Control of Solution MMA Polymerization in a CSTR

by

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ABSTRACT

Simulation studies on the on-line control of polymerization reactors are numerous, and have been extensively employed. Proportional-integral-derivative (PID) controllers have been and continue to be in wide spread use throughout industry. This thesis work was to review the literature on the control of polymerization reactors, and to experimentally verify a novel PID controller tuning method on a well studied polymerization system.

A continuous stirred tank reactor (CSTR) system for the solution homopolymerization of methyl methacrylate (MMA) in toluene was designed and built by a past Ph.D. student (Chien, 1992). He also developed a mechanistic model for the process. Several other students have studied system identification and control of this system.

Recently, Wang, Barnes and Cluett (1995a) proposed a novel PID controller design method, which will be referred to as the W-C method from now on. The W-C method could be applied to a PID conversion controller on the CSTR and compared to Chien's results.

PID control of conversion with differently tuned PID controllers was first simulated. Next, PID controllers tuned using the internal model control (IMC) tuning method and the W-C method were applied in a long experimental run to verify the simulation results.

The CSTR process had changed little in the five years since Chien's work. The IMC tuned controller was fast and able to control conversion at the desirable setpoints and recover from disturbances. The W-C tuned controller had a controller output signal as predicted by the simulations, and also controlled and recovered from disturbances well.

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TABLE OF CONTENTS

CHAPTER 1: INTRODUCTION 1

CHAPTER 2: CONTROL OF POLYMERIZATION REACTORS – AN UPDATED REVIEW 4

2.1 Introduction 4

2.2 Optimization 7
2.2.1 Periodic Operation 9
2.2.2 Steady-state Optimization 9
2.2.3 Optimal Start-up/Changeover Policies 9
2.2.4 Reactor Design 10
2.2.5 Batch Optimization 10

2.3 Modelling in Polymer Reactor Control 12

2.4 Online Monitoring 19
2.4.1 Monitoring Polymer Reactor Operation and Measuring Polymer Properties On-line 23
2.4.2 Product Property Trajectory Tracking 23

2.5 Batch and Semibatch Reactor Control 26
2.5.1 Molecular Weight Distribution Control 29
2.5.2 Conversion and Composition Control 32
2.5.3 Long Chain Branching Control 34
2.5.4 Particle Size Distribution (PSD) Control 35
2.5.5 Temperature Control 35

2.6 Continuous Reactor Control 39
2.6.1 Steady State Optimization 41
2.6.2 Dynamic Optimization 41
2.6.3 Start-Ups and Change of Grades 42
2.6.4 Reactor Design Optimization 42
2.6.5 Temperature Control 43
2.6.6 State Estimation / Filtering 45
2.6.7 Recent Efforts Towards On-Line Control 47

CHAPTER 3: PID CONVERSION CONTROL -- CONTROLLER TUNING AND SIMULATION STUDY 55

3.1 Introduction 55

3.2 Literature Survey of PID Controller Design 56
3.2.1 Performance Measures 58
3.2.2 Controller Selection 59
3.2.3 Tuning with Fundamental Models 59
3.2.4 Tuning with Approximate Models 60
3.2.5 Tuning with Frequency Response Models 61
3.2.6 Tuning without Models 63

3.3 Theoretical Background 64
3.3.1 Process Description 64
3.3.2 Wang and Cluett (W-C) Tuning Method 65
3.3.3 Controller Tuning 67
3.3.4 PID Simulation 70

3.4 Simulation Results 73
3.4.1 Open Loop Results 73
3.4.2 Closed Loop Results 75
3.4.3 Evaluation 85

3.5 Discussion of Simulation Results 86

3.6 Concluding Remarks 89

CHAPTER 4: EXPERIMENTAL VERIFICATION OF A PID CONTROL TUNING METHOD FOR CONVERSION IN A SOLUTION MMA CSTR 90

4.1 Introduction 90

4.2 Experimental System 91
4.2.1 Apparatus and Materials 93
4.2.2 I/O Interface and Process Computer 95
4.2.3 Operating Conditions and Procedures 97
4.2.4 Temperature Controller 98
4.2.5 Conversion Controller 99

4.3 Methodology 100
4.3.1 Experimental Run Objectives 100
4.3.2 Experimental Run Procedures 101
4.3.3 On-line Data Collection 102
4.3.4 Off-line Measurements 103

4.4 Experimental Results 104
4.4.1 Open Loop Results 104
4.4.2 Closed Loop Results 106
4.4.3 Off-line Results 110

4.5 Discussion of Results 113
4.5.1 Comparison of Chien (1992) Model with Experimental Results 113
4.5.2 Transfer Function Model Parameters 117
4.5.3 Quality of Control 125
4.5.4 Control Signal Specification 130
4.5.5 Off-line Gravimetry 132
4.5.6 Molecular Weight Measurement 132

4.6 Concluding Remarks 133

CHAPTER 5: CONCLUSIONS AND RECOMMENDATIONS 134

APPENDIX A: ABBREVIATIONS AND NOMENCLATURE 137

APPENDIX B: PROGRAM LISTINGS 140

BIBLIOGRAPHY 141

LIST OF TABLES

Table 2.1: Chronological list of past review papers on polymerization reactor control 6
Table 2.2: Recent work on optimization of polymer reactor systems 8
Table 2.3: Recent work on modelling issues related to polymer reactor control 17
Table 2.4: Recent work on modelling specific polymer reactor systems 18
Table 2.5: Recent work on monitoring polymer reactor operation and on-line polymer property measurements 21
Table 2.6: Recent work on state estimation and data filtering for polymer reactors 22
Table 2.7: Recent work on control of batch and semibatch polymer reactor systems 28
Table 2.8: Recent work on control of continuous polymer reactor systems 40
Table 3.1: Recent literature on the design and implementation of PID controllers 57
Table 3.2: Tuning constants for PID controller tuned using W-C rules 66
Table 3.3: Tuning constants for PID controller tuned by various tuning rules 69
Table 3.4: Time-integral performance measures of the simulated closed loop responses to setpoint change and load disturbances 85
Table 4.1: Opto-22 I/O module summary 95
Table 4.2: Nominal operating conditions 97
Table 4.3: Run procedure summary 101
Table 4.4: Sum of squared errors for different parameter estimates for the model 119
Table 4.5: Closed loop mode experimental performance measures 127

LIST OF FIGURES

Figure 3.1: Simulated open loop process response to a 2 ml/min increase in AIBN flow rate 74
Figure 3.2: Simulated closed loop process response with W-C tuned (α = 0.5) controller 76
Figure 3.3: Simulated closed loop process response with W-C tuned (α = 1.5) controller 77
Figure 3.4: Simulated closed loop process response with W-C tuned (α = 10.0) controller 78
Figure 3.5: Simulated closed loop process response with Z-N tuned controller 79
Figure 3.6: Simulated closed loop process response with C-C tuned controller 80
Figure 3.7: Simulated closed loop process response with ITAE tuned (setpoint change) controller 81
Figure 3.8: Simulated closed loop process response with ITAE tuned (disturbance rejection) controller 82
Figure 3.9: Simulated closed loop process response with IMC (λ/θ = 0.25) tuned controller 83
Figure 3.10: Simulated closed loop process response with IMC tuned (λ/θ = 0.9) controller 84
Figure 3.11: Comparison of closed loop controller performance by ISE 87
Figure 3.12: Comparison of closed loop controller performance by ITAE 88
Figure 4.1: Schematic of experimental apparatus 92
Figure 4.2: CSTR control program flow sheet 96
Figure 4.3: Conversion control block diagram 99
Figure 4.4: Open loop response of conversion process 105
Figure 4.5: Conversion control with W-C tuned controller 107
Figure 4.6: Conversion control with IMC tuned controller 109
Figure 4.7: Off-line conversion measurement 111
Figure 4.8: Off-line molecular weight measurement 112
Figure 4.9: Comparison of simulation with Chien (1992) model with open loop experimental results 114
Figure 4.10: Comparison of simulation with Chien (1992) model with W-C segment of closed loop experimental results 115
Figure 4.11: Comparison of simulation with Chien (1992) model with IMC segment of closed loop experimental results 116
Figure 4.12: Comparison of the Chien (1992) model with experimental results 120
Figure 4.13: Comparison of the open loop estimated model with experimental results 121
Figure 4.14: Comparison of the mean conversion adjusted model with experimental results 122
Figure 4.15: Comparison of gain adjusted model with experimental results 123
Figure 4.16: Comparison of dead time adjusted model with experimental results 124
Figure 4.17: Closed loop data regions for performance measure 126
Figure 4.18: Comparison of closed loop controller performance by ISE 128
Figure 4.19: Comparison of closed loop controller performance by ITAE 129

CHAPTER 1: INTRODUCTION

Recent literature available on the on-line control of polymerization reactors is dominated by simulations instead of experimental results. For polymer reactor control, effective algorithms require:

- a reliable model
- knowledge of the polymerization behaviour
- dependable on-line sensors
- good control theory and methods
- a process control data acquisition and computing system

This thesis begins with a literature review in these areas. Chapter 2 is a survey of recent papers after 1992, specific to the control of polymerization reactors. It is fashioned as an update to the review paper by MacGregor, Penlidis and Hamielec (1984) and builds on Chien (1992).

Methyl methacrylate (MMA) polymerization has industrial importance, its kinetics have been well studied and there is good documentation in the open literature. An on-line densitometer to measure the conversion of monomer to polymer was available. Proportional-integral-derivative (PID) controllers continue to be in widespread industrial use. Therefore, on-line PID control of MMA solution polymerization was chosen for this study.

