the various stages of the process to introduce as little variation as possible into the batch. However, unmeasured process disturbances are not considered in this scheme. The second approach deals with such disturbances by using a mid-course corrective action based on predictors that are developed from a first-principles model. The drawback is that such models are not typically available. Clarke-Pringle et al [88] proposes a nonlinear adaptive controller based on general energy balances coupled with an extended Kalman filter to control a semi-batch polymerization reactor. Although the nonlinear adaptive controller performs well in this application, fairly specific knowledge of the process is required for the heat transfer model. The approach did not utilize past data for process understanding in order to improve the process; rather, a specific controller was implemented in order to obtain tighter control.

Yabuki et al [89] control product quality in a semi-batch reactor by predicting the quality and, if necessary, using a mid-course correction policy to bring the quality closer to target. If the predicted quality is off-target, then a shot of either initiator or inhibitor is injected into the reactor during the batch to bring the quality closer to target. Predictive models are developed from both a theoretical model and from empirical regressions. Since theoretical models are not always available, the authors present the data-driven approach as an alternative. The mid-course corrective policy is a reasonable strategy to use because the natural course of the batch is not altered unless a problem occurs. The simple control move proves to be able to bring the quality closer to target when disturbances are

present. However, the method is demonstrated on a process with no time-axis variability, so potential problems arising from time synchronization issues are not discussed. Also, the predictive model depends on an intermediate measurement of the product quality, which is seldom available in industry. Furthermore, the control law came from the assuming that a database of batches is available in which the effects of control actions on the product quality are known a priori. This paper demonstrates a specific application of a control method rather than systematically mining the existing historical database of past batches to increase understanding of the process and leading to process improvement.

Batch-to-batch Control

The method of Vander Wiel et al [38] is applied to a batch polymerization process and removes batch-to-batch variability leaving the stochastic within batch variability. However, the method assumes that quality measurements are available before the next batch begins. Lee et al [90] develop a model based controller referred to as batch model predictive control (BMPC) for quality control. The method applies iterative learning from past batches for better profile tracking. However, the method applies to a fixed trajectory, and thus does not allow for time-axis variability. Chin et al [91] and Chae et al [92] expand on BMPC by incorporating quality into the framework. The unified framework for batch process control is referred to as quality batch model predictive control (QBMPC). However, as with BMPC, QBMPC assumes that no time axis variability occurs. Furthermore, the tuning is complicated as six separate tuning parameters must be set. Kaistha et al [93] demonstrated improved quality from batch to batch on both a simulated and industrial process by utilizing time synchronization for

quality predictions and control. However, the method assumes that the quality outcomes are known before the end of the batch.

Time Alignment Issues

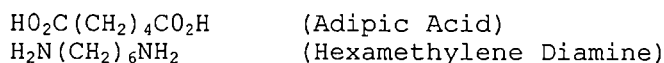
Time Alignment Literature

Methods in the literature, such as MPCA and MPLS, have the assumption that batches are of equal duration and are synchronized. This is not the case almost all of the time in real processes. Such things as varying levels of impurities in the initial raw materials, variation in the charge of raw materials, or heat transfer variation due to seasonal changes all contribute to batch profiles that are not aligned in time. Several works already mentioned, by Darnell [71] and Kaistha [72], address this problem of scaling the time axis. A description of Kaistha's method and its application for quality predictions is given in [93-94]. Martin et al [63] give a brief discussion on the practical limitations of applying MPCA to batches of unequal duration. Kassidas et al [95] provide extensive details about the DTW algorithm and apply the method to multivariate industrial batch data with good results. To handle the multivariate case, a weight matrix is calculated and used to give heavier weighting to those variables that are more consistent from batch-to-batch, and, thus, provides a better warping. In the case study, the data has the advantage of having a smooth and strictly monotonic variable upon which 85 % of the weight was given in the weight matrix. The problem with this study is that indicator variables (e.g., conversion) are not common in industrial data. A second criticism of the method is to raise the question of how the method performs with multivariate data in the absence of a smooth, monotonic indicator variable. Thirdly, Kassidas does not provide any

analysis of the warping function to characterize the variability along the time-axis. However, Darnell and Kaistha have shown that characterizing the time-axis variability is not only important, but may actually be the most important source of process variability. Nevertheless, DTW appears to be emerging as a useful technique in the batch synchronization problem. Further technical details of DTW are given in [96-98].

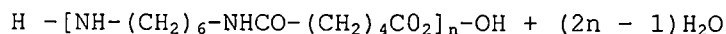
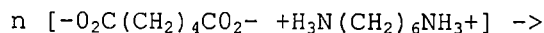
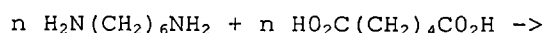
Batch Polymerization Reactor Simulation

Fogler [99] provides a general introduction to reactor engineering. Biesenberger et al [100] and Gupta et al [101] introduce polymerization engineering and step growth polymerization, respectively. The nylon-6,6 polymerization process is chosen to demonstrate the methodology presented in this work. Nylon 6,6 is a common name for poly(hexamethylene adipamide), or poly(iminohexamethyleneiminoadipoyl) [102]. The chemical formulas for the two main components of the nylon-6,6 reaction, adipic acid and hexamethylene diamine, are given as:



Where: H = Hydrogen atom
 O = Oxygen atom
 N = Nitrogen atom
 $(\text{CH}_2)_x$ = Chain of CH_2 groups of length x

The main reaction is written as [102]:



Where: n = Number of monomers present in solution
 $[-\text{O}_2\text{C}(\text{CH}_2)_4\text{CO}_2^- + \text{H}_3\text{N}(\text{CH}_2)_6\text{NH}_3^+] =$ Ammonium salt charged to reactor
 $\text{H} - [\text{NH} - (\text{CH}_2)_6 - \text{NHCO} - (\text{CH}_2)_4\text{CO}_2] - \text{OH} =$ Nylon-6,6 polymer
 $\text{H}_2\text{O} =$ Water

Steppan et al [103-105] provide a kinetic model of the nylon-6,6 process. Odian [102] and Jacobs et al [106] discuss the industrial preparation of nylon-6,6. Robertson et al [81] and Russell et al [83] develop a first principles model of a nylon-6,6 batch process from the work of Steppan et al. The work of Russell et al will be simulated in order to generate the batch data for this research. The reactor simulation will be described in chapter 4.

Summary of Batch Monitoring and Control

Batch monitoring and analysis methods have concentrated on the detection of special cause problems in batches. Data mining techniques are not typically utilized to understand common cause variability in batch processes. Further, much of the research in batch monitoring does not consider time alignment issues in batch profiles.

In the area of batch process control, many strategies for product quality control are developed with specific processes in mind. Several approaches do not allow for further process improvement due to the black box nature of the process models. Other approaches depend on normally unavailable first-principles models. Also, some quality control schemes are developed through normally unavailable prior knowledge about the effects of control moves on the process.

There is a gap in the literature between data mining techniques used for problem detection and the application of corrective policies for better quality control. Very little work has been done to increase process understanding of batch processes by systematically examining the historical database of profiles using data mining techniques and then applying that knowledge for process improvement of batch processes in general. Thus, a need exists for a general, data-driven methodology to

facilitate batch understanding leading to process improvement. This work is an attempt in that direction.

Chapter 4

Nylon-6,6 Simulation

Introduction

The purpose of the current research is to systematically explore the historical database of past batches using existing data mining tools for process understanding that can suggest directions for process improvement. An industrially relevant simulation of a nylon-6,6 process is taken from the literature [81,83] and used to generate a database for illustrating the approach. The simulation is coded for use in the Matlab® computing environment.

Reactions

The main reaction given in chapter 3 is a reversible, second order formation of nylon-6,6 polymer and water from both amine and carboxyl groups and is written symbolically as:



Where: A = Amine group derived from a component of the reaction, hexamethylene diamine (HMD).
C = Carboxyl group derived from a component of the reaction, adipic acid (AA).
L = Polymer link.
W = Water molecule.

Additional side reactions modeling degradation or depolymerization of the polymer are given as:



Where: SE = stabilized (cyclized) end groups [104-105].

The details of the kinetics and material and energy balances are given in Appendix A.

Process Description

The polycondensation reaction is performed in a batch autoclave. A jacket around the reactor supplies heat via Dowtherm. Dowtherm is a commercial heat transfer fluid that allows for higher temperatures than steam at a given pressure. The reactor has a vent valve for the venting of volatile reaction components. A diagram of the process equipment is shown in Figure 4.1.

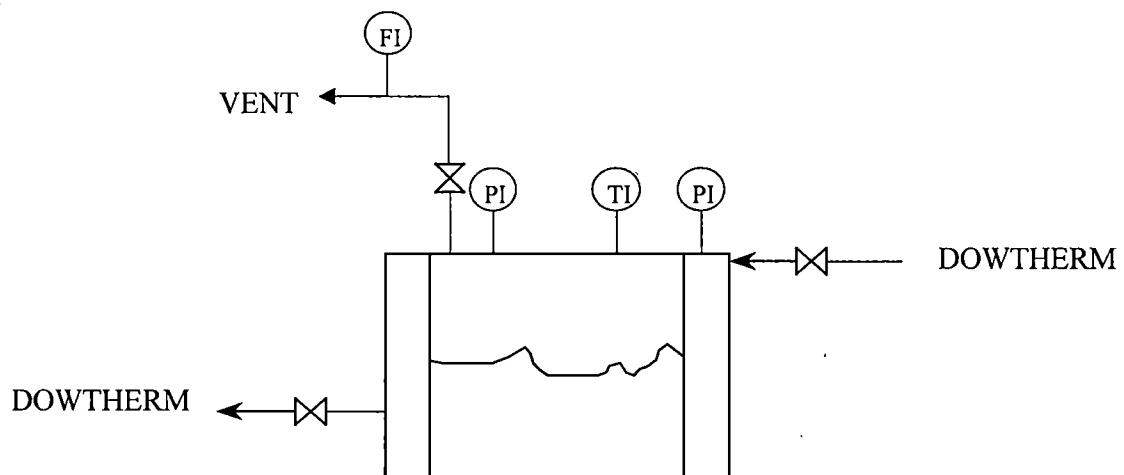


Figure 4.1 Nylon-6,6 Autoclave. Adapted from Fig. 1 of Russell, S. A., Robertson, D. G., Lee, J. H., and Ogunnaike, B. A., (1998), "Control of product quality for batch nylon 6,6 autoclaves", *Chem. Eng. Sci.*, 53(21), 3685-3702.

The reaction time is around 3-4 hours. The process has three heat transfer zones: 1. convection, 2. boiling, and 3. conduction. After the boiling phase begins, the reactor contents can be assumed to be well-mixed [107]. The process is conveniently divided into 5 phases as shown in Figure 4.2. Phase I is the convection zone. The middle 3 phases, II-IV, are the boiling zones. Phase V, the curing phase, is the conduction zone.

Available Measurements

The following variables are measured in the nylon-6,6 autoclave: 1. Reactor pressure (psia), 2. Jacket pressure (mmHg gauge), 3. Reactor liquid temperature (K), and 4. Vapor vent rate (g / h). The reactor pressure and jacket pressure trajectories are shown in Figures 4.3. and 4.4. The nominal temperature and vapor vent rate profiles are shown in Figures 4.5. and 4.6. The batch reactor that is being simulated does not have composition measurement capabilities. In the simulation, the jacket temperature is calculated from the jacket pressure.

Control

The manipulated variables for controlling the batch to the given recipe are the reactor and jacket pressures. Because pressure controller dynamics are fast, perfect control is assumed (i.e., the setpoint is equal to the current process value without any calculation of controller action). The control scheme is initiated based on the beginning of the boiling. As soon as boiling begins, the jacket pressure is ramped up from 1450 mmHg gauge to 1575 mmHg gauge. This is done to enhance the

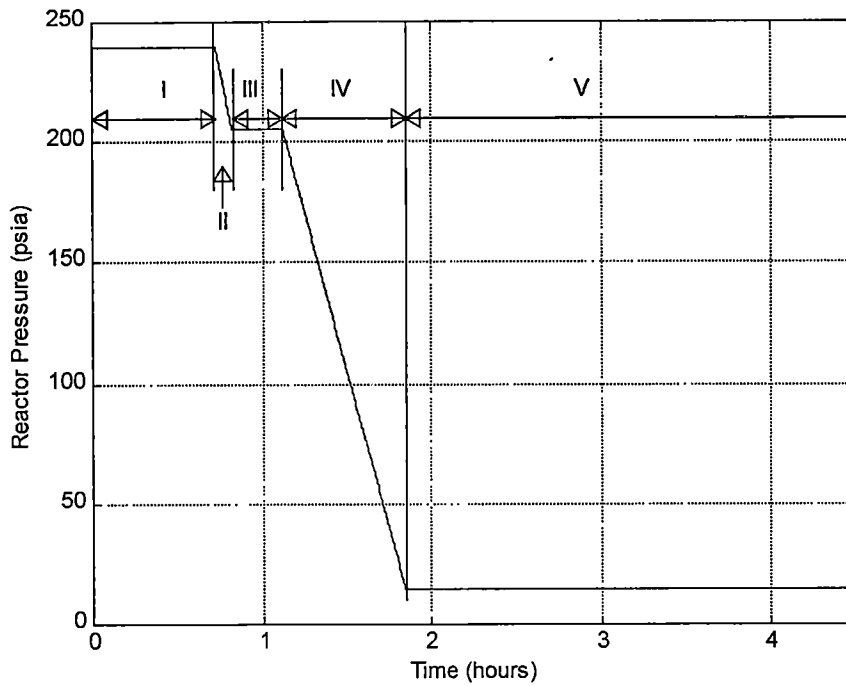


Figure 4.2 Reactor Phases

- Where:
- I. = Preheating
 - II. = Boiling, 1st Reactor Pressure Decrease
 - III. = Boiling, 2nd Reactor Pressure Setpoint
 - IV. = Boiling, Reactor Depressurization
 - V. = Curing Phase