The objective was to experimentally verify a novel PID controller tuning technique proposed by Wang, Barnes and Cluett (1995a), but not yet tested with an on-line polymerization control system. The controlled variable would be monomer conversion - or product quantity. Product quality (molecular weight) would be measured but not controlled.

Chapter 3 is a simulation study of the PID tuning method applied to the CSTR to predict expected behaviour as well as identify comparable PID tuning methods.

Chapter 4 covers experimental verification of the proposed PID controller tuning method as applied to conversion control of solution MMA polymerization in a CSTR. Simulations showed that the process could be controlled by a PID controller tuned using the Wang et al. method. Comparisons were made with an IMC tuned PID controller.

The laboratory, bench-scale, continuously stirred tank reactor (CSTR) system is used for studying solution polymerization of methyl methacrylate (MMA) in toluene with 2,2'-azobisisobutyronitrile (AIBN) initiator. Four graduate students have simulated and experimentally verified modelling and control of the system.

Chien (1992) completed theoretical and practical work that has been the basis for much of the subsequent work. He conducted an extensive literature review and developed a mechanistic model for the solution polymerization of MMA in a CSTR that included reactive impurities and predicted polymerization behaviour and important polymer properties. He also designed and built the automated, lab-scale CSTR polymerization system to experimentally verify the promising control studies identified in his simulation runs.

Conventional proportional-integral-derivative (PID) and stochastic control strategies were explored. Conventional PID strategies also included Smith Predictor and Dahlin's control. Specific stochastic control strategies were: unconstrained minimum variance control (MVC), constrained MVC , and one-step optimal control.

Ogunye (1994) emphasized computer-based linear algebra and simulation of different control strategies.

He extended the previously single input single output (SISO) control of conversion by manipulated initiator feed rate, to multiple input multiple output (MIMO) control by including on-line molecular weight estimation and manipulating reactor temperature. Ogunye also formulated a practical solution to input saturation and controller wind-up in advanced digital controllers.

Multivariable controllers studied were: linear quadratic Gaussian (LQG) which was ineffective and internal model control (IMC) which was more flexible. Both LQG and PID were effective in simulation runs and experiments confirmed the results.

Lawrence (1994) modelled the multivariable process and compared further application of SISO and MIMO control strategies. Her work in system identification and control investigated both open- and closed-loop behaviour of the reactor in terms of productivity (conversion) and quality (molecular weight). Two correlations were developed relating measured reactor variables: reactor or product temperature and solution viscosity to conversion and weight average molecular weight.

Using the correlations Lawrence implemented two model-based control algorithms using initiator flow rate and reactor temperature as the manipulated variables. The two controllers were: generalized predictive control (GPC) and model predictive control (MPC). GPC and MPC were implemented at the simulation

level and two SISO GPC controllers were verified experimentally. SISO GPC and MPC control produced similar results. The GPC control was less drastic and had smoother manipulated variable action than the MPC. GPC performed better for both disturbance rejection and set-point changed. MPC was easier to implement in the multivariable case.

Mutha (1996) presented a modified extended Kalman filter (EKF) and a non-linear model predictive control (NLMPC) algorithm. The fixed-lag smoothing-based EKF uses measurements multiple times to achieve good convergence and robustness to state and measurement noise.

The NLMPC was developed for the more difficult case of control non-affine (CNA) systems. Temperature affects the system nonlinearly through the Arrhenius relationship of the reaction rate constants so the system is CNA.

Both techniques were applied to a simulated acrylonitrile/butadiene batch emulsion copolymerization (NBR) system and the experimental continuous solution homopolymerization of methyl methacrylate (MMA) system.

A new control program was run under QNX, a real-time, multi-processing, UNIX-based operating system. This implementation was different from the previous work which was done in DOS using a single QuickBASIC program.

CHAPTER 2: CONTROL OF POLYMERIZATION REACTORS – AN UPDATED REVIEW

2.1 Introduction

This chapter is an updated review of polymer reactor control. It is modelled after MacGregor et al. (1984) and builds on Chien (1992).

Polymerization systems are characterized by many complex, non-linear, interacting phenomena, and a great deal of effort has been invested in the development and application of modern control techniques that can cope with the difficulties inherent in such complex processes.

With the advances in present-day computers, direct digital control (DDC) has evolved to a more mature stage for practical applications. On-line monitoring of process variables coupled with state estimation techniques and control laws that may have adaptive parts are examples of the complexity and capacity that a computer is called upon with which to deal.

Numerous publications on modelling, optimization, and control of polymerization processes can be found in the open literature, especially in the last twenty years or so. These can be further classified into papers dealing with homogeneous or heterogeneous polymerizations, including various reactor types (batch, semibatch, and continuous). However, as mentioned earlier, the great majority of these papers deal with the simulation stage.

There are only a few review papers on recent developments in the area of polymer reactor control, which have appeared since the MacGregor et al. (1984) review. These papers are summarized in Table 2.1 and discussed below.

Ray (1985) illustrates the principal difficulties in achieving good control of polymerization reactors. These are related to inadequate, or even unavailable, on-line measurements, a lack of understanding of the dynamics of the process, the overly sensitive and nonlinear behaviour, which is strongly dependent on the operating conditions of these reactors, and the lack of well-developed techniques for the control of nonlinear processes.

Elicabe and Meira (1988b) emphasize the problems from the point of view of optimal control, in an attempt to classify the tools that modern control theory has to offer. They cover in detail issues related to the formulation of meaningful control objectives, and discuss problems with process dynamics and

measurements. Then, a review of the solutions attempted with respect to state estimation and control is provided. In their review, considerable emphasis has been put on optimal and adaptive control strategies.

MacGregor (1986) is concerned more with the control of polymer quality and how it is related to other process variables. He points out that the presence of stochastic disturbances (usually varying amounts of reactive impurities) needs to be fully appreciated and somehow modelled, as to its final effect on polymer production and quality.

An industrial perspective on reactor control has been presented by Schnelle and Richards (1986) and Richards and Schnelle (1988). They point out that the trend in both literature and industry is to view reactor control as a multivariable problem, while modelling and control system analysis are becoming standard parts of the general reactor control design methodology.

Schork, Deshpande and Leffew (1993) have written a textbook on the control of polymerization reactors. They cover from basic kinetics of polymerization, through single-loop, multi-loop and non-linear control strategies to polymer processing. Important aspects of polymer reactor engineering, details of problems with measurement and estimation, and control strategies are covered.

Dimitratos, Eliçabe and Geogakis (1994) review the major issues related to control of emulsion polymerization – the process, modelling, sensors and control techniques. They place an emphasis on reviewing control relevant literature.

Embiriçu, Lima and Pinto (1996) surveyed recent papers in advanced control of polymerization reactors. Emphasis was on optimal control theory, nonlinear control, adaptive control and predictive control. The review also covered optimization and state estimation for polymerization systems.

Abbreviations and symbols in tables are explained in Appendix A. "Work" can be algorithmic, mathematical modelling, or other theoretical work (T), simulation or computer programs (S), and experimental, industrial or applications (X).

Table 2.1: Chronological list of past review papers on polymerization reactor control

Reference	Notes
Amrehn (1977)	emphasis on digital computer control for reactor environment
MacGregor et al. (1984)	large review; emphasis on polymer properties
Ray (1986)	ACC 1985; difficulties in control & some techniques to apply
MacGregor (1986)	polymer quality
Schnelle & Richards (1986)	industrial perspective
Eliçabe & Meira (1988b)	estimation and control; emphasis on optimal control
Richards & Schnelle (1988)	hierarchical building block approach to industrial reactor control
Ray (1989)	computer-aided monitoring, design and control
Chien (1992)	Chapter 6 in Ph.D. thesis
Schork et al. (1993)	textbook on control of polymerization
Dimitratos et al. (1994)	control of emulsion polymerization reactors
Embiruçu et al. (1996)	survey of advanced control of polymerization reactors

2.2 Optimization

Tieu, Cluett and Penlidis (1994) reviewed optimization of polymer reactor operation. Table 2.2 lists recent work that updates the review with information generally published since 1994.

There is some overlap of this section with the sections on batch/semi-batch and continuous control. Papers listed here are less specific to a particular polymer system or type of operation.

Table 2.2: Recent work on optimization of polymer reactor systems

Reference	System	Work	Notes
Brooks (1997)		T	review of special aspects of polymer reactor design
Chakravarthy et al. (1997)	MMA	T,S	genetic algorithm, minimum time, gel effect
Choi & Butala (1989)	SAN batch	T,S	reactor optimization
Choi & Butala (1991)	MMA/VAc	S,X	optimal open loop control strategy
Cozewith (1988)			optimal start-up/changeover policies
Debling & Ray (1995)	olefins		dynamic modelling of product grade transitions (PP, LLDPE, HDPE, EPR)
Fan et al. (1995)	CSTR	T,S	start-up residence time distribution
Fernandez-Sempere et al. (1995)	various reactors	S	residence time distribution
Fuchigami et al. (1996)	bulk MMA CSTR	X	short life initiator, mixing effects and reactor scale-up
Hugo & Steinbach (1986)	SBR/CSTR	T	safe operating limits
Kataoka et al. (1995)	STY/emulsion	X	Taylor vortex flow reactor
Keskinen (1993)	PE	T	high pressure; modelling and flowsheeting
Kim et al. (1991)	PS, 2 CSTRs	S	bifurcation study with binary initiator mixture
Kiparissides et al. (1994)	LDPE/tubular	T,S,X	online optimization procedure
Kohli et al. (1996)	nylon 6/ semibatch	X	industrial batch time optimization
McAuley et al. (1990)			modelling
McAuley & MacGregor (1992)	LLDPE/gas phase	T,X	optimizing grade transition
McAuley & MacGregor (1993)	gas phase PE	S	NL controller, global input/output linearization, MI & density control, dynamic grade change optimization
Mrázek et al. (1991)	PVC/suspension	X	gradual initiator dosage study
Pinto & Ray (1995a)	VA-MMA	X	validating model for bifurcation study
Pinto & Ray (1995b)	VA-MMA	S	bifurcation analysis
Pinto (1995)	copolymer	X	bifurcation analysis in plant
Rahman & Palanki (1996)	batch	T	on-line end-time optimization
Russo & Bequette (1995)			CSTR multiplicity behaviour
Sajjadi & Jahanzad (1994)	styrene/batch	S	minimum reaction time
Salaün et al. (1996)	terpolymer	T,S,X	sequential method tendency modelling; optimizing for end use properties; industrial
Secchi et al. (1990)	semi-batch bulk PMMA	S	constained optimal control
Soroush & Kravaris (1993b)	batch	T	optimal design - theory
Soroush & Kravaris (1993c)	batch, MMA	T,S	optimal design - case study
Terwiesch (1995)			parameter switch from off-line to on-line optimization
Thiele (1986)		T	dynamic behaviour, stability, optimal start-up
Tieu et al. (1994)	various	T,S	review and case studies
Tieu et al. (1995)	general	T	comparison of collocation methods
Verwijs et al. (1996)			start-up design, temperature, safety
Wu et al. (1982)	polystyrene	S,X	Pontryagin's Maximum Principle

2.2.1 Periodic Operation

Kim, Choi and Alexander (1991) studied the simulated, free-radical, solution polymerization of styrene in a series of two CSTRs with a binary initiator mixture. They found many complex, periodic operating modes using different process parameters.

Some recent work on dynamic modelling and bifurcation has been done by Pinto and Ray (1995a,b) on the VA-MMA copolymer system and was tested industrially by Pinto in 1995.

2.2.2 Steady-state Optimization

Kiparissides, Verros and Pertsinidis (1994) developed an on-line multi-level optimization procedure for high pressure tubular LDPE (low-density polyethylene) reactor based on a comprehensive steady-state model of the process.

Tieu, Cluett and Penlidis (1994) reviewed optimization of batch, semi-batch, and continuous polymerization reactors. The OPTPAC software package by Tieu was evaluated against experimental results from literature.

Tieu, Cluett and Penlidis (1995) compared an end-point collocation method with the simultaneous optimization and collocation (SOCOLL) method for dynamic optimization problems. They found that their method yielded state estimates with smaller overall deviation than the SOCOLL method. They also found that the method gave a better solution than previous literature values for a terminal state constraint example.

2.2.3 Optimal Start-up/Changeover Policies

McAuley and MacGregor (1991, 1992, 1993) studied an industrial polyethylene reactor. They modelled the system, studied nonlinear control and optimized grade changes. The industrial setting had product parameters measured by melt index and density instead of the more academic molecular weight distribution and copolymer composition.

Fan, Shen and Chou (1995) have modelled transient residence time distributions for the study of reactor start-up. They derived a mathematical model for the residence time distribution (RTD) of a CSTR during process start-up. They used stochastic population balances to visualise the fluid flow as a system of discrete entities such as molecules.

Verwijs, van den Berg and Westerterp (1996) present a procedure for an industrial adiabatic chemical reactor to achieve an initial temperature profile. They emphasise the safety of any strategy.

2.2.4 Reactor Design

Soroush and Kravaris (1992) modelled the continuous free radical solution polymerization of MMA as a two layer system. Their first model was a reduced order model based on Kelley (1964) that was used for the dynamic single index optimization for batch time and product concentration at the end of the batch cycle. The second model was the overall model used for assessment of feasibility, flexibility, safety and controllability as described in depth in the article. The control problem and a control law were formulated with a nonstationary, MIMO, non-linear dynamic model. They propose use of globally stabilising control (GLC) to handle MIMO, nonlinearities, and modelling errors and disturbances.

Soroush and Kravaris (1993b,c) published two articles on the theoretical framework and a case study on the optimal design and operation of single-stage or semi-batch reactors. The first paper deals mostly with the equations and requirements of optimization of their inner and outer system. The second paper follows the case study of solution polymerization of MMA in toluene with an AIBN initiator. Their objectives are low PDI, high conversion, and MW of 400,000. Their process uses the typical free-radical homopolymerization assumptions. They also take the gel effect/auto-acceleration and volume shrinkage into account. From their process model, they design a reactor, assess for feasibility, flexibility, controllability, and safety of the design and formulate a control law for the system – PI control for the SISO temperature regulation problem.

Keskinen (1993) proposed flowsheeting and design optimization practice using various polymerization processes such as high pressure polyethylene production as examples.

Russo and Bequette (1995) reviewed and studied the influence of design parameters on the multiplicity behaviour of the classic continuous stirred tank reactor model when a third state, cooling jacket dynamics, is included. They found that certain design parameter changes that remove multiplicities in the two-state model can not remove multiplicities in the three-state model.

Pinto (1995c) analyzed the dynamic behaviour of the solution polymerization of AIBN initiated vinyl acetate - MMA copolymerization in tert-butanol in a CSTR. They found the dynamic behaviour of a full scale reactor had complex behaviour when pressure was increased beyond those analyzed experimentally.

Fuchigami, Nagai and Sugiyama (1996) studied CSTR scale-up to an industrial scale from a novel operational design. They used a very short life initiator, and empirically modelled mixing effects they had developed (Fuchigami and Inami, 1995) to design a safer reactor operating mode.

2.2.5 Batch Optimization

Semibatch optimization typically is the minimization of batch time while attaining a specified molecular weight distribution (Chen and Jeng, 1978).

Secchi, Lima and Pinto (1990) studied constrained optimal control for the bulk polymerization of MMA initiated by tert-butyl peroxide in a semi-batch polymer reactor. They defined a performance criterion based on squared errors of conversion and molecular weight averages from target values. They used the coordinate-transformation method solve some of the numerical problems. Conversion was limited to 70% to avoid the gel effect.

Soroush and Kravaris (1993b) proposed a framework for the optimal design and operation of batch reactors using polymerization reactors as some of their examples. Typical quality indices such as flow and strength properties were tied to performance indices such as polydispersity index (PDI) and average molecular weight which could be optimized along with other performance indices like end time and conversion drift by varying operating conditions such as temperature and initiator concentration. Key areas of modelling, dynamic optimization, design, feasibility, and control schemes were discussed.

They also presented an example of a MMA batch reactor case study (Soroush and Kravaris, 1993c) that showed the steps in modelling to optimizing the reactor design and operation for building a feasible , flexible, controllable and safe system.

Sajjadi and Jahanzad (1994) optimized the simulated, isothermal, bulk, free radical polymerization of styrene for minimum reaction time. The final product had conversion and number average chain length (NACL) targets.

Terwiesch (1995) proposes a single scalar parameter to cautiously switch from an off-line optimized input profile to on-line optimization for batch process control. This parameter is used to estimate uncertainty of model mismatch, and measurement and parameter error.

Kohli, Sareen and Gupta (1996) have studied optimal vapour release rate (pressure) histories for batch time minimisation in industrial semi-batch nylon-6 reactors, with end-point constraints. Optimal pressure histories were qualitatively similar to standard operating conditions but quantitatively different and predicted up to 50% improvement.

2.3 Modelling in Polymer Reactor Control

Modelling issues for various polymerization reactors are reviewed here. Modelling and reviews of models of polymerization systems abound. A comprehensive review was published by Gao and Penlidis (1996). This section covers the last decade or so, since it is not the primary focus of the review, but is strongly related to controller design.

Table 2.3 summarizes some recent literature on modelling issues specific to polymer reactor control. Several recent representative pieces of work from the literature, in which polymerization models have been developed in order to aid the polymer reactor control stage, are listed in Table 2.4. Detailed reviews can be found in Penlidis et al. (1985a), Hamielec el al. (1987a), Tirrell et al. (1987) and Rawlings and Ray (1987a,b).

Fundamental models play a very important role for the control of polymerization processes. This is due to a number of reasons: the lack of available on-line sensors, the complexity of the polymerization processes, the highly sensitive and nonlinear behaviour of these reactors, and the lack of well-developed techniques for the control of nonlinear processes. Therefore, if a model can provide reliable information about the process states from the measured operating conditions, it can be used for process simulation, design (scale-up), optimization and control.

The primary objective of polymer reactor modelling is to develop mathematical equations that are capable of explaining the polymerization rate behaviour and predict polymer or latex properties accurately. Since polymerization reactions are extremely complex, involving interactions between species of different chain lengths, a rigorous mathematical analysis will result in an infinite number of differential-algebraic equations (DAE). The mathematical description becomes even more complicated due to diffusion-controlled reactions. Therefore, if one's basic objective is to develop mathematical models to account for various kinetic and thermodynamic factors involved in polymerization reactors, then the model should take all of these factors into account.

However, in developing models for control studies, the motivation is different from that of pure mathematical modelling as practised in basic science and engineering (Franklin and Powell, 1980). In control studies, one is more interested in understanding and appreciating the effect of different input variables on the properties of the polymer product, so that one can devise a strategy suitable for controlling these properties. Furthermore, it is desirable to develop models that are simple in mathematical structure; this facilitates the solution of the control or optimization problem, and hence the subsequent on-line implementation of the optimal control policies.

There are a number of different approaches that have been taken in developing expressions for the molecular weight part in polymerization (e.g. statistical methods, instantaneous property balances, transform methods and the method of moments). From the viewpoint of reactor control, one useful description is via a set of differential equations describing the low order moments of MWD. From these moment states the number and weight average molecular weights are easily calculated.