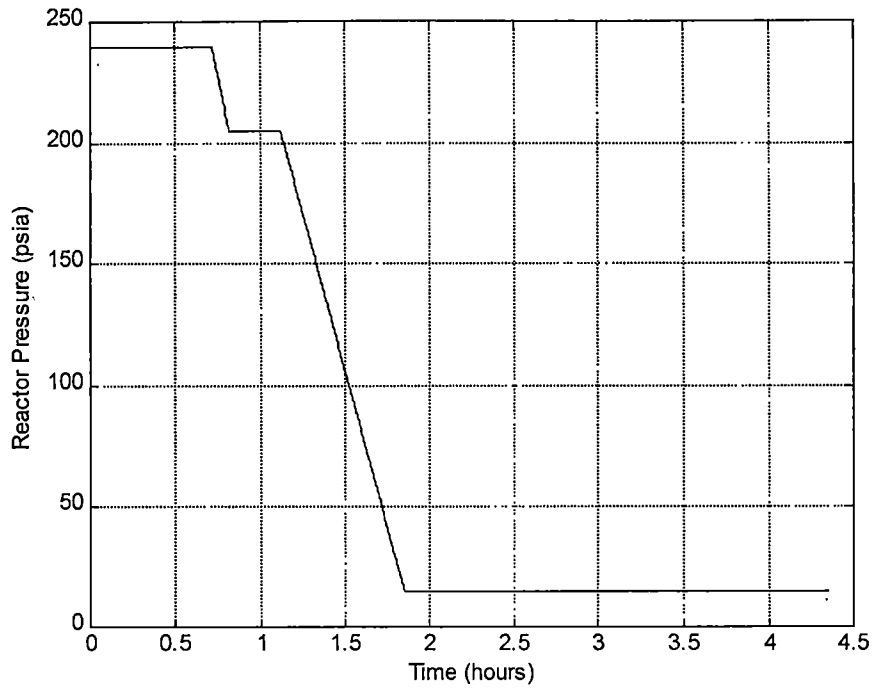


Figure 4.3 Reactor Pressure Profile

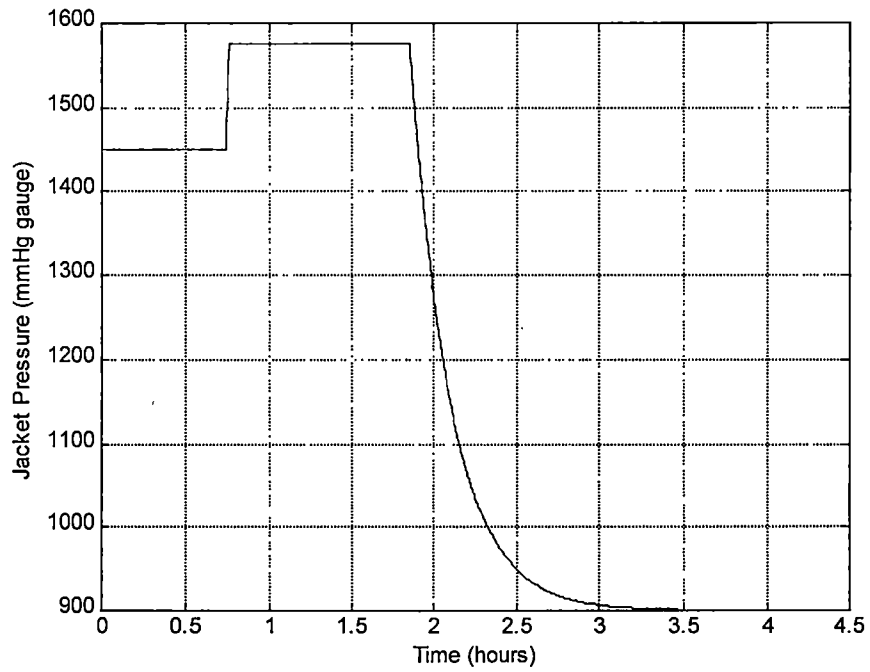


Figure 4.4 Jacket Pressure Profile

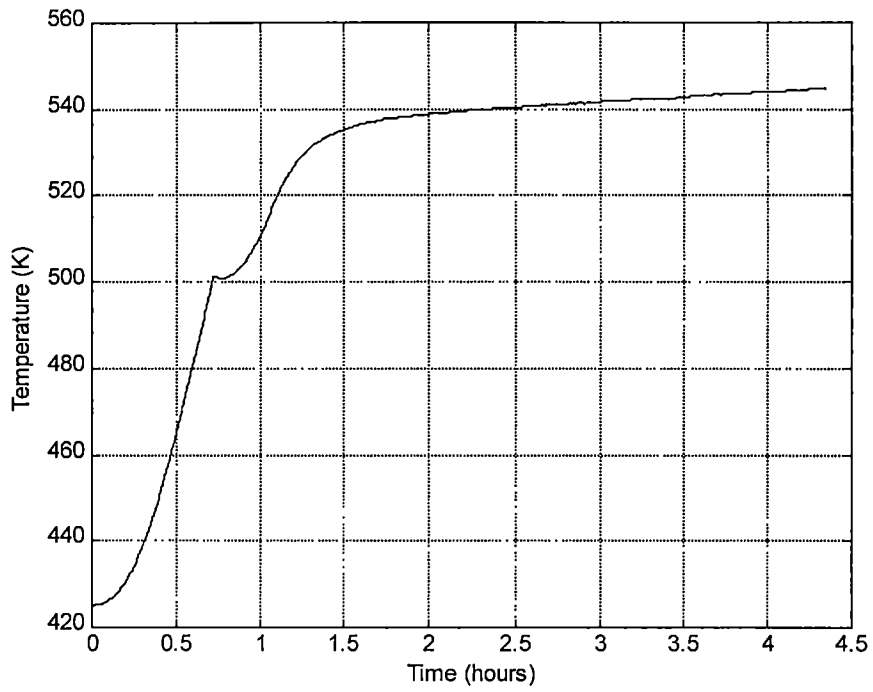


Figure 4.5 Temperature Profile

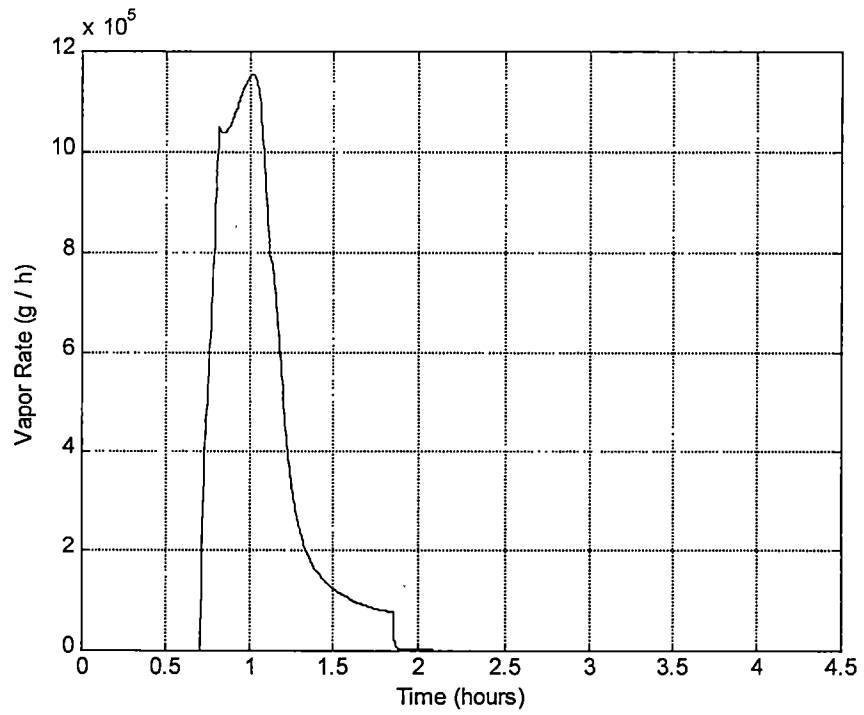


Figure 4.6 Vapor Rate Profile

boiling. The reactor pressure setpoint is immediately ramped down at a fixed rate from 240 psia to 205 psia. The reactor pressure remains at this setpoint until the reactor temperature reaches 521 K. After the 521 K temperature is reached, the reactor pressure is ramped down at a fixed rate to atmospheric pressure. The jacket pressure is held at 1575 mmHg gauge until the reactor drops to atmospheric pressure. The Dowtherm inlet valve is then closed and the jacket pressure is allowed to decay. This is modeled as a first order response. The final jacket pressure is about 900 mmHg gauge.

Autocorrelated noise is added to the measurement data to more closely simulate a real industrial process. The autocorrelated noise is generated by passing identical independently distributed (iid) Gaussian noise through a first order transfer function.

Quality Measures

The primary quality variable is the number average molecular weight (MW). The maximum number average molecular weight attained by a normal batch is 13,222 g/gmol. The MW from a nominal batch is shown in Figure 4.7. The degradation reactions (4.1-4.2) are the reason that the MW goes through a maximum and then begins to decrease.

Nominal and Disturbance Batches

The nominal case is generated in a reaction time of 4.5 hours. In the process, excess water in the reactor charge and reduced heat transfer are considered to be the primary disturbances for the purposes of this work. The water disturbance is randomly varied between 0 - 5% of the initial charge of water (350 kg). A 0 - 5% reduction in the heat

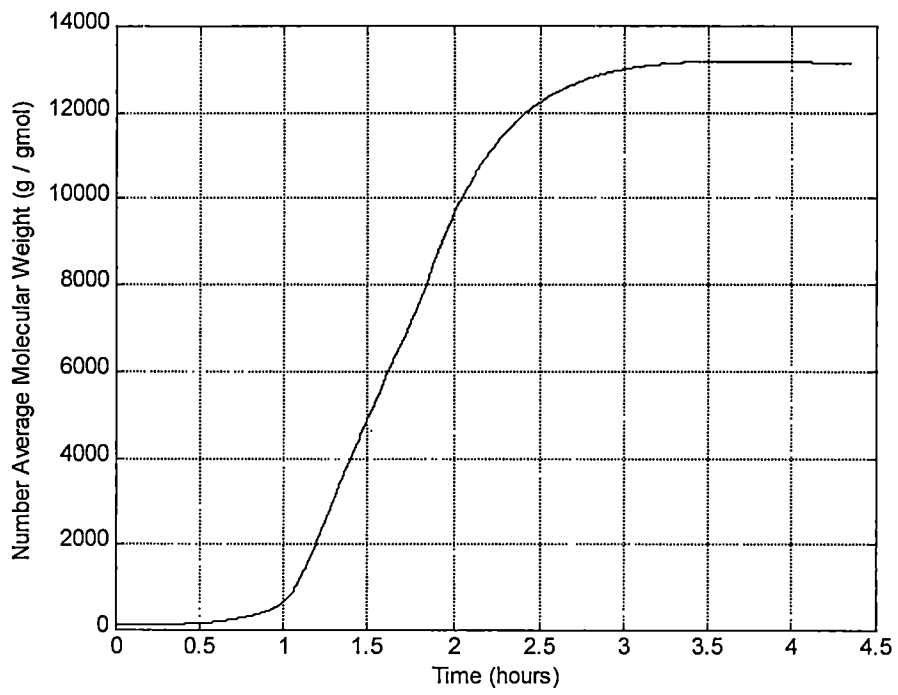


Figure 4.7 Molecular Weight Profile

transfer coefficient is effected through a scaling factor, f , which varies between 0.95 and 1.

Historical Database

A simulated database of batches covers. 100 batches are simulated in which the primary disturbances were randomly chosen. The disturbances are uncorrelated and randomly chosen to illustrate the expected nominal range of primary disturbances in the process. The space of the disturbances is shown in Figure 4.8. As shown by Figure 4.8, the space is spanned fairly well by the randomly selected disturbances.

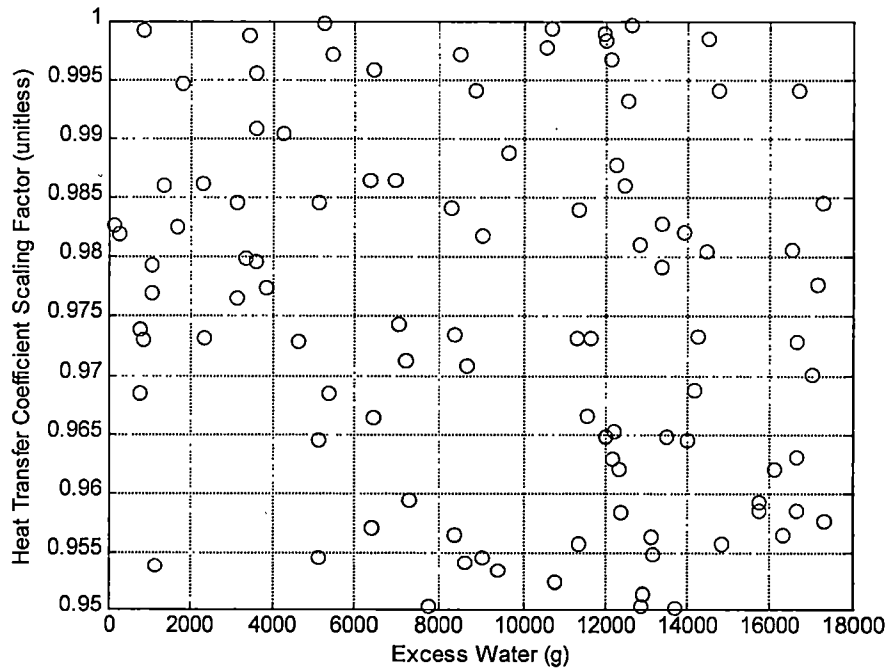


Figure 4.8 Disturbance Space

Batch Ending Recipe

The batches end at a fixed hold time of 1.84 hours after the end of the reactor depressurization. A fixed hold is typical for industry, as discussed by Jacobs et al. [104]. After the batch reactor is depressurized, the contents are allowed to cure for a set time to achieve the desired molecular weight. The hold time selected in this work is based on the time that a nominal batch takes to reach its maximum molecular weight.

Summary

A nylon-6,6 simulation is used in this research to generate batch data. The results are qualitatively similar to Russell et al [83] in both nominal and disturbance cases. A database is generated which is subjected to data mining to uncover the disturbances and explore opportunities for within batch control.

Chapter 5

Process Understanding

Introduction

In order to gain process insight, data mining tools must be utilized to explore the historical database. The common cause variability caused by the process disturbances may be reflected in these profiles. The goal of mining the data is to uncover the signature of these sources of common cause variability in the routine online measurements. Analysis of the profile data via the data mining tools can lead to increased process insight. Such process understanding can then point to directions for improving the process. The objective of this chapter is to demonstrate the application of data mining tools for analyzing the historical database in order to gain process insight.

The chapter is arranged as follows. Background on data mining of batch profiles is given. The variability along both the time and measurement axes is extracted using the method of Kaistha [72]. The data mining results are then interpreted using process knowledge to identify the fingerprint of the primary disturbances. Physical understanding is emphasized. Subsequently, correlations of the data mining results with quality are presented and offline quality predictions are developed. Predictions of the quality are compared with quality predictions developed from other multivariate techniques.

Data Mining Background

As discussed in chapter 2, batch profiles exhibit two types of

variability, consistent and inconsistent. The consistent variability repeats from batch to batch in a particular way, while the inconsistent arises from process noise and special causes. The consistent variability occurs along both the time and measurement axes. The variability along each axis can be characterized by scale parameters.

Data mining must begin by characterizing the time-axis variability because variability in time is not properly characterized by projection-based methods. In this work, the time-axis variability is characterized via the time-scale (TS) parameters of [71,72]. These time-scale parameters reflect the variation in time of segments of the batch profiles. The segments are determined from a study of the important events in the batch. After the time-axis variability is characterized, the remaining consistent variability is due to variation along the measurement axis. This measurement variability can be characterized by projection-based methods, e.g., principal component analysis (PCA). In this work, measurement variability is characterized by the magnitude-scale (MS) parameters of [71,72]. Correlations of the extracted TS and MS parameters can be developed with other variables of interest, such as quality or known disturbances. These correlations can then be studied by utilizing prior process knowledge in order to gain insight.

Characterization of Profiles

Time Axis Variability

The raw measurement profiles from the process are shown in Figure 5.1. As shown by the figure, these profiles exhibit clear variability on the time axis. Significant events in the batch profiles are easily identified. These events correspond to the phase behavior, as shown in Figure 4.1. The event times for 5 events are recorded: 1. Time that vaporization begins, 2. Time that reactor pressure reaches second

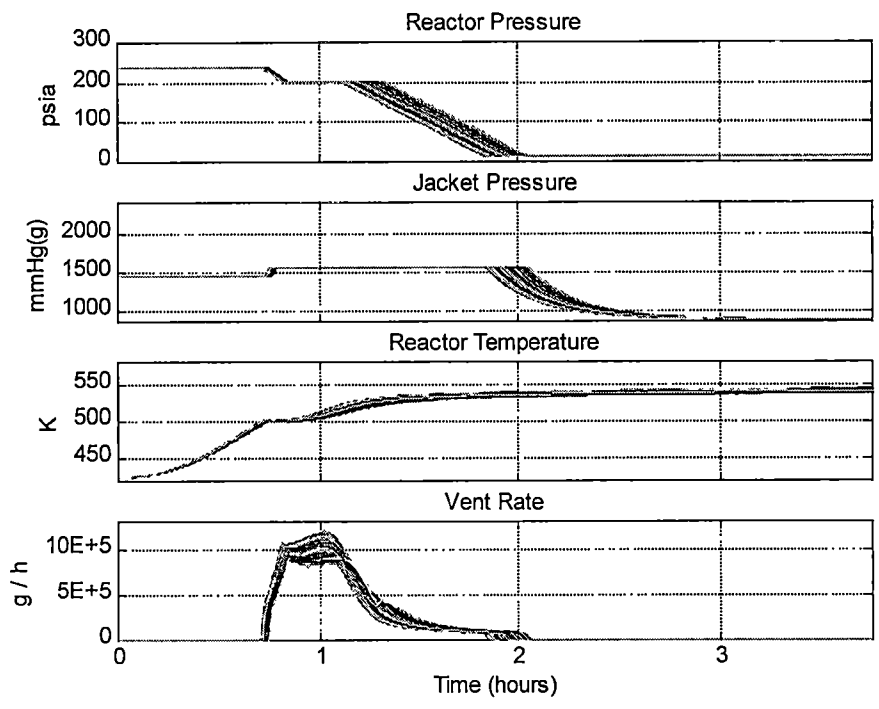


Figure 5.1 Raw Profiles

setpoint (= 205 psia), 3. Time that polymer solution in the reactor reaches a temperature setpoint (521 K), 4. Time at which the reactor pressure reaches atmospheric pressure, and 5. Time at which the batch ends.

The raw profiles are time-scaled by linearly interpolating each phase of each profile to a common length between the events [72, 93]. Reference lengths are determined for each phase. The change in length of the raw profile relative to the common or reference length provides the time scale. Phases I and III vary in length, and thus, contain useful time-scale information. However, phases II, IV, and V are fixed in length, and so the time scale is the same for all the batches. Time scales for phase I (ts_1) and phase III (ts_2) are shown in terms of phase ending time in Figures 5.2 and 5.3. The time-scaled profiles are shown in Figure 5.4. The time axis shown is the reference time; that is, the average time that the 100 batches take to finish.

Measurement Axis Variability

After time-scaling the profiles, the MS parameters are extracted in similar fashion to [72] by identifying the systematic zones of variability, or factors, in the time-scaled measurement profiles. Since the reactor pressure and the jacket pressure are assumed to be maintained with perfect control, no new magnitude information is contained in these profiles, as shown by Figure 5.2. Thus, the temperature and vapor rate profiles are used to characterize the measurement axis variability.

The methodology to extract the MS parameters is as follows. Let row vector \mathbf{p}_i be the i^{th} time-scaled profile. A reference trajectory, row vector \mathbf{r} , is the mean of the time-scaled profiles. The measurement variability remaining in $\mathbf{x}_i = (\mathbf{p}_i - \mathbf{r})$ is found by projecting \mathbf{x}_i onto the