For free radical polymerization mathematical models appear to be reasonably well developed (e.g. Hamielec and MacGregor, 1984; Hamielec et al., 1987b). The major uncertainties lie in obtaining reasonable estimates of the termination rate parameters (including the gel effect parameters) and the rate constants for transfer reactions, especially the ones that represent transfer to large polymer molecules. Obtaining good estimates of these parameters is far from straightforward. Systematic kinetic investigations are required, and often the type of data required (GPC, NMR) is very difficult to obtain due to poor precision, lack of well-developed procedures, and frequently practical problems like the insolubility of branched polymers in common solvents, etc.

Seth and Gupta (1995/96) improved on an earlier semi-empirical model to predict the Trommsdorff effect on the rate constants in a non-isothermal semibatch reactor for the polymerization of MMA. The newer model was less sensitive to small variations in the parameters. Systems were benzoyl peroxide (BPO) and AIBN initiator with MMA in benzene.

Nair, Chaumont and Colombani (1995) proposed a kinetic model for the free-radical polymerization in the presence of added initiator and an addition-fragmentation agent. Their model accounts for chain termination by primary radicals but excludes the mutual termination of primary radical. Their general model was applied to MMA polymerization in the presence of an addition-fragmentation agent. The agent retarded the reaction up to 70°C beyond which, the thermal decomposition of the peroxy group in the agent increased the reaction rate. This study discusses the reactions and control of chain transfer in free-radical polymerization.

For Ziegler-Natta (Z-N) systems the understanding of the molecular weight development is much less well understood. Again, modelling ionic systems involves a more detailed understanding of the nature of the active catalyst sites, and of the effect of impurities. Diffusional effects in solid catalyzed gas phase reactions may also play an important role, which makes the overall picture even more complicated.

In heterogeneous polymerization such as emulsion or dispersion systems where the particle size distribution plays an important role in both the final product quality and the rate of reaction, population balances accounting for the birth, death and growth of polymer particles constitute an important part of the model. Penlidis et al. (1986a,b) and Rawlings and Ray (1987a,b) have summarized the population balance and age distribution approaches. Major uncertainties still lie in modelling the particle nucleation and coagulation

phenomena, particularly in the presence of non-ionic stabilizers. Usually these phenomena are so unpredictable and occur so rapidly that little can be done to actively control them during a polymerization. The best that can be done is to try to avoid having to deal with them by using seeded emulsions, and an adequate amount of stabilizer.

Semino and Ray (1995) analyzed the modelling and control of systems described by population balances. Their examples were human populations, emulsion polymerization, and crystallisation. Control of particle size distribution by manipulating the feed concentrations is suggested as a practical solution.

Chen, Lee and Chiu (1996) developed a general mathematical model to estimate the particle concentration during the course of soapless emulsion polymerization of methyl methacrylate (MMA). Rate of polymerization, number of radicals in each particle, instantaneous average molecular weight, and termination rate constants were calculated and agreed with experimental results.

Over the last quarter century or so, Ray (1972), Ray and Lawrence (1977), Hamer, Akramov and Ray (1981), Ray (1983), Arriola and Ray (1987), Stevens and Ray (1989) and Congalidis, Richards and Ray (1989) have developed extensive mathematical models to describe various polymerization systems: step and addition homo- and multi-component polymerization and polyolefin systems. Multiplicity in continuous reactors is a phenomenon of continuous polymerization systems that has also been modelled. Predictions of conversion, molecular weights, composition and sequence length are some properties that are predicted by these models. Some recent work is summarized in Hutchinson and Ray (1987), and de Carvalho et al. (1990). Coyle et al. (1985) discussed the high conversion free radical polymerization of methyl methacrylate (MMA) using a finite element analysis, and Gonzalez-Romero and Rodriguez (1987) presented an alternative using orthogonal collocation solutions. Hamielec el al. (1987b) presented mathematical models for multicomponent polymerization in order to control composition, chain microstructure, molecular weight, and long chain branching and crosslinking.

Litvinenko (1996) reviewed chain transfer reactions in batch and continuous non-terminating catalytic polymerization reactions. The effect of chain transfer to solvent, monomer and polymer on molecular weight distributions were discussed.

As an example of a recent, practical and direct application of modelling, Polacco, Semino and Rizzo (1994) used modelling of suspension polymerization of MMA to study acrylic bone cement applications. PMMA is used to join the bone and metallic joint with a polymerization process in the surgery room. In a new process, a suspension of monomer in agarose and water in the form of a gel is used to hold the monomer in place. Modelling of the reaction was done to analyze the influence of system geometry and the temperature distribution. Monomer droplets were treated as small batch reactors. Mass balances were determined for

each drop and an overall energy balance applied. Benzoyl peroxide was the initiator. Mechanistic models developed by Chiu, Carratt and Soong (1983) were used with empirical rate constants.

One of the most widely applicable techniques used to derive a simpler model for control purposes is model reduction. During model reduction one's aim is to simplify a higher order detailed model by a lower order simpler model such that the simpler model captures the essential features of the original process adequately. Model reduction is a common procedure in engineering practice. Its main objectives are to simplify the analysis and simulation of the original complicated system, to reduce the computational load in the design of controllers, and finally, to derive simpler control system structures (Paraskevopoulos, 1986).

Penlidis, Ponnuswamy, Kiparissides and O'Driscoll (1992) reviewed modelling specifically for reactor control. They simplified the polymerization model in five steps, starting with the basic kinetic equations and assumptions. They next used the method of moments, applied the quasi-steady state approximation, simplified with an order of magnitude study, and converted from continuous to discrete time. Thus, for control studies in polymer reaction engineering purposes, this simplified model adequately described the effect of input variables on the process and final polymer properties as required.

Another area of modelling, is to empirically model only the required area of study. Fuchigami and Inami (1995) modelled the solution polymerization in a CSTR using very short life initiators of 2 to 30 seconds for an industrial reactor. They continued the work (Fuchigami, Inami and Sugiyama, 1996) to study mixing effects to evaluate a double helical impeller design at the pilot plant stage.

They first studied continuous bulk polymerization of methyl methacrylate using very short-life initiators (<10 sec). Their pilot plant study was for the experimental verification of initiator and mixing effects for a commercial plant. They combined initiator and mixing effect in an effectiveness factor.

Higher mixing speed resulted in higher conversion and was more pronounced for shorter life initiators. It was easier to control conversion with a shorter life initiator with initiator concentrations over 45 ppm, where increased temperature decreased conversion (key!) and increased the F-value. With initiator half-lives shorter than mixing times, initiator decomposes before complete mixing. As temperature increases, the half-life decreases and conversions decrease. This technique was used as a basis for a plant producing 6000 tons/yr of PMMA that was constructed in June 1991.

Feliu, Sottile, Bassani, Ligthart and Maschio (1996) have investigated the thermal behaviour and parameters for bulk MMA polymerization. Correlations and mathematical models were proposed to predict the dynamic behaviour which emphasis on safety.

Chemical processes are often modelled by a combination of algebraic and ordinary differential equations (ODE). These differential algebraic equations (DAE) describe a wider range of systems. The differential index is the minimum number of differentiation operations required to convert the DAE system to a purely ODE system. Kumar and Daoutidis (1995) discuss output feedback control of high index, multivariate, non-linear DAEs. They developed an algorithmic procedure to derive a state space realisation, and then control with state feedback and state observers. A two phase reactor simulation was used as an example.

The intent here is not to form a detailed review on the mathematical modelling of polymerization processes, but to simply mention a few representative recent efforts. Detailed reviews can be found in Ray (1972), Penlidis et al. (1985a) and Hamielec et al. (1987a).

Table 2.3: Recent work on modelling issues related to polymer reactor control

Reference	System	Work	Notes
Burke et al. (1995)	copolymer	S	model discrimination
Burke et al. (1997)	copolymer	T	discriminating between terminal and penultimate models
Canu & Ray (1991)	polymer	T	discrete weighted residuals method (DWRM) for large sets of discrete DAE
Chien & Penlidis (1994b)	MMA	S,X	effect of impurities
Chylla et al. (1997)	STY-αmethyl-STY - AA	X	dynamics of semibatch
Debling et al. (1997)	PP	S	residence time distribution effects
Dubé et al. (1997)	general	T	multicomponent chain-growth polymerization modelling review
Fuchigami et al. (1996)	MMA/CSTR	X	mixing effects
Harismiadis et al. (1996)	general		miscibility models
Hong (1997)	polymer/solvent		estimate free volume parameters
Kantzas & Kalogerakis (1996)	polyolefin	X	fluidization characterisation using x-ray computer assisted tomography scanning
Kwag & Choi (1994)	PE	T,S	initiator characteristics at high T & P
Litvinenko (1996)	batch & continuous	T	steady-state model of conversion, PDI, and MW, with chain transfer
Palavajjhala et al. (1996)	black box	T	PRBS signal design
Penlidis et al. (1992)	general	T	modelling for control, review
Powell & Brooks (1995)	vinyl/semibatch		steady & stationary state assumptions, reactor performance
Russo & Bequette (1995)	jacketed CSTR	T	process design impact on multiplicity
Russo & Bequette (1996)	jacketed CSTR	T	cooling jacket dynamics
Srinivas et al. (1996)	semibatch	X	gel effect
Teymour (1997)	semibatch	T	analysis of dynamics
Tobita (1997)	CSTR	T	LCB control using chain transfer monomer
Yamamoto et al. (1996)	batch	X	mixing effects on T distribution
Yang & Suspene (1991)	polyester	S,X	viscosity & gel point of curing study
Zacca et al. (1996)	olefins		residence time distribution
Zacca et al. (1997)	PP, PE	S	residence time distribution effects

Table 2.4: Recent work on modelling specific polymer reactor systems

Reference	System	Work	Notes
Bhagwat et al. (1994)	slurry PE	T	solid Z-N catalyst; MW, PDI; mass transfer explanation for high PDI with single-site
Chen et al. (1996)	MMA	T	soapless emulsion, particle formation
Dairanieh (1991)	condensation	T,X	viscosity buildup of sulfonated melamine-formaldehyde resins
Dubé et al. (1996)	Acrylonitrile-Butadiene	T	modelling of emulsion copolymerization
Feliu et al. (1996)	MMA	X	thermokinetic parameters, stability
Fuchigami & Inami (1995)	MMA-EA	T,X	steady-state CSTR model of reaction rates and mixing effects using short life initiator
Gao & Penlidis (1996)	general		review
Gao et al. (1997)	PET	T,S	modelling solid state polycondensation
Jacob et al. (1997)	PMA		microwave polymerization
Kiparissides (1996)			review
Kiparissides et al. (1997)	PVC/batch	T,X	predictive model
Lei et al. (1992)	PET	T,S	CSTR and extruder modelling
Madras et al. (1996)	PMMA	X	degradation
Maschio et al. (1994)	MMA	S,X	batch/suspension and tubular/solution models to study MWD control
McKenna et al. (1996b)	PE/slurry	X	transfer phenomena, hetero Z-N catalyst
Mutha (1996)	NBR, MMA	T	model conversion, composition, and MW
Nair et al. (1995)	MMA/bulk	X	reaction rates
Pinto (1990)	VC	T,S	continuous suspension & bulk systems, study of multiple steady states
Pinto & Ray (1996)	VA-MMA	T,X	inhibitor effect
Polacco et al. (1994)	MMA suspension	T	bulk suspension, temperature profile
Polacco et al. (1996)	MMA suspension	T	temperature profile during reaction
Roy & Devi (1996)	MMA emulsion	X	mechanism in emulsion and microemulsion
Sayer et al. (1997)	SBR emulsion	T	dynamic modelling of cold emulsion polymerization
Semino & Ray (1995a)	emulsion	T	population balance model
Seth & Gupta (1995/96)	MMA	T	gel effect in semi-batch
Soares & Hamielec (1996a)	olefins CSTR train	T	dynamic modelling with copolymerization, Z-N & metallocene catalysts
Soares & Hamielec (1996b)	PE		catalyst pre-polymerization effect
Soares & Hamielec (1996c)	PP		kinetics, effect of hydrogen
Sun et al. (1997)	butadiene	T	gas phase, particle growth
Vega et al. (1997)	NBR	T,X	modelling an industrial reactor
Vivaldo-Lima et al. (1997)	styrene/divinyl-benzene	T	review of suspension polymerization modelling for PSD
Wolff & Bos (1997)	PS	S	modelling MWD
Xie et al. (1994)	gas phase PE		review

2.4 Online Monitoring

An integral element in the control of polymerization processes is that the controlled properties of the process be measured or at least estimated. Some of the most common measuring instruments include thermocouples, densitometers, and gel permeation chromatographs for temperature, conversion, and molecular weights, respectively. Sensors can in general be implemented in two ways: on-line and off-line. It is desirable, particularly for control purposes, to choose sensors which can take rapid measurements of changes occuring in the process. In other words, delay times should be small. These types of measurements are available through on-line sensors, and typically reflect the operating conditions of the process. However, for processes in which it is necessary to also maintain a particular quality of the product, other measurements are usually needed, However, since these measurements are performed off-line, significant delays may result. From a control point-of-view, this is undesirable because delays may cause instability to a controlled process and, unless very good state estimation (or prediction) schemes can be implemented, the control objectives should be reconsidered. A concise review of on-line sensors for polymerization reactors was prepared by Chien and Penlidis (1990). This section updates this review and discusses the technology available for on-line monitoring of polymer reactor operation and polymer properties (quality). Also, a brief discussion is given on several promising off-line techniques that some day may be implemented on-line.

Monitoring can be broken down into sensors of reactor operation and polymer property estimation. Monitoring techniques for reactor operation are typical of chemical engineering measurements: temperatures, pressures, flows, levels and heat balances.

On-line polymer and latex property measurement includes density, viscosity, gas chromatography (GC), hydrodynamic chromatography (HDC), turbidity spectra, and surface tension. Off-line measurements can also supplement the on-line measurements to give better measurement for control purposes such as fast feedback, robustness and accuracy:

- gravimetry for conversion
- gel permeation chromatography (GPC) for molecular weight distribution
- nuclear magnetic resonance (NMR) for composition, sequence length, and structural information
- low angle laser light scattering (LALLS) for precise measurement of weight average molecular weights
- hydrodynamic (HDC) or size exclusion chromatography (SEC) for particle size distributions

Table 2.5 summarizes recent work on monitoring techniques for reactor operation and on-line monitoring of polymer product properties. Polymer property measurement requires state estimation and data filtering techniques due to lack of on-line sensors. Literature on these topics is summarized in Table 2.6.

Table 2.5: Recent work on monitoring polymer reactor operation and on-line polymer property measurements

Reference	System	Work	Notes
Arola et al. (1997)		X	NMR as a viscometer
Chatzi et al. (1997)	copolymer /emulsion	X	midrange IR optical fibre probe for conversion measurement
Chien & Penlidis (1990)	all		review of online sensors
Dumoulin et al. (1995)			techniques for real-time measurement
Dumoulin et al. (1996)			ultrasound
Gagliano (1996)		X	tape level sensor for viscous systems
Ginesi (1991)			flow measurement
Gossen & MacGregor (1993)	VA/latex	X	online particle size measurement
Gretzinger et al. (1995)			optimal sensor selection
Hansen & Khettry (1995)			probes for inline polymer melt analysis
Kim et al. (1992)	MMA batch		EKF to monitor heat transfer coefficient
Kourti et al. (1990)			particle size from tubidity
Martin (1992)	various	news	near-infrared reflectance spectroscopy
McKenna et al. (1996a)	copolymer	X	calorimetry, densimetry, gravimetry
Nicoli et al. (1990)			remote sensor for particle sizing
Nomikos & MacGregor (1994)	SBR	T,S	batch process monitoring through principal component analysis
Nomikos & MacGregor (1995)	batch		SPC
Pabedinskas et al. (1991)	extruder	T,X	in-line rheometer development
Pabedinskas & Cluett (1992)	extruder	X	signal processing of pressure measurements
Schlenoff et al. (1995)	emulsion/ bulk		fibre optic Raman spectroscopy
Schuler & Schmidt (1992)			review of calorimetric state estimation for reactor diagnosis and control
Tham & Parr (1994)		T	validation & reconstruction of data
Thomas & Dimonie (1990)	emulsion	X	fibre optic dynamic light scattering; measures particle size during latex emulsion polymerization
Urretabizkaia et al. (1993)	copolymer batch	X	conversion & cumulative composition monitoring via calorimetric measurements

Table 2.6: Recent work on state estimation and data filtering for polymer reactors

Reference	System	Work	Notes
Adebekun (1996)			output feedback control
Albuquerque & Biegler (1995)	nonlinear	T,S	decomposition algorithm for on-line estimation
Appelhaus & Engell (1996)	PET		design and implementation of an extended observer
Brandolin & Garcia-Rubio (1990)			PSD measurement
Chang & Chen (1995)	fault diagnosis	T	implementation issues with EKF-based
Choi et al. (1997a)	olefin	T	estimation of kinetic parameters
Dong & McAvoy (1996b)	SBR	T	batch tracking, NL-PCA monitoring SBR example
Ellis et al. (1994)	MMA batch	X	MWD estimation with EKF and SEC; experiments with on-line SEC and density
Karjala & Himmelblau (1996)			general RNN, EKF data rectification
Kim et al. (1992)	MMA batch		EKF to infer MW
Kozub & MacGregor (1992)			
MacGregor et al. (1994)		T,S	multiblock PCA/PLS
McAuley & MacGregor (1991)	PE	T,X	MI and density inference from on-line data
McKenna et al. (1996)			calorimetry, densimetry, gravimetry
Mutha et al. (1997)	batch copolymer	T,S	multirate-measurement-based estimator
Myers et al. (1996)			EKF, delayed off-line measurement
Nomikos & MacGregor (1994)	SBR	T,S	batch process monitoring through principal component analysis
Régnier et al. (1996)	MMA/VA & MMA; batch		EKF, temperature control
Robertson et al. (1996)			MHE comparison
Schuler & Schmidt (1992)			calorimetric state estimator review and application
Schuler & Schmidt (1993)	general	T	state estimation review with example in polymerization
Shyichuk (1996)		T	degradation index from viscometry
Soroush (1997)	general	T	NL state observer design
Tan et al. (1996a)			Smith predictor
Tatiraju & Soroush (1997)	MMA	T,S	nonlinear state estimator for initiator and solvent concentrations and leading moments of MWD
Terwiesch (1995)	batch	T	on-line correction mechanism
Terwiesch & Agarwal (1995b)			EKF state-space grid
Tham & Parr (1994)	general	T	filtering and intelligent reconstruction from noisy plant data
Tsen et al. (1996)	VA/batch	S,X	hybrid ANN models
Valluri & Soroush (1996)	CSTR		nonlinear, inadequacy of linear observers
Viel et al. (1995)	STY	S	stability using I/O linearization and high gain observer
Wang et al. (1997a)	nonlinear		sliding observer

2.4.1 Monitoring Polymer Reactor Operation and Measuring Polymer Properties On-line

Sensors to monitor reactor operation (temperature, pressure, flows, level) are similar to sensors for other chemical processes although high viscosity polymeric streams may have mixing, clogging, reaction run-away, heat removal and other problems that are specific to polymer reactors. For example, a resistance tape sensor can be used to track tank levels for sticky or viscous fluids as described by Gagliano (1996). Ginesi (1991) described some flow measurement techniques.