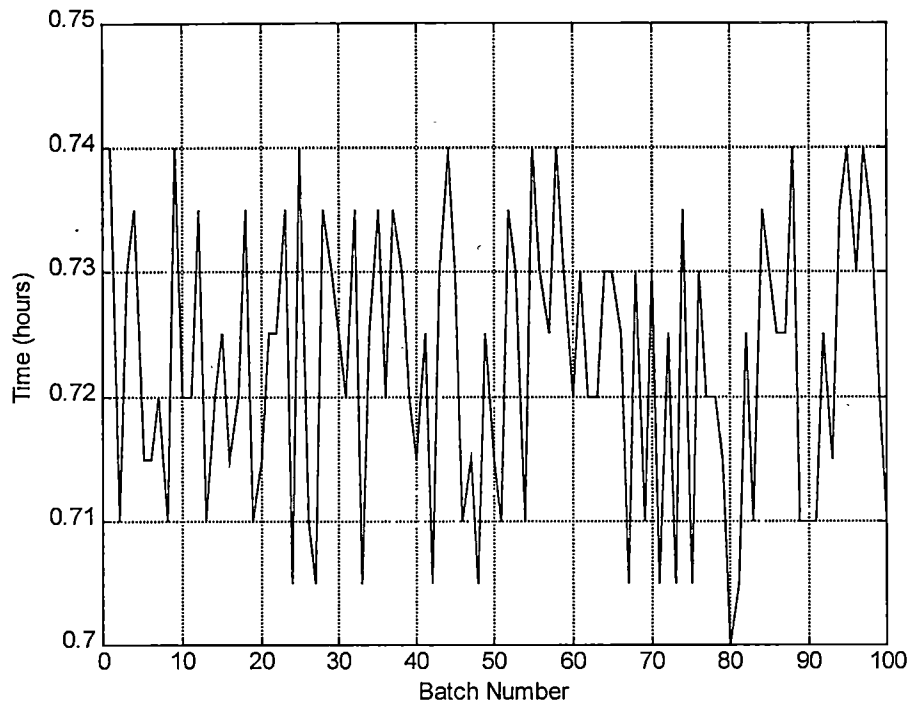


Figure 5.2 Phase I Length

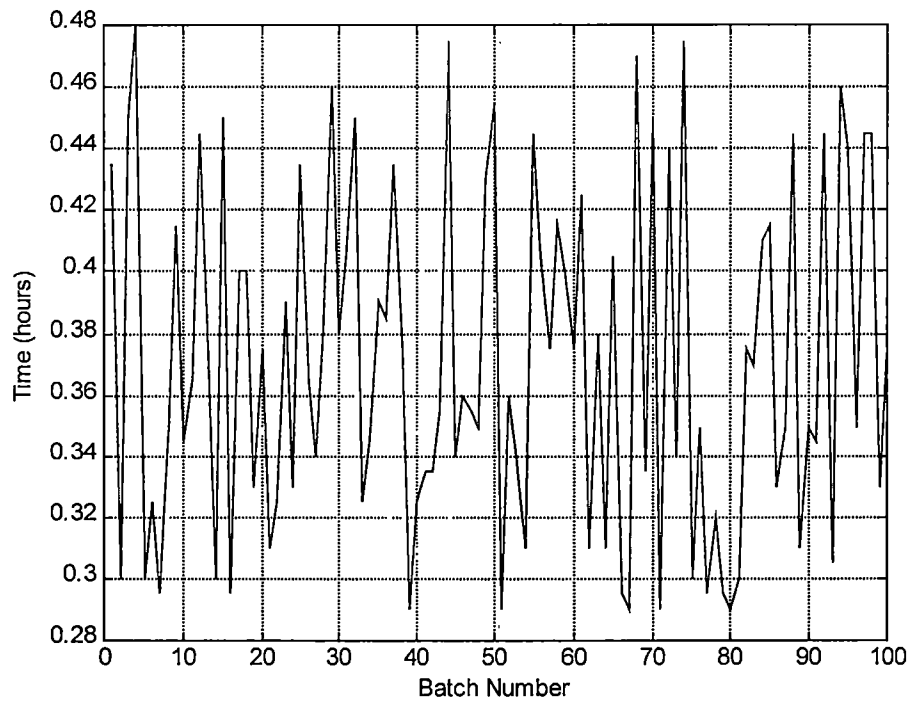


Figure 5.3 Phase III Length

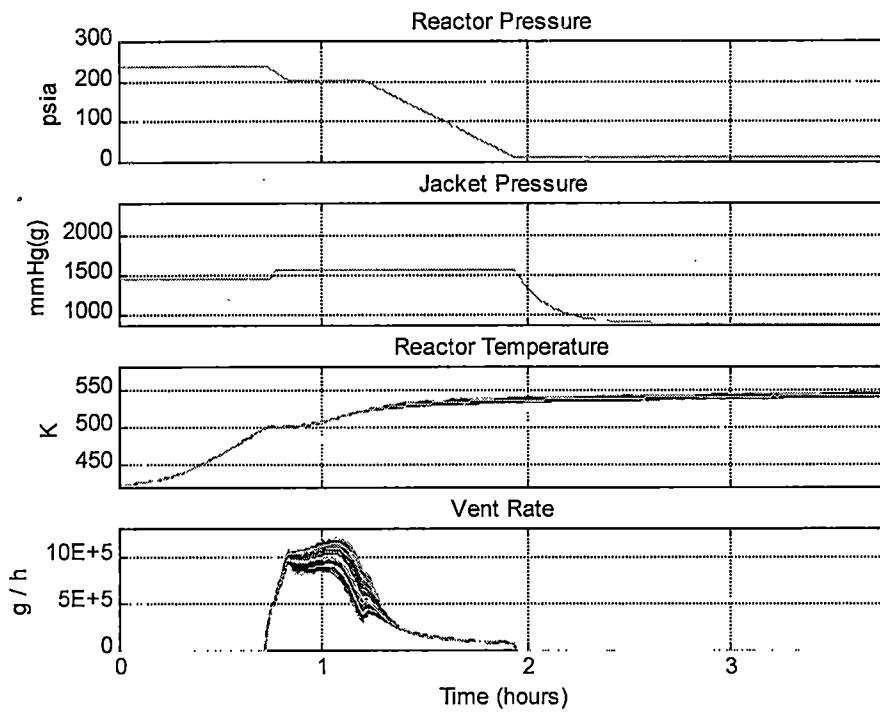


Figure 5.4 Time-Scaled Profiles

columns of H , which contain the principal directions of variation. In this work, $H = F$, the factor matrix. The factors are systematic zones of variability observed in \mathbf{x} . The MS scale parameter vector for \mathbf{x}_i is found by:

$$\mathbf{ms}_i = \mathbf{x}_i F \quad (5.1)$$

The residual error unexplained by projection is given by:

$$\mathbf{e}_i = \mathbf{x}_i - \mathbf{ms}_i F^T \quad (5.2)$$

For the whole set of i batches, the residual error is given by:

$$\mathbf{e} = \mathbf{x} - \mathbf{ms} F F^T \quad (5.3)$$

Figure 5.5 shows \mathbf{p} , \mathbf{e} , and F for each respective variable. As shown by the figure, the systematic measurement axis variability is explained very well by the factors in each measured variable since the residuals are small. The MS scale parameters for the vapor rate, \mathbf{ms}_1 - \mathbf{ms}_3 , and the temperature, \mathbf{ms}_4 - \mathbf{ms}_6 , correspond to the respective numbered factors shown in the figure.

Data Mining for Process Insight

Since the goal of this work is to utilize data mining tools for process insight, the scale parameters are interpreted using prior process knowledge and engineering judgment. A greater understanding of the process can then lead to process improvement by suggesting routes of corrective action to reduce the common cause variability.

Since the idea of data mining is to connect the scale parameters with the physics of the process, previous process knowledge is required to interpret the scale parameters. The developed TS and MS parameters are highly collinear so the interpretation of the scale parameters is facilitated by performing PCA. Since PCA identifies the principal directions of variation exhibited by a dataset, the technique may be useful in determining the underlying sources of variation captured in

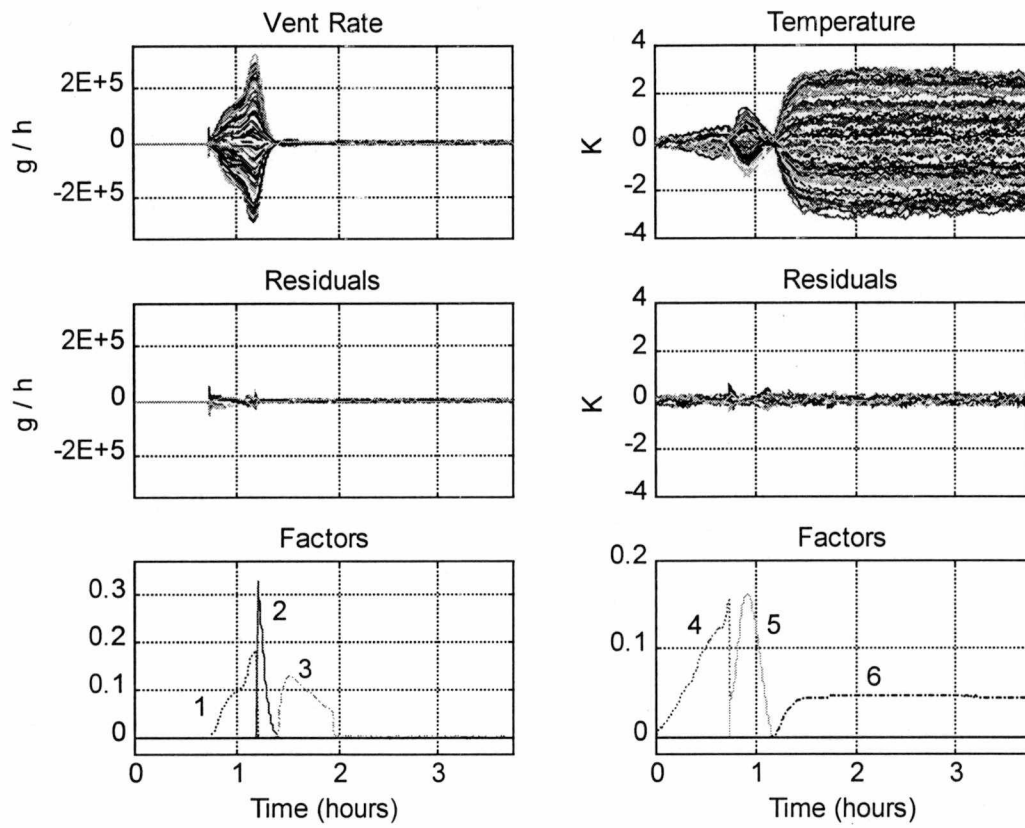


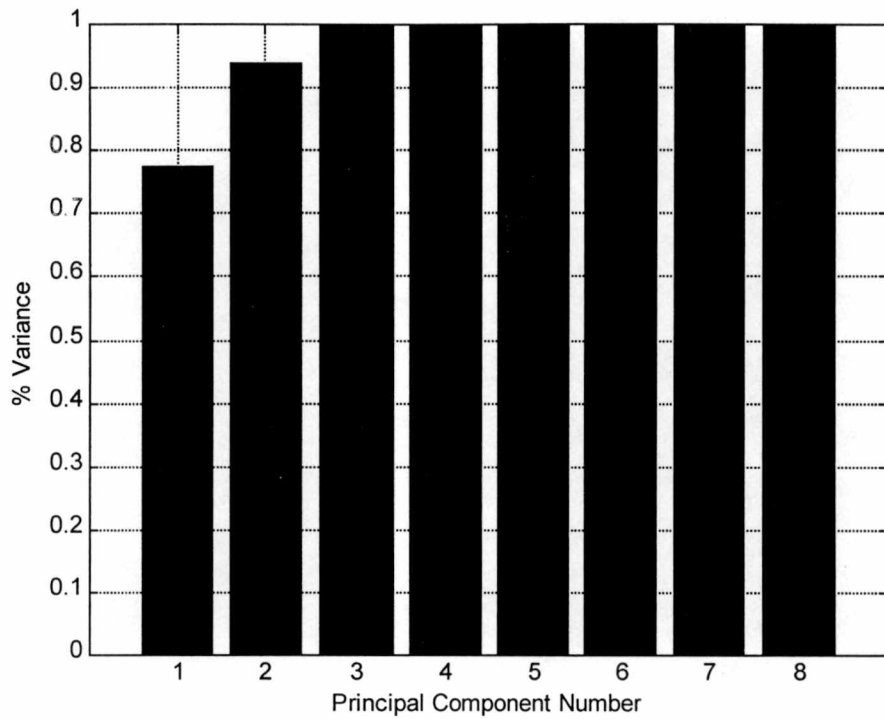
Figure 5.5 Time-scaled, Mean-centered Profiles, Residual Errors After Projection, and Factors for Vent Rate and Temperature

the scale parameters.

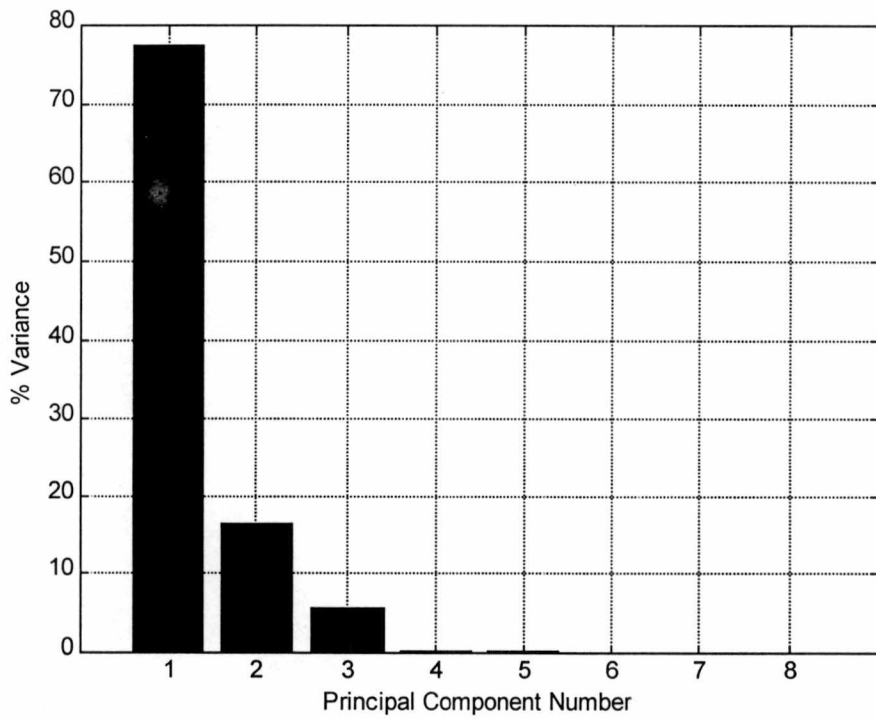
The scale parameter matrix subjected to PCA is arranged as row vectors (i.e., [ts1, ts2, ms1 ms2 ms3 ms4 ms5 ms6]). The scale parameters are first mean-centered and scaled to unit variance in order to compensate for the difference in engineering units. Figure 5.6 shows percentage variance scree plot obtained from an SVD analysis of the mean-centered, unit variance data. As shown in the figure, 2 PCs are sufficient to explain 93% of the variance in the scale parameter data. 3 PCs explain 99.7% of the variance. The PCs developed from this analysis are interpreted below in light of process knowledge.

The first two PCs are shown in Figure 5.7. PC 1 has high positive loadings for ts1, ts2, and ms3 and low negative loadings for ms1, ms2, ms5, and ms6. Physically speaking, the high positive loading on ts1 reflects a longer preheat time. The high positive loading on ts2 signifies a longer boiling phase before the reactor depressurization. The vapor rate is less as seen from the low negative loadings on ms1 and ms2. The temperature is less as seen in the low negative loadings on ms4 and ms5. The most likely cause for the combination of longer preheat and boiling phases, coupled with low vaporization and low temperature is low heat transfer. It is noted that the high loading on ms3 can be interpreted as a consequence of low heat transfer. Since the vaporization in the first part of the batch is very low, more material is vaporized later in the reactor depressurization phase.

The second PC shown in Figure 5.7 shows that ts1 has a high positive loading and ms4 and ms5 have low negative loadings. In physical terms, a high loading on ts1 signifies that the preheat phase is longer. The low loadings on ms4 and ms5 reflect a lower preheat temperature and a temperature dip following the beginning of vaporization. The presence of excess water in the reactor may possibly



(a)



(b)

Figure 5.6 Scree Plot from PCA of Scale Parameters. (a) Cumulative sum Scree plot. (b) Percentage variance per PC Scree plot.

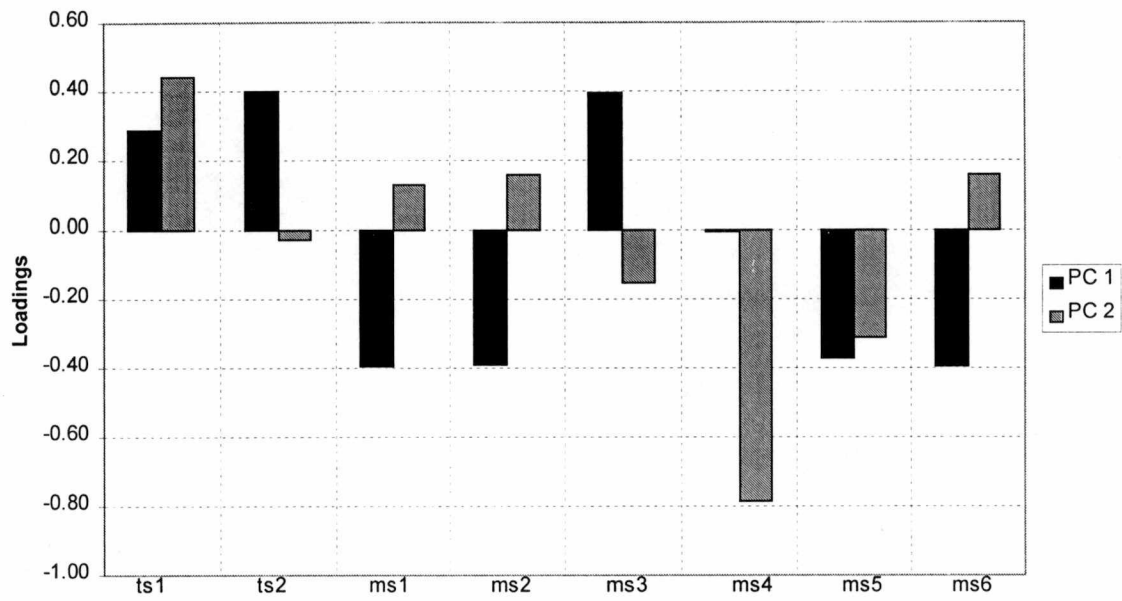


Figure 5.7 First Two Principal Components of Scale Parameter Matrix

explain the pattern of these loadings. If water is present, the reversible reaction (4.1) is shifted to the left, so HMD is consumed less which initially reduces the reaction rate. As a result, the heat released by the reaction is less, leading to a longer preheat time that is reflected in ts1. Scale parameters ms4 and ms5 corroborate this effect, as the temperature is lower, reflecting a slower reaction rate. Since the loadings on ms1-ms3 are close to normal, a heat transfer problem is not considered to be the dominant source of variation. Furthermore, ms6 is higher, which shows that the reaction rate eventually recovered. These effects are consistent with those expected from a water disturbance.

The third PC is shown with the first 2 PCs in Figure 5.8. The third PC shows high positive loadings on ts1 and ms4. Physically, a high positive loading on ts1 reflects a longer preheat time. A high positive loading on ms5 shows a temperature rise after the beginning of vaporization. Initially, these loadings do not conclusively point to a particular disturbance. The long preheat phase suggested by ts1 is not distinctive, since ts1 is long in both the heat transfer and water disturbance cases. This PC will be further considered below.

The first two PCs have been tentatively identified as the fingerprints of low heat transfer and excess water. The scores, or projection of the data onto the respective PCs 1 and 2, are correlated with the product quality to attempt to corroborate the hypotheses formed from studying the loadings.