Marten (1991) reviewed advances in near infrared reflectance spectroscopy (NIR). It could be used to measure moisture, as well as end and functional groups.

Gossen and MacGregor (1993) outlined the design and testing of an analyzer system for poly(vinyl acetate) that measures solid fractions and mean particle diameters. They measured changes in solids fraction of less than 5% of the value (0.5% to 10% solids range). Changes in the particle diameter of 10 nm were also measured.

However, most papers refer to some form of estimation or inference of the polymer properties from measured values. A fundamental problem with polymer property measurements is that end-use product specifications are not always related to molecular architecture. Ray (1985) reviewed polymer property observability and detectability with various sensors.

2.4.2 Product Property Trajectory Tracking

Since most polymer properties must be estimated and inferred from available on-line and off-line measurements, state estimation is important. State estimation, date validation, diagnosis, and other topics not as clearly categorized into the later sections have been covered in this section.

Kozub and MacGregor (1992) have applied a non-linear estimator to infer copolymer properties in a simulated styrene/butadiene rubber (SBR) polymerization. Optimal open loop control strategies based on established conditions were used and feedforward control trajectories recomputed with updated state estimates. Feedback control was added to correct copolymer properties.

Schuler and Schmidt (1992, 1993) have published reviews on calorimetric state estimation and application to various chemical processes including polymerization.

MacGregor, Jaeckle, Kiparissides and Koutoudi (1994) extended process monitoring of large continuous reactors using multivariate statistical projection methods by blocking the system into subsections. Multiblock projection to latent structures (PLS) was developed. Faults and events in blocks were identified

earlier and with more precision than without blocking. The technique was applied to a simulated multi-section tubular reactor for the production of low density polyethylene.

Ellis, Taylor and Jensen (1994) developed a model for the batch polymerization of MMA in ethyl acetate with AIBN initiator. The emphasis was on the prediction of MWD with periodic size exclusion chromatography (SEC) readings. This model would then be used for feedback control of the MWD. Their model was tested on an experimental system with an on-line SEC and on-line density meter. The manipulated variables were batch temperature, monomer addition, or both. The model performed well and was used in a PID scheme to control molecular weight.

Terwiesch and Agarwal (1995b) advocate use of a state-space grid of the Bayes' rule over the extended Kalman filter (EKF) for state estimation of non-normal probability distributions or non-linear processes.

Chang and Chen (1995) presented multi-parameter EKFs designed to detect and identify faults. They addressed the problems in identifying multiple fault origins. Their procedure to evaluate the performance of a given system was tested via simulation studies.

Valluri and Soroush (1996) showed the inadequacy of the Luenberger observer and the extended Luenberger observer with constant gain for state estimation of non-linear deterministic processes. The global asymptotic convergence of a non-linear design method was proposed. The example process was a classic exothermic CSTR with multiple steady states.

Dong and McAvoy (1996b) showed application of non-linear principal component analysis (NL-PCA; Dong and McAvoy, 1996a) to monitor a batch process. Their first example was the monitoring of the emulsion polymerization of an SBR process described by Nomikos and MacGregor (1994).

Tan, Wang, Lee and Bi (1996) reviewed the Smith predictor. It has structure similar to the internal model control (IMC) structure (Morari and Zafiriou, 1989). They use the area enclosed within the Nyquist curve of the feedback element to assess the achievable closed loop performance and formulate an optimization objective function.

Adebekun (1996) analyzed output feedback control of a CSTR. Input-output linearization was used to control conversion and temperature. A reduced order state estimator was used to estimate conversion. Even with an arbitrary initialization of the state estimator, convergence to the desired equilibrium point was achieved.

McKenna, Fevotte, Graillat and Guillot (1996) studied calorimetry, densimetry, and gravimetry in modelling batch and semi-batch polymerization reactors. They found that heat balances and calorimetry combined with off line gravimetry gave satisfactory on-line estimation of individual monomer conversions. The combination of densimetry and calorimetry was reported as not satisfactory. They further noted that

densimetry measurements to 3 or 4 significant digits of accuracy would be required for satisfactory estimation.

Myers, Kang and Lueke (1996) applied EKF state estimation for a system with delayed off-line measurements.

Robertson, Lee and Rawlings (1996) presented the general formulation of the moving horizon estimator. It is compared to the extended Kalman filter (EKF), iterated EKF, Gaussian second-order filter ,and the statistically linearized filter.

Karjala and Himmelblau (1996) proposed the use of recurrent neural nets (RNN) and the extended Kalman filter (EKF) for the dynamic rectification of measurement data. The two are used for data rectification of badly autocorrelated measurement data as well as to estimate measurement bias.

Mutha, Cluett and Penlidis (1997) proposed a fixed-lag smoothing-based extended Kalman filter algorithm for systems with multirate measurements. Their estimator was evaluated through simulation studies of an acrylonitrile/butadiene emulsion copolymerization batch process. The proposed estimator had robustness to state and measurement noise and could handle systems with large measurement delays. Performance of the algorithm using delayed measurements was superior to that of the standard extended Kalman filter.

2.5 Batch and Semibatch Reactor Control

Control policies for batch and semibatch reactors are surveyed with respect to the property being controlled (molecular weight, branching, composition, conversion, particle size, etc.).

Batch and semibatch polymerization systems have been the most widely investigated from the control point of view, and their major advantages, great flexibility and rapid response to changing market conditions, are well-known. In contrast to continuous manufacture of commodity products, batch processes are characterized by both a very different manufacturing environment and a different dynamic behaviour. For example, the multiple product environment of batch processes requires that ingredients, control loop set-points, and tuning parameters must be changed frequently (Shaw, 1983). The frequent product and process changes associated with a batch chemical manufacturing facility also present some special challenges for control system design (Smith, 1984). For example, the requirement for startup and shutdown regulatory control in batch polymerization processes demands good dynamic response over the entire operating range of the controlled variable. This contrasts with the precise control over a small range that is required in many continuous processes.

The wide operating ranges and nonstationary behaviour that cause difficult sensor problems in batch reactors also influence control system design (Juba and Hamer, 1986). For example, during process identification, the linearized approximate models, so common for continuous processes, are not applicable, because of the lack of a nominal steady state about which the process can be linearized. As a result, non-linear models based on actual values rather than deviation variables must be used. Similar difficulties are encountered during controller design, where variable-gain controllers are often required to compensate for time-varying process dynamics. The controller design is further complicated by asymmetric penalties, such as in composition control where the formation of unwanted byproducts is irreversible. This contrasts with continuous processes, where upsets eventually wash out of the system and the process may return to the desired steady-state.

In batch polymerization, the optimization problem is identified as the time dependent control actions which maximize product quality and minimize production time (Amrehn, 1977). Different techniques have been suggested and applied to calculate and control to the optimal or sub-optimal trajectories.

Some opimization problems should not be solved using controllers and controller trajectories alone but by using reactor design. Recent papers on batch optimization, reactor design and selection were reviewed in Tieu et al. (1994, 1995). Some more papers were discussed in Sec. 2.2.

MacGregor et al. (1984) reviewed batch polymerization control divided into five categories:

- molecular weight distribution (MWD) control
- conversion and composition control
- long chain branching (LCB) control
- particle size distribution (PSD) control
- temperature control

Recent literature on control of batch and semibatch polymerization reactors is summarized in Table 2.7.

Table 2.7: Recent work on control of batch and semibatch polymer reactor systems

Reference	System	Work	Notes
Arzamendi & Asua (1991)	VAc/MMA emul	S,X	copolymer composition control; optimal monomer feed rate; limited heat removal
Chang et al. (1996)			temperature trajectory
Choi et al. (1997b)	PE	S	prediction of MW and PDI
Davidson (1987)		X	intelligent temperature controller for jacketed reactor
de Buruaga et al. (1996)	emulsion		on-line calorimetric control
de Buruaga et al. (1997)	VAc/BuA	X	calorimetric measurement optimize feed profile versus conversion
Defaye et al. (1993)	VAc/2-ethyl-hexyl acrylate	T,X	adaptive-predictive temperature control, data filtering
Ellis et al. (1994)	MMA	X	estimation with EKF, MWD control
Gridnev (1992)	MMA	X	cobaloximes and BPO CTA, MWD
Hidalgo & Brosilow (1990)	STY	S	NL-MPC at unstable points
Infante et al. (1996)	VA.	X	temperature control in microemulsion batch reactor
Kim et al. (1992)	MMA	X	EKF to estimate changing heat transfer coefficient and improve temperature control
Kiparissides et al. (1990)	bulk MMA	S	DMC and ESTR applied for conversion and number average MW trajectories
Kozub & MacGregor (1992)	semibatch SBR	S	inferential FB control of polymer quality
Maschio et al. (1994)	MMA	S,X	suspension polymerization, MWD
Mayer et al. (1995)	STY	X	large scale commercial, high conversion
Nagy & Agachi (1997)	PVC	S	NL-MPC, temperature control
Ni et al. (1997)	acrylamide		fuzzy-hybrid-PID, high exothermic temperature control
Peterson et al. (1992)	free radical MMA	T,S	NL-MPC (DMC) applied to simulated jacketed CSTR, unconstrained
Powell & Brooks (1995)	PS	S	filling policy, conversion and cumulative MWD effects, styrene example
Rawlings et al. (1989)			endpoint control
Sajjadi & Jahanzad (1994)	STY	S	minimum reaction time
Soares et al. (1997)	PE	T	measurement & control of MWD & CCD
Soroush & Kravaris (1992)	batch MMA	T,X	modelling and application of global linearizing control (GLC)
Tsen et al. (1996)	VA/batch	S,X	hybrid ANN models, predictive control, MWD control
van Doremaele et al. (1992)	STY/MMA emulsion	X	composition control by optimal addition profile
Yabuki & MacGregor (1997)	SBR	S	practical approach to product quality control

2.5.1 Molecular Weight Distribution Control

Perhaps the single most significant property that directly influences the end-use characteristics of a polymer is its MWD. For instance, the MWD of the polymer can be an important variable as far as further processing is concerned. Ultimately, many of the mechanical properties of the polymer, such as strength and impact resistance (Ray, 1985), are directly related to the MWD. For the majority of chemical processes the requirements are to simply maintain specified operating conditions, such as temperature and conversion (ie., production rate). However, with polymerization processes, it is necessary to control product quality also (ie., MWD) in addition to process operating conditions. Although the process operating conditions reflect the economic aspects of the process (production rate, energy consumption, etc.), it is the tight control over the MWD that determines not only product quality but whether or not the product is suitable for the intended end-use. The initial step towards control of the product quality is to have some understanding and subsequently access to the variables which govern the MWD.