Correlations of Scores with Quality

The correlation of the scores on the first two PCs with the MW is shown in Figure 5.9. As shown by column 1 in Figure 5.9, the MW is higher when low heat transfer is suspected. This corroborates with

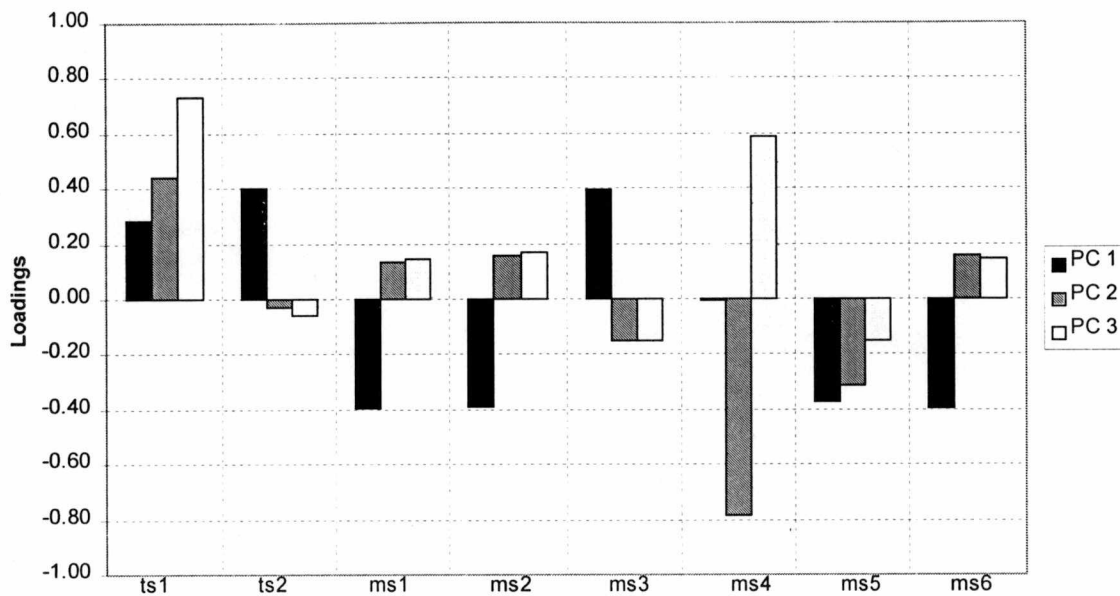


Figure 5.8 First Three Principal Components of Scale Parameter Matrix

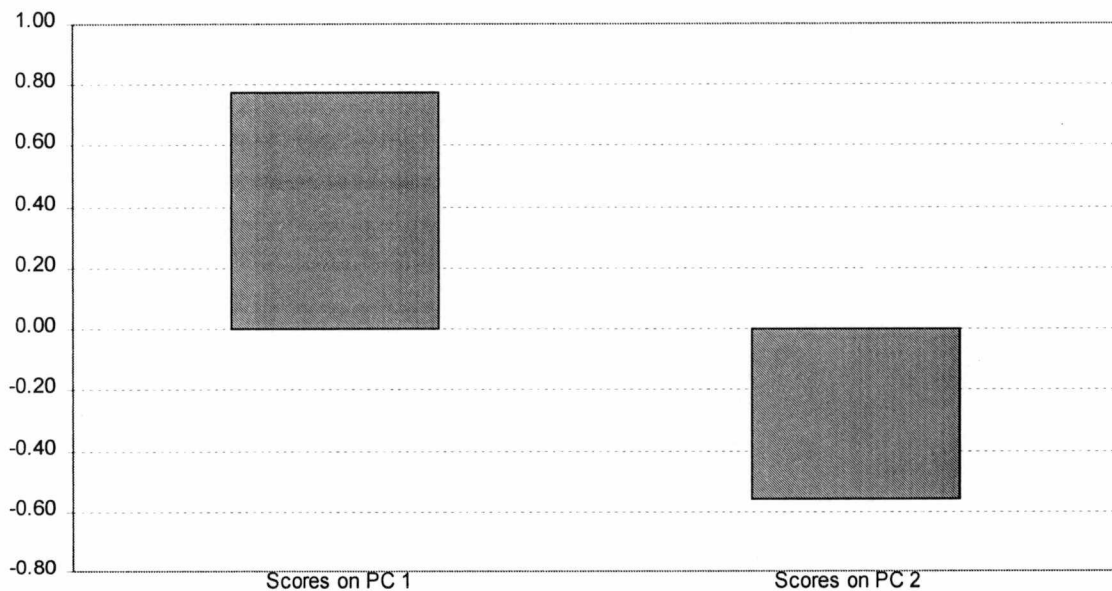


Figure 5.9 Correlation of Scores on First Two PCs of Scale Parameter Matrix with Molecular Weight

process knowledge. Since the vapor rate is lower, less HMD vaporizes. Since more monomer stays in solution, the polymerization continues resulting in a higher MW than the target.

When a suspected water disturbance is present, i.e., the scores on PC 2 are high, the MW is lower. HMD is consumed less due to the shift in the reversible reaction (4.1). Additional monomer in solution results in a greater loss of HMD during the boiling phase. Since less monomer is present following the vaporization, the achievable MW is less.

Figure 5.10 shows the correlation of the scores on the first 3 PCs with the MW. The scores on PC 3 are negatively correlated to the MW, as are the scores on PC 2. This similarity in the correlations of scores on PCs 2 and 3 with the MW may indicate that the scores on PC 3 also suggest the presence of a water disturbance. However, from the data, this conclusion is tentative at best.

In summary, the scores on PC 1 seem to indicate a heat transfer disturbance and the scores on PC 2, and possibly those on PC 3, seem to indicate a water disturbance. The disturbances hypothesized to be present in the process and reflected by the data correspond to the effects shown by the actual water and heat disturbances on the quality outcomes. Thus, these hypotheses seem very reasonable.

Correlations from Process Studies

The disturbance hypotheses are verified from process studies. Figure 5.11 shows the correlations of the scores on the first two PCs with the disturbances generated in the process. As shown in Figure 5.11, the scores on the first PC are almost perfectly correlated with the heat disturbance. If the heat transfer is less, the score on the first PC is higher; i.e., the greater that the heat disturbance is, the

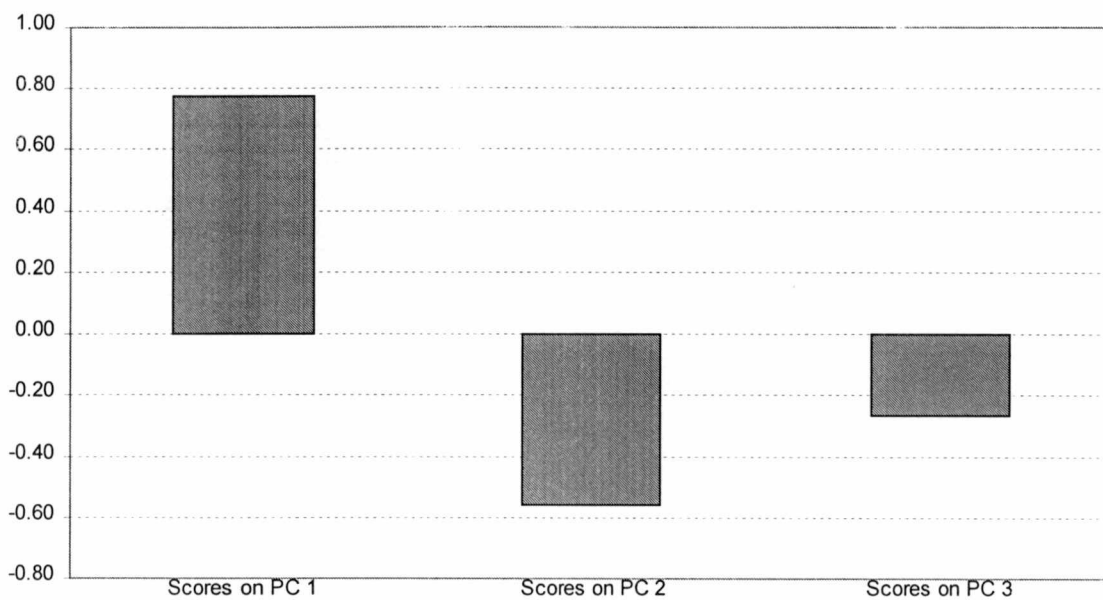


Figure 5.10 Correlation of Scores on First Three PCs of Scale Parameter Matrix with Molecular Weight

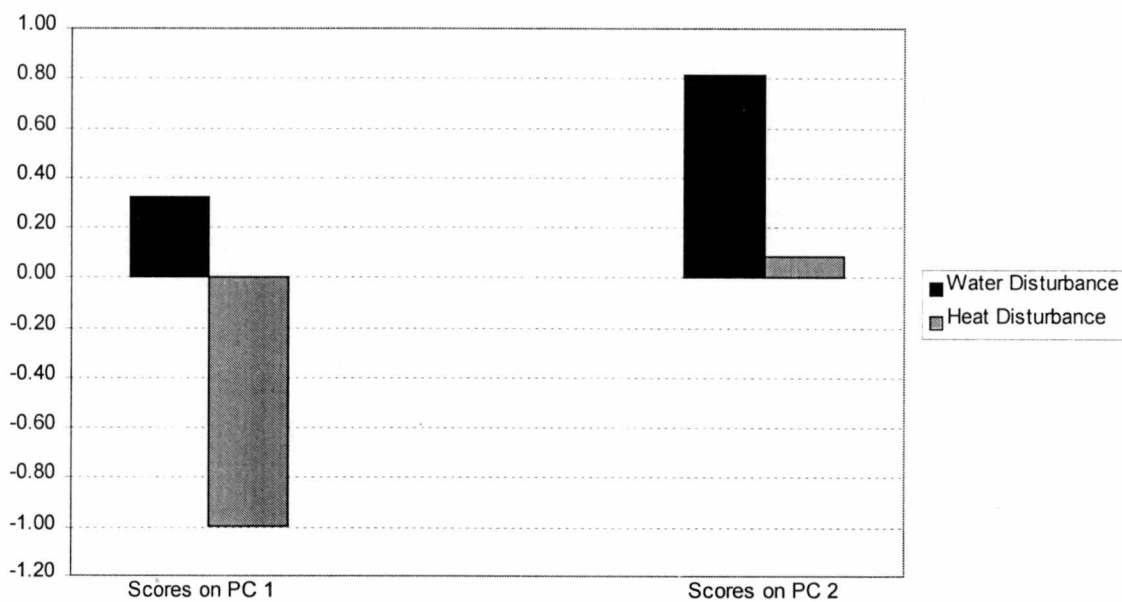


Figure 5.11 Correlation of Scores on First Two PCs of Scale Parameter Matrix with Disturbances

lower the heat transfer rate is. This accounts for the difference in sign. The scores on the second PC are strongly correlated to the water disturbance. The greater the water disturbance is, the higher the score on PC 2. A slight interaction with the heat disturbance is also seen, but the dominant effect explained by PC 2 is that of a water disturbance. Figure 5.12 shows the correlation of the first 3 PCs with both simulated disturbances. The dominant effect of PC 3 is clearly that of water disturbance.

To verify these observations, a linear regression model with each disturbance is built from the scores of the first three PCs. The purpose of the regression is simply to see the relation of each regression coefficient to the scores on the PCs of interest, not to build a robust predictive model. The regression relation is expressed by:

$$\underline{Y} = \underline{X}\beta \quad (3.3)$$

with
$$\underline{\beta} = (\underline{X}^T\underline{X})^{-1}\underline{X}^T\underline{Y} \quad (3.2)$$

Where: \underline{X} = Scores
 \underline{Y} = Simulated disturbances
 \underline{Y} = Predicted disturbances

Table 5.1 shows the regression coefficients from these predictions. As shown by Table 5.1, PC 1 primarily explains the heat disturbance since the magnitude of the regression coefficient for score 1 is significantly higher than the magnitudes of the coefficients for scores 2 and 3. As shown by the regression coefficients for scores 2 and 3 as compared to the coefficient for score 1, PCs 2 and 3 primarily explain the water disturbance. Figure 5.13 shows that the scores on PC 1 and the

Table 5.1. Predictions of the Disturbances Using Scores from the First 3 PCs

Disturbance Type	Regression Coefficients		
	Score 1	Score 2	Score 3
Heat Transfer	-0.4071	0.0690	0.0467
Water	0.0509	0.7212	0.6499

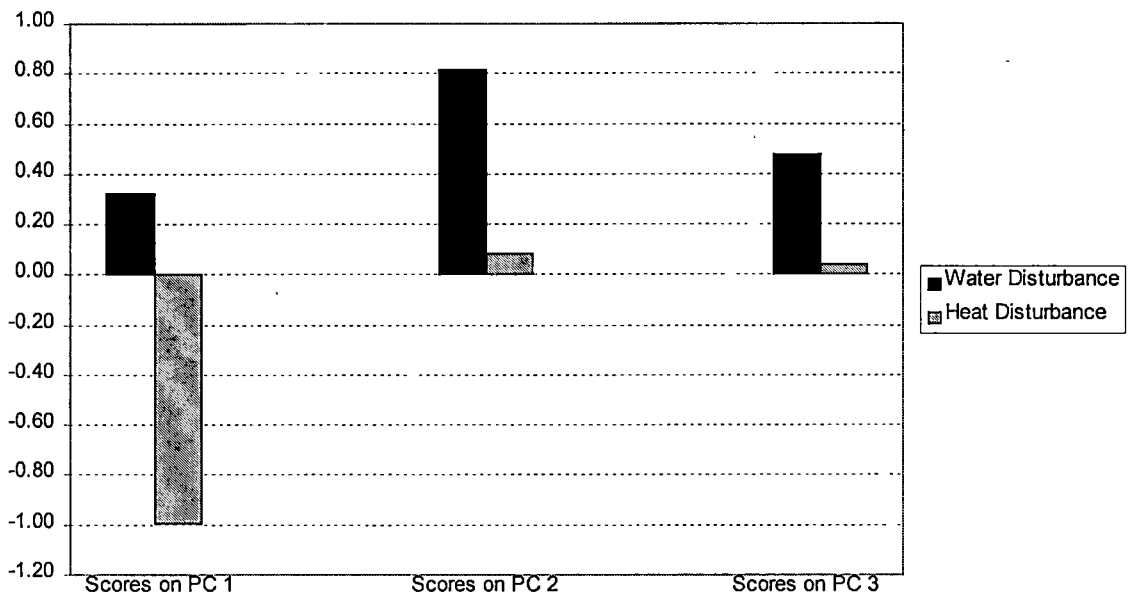


Figure 5.12 Correlation of Scores on First 3 PCs of Scale Parameter Matrix with Disturbances

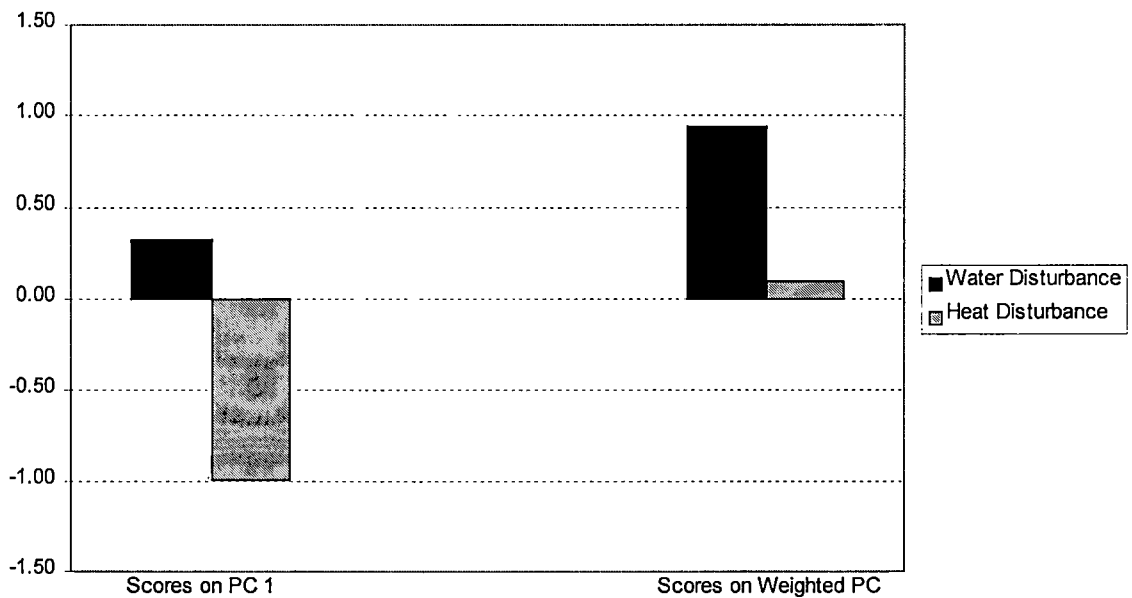


Figure 5.13 Correlation of Scores on First PC and Weighted PCs 2 and 3 of Scale Parameter Matrix with Disturbances

scores on a weighted PC, constructed by weighting PC 2 and 3 with their respective regression coefficients, are strongly correlated with the heat transfer and water disturbances, respectively. This again verifies that PC 2 and PC 3 explain the water disturbance. The disturbances are thus tentatively identified from data mining coupled with process knowledge as excess water and low heat transfer, and verified from process studies.

These disturbances are known to have definite effects on the product quality. Since the effects of disturbances are seen in the MW, in order to effect process improvement, the MW must be predictable from the profile data. Predictions of the quality are now demonstrated using the scores from the scale parameter data.

Quality and Disturbance Predictions from the Scores

A principal component regression (PCR) is used to develop a predictive model of the quality from the scores. The measure of the accuracy of the predictions is the commonly used root-mean square error of prediction (RMSE). The RMSE is calculated as follows:

$$RMSE = \sqrt{(\sum (Y_{\text{predicted}} - Y_{\text{actual}})^2 / n)} \quad (5.1)$$

Where: $Y_{\text{predicted}}$ = the predicted value of the outcome
 Y_{actual} = the actual outcome
 n = number of samples

A training set of 50 batches is used to build the model. All of the training inputs used in the predictions are first standardized by mean-centering and scaling to unit variance to account for large magnitude differences in the scores. After using the training set to develop the model, the model is tested on the remaining 50 batches. Figure 5.14 shows the MW predictions from the scores from six retained PCs. The addition of quadratic terms, as in equation (3.5), improved the predictions significantly. Table 5.2 summarizes the results.

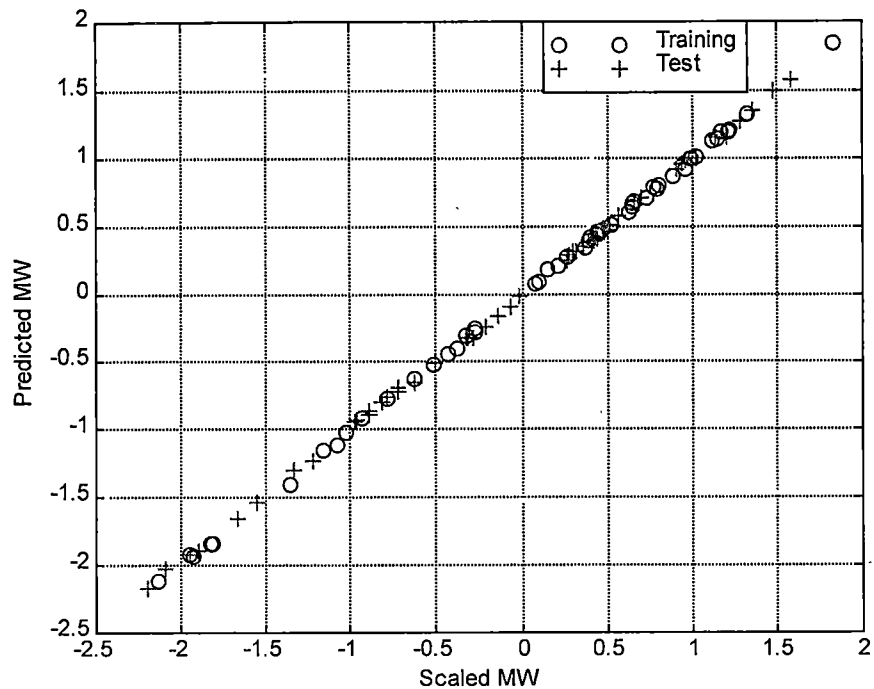


Figure 5.14 Offline Predictions of Molecular Weight Using Autoscaled Scores on 16 Retained PCs of Scale Parameter Matrix

Table 5.2. Predictions of the Molecular Weight at Fixed Batch End Time Using Scores (All PCs Retained: 8 - Linear; 16 - Non-linear) (Autoscaled Data)

Data Type	Training RMSE	Test RMSE
With Linear Scores Only	0.0515	0.0626
With Quadratic Terms	0.0187	0.0230

As shown by Table 5.2, the MW at the end of the batch is predictable using the scores. The predictions are very good as seen in Table 5.2 and Figure 5.14. Since the quality is predictable from the scale parameter information, process improvement becomes a practical possibility.

For further insight into the process, the simulated disturbances are found to be predictable from the scores also. Figures 5.15 and 5.16 show the predictions from the scores of both the water and heat transfer disturbances used in the process. Table 5.3 summarizes the results of both the score predictions. As shown in Table 5.3, the predictions are very good.

Although primarily presented here for academic interest, these predictions may have a practical value in industry. It is not an unreasonable suggestion in industry to catch a sample of the reactor contents and measure for excess water. If samples of the reactor contents can be obtained, a predictive model for the water disturbance is very practical. Similarly, if a known quantity, e.g., the scores on PC 1, is positively identified to be a signature of a heat transfer disturbance, a predictive model of the heat transfer disturbance is also very practical for use in process operation and process improvement.

Table 5.3. Predictions of the Actual Disturbances Using Scores (All PCs Retained: 8 - Linear; 16 - Non-linear) (Autoscaled Data)

Data Type	Disturbance Type	Training RMSE	Test RMSE
With Linear Scores Only	Heat Transfer	0.0069	0.0079
With Additional Quadratic Terms	Heat Transfer	0.0059	0.0068
With Linear Scores Only	Water	0.0344	0.0404
With Additional Quadratic Terms	Water	0.0143	0.0199

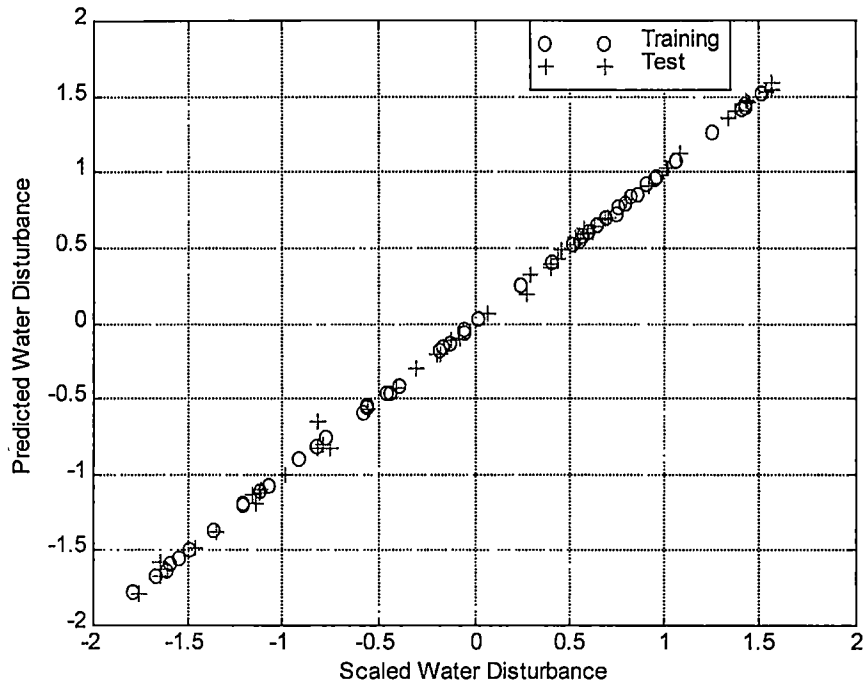


Figure 5.15 Offline Predictions of Water Disturbance Using Autoscaled Scores on 16 Retained PCs of Scale Parameter Matrix

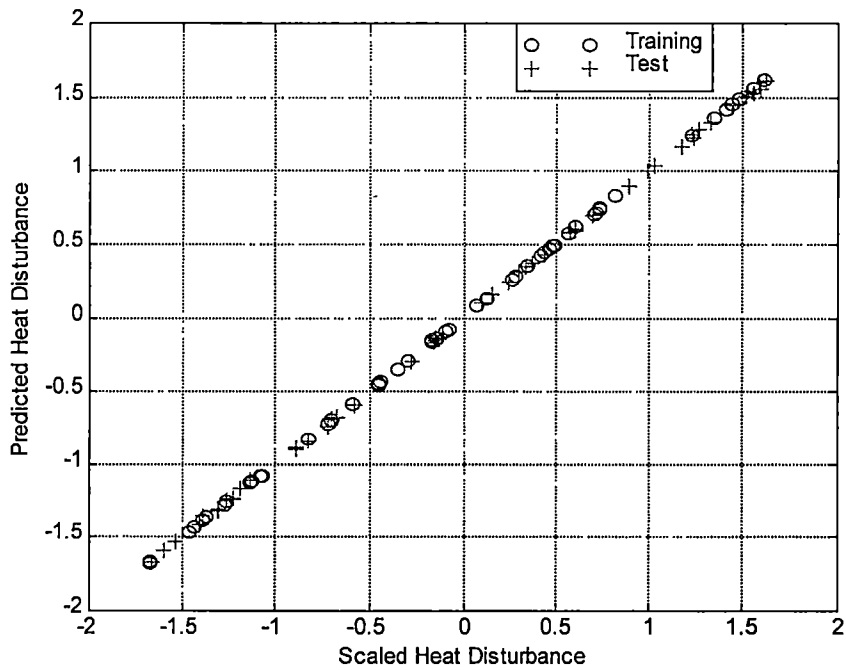


Figure 5.16 Offline Predictions of Heat Disturbance Using Autoscaled Scores on 16 Retained PCs of Scale Parameter Matrix

Comparison with Predictions from Other Data Mining Methods

Many data mining techniques can be utilized in the monitoring and analysis of batch profiles. The technique described above is based on the method of Kaistha [72]. Two other methods are investigated in order to compare their predictive methods to that described in this work. The other methods studied in this work are multi-block multiway partial least squares (MPLS) and multi-block MPCA. The loadings are determined from time-scaled temperature and vapor rate profiles that are mean-centered and scaled to unit variance, to account for differences in engineering units. Only the non-zero portions of the vapor rate profiles were used in building the model. The second block of predictors used is a matrix of the phase ending times. These times provide the time-scaling information. For the MPCA study, PCR is used to develop the predictions. This is referred to as multiway principal component regression (MPCR). In the predictions, two latent variables (MPLS) or three retained PCs (MPCR) are used to obtain the best predictions. The predictions for each method during the batch are shown in Table 5.4. As shown in Table 5.4, the predictions of the scale parameter-based method are much better than the predictions of other multivariate techniques. Further, the scale parameter-based method described in this work has the advantage that the scale parameters can be interpreted more easily when attempting to understand the physics of the process.

Table 5.4. Offline predictions of the MW Using MPCR (3 retained PCs), MPLS (2 latent variables), and Scale Parameter-Based Method. (All PCs Retained: 8 - Linear; 16 - Non-linear) (Autoscaled Data)

Method	Training RMSE	Test RMSE
MPCR	0.3135	0.3406
MPLS	0.2244	0.2657
Scale Parameter-Based Method	0.0187	0.0230

Summary

This chapter discusses and demonstrates the use of a data mining technique applied to batch profiles for increased process understanding. The parameters extracted using the method of Kaistha [72] are studied to determine their relationship to the quality and the primary disturbances in the nylon-6,6 process. As a result of performing a PCA on the scale parameters, and interpreting the loadings using process knowledge, the sources of common cause variability affecting the process are better understood. The scores of the data on the PCs are used to build offline models for prediction of quality and the primary disturbances. The scale parameter-based method for offline predictions furnishes better predictions compared to those developed from other data mining techniques, with the added benefit that the scale parameters are more easily connected with the physics of the process. The subsequent chapters 6 and 7 discuss online predictions and the development and implementation of recipe adjustments to improve the process.

Chapter 6

Within Batch Quality Control

Introduction

Improved process understanding can be obtained by mining the historical database of past batches and utilizing prior process knowledge to interpret the results. If the final quality can be predicted online, this increased understanding directly leads to suggestions for process improvement in the form of online recipe adjustments. These recipe adjustments also require that suitable manipulated variable handles be available. The goal of the online recipe adjustments is tighter product quality control. This is referred to as within batch quality control.

In this chapter, online quality predictions are demonstrated. Suggestions for process improvement are developed from the results of the data mining study on the offline data. A within batch quality control strategy is presented and subsequently implemented for the nylon-6,6 process. The control strategy is presented along with a discussion of the necessary handles for adjusting the recipe. In specific, a linear MW-based controller is implemented and the results are studied.

Soft-Sensor Quality Predictions

The goal of online recipe adjustments is to correct for the effects of disturbances on the batch. To determine the need for such adjustments, the quality must be predictable well before the end of the

Table 6.1. Available Scale Parameters by Phase

Batch Phase	Available Parameters at End of Phase		
	Time Scale (TS)	Vapor Rate (V)	Temperature (T)
Preheat	ts1		ms4
Boiling, 1 st Pressure Decrease	ts1		ms4
Boiling, 2 nd Reactor Pressure Setpoint	ts1, ts2	ms1	ms4, ms5
Boiling, Reactor Depressurization	ts1, ts2	ms1, ms2	ms4, ms5
Curing	ts1, ts2	ms1, ms2, ms3	ms4, ms5, ms6

batch. In developing online predictions of the MW, consideration must be given to phase divisions. The studies in chapter 5 are done on completed profiles. During the batch, only certain scale parameters will be available to use in a predictive quality model. The scale parameters that are available by phase are shown in Table 6.1.

Systematic Methodology

The best online predictions of the molecular weight should be used as a basis for applying a corrective action to the batch. Thus, predictions at the end of each phase should be done as described below to determine when the best predictions are available. However, a trade off exists between the availability of accurate predictions of the quality and the physics of the process. A corrective action must be applied with consideration to operational constraints of the given batch process.

In the nylon-6,6 process, operational difficulties with viscous polymer late in the batch require that adjustments to the recipe be done by no later than the end of the first boiling phase (phase III), since the reactor is still well-mixed [81,107]. Therefore, it is imperative that the MW at the end of the batch must be predictable by the end of phase III. From Table 6.1, 5 scale parameters are available by the end of phase III: ts1, ts2, ms1, ms4, and ms5.

Since only a partial number of scale parameters are available, partial scores from the completed profiles have to be used in the online quality predictions. The partial scores are calculated as follows:

$$T_p = X * V_p \quad (6.1)$$

Where: T_p = Partial Scores
 X = Available time-scaled data
 V_p = PCs with partial loadings

The partial loadings omit contributions from scale parameters that are not available by setting the respective loading(s) in each PC to zero. It should be noted that if a predictive model is developed using only the available parameters instead of zeroing the unavailable loadings from the offline case, the model is different, but the predictions are similar.

To illustrate online predictions at the end of phase III, the contributions from ms2, ms3, and ms6 are set to zero because they occur after the phase is over. Since the loadings in V_p are the columns, rows 4, 5, and 8 are set to zero. If additional quadratic terms are used, then rows 12, 13, and 16 are also set to zero. The partial scores obtained by projection of the online data onto the partial PCs are used as a 'soft-sensor' for quality predictions.

Online Prediction Results

Table 6.2 shows a summary of the prediction results at the end of each phase.

Table 6.2 Summary of Predictions at the end of each phase using PCR. (Autoscaled Data)

Batch Phase	Retained PCs	Training RMSE	Test RMSE
Linear Terms Only			
Preheat	2	0.9370	0.9120
Boiling, 1 st Pressure Decrease	2	0.9370	0.9120
Boiling, 2 nd Reactor Pressure Setpoint	5	0.067	0.0877
Boiling, Reactor Depressurization	6	0.0659	0.0842
Curing	8	0.0515	0.0626
Linear and Quadratic Terms			
Preheat	3	0.9363	0.9143
Boiling, 1 st Pressure Decrease	3	0.9363	0.9143
Boiling, 2 nd Reactor Pressure Setpoint	9	0.0261	0.0360
Boiling, Reactor Depressurization	12	0.0247	0.0352
Curing	16	0.0187	0.0230

As shown in Table 6.2, the predictions through the end of phase II exhibit too high a spread to allow for reliable early verification of a deviation of the MW from the target. Therefore, due to the operational constraints in the process, the predictions are performed at the end of phase III. Figure 6.1 shows the predicted values at the end of phase III of the MW closely match the actual MW values. Note that the addition of quadratic terms dramatically reduces the RMSE. In the quadratic case, the scores are mean-centered and scaled to unit variance before performing the predictions.

The partial scores are also used to predict the simulated disturbances at the end of phase III. Figures 6.2 and 6.3 show the predictions from the scores on the partially completed data for both the water and heat transfer disturbances used in the process. Table 6.3 summarizes the results of the predictions from the partial scores. As shown in Table 6.3, additional quadratic terms improved the predictions.

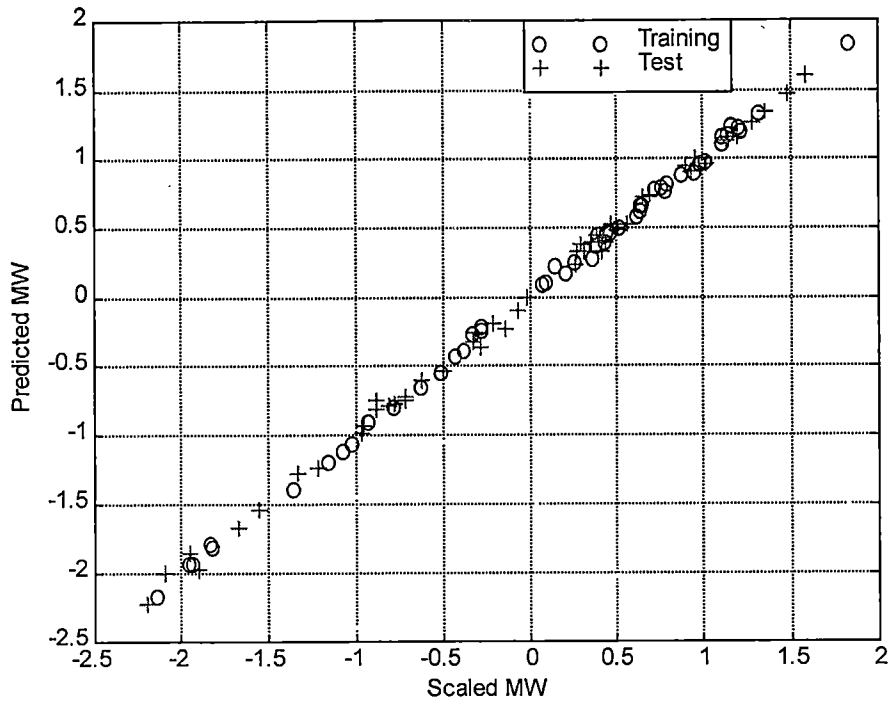


Figure 6.1 Online Predictions of Molecular Weight Using Autoscaled Scores on 9 Retained PCs of Scale Parameter Matrix

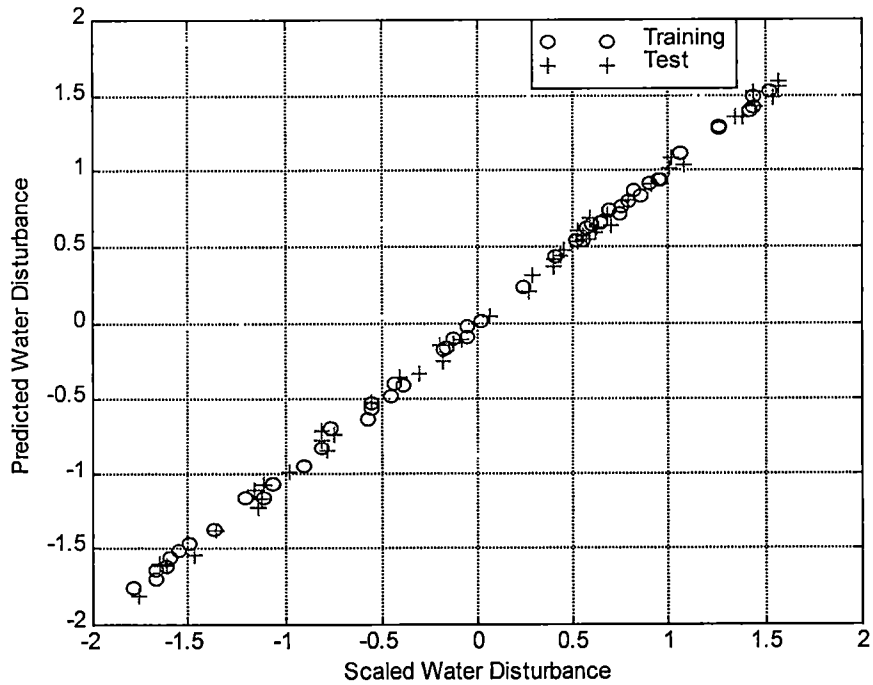


Figure 6.2 Online Predictions of Water Disturbance Using Autoscaled Scores on 9 Retained PCs of Scale Parameter Matrix

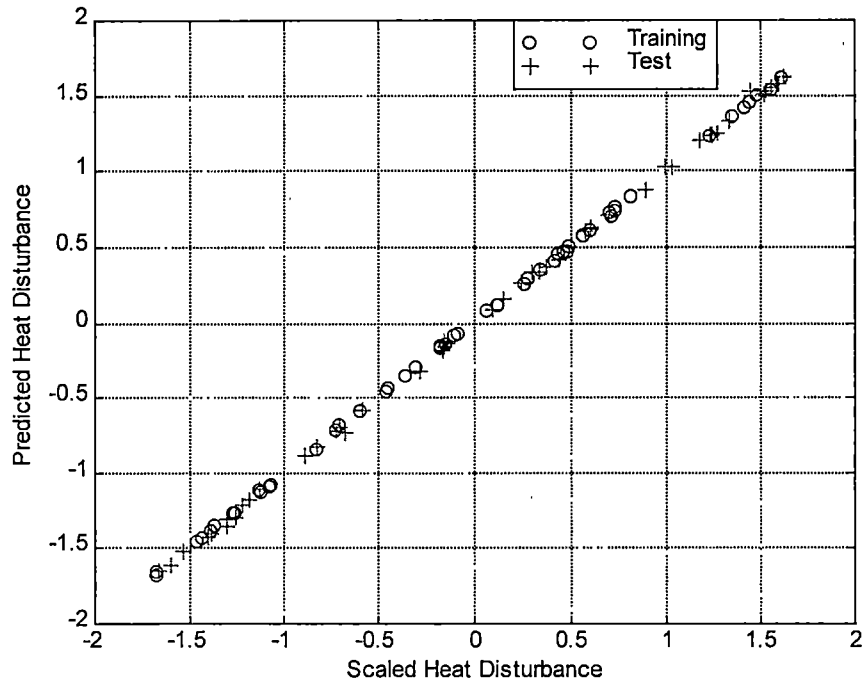


Figure 6.3 Online Predictions of Heat Disturbance Using Autoscaled Scores on 9 Retained PCs of Scale Parameter Matrix

Table 6.3. Online predictions of the disturbances at end of Phase III using partial scores (9 Retained PCs). (Autoscaled Data)

Data Type	Disturbance Type	Training RMSE	Test RMSE
With Linear Scores Only	Heat Transfer	0.0126	0.0193
With Additional Quadratic Terms	Heat Transfer	0.0130	0.0174
With Linear Scores Only	Water	0.0418	0.0469
With Additional Quadratic Terms	Water	0.0247	0.0331

These predictions from the partially completed data show that the disturbances can be reasonably estimated during the batch. As mentioned earlier, these predictions may turn out to be practical in industry. A comparison with online predictions developed using MPCA and MPLS is now done.