The paper by Choi and Butala (1987) describes a new method to synthesize open-loop control policies for composition and MWD for batch and semibatch polymerization reactors in which no on-line sensors exist. The method is based on the observation that the time varying trajectories of the manipulated variables obtained from a fictitious feedback control system are equivalent to the trajectories of the control variables required for control in open-loop systems in which no on-line sensors are available. The following sequence of events is proposed. First, dynamic simulation of the process model plus controller (fictitious feedback control system) is performed until the best (most desirable) closed-loop response is obtained. At the same time the transient path of the manipulated variables is recorded. Then, using this control trajectory in the actual open-loop process, will hopefully result in similar (if major disturbances do not enter the system) performance as in the fictitious feedback (closed-loop) control system. The advantages of this type of controller development are several: more elaborate process models can be used in the closed-loop simulations; the effect of various operating conditions can readily be evaluated by dynamic model simulations and, there is no need to introduce any weighting factors when more than one control variables are involved.

The papers by Gonzalez et al. (1986), Taylor et al. (1986), Ellis et al. (1987), Ellis et al. (1988a), and Ellis et al. (1988b) all deal with essentially the same subject matter. These authors present an innovative procedure by which continuous on-line estimation of the MWD is possible. The particular system analyzed is the isothermal solution polymerization of MMA in a batch reactor. The polymerization (kinetic) model employed in their investigation was developed by Tulig and Tirrell (1981, 1982) and Tulig (1983) and is used to estimate the complete MWD from simple on-line measurements of temperature, density, and off-line delayed GPC measurements. In addition, the model is valid over the entire conversion range. The polymerization model utilizes chain length as the spatial parameter, which includes a chain length dependent termination rate constant, so that the entire MWD of both living and dead polymer may be

evaluated even in the case of strong gel effect. In this way it is possible to observe or detect multimodal or skewed distributions, which is not possible if one considers only the moments of the distribution. Put it differently, it is not possible to infer the structure of the MWD from knowing only the moments of the distribution if the actual MWD is multimodal or skewed. Only if we know the MWD to be approximately normally distributed (unimodal) can we consider the moments of the distribution to be an accurate representation of the MWD. The moments of the MWD represent important characteristics of the MWD. For instance, the most frequently specified first two moments of the MWD are the number- and weight-average molecular weights.

Ray (1985) has classified the reactor operating variables as either observable or detectable. For example, monomer and initiator conversion are readily observable from temperature measurements, whereas molecular weight properties are only detectable. With temperature and bulk viscosity (or GPC) measurements, the weight-average molecular weight becomes observable. Interesting examples of state estimation techniques, pertaining to styrene polymerization in both batch and continuous stirred tank reactors, are presented by Schuler (1980) and Schuler and Zhang (1985). The aim in these papers is to infer the polymer chain length distribution from temperature and refractive index measurements.

Thomas and Kiparissides (1984a) and Secchi et al. (1990) used optimal control theory to analyze the performance of a batch PMMA polymerization reactor. Their formulation was based on Pontryagin's Minimum Principle, which leads to a mixed initial/final value optimization problem. Optimal dynamic profiles of temperature, initiator feed rate and initiator concentration in the reactor are generated in order to obtain specified conversion, number-average molecular weight and polydispersity of the final product.

Thomas and Kiparissides (1984b) subsequently performed a sensitivity study of a batch polymerization reactor. They considered the free radical polymerization of MMA and their analysis focused on the sensitivity of several output variables (conversion, zeroth and second moments of the dead polymer distribution) subject to uncertainties in kinetic parameters and initial conditions. The sensitivity coefficients which result can provide insight into the cases for which parameters can and cannot be estimated. It would also seem reasonable for control purposes that one could obtain information about input/output interactions that would enable one to select suitable input/output pairings.

Couso et al. (1985) applied an open-loop control strategy to obtain the desired MWD by periodically cycling the monomer solution feed in a semibatch 'living' anionic polymerization reactor. In their simulation study, two techniques were developed. In the direct form of the technique, the monomer solution flow rate and an intentional 'killing' agent flow rate are simultaneously added to the initiator solution. Alternatively, by controlling the reactor outlet flow instead of the 'killing' agent feed, an external deactivation of 'living' ends can be implemented. The attempt is to obtain polymers with a pre-specified MWD shape.

Later, Alassia et al. (1988) experimentally verified the above techniques by using two pumps to maintain the necessary flow profile of monomer and solvent in order to control the molecular weight distribution. They showed that it is impossible to eliminate the impurities present in the monomer solution. For this reason, they proposed a theoretical technique to allow the attainment of polymers with any "tailor made" MWD shape. The main difficulty was the estimation of the initial moles of initiator and of the concentration of impurities in the monomer solution.

Louie and Soong (1985) proposed a solvent injection program for controlling the average molecular weight and narrowing the polydispersity of products from a free radical polymerization process with an inherent tendency to exhibit strong gel and glass effects. Their experimental verification of this idea on MMA showed that the product MWD was indeed narrowed.

Kumar and Sainath (1987) investigated the polycondensation stage of polyethylene terephthalate (PET) formation. In this stage, many side reactions are known to take place, in addition to the main polymerization reaction, and the quality of polymer produced is largely dependent on the various side products formed. A flexible objective function was proposed with temperature and pressure as the control variables. Three weighting parameters were used to represent the relative importance of diethylene glycol in the product, number-average chain length of the product, and the amount of side products formed. Since water and perhaps other volatile components are naturally produced species of polycondensation reactions, it is necessary to vary the pressure within the reactor as the polymerization proceeds in order to drive the polymerization reaction to completion. On the other hand, to obtain a polymer quality variable such as number-average molecular weight with a desired value, the temperature of the reactor will have to follow some specific trajectory. In their paper, the authors performed optimization studies using the gradient method or control-vector iteration procedure in which both temperature and pressure were considered as the control variables. The results show that the pressure falls first and settles down to a preset lower limit, only after which the temperature begins to be adjusted. In view of this, the analysis is simplified by considering temperature as the only control variable, while carrying out the polymerization reaction at the lowest possible pressure.

Huang and Lee (1989) used a set of parabolic differential equations to seek the optimal wall temperature in order to minimize either reaction time or MWD polydispersity for the free radical polymerization of styrene in a casting process.

Peterson et al. (1992) developed a nonlinear model-predictive control (MPC) strategy using an enhanced version of the dynamic matrix control (DMC) algorithm for a semibatch MMA polymerization reactor. Their simulation results show that, with proper manipulation of initiator flow rate and cooling jacket temperature, the reactor temperature can be effectively regulated and the number average molecular weight can be forced to meet a target value at the end of the batch despite strong nonlinearities due to the reaction rates and the gel effect.

Maschio, Bello and Scali (1994) extended the model for bulk and suspension polymerization of MMA to solution polymerization and verified this model experimentally. Their objective was to model the polymerization well enough to control it to minimise the broadening of the MWD and achieve an assigned M_w. Their modelling and control of the MWD was successful in suspension polymerization with batch reactors. Their solution polymerization was in concentrated form (5-15% of solvent) and solvent addition during the course of reaction was necessary to offset the gel effect which broadened the MWD.

Powell and Brooks (1995) compared filling policies for simulated, free-radical, solution in semibatch reactors. The regions of validity for stationary state assumptions were determined. Results were presented on the effect of filling policies and initiator stability on the conversion and cumulative molecular weight distribution for a styrene example process.

Tsen, Jang, Wong and Joseph (1996) studied the use of an artificial neural network (ANN) as a model for product disparity and molecular weight distribution in a batch reactor for emulsion polymerization of vinyl acetate. The hybrid ANN was constructed using both experimental information and knowledge from a mathematical model. It performed better than a parameter fitting theoretical model when used in model predictive control of MWD, both in simulation and experiments.

2.5.2 Conversion and Composition Control

Copolymer composition governs physical/mechanical/optical properties and determines the economic uses to which a copolymer may be applied. Due to the fact that in a copolymerization reaction the monomers involved react at unequal rates, the monomer mixture is progressively depleted in the faster monomer, whereas the copolymer formed becomes saturated simultaneously in the slower monomer. This phenomenon, described as "Composition Drift", is usually unacceptable.

Budde and Reichert (1988, 1991) presented an on-line measurement scheme for conversion, viscosity, and MWD via density, viscosity, and GPC data for the batch and semibatch solution polymerization of MMA. Both open- and closed-loop initiator addition policies were evaluated for control of conversion under the constraint of a constant reaction rate.