Comparison with Predictions from Other Data Mining Methods

The loadings are determined from time-scaled temperature and vapor rate profiles through the end of phase III that are mean-centered and scaled to unit variance, to account for differences in engineering units. Only the non-zero portions of the vapor rate profiles were used in building the model. The second block of predictors used is a matrix of the phase ending times through the end of phase III. Three latent variables (MPLS) and seven retained PCs (MPCR) were used for the predictions. The predictions for each method during the batch are shown in Table 6.4. As shown in Table 6.4, the predictions are better for the scale parameter-based method in this work. As shown in Table 6.4, the online scale parameter-based method of predicting the quality provides better online predictions than those from MPCR and MPLS. As

Table 6.4. Online predictions of the MW at the end of Phase III using MPCR (7 Retained PCs), MPLS (3 Latent Variables), and Scale Parameter-Based Method (9 Retained PCs). (Autoscaled Data)

Method	Training RMSE	Test RMSE
MPCR	0.2543	0.2773
MPLS	0.2107	0.2711
Scale Parameter-Based Method	0.0261	0.0360

from MPCR and MPLS. As mentioned in chapter 5, the scale parameter-based method described in this work also has the advantage that the scale parameters can be interpreted more easily when attempting to understand the physics of the process.

Recommendations for Process Improvement Through Recipe

Adjustments

Since the quality is predictable online and since the scale parameters have been tentatively connected to the physics of the process, directions for process improvement are clearly indicated. If the process has low heat transfer, the data mining study in chapter 5 suggests that more HMD remains in solution to polymerize, resulting in a higher MW. To prevent this overshoot, the heat must be further *reduced* in order to reduce the rate of reaction. A reduced reaction rate means that the polycondensation proceeds slower, resulting in a lower MW when the batch is ended. In this case, the appropriate adjustment handle, or manipulated variable, for improving the process is the jacket pressure setpoint.

If a water disturbance is present, the study in chapter 5 suggests that more HMD will volatilize, resulting in a lower MW. Therefore, more monomer must be added to replace the volatilized HMD. The addition of more HMD means that more monomer is present to replace that lost due to vaporization. Adding HMD raises the achievable MW and allows the target to be attained. In this case, the appropriate adjustment handle for improving the process is the amount of HMD.

However, correcting for the presence of water and low heat transfer is more subtle than simply adding HMD for a water disturbance and reducing the heat for low heat transfer. Interactions between the

disturbance effects must also be considered to give guidance on when to perform the respective corrections for low heat transfer and water disturbances. For example, if low heat transfer occurs, yet a sufficiently high water disturbance is present, the batch may still reach the target. Low heat transfer reduces the amount of monomer that is volatilized, but the presence of water consumes HMD less, allowing more monomer to volatilize. These effects tend to cancel each other. The batches as implied by the data mining study naturally subdivide between those batches that would reach the target without a recipe adjustment, and those that would not. This insight, provided by the data mining study, gives the key to implementing the online recipe adjustments.

Figure 6.4 confirms these effects in the disturbance space with each disturbance combination labeled as to whether the batch reached the target or not. As shown by the figure, a high degree of interaction is present among the disturbances. Note that the disturbance space naturally subdivides into those batches in which the MW reached the target and those that did not. Therefore, this interaction is easily taken into account by a prediction of the MW for a given batch.

The suggested manipulated variables, the jacket pressure setpoint and the amount of HMD, are readily available for performing the recipe adjustments. Furthermore, the appropriate corrective action is suggested by a prediction of the MW. Thus, online recipe adjustments for the process are a practical possibility. A strategy for performing the online recipe adjustments based on the MW is now presented.

A MW-based Approach for Recipe Adjustments

The MW can be predicted before the end of the batch. Furthermore, the appropriate corrective action to apply depends on whether the fixed

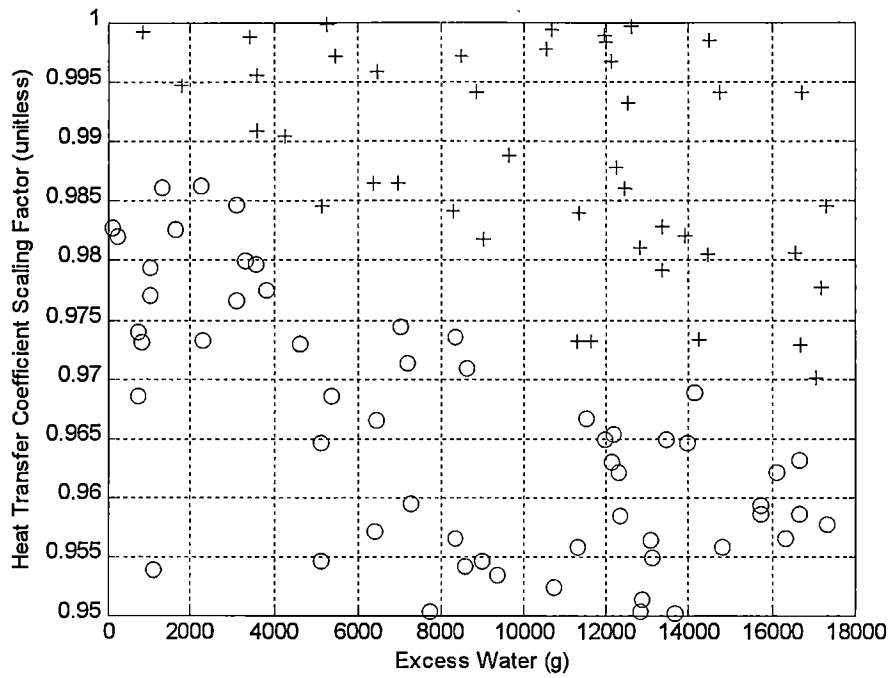


Figure 6.4 Division of Disturbance Space

Where: + = Batches with $MW < MW_{target}$
 o = Batches with $MW \geq MW_{target}$

recipe batch quality will reach the target or not. Therefore, the deviation of the predicted fixed recipe MW from the target MW is a natural choice for performing a recipe adjustment. The simplest recipe adjustment based on such a deviation is a linear correction. Expressed mathematically,

$$\Delta U = K_c * \Delta Y \quad (6.2)$$

Where: ΔU = Calculated change in the manipulated variable, or handle
 K_c = Controller Gain
 ΔY = $MW_{\text{predicted}} - MW_{\text{target}}$

In the absence of a first-principles model, the controller gain must be determined from experimentation, as detailed below.

Since different corrective actions are required depending on the direction of the deviation from the target in the nylon-6,6 process, a split-range controller is proposed to handle each type of deviation. If the deviation is negative, a shortage of HMD is suggested from the data mining study. If the deviation of the predicted MW from the target is positive, a dominant low heat transfer effect is indicated from the data mining study. The appropriate manipulated variables identified above for correcting these deviations are the amount of HMD and the jacket pressure setpoint, respectively.

The split-range controller is proposed mathematically as follows:

$$\Delta HMD = K_{HMD} * \Delta MW_{\text{est}}, MW < \text{target} \quad (6.3)$$

$$\Delta Pj_{\text{sp}} = K_{Pj} * \Delta MW_{\text{est}}, MW \geq \text{target} \quad (6.4)$$

Where: ΔHMD = Amount of HMD to add
 ΔPj_{sp} = Amount to change jacket pressure setpoint
 K_{HMD} = HMD gain
 K_{Pj} = Pj gain
 ΔMW_{est} = $(MW_{\text{predicted}} - MW_{\text{target}})$

Two gains are necessary for the MW deviation-based strategy: 1. The gain to adjust the amount of HMD to be added, if the MW is predicted to be less than the target, and 2. The gain to lower the jacket pressure setpoint, if the MW is predicted to be higher than the target. Since

the methodology for gaining process insight is data driven, experimentation on real batches is the principal option for finding the gains necessary.

Gain Determination

A simple way to provide a reasonable initial estimate for each gain is a process step test. A manipulated variable, U is varied in order to see the effect on the quality, Y . Since good final quality predictions are available, the quality for the given process conditions in a fixed recipe batch is known. The real outcome of the batch reflects the effect of varying U . The process gain, K , can be determined from the step test. The process gain is given by:

$$K = \Delta Y / \Delta U \quad (6.5)$$

Where: $\Delta Y = MW_{\text{predicted}} - MW_{\text{actual}}$
 $\Delta U = U_{\text{adjusted}} - U_{\text{fixed-recipe}}$

The controller gain is related to the process gain by:

$$K_c = 1 / K \quad (6.6)$$

yielding:

$$K_c = \Delta U / \Delta Y \quad (6.7)$$

The controller can then be implemented using the estimated gains. As the controlled batches accumulate, the tuning can be refined by re-estimating the gain(s) from recent batches.

Practically speaking, an estimate of a single gain can be made from a step test on a single batch. Two gains are necessary for the split-range controller, so step tests on two batches are required for initial estimates of K_{Pj} and K_{HMD} . Two step tests are very reasonable in an industrial setting since only two batches need to be altered to obtain an estimate of the controller gain. The best conditions to determine K_{Pj} are during a batch with quality that is predicted to be

greater than the target; i.e., a dominant low heat transfer effect is present. The reason is that a conservative step test should yield improved quality compared to fixed recipe operation, since the disturbance is slightly compensated for by the step test. Similarly, determining K_{HMD} during a batch with predicted quality less than the target should improve the final quality that would result from the fixed recipe. Very little off-spec product would be generated as a result, so plant management can be sold on the idea more readily given that much better quality control may result. In case the controller does happen to fail, there is minimal waste of product involved and insignificant loss of profitability.

The gains are estimated and used in the split-range controller. The linear split-range controller for adjusting the recipe during a batch is implemented to illustrate recipe adjustments for the nylon-6,6 process.

Linear Split-Range Recipe Adjustments

Table 6.5 shows the results of the linear split-range controller for a set of 100 batches having the same disturbances as the fixed recipe. The batch is ended at the fixed hold. The results are compared with the fixed recipe case in Table 6.5. As shown by Table 6.5, the linear split-range controller was able to significantly reduce variability seen in the MW from the fixed recipe case.

Table 6.5. Results of Linear Split-Range Controller

Method	RMSE from Target (= 13,250 g/gmol)
Fixed Recipe	55.19
Linear Split-Range Controller	9.93
Gains Used	
Pressure Gain (mmHg gauge / (g/gmol))	-0.68
HMD Gain (g HMD / (g/gmol))	-4.72

Figure 6.5 shows a plot of the molecular weights obtained from control with those of the fixed recipe case. The reduction in variability is clearly illustrated by the figure.

Extension of the Linear Split-Range Controller

An extension to the linear split-range controller is to incorporate quadratic terms into the prediction of the pressure adjustment. The relationship of the change in jacket pressure to the change in the molecular weight is non-linear. Testing has shown that quadratic MW terms slightly improve the split-range controller's performance. A secondary non-linear effect is explained by the water disturbance. Since the water disturbance used in the process can be reasonably predicted using the scores on PCs 2 and 3, these estimates are available for incorporation into the split-range controller. When both linear and quadratic MW and water disturbance terms are used to create a non-linear split-range controller, the performance noticeably improves, as shown in Table 6.6. Figure 6.6 shows a plot of the molecular weights obtained from control with those of the fixed recipe case. The reduction in variability is clearly illustrated by the figure. The difficulty with implementing the non-linear, split-range controller in industry is twofold. Unless the water disturbance at the

Table 6.6. Results of Non-Linear Split-Range Controller. (Autoscaled Data)

Method	RMSE from Target (= 13,250 g/gmol)
Fixed Recipe	55.19
Non-linear Split-Range Controller	3.67
Regression Coefficients By Term (on Autoscaled Predictors)	
Linear Molecular Weight	0.3702
Non-linear Molecular Weight	-1.0199
Linear Water Disturbance	-0.0324
Non-linear Water Disturbance	0.4842

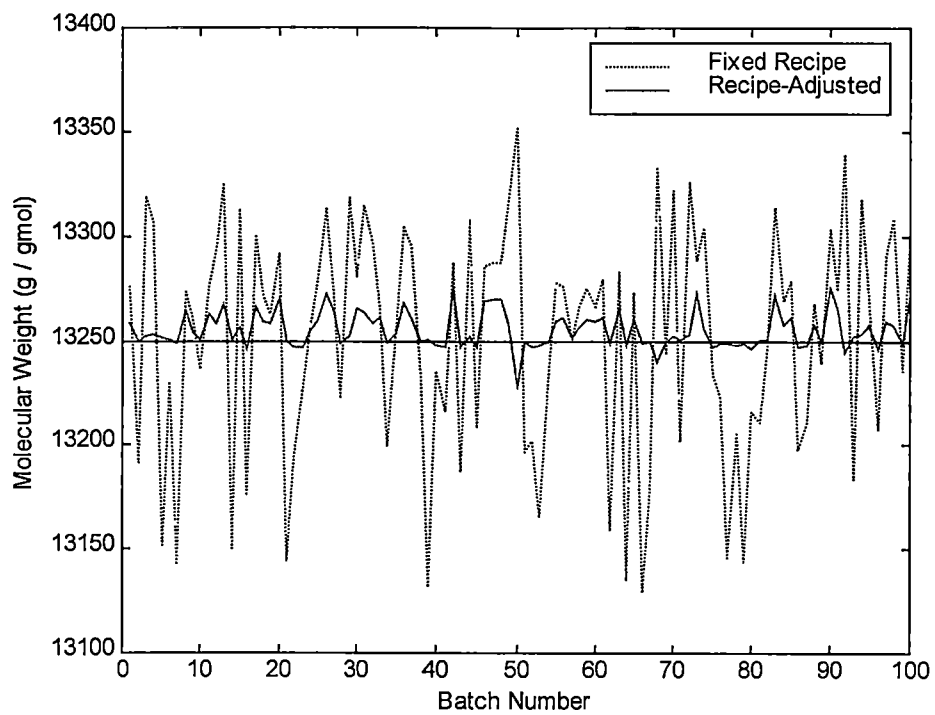


Figure 6.5 Linear Split-Range Control of Molecular Weight

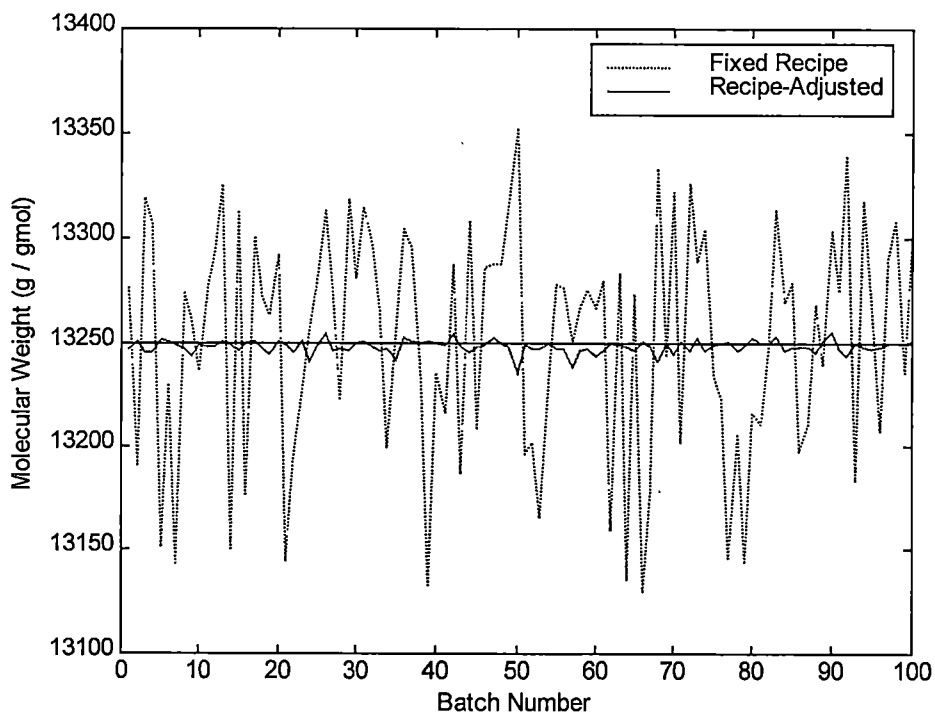


Figure 6.6 Non-linear Split-Range Control of Molecular Weight

beginning of the batch is known, it cannot be confidently predicted during the batch. As mentioned in chapter 5, though, it is reasonable in industry practice to request catching a sample of the reactor contents and measuring for excess water. If samples are collected, a predictive model for the water disturbance can be constructed. Non-linear adjustments may become possible if sufficiently rich data become available. Therefore, until further studies are done, this controller is not practical for industry.

Discussion

From a control perspective, the within batch recipe adjustment strategy is feedforward in nature. That is, the corrective action is based on knowing the product quality that is likely to result from the current batch. In this case, the MW is predicted before the batch ends, and a subsequent recipe adjustment is performed. Further recipe adjustments after a midcourse corrective action are not possible since the correlation structure of the profiles changes due to the recipe adjustment. In order to perform other recipe adjustments subsequent to the midcourse correction, a new predictive quality model is required that incorporates the effects of the various recipe-adjusted batches on the product quality. Such a model for predicting quality allows for further recipe adjustments and is possible as sufficient data is available.

Summary

Online quality predictions are demonstrated well before the batch ends. The scale-parameter method for online predictions outperforms online predictions developed from comparable data mining techniques. Recommendations for improving the process are suggested as well as the

appropriate manipulated variable handles. These handles are readily available, which make online recipe adjustments possible. A practical approach for performing the online recipe adjustments is a split-range controller based on the deviation of the predicted molecular weight from the target molecular weight. The online recipe adjustments performed by the linear split-range controller are able to reduce the variability in the product quality as well as to improve the consistency in reaching the desired target molecular weight. Chapter 7 discusses the next direction for further process improvement: reduction in the batch production time in addition to good quality control.

Chapter 7

Cycle Time Reduction

Introduction

Industry practice for ending batches is to stop the nylon-6,6 batch at a fixed hold time after the depressurization of the reactor. The reactor contents are cured during phase 5 and the batch is ended after this fixed hold time. Therefore, each batch ends at roughly the same time, with any variability in the total duration of the batch being due to the effects of various disturbances on the batch, or from recipe adjustments to the batch. The controller strategy developed in chapter 6 is for the fixed hold case. No further consideration is given to reducing the batch production time. However, an opportunity exists to further improve the process by reducing the batch production time in addition to tight quality control. Pursuing reduced cycle time for the batch results in a departure from the standard industry practice of stopping batches at fixed hold. Specifically, for a fixed recipe, the target quality is exceeded many times due to low heat transfer. Stopping such batches at a fixed hold time after reactor depressurization prevents the possibility of minimizing batch production time.

In this chapter, issues in potentially reducing the batch production time are discussed. An alternative recipe adjustment strategy for reducing the cycle time and the accompanying necessary batch ending strategy are proposed. In developing the ending strategy, information from controlled batches is utilized in estimating the

appropriate ending time for the batch. An extension to the proposed controller is also presented. Before discussing the issues in reducing the batch production time, some further process information concerning the maximum achievable MW in a batch is given.

Peak Molecular Weight

A maximum achievable, or peak, MW is always observed in batches generated by this process. In a batch, the polymer begins to degrade if the cycle time is too long. For batches with quality that either meets or falls short of the target MW, the time that the peak MW occurs corresponds to the minimum possible time that the MW can achieve the target. For example, for a batch with predicted quality that will not reach the target, the addition of sufficient monomer allows the batch quality to achieve the target MW. In such a case, the batch quality has its peak MW at the target MW, and the corresponding peak time is the same as that of a fixed recipe batch which reaches the target, but does not exceed it.

However, for batches with quality that exceeds the target MW, the peak MW is higher and occurs later. The peak MW is higher since more monomer stays in solution to polymerize. The peak MW occurs later because low heat transfer slows the rate of reaction.

An additional side benefit occurs at the peak MW. A secondary quality variable, the amine end group concentration, is found to be minimized whenever the MW reaches its peak. This occurs because, at the peak, the maximum amount of amine ends is consumed in the polymerization. As degradation begins, more monomer returns to the solution as given by (4.2-3).

Issues in Reducing Batch Production Time

A trade off exists between quality and production time. For fixed recipe batches with low heat transfer, the final quality exceeds the target MW. Yet, standard practice is to stop the batch at a fixed hold time after the end of reactor depressurization. If such low heat transfer batches are stopped when the quality reaches the target, quality control can be accomplished along with decreased production time. Furthermore, an additional opportunity to reduce the cycle time is evident, even if a low heat transfer batch is stopped at the target. For such batches, the reaction rate is proceeding slower. In order to reduce the cycle time further, the reaction must be carried out at a faster rate.

To increase the rate of reaction, the temperature must be increased. Increasing the temperature causes the batch to achieve its peak MW earlier. For a precise temperature adjustment, the peak MW occurs at the target MW. Furthermore, this peak MW occurs in the minimum time that the target MW can be reached. However, if the temperature is too high, two effects will prevent the polymer peak MW from reaching the target MW: 1. Too much monomer will be driven off, thus lowering the peak MW, and 2. Thermal degradation will accelerate, also lowering the peak MW. These effects must be considered in implementing a recipe adjustment for reducing the cycle time with tight quality control.

To minimize the batch production time, the batch must be stopped at precisely the point that the MW reaches the target. Thus, an online measurement of the molecular weight must be available. This is seldom the case in industry. However, better instrumentation is being developed that can measure quality in batch processes online [108 - 111]. At present, such instrumentation is either unavailable, or is

expensive to purchase and maintain. Companies that may have access to such instrumentation can utilize the techniques developed in this chapter. Situations may even occur within a company in which an advanced online quality measurement system may be available on limited basis within a particular plant or process unit. In such cases, the methods developed herein apply.

For batches adjusted by increasing the heat, the batch ending strategy is simply to stop the batch whenever the MW reaches the target. As mentioned above, even if the instrumentation is available, it may only be available for limited time for a given process. The strategy outlined below proposes to collect data from batches in which the MW trajectory is measured, and utilize the regular available measurements (temperature, vapor rate, etc.) to predict the time at which the batch should end. Once a good model to predict the ending times is built, the online analyzer may be taken offline. Predictions of the MW are necessary in order to determine if a corrective action is needed for a particular batch. The MW predictions are now considered.

MW Predictions

Predictions of the MW at the fixed hold time are used to decide whether the recipe should be adjusted or not. The reason the hold MW predictions are used is that, unlike the hold MW, the peak MW cannot be predicted from fixed recipe data. The peak MW cannot be predicted because, for batches in which the quality exceeds the target MW, the peak occurs at a later time than the fixed hold. Therefore, the peak MW is unavailable for these batches and so it cannot be predicted.

As demonstrated in chapters 5 and 6, the MW at the fixed hold time is predictable from the measurement profiles normally available. Furthermore, online predictions can be done by the end of phase III.

The alternative recipe adjustment strategy is now presented in detail.

Recipe Adjustments for Reducing Cycle Time

As mentioned earlier, to reduce cycle time, the heat input to the reactor must be increased. As studied in chapters 5 and 6, a shortage of HMD means that the MW cannot achieve the target. Furthermore, sufficiently low heat transfer means that the MW will exceed the target. As formulated, the strategy for reducing the cycle time by increasing the heat input to the reactor applies only to batches in which the MW would have exceeded the target.

For a fixed batch ending time, the strategy used to compensate for batches with quality that exceeds the target is to decrease the heat. This strategy assumes that the batch ends at a fixed time. So, the recipe adjustment strategy calls for decreasing the heat to further slow the reaction in order to force the MW to reach the target *at the fixed hold time*.

The strategy for reducing the cycle time calls for the heat to be *increased*. This is reasonable, since the peak MW in a heat transfer disturbance batch is higher than the target and occurs later. The resultant MW is higher since more HMD remains in solution and polymerizes. The peak MW occurs later since the reaction rate is slower. In effect, by increasing the heat to compensate for such a disturbance, more of the HMD remaining in solution is vaporized, which decreases the maximum achievable MW. Furthermore, the reaction rate is increased, causing the peak to occur sooner.

Cycle time reduction along with tight quality control can be done via a split-range controller for adjusting the batches in midcourse based on the hold MW. This is reasonable since different control actions are taken depending on whether the MW deviation from the target

is positive or negative. If negative, there is a shortage of HMD, so the MW cannot reach the target unless more HMD is added. Since the minimum time possible to reach the target MW occurs at the peak MW for batches with predicted quality at or below the target, the cycle time cannot be further reduced for a shortage of HMD. If the deviation of the predicted MW from the target is positive, a recipe adjustment can be used to reduce the cycle time by increasing the heat input.

The controller is implemented in similar fashion to the linear split-range controller in chapter 6. The deviation of the predicted fixed recipe MW from the target MW is used for adjusting the recipe and is expressed in the equation:

$$\Delta U = K_c * \Delta Y \quad (6.2)$$

Where: ΔU = Calculated change in the manipulated variable
 K_c = Controller Gain
 $\Delta Y = MW_{\text{predicted}} - MW_{\text{target}}$

The split-range controller is as follows:

$$\Delta HMD = K_{HMD} * \Delta MW_{\text{est}}, MW < \text{target} \quad (6.3)$$

$$\Delta P_{jSP} = K_{Pj} * \Delta MW_{\text{est}}, MW \geq \text{target} \quad (6.4)$$

Where: ΔHMD = Amount of HMD to add
 ΔP_{jSP} = Amount to change jacket pressure setpoint
 K_{HMD} = HMD gain
 K_{Pj} = Pj gain
 $\Delta MW_{\text{est}} = (MW_{\text{predicted}} - MW_{\text{target}})$

The selection of appropriate gains for the controller is now considered, before discussing the ending strategy for the heat-adjusted batches.

Gain Selection for the Cycle Time Reduction Split-Range Controller

In considering the gain for the HMD addition in the split-range controller, the time of the peak MW for a nominal batch also corresponds

to the time of the fixed hold (=1.84 hours) past depressurization. Thus, the gain for the HMD addition in the split-range controller for the hold case needs no adjustment. However, the jacket pressure gain requires more attention.

The appropriate jacket pressure gain cannot be found from a simple step test since no guidance is available to select a suitable pressure. The consequences of blindly selecting a high pressure could be severe. Therefore, a search is used to find the gain. The search criteria is that the batch reach the target MW. The reason that the batch must reach the target concerns the implementation of the batch ending strategy. Since the batch MW must be known, an online MW analyzer is necessary to do the search. Because a measurement of the MW is taken, nominal noise is added to the molecular weight profiles in the gain determination.

The search procedure is as follows. A gain of zero is used to start the search; i.e., no jacket pressure adjustment during the batch. If the batch reaches the target, the batch is stopped then and the time is recorded. Subsequently, the gain is conservatively increased and the procedure is repeated. If a batch does not reach the target, then it is stopped at the peak to reduce quality variation.

When a batch does not reach the target, the gain is too high for the given disturbance combination and is reduced to the previous gain. The gain is held at this value for subsequent batches until another batch does not reach the target. The procedure is repeated until no batches fall below the target.

Since the batches are stopped at the target during the test, there will be very little off-spec product generated. This is a significant selling point in getting plant management and operations to buy into

such testing, given that the production time may be decreased while maintaining good quality control.

Discussion

The goal of this procedure is to search for the worst-case gain in the presence of random disturbances. If, however, the disturbances are autocorrelated instead of purely random, further opportunities may exist to reduce the cycle time. In such cases, the best gain should vary with the given disturbance combinations. The best gain could then be found by a continuous search based on the quality results of previous batches. In such a case, the cycle time can be reduced even further.

A second important issue concerns the batch ending strategy in the absence of an analyzer. Since the worst-case gain is found, this procedure provides a basis for accommodating times that the online MW analyzer is available only temporarily. In implementing a complete ending strategy, a method is needed to predict when to stop the batch. Furthermore, even if online MW measurements continue to be available, an estimate of the batch ending time in the cycle time reduction case would provide additional guidance for operating the process. A model can be built to predict the stopping times for future batches from the regular measured variables. The methodology for predicting the batch ending times is now described.

Methodology for Constructing a Predictive Model of the Batch Ending Time

After many batches are collected in which the cycle time is reduced by recipe adjustment of the jacket pressure, the times at which the quality reached the target MW are known. Subsequently, a model can be built to predict the ending times from such batches. The methodology

for building a model to predict the ending times assumes that the quality in all batches actually reaches the target. If the quality of a batch does not reach the target, and if an online analyzer is unavailable, the batch ending strategy cannot accurately predict the appropriate ending time.

The methodology for building the ending time predictive model is to apply Kaistha's method as illustrated in chapter 5 to the profiles of these batches. By including batches with recipe adjustments, the variability introduced to the measurements by the recipe adjustments is captured in the systematic zones of variability. Because the correlation structure of the process is changed in recipe-adjusted batches, new factors and scale parameters for the modified batches must be extracted in building a model for predicting the batch ending time. It is important to note that only heat-adjusted batches should be used in building the ending time prediction model. The reason is that increasing the heat has a different effect on the profiles than the addition of HMD. If the effects of HMD addition on the profiles are studied, a separate model must be built.

The batches are first time-scaled and mean-centered. Figures 7.1 and 7.2 show the raw controlled batch data and the time-scaled, controlled data for heat-adjusted batches in the cycle time reduction case. The mean-centered, time-scaled data, residual errors after projection, and the new factors are shown in Figure 7.3. The effects of the recipe adjustments are captured by the factor(s) in phase IV as seen in Figure 7.3. Since the correlation structure only changes after the recipe adjustments, the factors prior to the beginning of phase IV are the same as in the fixed recipe case.

In the prediction of the batch ending time, only factors through the end of phase IV are considered. This is due to the fact that the

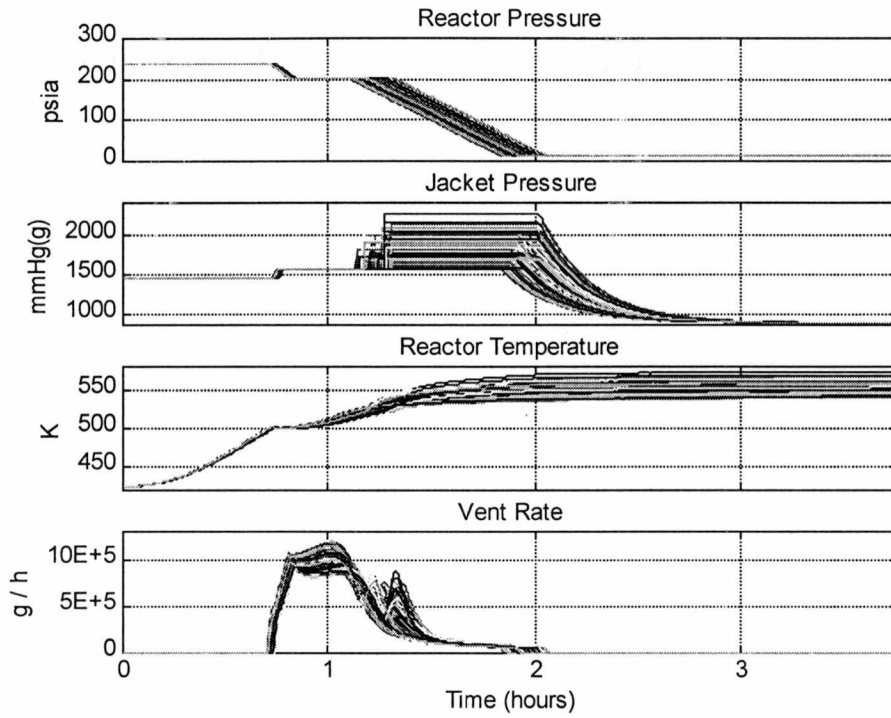


Figure 7.1 Raw Recipe-Adjusted Profiles

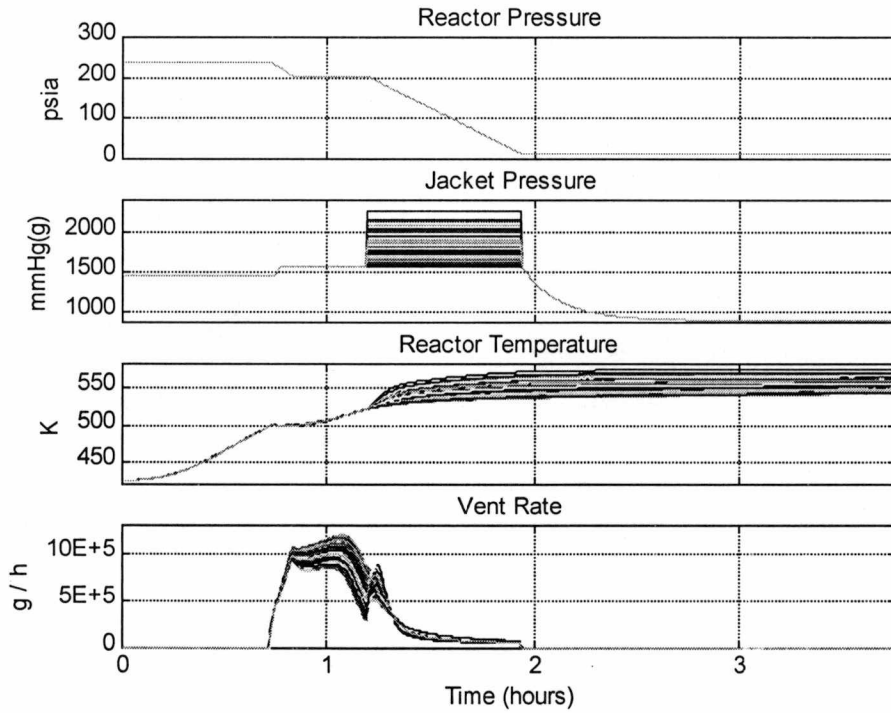


Figure 7.2 Time-Scaled Recipe-Adjusted Profiles

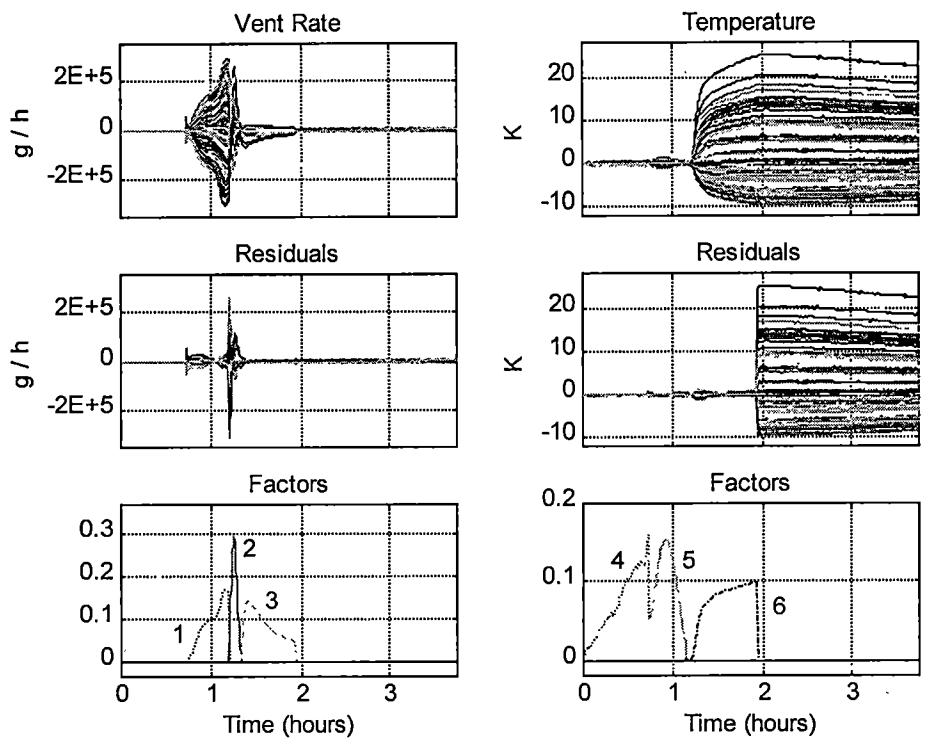


Figure 7.3. Time-scaled, Mean-centered Recipe-Adjusted Profiles, Residual Errors After Projection, and Factors for Vent Rate and Temperature

Table 7.1. Available Scale Parameters by Phase for the Prediction of the Batch Ending Time

Batch Phase	Available Parameters at End of Phase		
	Time Scale (TS)	Vapor Rate (V)	Temperature (T)
Preheat	ts1		ms4
Boiling, 1 st Pressure Decrease	ts1		ms4
Boiling, 2 nd Reactor Pressure Setpoint	ts1, ts2	ms1	ms4, ms5
Boiling, Reactor Depressurization	ts1, ts2	ms1, ms2*, ms3*	ms4, ms5, ms6*
Curing	ts1, ts2	ms1, ms2*, ms3*	ms4, ms5, ms6*

batch ends in phase V. The scale parameters that are available by phase are shown in Table 7.1. The scale parameters labeled with a * reflect projections onto factors that are different from the respective factors seen in chapter 5.

Results of the Cycle Time Reduction Split-Range Controller with Batch Ending Strategy

A PCR is used to develop a predictive model of the known batch ending times from the scores through the end of phase IV. In the prediction, the scale parameters are used as well as the jacket pressure adjustment. The jacket pressure adjustment is included since it is the primary variable affecting the ending time. As shown by Figure 7.4, the predicted values of the batch ending times closely match the actual times. The RMSE statistics are summarized in Table 7.2.

Figure 7.5 shows the quality obtained from the cycle time reduction split-range controller coupled with the batch ending strategy for a set of 100 batches having the same disturbances as the fixed

Table 7.2. Online Predictions of the Batch Ending Times (Autoscaled Data)

Data Type	Training RMSE	Test RMSE
With Linear Scores Only	0.0589	0.0867
With Quadratic Terms	0.0245	0.0357

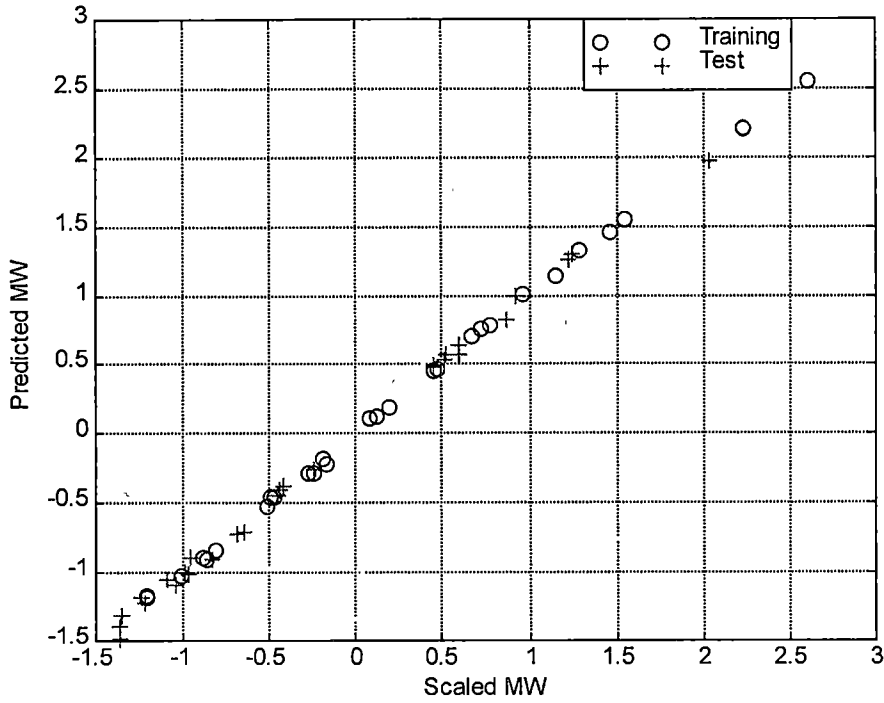


Figure 7.4. Online Predictions of Batch Ending Time with 14 Retained PCs on Autoscaled Data from Scale Parameter Matrix

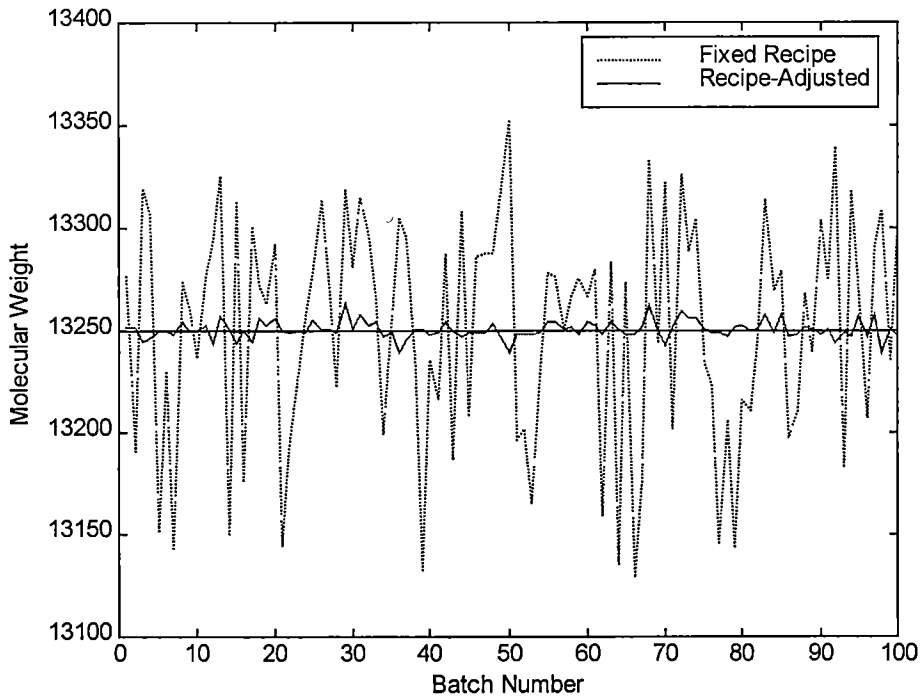


Figure 7.5. Demonstration of Cycle Time Reduction Linear Split-Range Controller

recipe. Table 7.3 shows the RMSE statistics. As shown by the figure and table, the alternative recipe adjustment strategy controls the quality very well.

Table 7.4 shows the reduction in cycle time for the heat-adjusted batches. Since the cycle time for batches requiring the addition of HMD cannot be further reduced, the net effect of the cycle time reduction will be diluted. For a typical set of batches including both heat-adjustments as well as HMD adjustments, Table 7.5 shows the net effect of decreasing the cycle time for the heat-adjusted batches on the average time to completion for all batches.

As shown by Tables 7.4 and 7.5, the split-range controller was able to reduce the cycle time by around 50 minutes for those batches which would have exceeded the target MW, and around 30 minutes for a typical set of batches with both types of recipe adjustments. Thus, the cycle time reduction split-range controller coupled with the batch ending strategy is a practical alternative to the hold strategy.

Summary

An alternative recipe adjustment strategy for reducing the cycle time and the accompanying batch ending strategy is demonstrated by utilizing online MW measurements. In developing the batch ending strategy, information from controlled batches is utilized in estimating the appropriate ending time for the batch. The controller performs very well in reducing the variability seen in the final quality as compared to the fixed recipe. Lastly, the cycle time is significantly reduced using a conservative gain setting.

Table 7.3. Results of Cycle Time Reduction Linear Split-Range Controller

Method	RMSE from Target (=13,250 g/gmol)
Fixed Recipe	55.18
Linear Split-Range Controller with Batch Ending Time Predictions	4.58
Gains Used	
Pressure Gain (mmHg gauge / (g/gmol))	6.60
HMD Gain (g HMD / (g/gmol))	-4.72

Table 7.4. Reduction in Cycle Time for Heat-Adjusted Batches

Reduction for Batches Requiring Heat Manipulation Only	Average Ending Time (hours)
Fixed Recipe	3.80
Cycle Time Reduction	2.90
Percentage Reduction in Cycle Time	
	23.68

Table 7.5. Overall Reduction in Cycle Time

Overall Reduction (All Batches)	Average Ending Time (hours)
Fixed Recipe	3.78
Cycle Time Reduction	3.25
Percentage Reduction in Cycle Time	
	14.02

Chapter 8

Summary and Future Work

Summary and Conclusions

In this work, a historical database of past batches generated from a nylon-6,6 simulation is subjected to data mining techniques and the results are interpreted using process knowledge and engineering judgment. The analysis of the database results in a greater understanding of the principal sources of common cause variability affecting the process. This understanding leads directly to process improvement by suggesting appropriate corrective actions to compensate for the sources of variability affecting the process. The relevant quality parameter, the MW, is shown to be predictable well before the batch concludes which allows midcourse recipe adjustment strategies to become a practical possibility.

The process is improved by implementing the suggested appropriate corrective actions as a within batch recipe adjustment based on the deviation of the predicted quality from a fixed recipe with the desired target. Different control actions are required depending on whether the deviation is positive or negative. The resultant approach to the recipe adjustment strategy is a linear split-range controller that takes appropriate corrective action depending on the direction of the deviation. The strategy is demonstrated on the nylon-6,6 process for a fixed batch ending time.

An alternative approach to performing recipe adjustments is implemented in order to further improve the process by reducing the

batch cycle time in addition to tightly controlling quality. This approach is developed assuming the availability of online MW measurements which is an emerging area of research and development. The recipe adjustments are coupled with the necessary batch ending strategy, in which prior controlled batches are used in constructing a model to predict the appropriate batch ending time. The strategy demonstrates that the cycle time can be reduced by increasing the heat to the nylon-6,6 reactor and stopping the batch at the appropriate time. The ending time is variable in this case.

Thus, data mining coupled with engineering judgment and process knowledge is demonstrated to be a systematic approach to examining a historical database of past batches for the purpose of greater process understanding leading to recommended directions for process improvement using within batch control. It is emphasized that the recipe adjustment strategies were developed using only fixed recipe data; that is, no prior control moves are reflected in the dataset. Both within batch control strategies prove to be very successful in improving the example nylon-6,6 process.

It must be mentioned that, even though demonstrated on a specific process, the methods used to study the historical database are general in nature. These methods, when coupled with process specific knowledge, can be applied to a wide variety of batch processes, not limited to reactive systems.

Future Directions for Research

The recommended improvements to the process are proposed in a within batch structure; that is, the corrective actions are performed based on the current batch only for uncorrelated disturbances. Consideration needs to be given to autocorrelation in the disturbances

that are present. The scope of the recipe adjustments can, therefore, be broadened to include both batch-to-batch adjustments and within batch adjustments. An integrated framework to integrate both types of adjustments is a clear future direction to this research. The framework should be developed by utilizing the same approach to process improvement as that described herein: data mining coupled with process knowledge and engineering judgment leading to process improvement.

A second area of research that remains to be explored is multivariable control. In addition to the MW, other quality measurements are used to measure the performance of the nylon-6,6 process, such as the concentration of amine end groups and the viscosity. Minimization of the amine end group concentration was a by-product of the cycle time reduction strategy, given that the peak MW occurs at the target. However, full multivariable control was not investigated.

For the nylon-6,6 process, an area to explore involves the availability of online molecular weight measurements. The availability of such measurements provides an additional profile to analyze using Kaistha's method. Scale parameters can be easily obtained from MW profiles and studied for insight into the process. Furthermore, since a MW profile is available, alternative control schemes can be explored. As an example, the batch could be forced to match a reference MW trajectory through continuous or discrete manipulations of an appropriate handle(s).

Additional consideration needs to be given to an ending strategy that is robust to batches with quality that does not reach the target. The current strategy was accomplished by using a conservative gain setting which reflects the worst-case gain in which no batches are allowed to fall below the target. Improvements in the batch ending

strategy provide a basis to optimize the gain for both uncorrelated and autocorrelated disturbances.

The current ending strategy assumes that uncorrelated disturbances are present. However, further reduction in the cycle time should be possible in batches with autocorrelated disturbances. In this case, the autocorrelation in the disturbances may be useful in developing an online strategy to find the optimal gain for cycle time reduction.

Two disturbances are investigated in this work. However, many disturbances occur in real processes. Additional disturbances known to affect the process should be enumerated and studied to understand their effects on the process. Special attention must be given to the selection of appropriate recipe adjustments that do not adversely affect the quality in the presence of multiple disturbances.

Finally, disturbance based control can be investigated. As discussed, good disturbance predictions can be made during the batch, if sufficient data are available to construct a predictive model. Disturbance based control is conceptually and practically familiar in industry and provides a more intuitive basis for control. Care must be taken, though, to prevent overcontrol, since the effects of low heat transfer and excess water tend to cancel each other in certain regions of the disturbance space.

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APPENDIX

Appendix

Nylon-6,6 Reaction Model

This appendix gives the details of the nylon-6,6 simulation that is adapted from Robertson et al [81] and Russell [83]. The work of Stepan et al [102,103] is also cited.

The main reaction given in chapter 3 is a reversible, second order formation of nylon-6,6 polymer and water from both amine and carboxyl groups:



Where: A = Amine group derived from a component of the reaction, hexamethylene diamine (HMD).
C = Carboxyl group derived from a component of the reaction, adipic acid (AA).
L = Polymer link.
W = Water molecule.

Additional side reactions modeling degradation or depolymerization of the polymer are given as:



Where: SE = stabilized end groups (cyclized) [104-105].

The kinetics of the simulation are based on the work of Stepan et al [102-103]. In the first work [102], an activity-based kinetic model of the nylon 6,6 polymerization reaction is given. In developing equilibrium and rate constants, the equal-reactivity assumption is used. In the equal-reactivity assumption, each end-group is assumed to react at the same rate independent of the size of the end-group [102]. However, the authors also note that the nylon 6,6 solution is highly non-ideal and this fact is accounted for in the model by composition

dependence on the mole fraction of water. The reaction rate expressions for the above equations (4.1-4.3) are as follows:

$$R_1 = CT k_1 X_c \quad (A.1)$$

$$R_2 = X_L (k_2 + k_{2C} X_A) \quad (A.2)$$

$$R_3 = CT k_{app} \left(X_A X_c - \frac{X_L X_W}{K_{app}} \right) \quad (A.3)$$

Where: R_i = Reaction rate (gmol / h) of reaction i.
 $CT = C_A + C_c + C_L + C_W + C_{SE}$.
 X_j = Mole fraction of component j given by [102] $X_j = C_j / CT$.
 k_k = Kinetic parameter ($k = 1, 2, 2C$) (h^{-1}).

The apparent rate constant, k_{app} (gmol / total gmol-h), and the apparent equilibrium constant, K_{app} (dimensionless), are given by empirical relations derived from various experimental data. K_{app} depends on the apparent heat of reaction, ΔH_{app} (cal / gmol). The apparent heat of reaction is expressed in the following equation:

$$\Delta H_{app} = \underline{\Delta H_L} + \underline{\Delta H_W} - \underline{\Delta H_A} - \underline{\Delta H_C} \quad (A.4)$$

Where: ΔH_{app} = The apparent heat of reaction (cal / gmol).
 $\underline{\Delta H_i}$ = Partial molar heat of mixing of component i in the liquid phase.

An empirical relationship for the heat of reaction that expresses the non-ideality of the solution environment is developed from experimental data. The heat of reaction is found to be a function of the mole fraction of water present. The expression is given as:

$$\Delta H_{app} = 7650 \tanh[6.5(x_w - 0.52)] + 6500 \exp\left(-\frac{x_w}{0.065}\right) - 800 \quad (A.5)$$

Where: x_w = Mole fraction water.

The main reaction appears to be endothermic for $x_w > 0.52$, and exothermic at lower concentrations [102].

Material Balances

The material balance relations are given below:

$$\frac{dVC_A}{dt} = VR_2 - VR_3 - 2 \frac{\omega_{HMD}^e}{M_{HMD}} v \quad (A.6)$$

$$\frac{dVC_C}{dt} = -VR_3 - VR_1 \quad (A.7)$$

$$\frac{dVC_L}{dt} = VR_3 - VR_2 \quad (A.8)$$

$$\frac{dVC_w}{dt} = VR_3 + VR_1 - \frac{\omega_w^e}{M_w} v \quad (A.9)$$

$$\frac{dVC_{SE}}{dt} = VR_1 + VR_2 \quad (A.10)$$

$$\frac{d\rho V}{dt} = -v \quad (A.11)$$

Where:

ω_i^e = The equilibrium mass fraction of component i in the vapor phase

M_i = The molecular weight of component i (g / gmol).

$$v = K(P - P_{vap}) = \text{Mass vaporized}, \quad (A.12)$$

with $K = 25,000$ gmol / psi-h, P = reactor pressure (psia), and P_{vap} the vapor pressure above the reaction mixture (psia).

The heat balance is given by:

$$\frac{dT}{dt} = -\frac{\Delta H_{app}}{\rho C_p} R_3 - \frac{\Delta H_v^{HMD} v_{HMD}^M}{\rho C_p V} - \frac{\Delta H_v^w v_w^M}{\rho C_p V} + \frac{Q_{heat}}{\rho C_p V} \quad (A.13)$$

Where: T = Temperature (K)

ΔH_v^i = heat of vaporization of component i (cal / gmol).

v_i^M = molar vaporization rate of component i (gmol / h).

ρ = density (g / l); inverse of specific volume.

C_p = heat capacity (cal / g-°C).

In this research, a cylindrical tank of equal height / diameter ratio is assumed. The diameter was calculated by setting the initial volume of the reaction mixture equal to the volume of the cylinder, and solving for the diameter. Expressions for the specific volumes of water, polymer, and the solution of water and polymer are supplied in [83]. Two vapor pressure relations are given, with the larger of the two

being kept as the estimate for the vapor pressure of the water / HMD mixture.

Initial Concentrations

The initial reaction mixture is 20% by weight water and 80% by weight equimolar adipic acid and HMD. The initial weights used are 350 kg water and 1,400 kg of the equimolar mixture [105]. To account for the loss of HMD due to vaporization during the reaction, an additional 1 kg of HMD is added.

Quality Model

Three variables are given for the M_n quality model: the feed ratio, r , of carboxyl groups to amine ends, the reaction extent, p (or ϵ), and the fraction, n_1 , of polymer molecules that are HMD. The feed ratio is set up in terms of the limiting end group. If r is less than one (i.e., more amine than carboxyls), then one set of relations for p and n_1 is used. However, when enough HMD vaporizes during the reaction, and r becomes greater than one, r is recalculated by a different relation and two different relations are used to calculate p and n_1 . Subsequently, M_n is given as follows:

$$M_n = \frac{M_0(1+r)}{(1+r-2rp)} \quad (\text{A.14})$$

Where: M_n is the number average molecular weight (g / gmol).
 M_0 is the mass of one unit of the polymer chain (g / gmol).

The quality parameter model for M_n is explained in greater detail in [81,83].

Vita

The author was born in Johnson City, Tennessee, on November 2, 1969. He attended Daniel Boone High School in Gray, Tennessee, from 1984-1988. In September of that same year, he entered Tennessee Technological University and graduated in 1993 with a B.S. in Chemical Engineering. In June of that same year, he went to work for Amoco Chemical Company as an engineer. In August of 1996, he left the company to pursue a degree at the University of Tennessee and graduated in August of 2001. The author and his family then moved to Baton Rouge, Louisiana, where he went to work for ExxonMobil.