Chen and Lee (1987) utilized initiator addition policies and temperature as the two manipulated variables to obtain a copolymer with pre-specified values for the final conversion of the less reactive monomer and for the cumulative copolymer composition of the more reactive monomer in the minimum time under the constraint that the copolymer composition is maintained constant. The policy was experimentally verified using the acrylonitrile/styrene system. The optimal initiator addition policy for the nonisothermal case is to

make the rate of initiation constant and for the isothermal situation the number of moles of initiator in the reactor must be maintained constant.

Choi (1989) presents a solution to the copolymer composition control problem which seeks to find the monomer(s) addition rate for two cases: varying initiator concentration and constant initiator concentration. The theoretical development is based on a general copolymerization system involving vinyl monomers.

A very thorough treatment of emulsion polymerization systems in general is presented by Dimitratos et al. (1989). The theoretical development of the dynamic modelling and state estimation schemes for an emulsion copolymerization in a semibatch reactor is explained in great detail. The control of copolymer composition for a vinyl acetate/n-butyl acrylate system is investigated. An extended Kalman filter (EKF) is used for the nonlinear state estimation problem, which is based on local linearization of the process model around the suboptimal filter estimates.

Kravaris et al. (1989) developed a nonlinear feedforward/feedback controller to obtain constant copolymer composition for the solution copolymerization of styrene/acrylonitrile in a batch reactor. Two different control policies are evaluated: the temperature profile is computed for a batch reactor and the feed policy of the more reactive monomer is determined for an isothermal reactor. In both cases above, the objective is to keep the copolymer composition constant.

Houston and Schork (1987) employed a time series model to study adaptive/predictive control schemes on a simulated methyl methacrylate semi-batch reactor. Their controls are optimal for the current control only, subject to the constraints of the process model and objective function. Both temperature regulatory control and trajectory tracking were implemented to reduce initiator consumption, batch time, and product polydispersity.

Ponnuswamy et al. (1985) investigated (by simulation only) linear feedback control policies in a MMA batch reactor, derived from a quadratic performance criterion. These policies maintain the state variables (ie., monomer concentration in the reactor and the zeroth and second moments of the dead polymer distribution) along desired trajectories in the presence of disturbances (deviation of the initiator concentration in the reactor from its nominal (optimal) condition). The manipulated variable was the reactor (polymerization) temperature.

Later, Ponnuswamy et al. (1987) applied linear quadratic feedback control to the above system. Temperature was utilized to control conversion and the zeroth and second moments of the molecular weight distribution. The initial initiator concentration was treated as a disturbance. This algorithm was found to give good MWD control. In their paper, three different optimization problems are proposed and analyzed: minimum-time optimal initiator policy; minimum-time optimal temperature policy; and the minimum polydispersity temperature policy. These policies are all derived based on the Maximum Principle applied to the process model.

Simulation of the solution polymerization of MMA in a batch reactor during the final stages of the reaction was performed by O'Driscoll et al. (1988). The objective was to minimize the residual monomer concentration as fast as possible, using a time optimal problem formulation. Both isothermal and nonisothermal cases were studied. An extension to this work was presented later by O'Driscoll and Ponnuswamy (1990), including molecular weight considerations.

Tzouanas and Shah (1985) applied an adaptive pole-placement controller to control the conversion of a batch polymer reactor. In their work, the flow rate of the reactor cooling water was adjusted in a cascade control configuration, in such a way that the reactor temperature was kept close to a desired one specified by the master controller such that the monomer conversion followed a desired trajectory. A PID controller was used as the slave controller. The adaptive pole-placement controller was used as the master controller.

Tzouanas and Shah (1989) implemented their adaptive pole-placement controller to a batch reactor for the solution polymerization of MMA. By manipulating the reactor temperature, they forced the monomer conversion to follow a particular trajectory in simulation and experimental studies. The adaptive controller's performance was similar to that of a generalized minimum variance controller and a PID controller.

Cawthon and Knaebel (1989) analyzed semibatch copolymerization systems using vector-objective techniques to determine the tradeoffs between the different goals of narrowing copolymer composition and polydispersity, and minimizing the reaction time.

Arzamendi and Asua (1991) presented a method to determine the optimal monomer addition strategy to produce homogeneous copolymer under conditions in which the reactor has limited capacity for heat removal. This method allowed for time dependent heat removal rates to be specified as well. The emulsion polymerization of MMA/VAc in an artificially heat removal limited, laboratory reactor produced homogeneous copolymer when the monomer feed rate profile was calculated with the heat transfer limitations taken into account. On the other hand, significant composition drift was observed if the feed rate policy had been calculated assuming all the reaction heat could be removed.

Mayer, Meuldijk and Thoenes (1995) reported on the procedure to control emulsion polymerization to complete conversion while operating at relatively low temperatures and isothermal operation. This technique applied in large scale commercial reactors was described in a short communication.

2.5.3 Long Chain Branching Control

For polymers in which any of the reactions, radical transfer to polymer, terminal double bond polymerization, or internal double bond polymerization, are important, then branched polymers will be formed. Since the branching frequency (i.e., average number of branch points per molecule) usually has a pronounced effect on the polymer properties and particularly polymer processability, it is desirable to investigate policies for controlling it. The effect of a number of reactor operating policies on branching

frequencies has been reported by Hamielec and MacGregor (1983) for styrene/butadiene and vinyl acetate/vinyl chloride copolymers. In general, a batch (or plug-flow) reactor operation will minimize branching frequencies compared to a CSTR or semibatch operation at the same conversion. In fact, one should be aware that different semibatch coplymer composition control policies may give widely different branching behaviour. Further details and examples can be found in Hamielec et al. (1987a).

2.5.4 Particle Size Distribution (PSD) Control

In general, the particle size distribution in semibatch emulsion polymerization can be controlled through seeding of the reactors, or through controlling particle nucleation by manipulating the emulsifier and/or initiator feedrates. Some policies using both of these methods have been investigated. Min and Gostin (1979) used initial seeding of the reactor, followed by a controlled emulsifier feedrate to produce a secondary particle generation. The bimodal PSDs obtained in a pilot plant reactor for polyvinyl chloride emulsion polymerization were well predicted by their population balance model. The effects of the following variables on the final latex PSD were investigated: seed particle size, quantity of seed, solid contents of the seed, and the initial amount of initiator.

Gordon and Weidner (1981) used the ideas of Min and Gostin to control the PSD in the emulsion polymerization of vinyl chloride by manipulating the emulsifier feedrate as a function of conversion. An on-line measure of conversion was obtained from an unsteady-state heat balance and then used as the feedback variable to control the emulsifier feedrate.

Emulsifier feedrate policies which produce multiple particle generations, or extend the period of particle generation have been studied by Penlidis (1986) for vinyl acetate emulsion polymerization. These policies significantly broaden the resulting PSD over that of a batch reactor operation. In a similar study, Lin and co-workers (1980) tried to control the particle size in vinyl chloride polymerization by varying the ratio of monomer to water at the beginning of the polymerization.

2.5.5 Temperature Control

Polymerization reactions are exothermic in nature. The amount of heat released as monomer is converted to polymer is considerable. Temperature variations greatly affect the kinetics of polymerization processes, and through the kinetics they have a strong impact on the way the produced polymer is structured and thus, on its physical properties and quality characteristics. Temperature control is, therefore, critical in the production of high quality polymers and specialty products of desired molecular structures.

Temperature control of a large, highly exothermic, batch or semi-batch, chemical or polymer reactor can be an involved problem. The reaction may be auto-accelerating. Heat transfer rates can vary during the

process. Random disturbances can enter the process from many sources. Changes in reactant feed rate often produce an inverse temperature response, since the cooling effect of the increased feed precedes the increase in the reaction rate. This is especially true for polymerizations. Changes in temperature can alter the reaction rate, resulting in poor molecular weight control, and, in severe cases, an entirely different polymer product. If control deteriorates badly enough, the tendency to autoacceleration can result in an uncontrolled and extremely hazardous runaway polymerization. Since temperature control can be critical to safety and quality, highly trained operators are often assigned to control the polymerization process.

Control of reactor temperature is also critical from a purely operational point of view. If the polymerization temperature is allowed to increase, monomer conversion increases and more polymer is produced. Hence, the polymerization mixture becomes more viscous and heat removal becomes difficult. Therefore, reactor temperature must be kept within the limits that allow one to carry out a safe polymerization, ie., within the system's heat removal capabilities.

Many papers have been published from both the academic and industrial view on temperature control of polymerization reactors.

Garcia (1984) applied quadratic dynamic matrix control to regulate the temperature of a synthetic rubber process in a semibatch reactor. This method used a linearization of a predetermined process model to predict the reactor temperature over a time horizon. The model was linearized around each point and optimal cooling applied to control the temperature along the desired trajectory. Control was effective and minimal on-line tuning was required.

Cluett et al. (1985) applied an adaptive-predictive control scheme with a dead-zone to temperature control, temperature set-point tracking and constant reaction rate control of a simulated batch suspension poly(vinyl chloride) (PVC) reactor. The adaptive controller compared favourably with the self-tuning controller of Astrom and Wittenmark (1973) and with a well-tuned PID controller.

Niederlinski et al. (1987) implemented a self-tuning temperature controller on an industrial poly(vinyl chloride) batch reactor. Modelling of the process was performed by empirical on-line system identification. The manipulated variable was the jacket temperature.

Takamatsu et al. (1986a,b) proposed two kinds of model-reference adaptive control schemes for the control of a batch suspension styrene polymerization reactor. At the simulation level, the two schemes seemed promising for the control of reactor temperature and average degree of polymerization.

Later, Takamatsu et al. (1988) used an adaptive version of the internal model control of Garcia and Morari (1982) for a temperature profile tracking problem in a simulated batch suspension styrene polymerization. Their approach was to maintain the reactor temperature and initiator concentration along predetermined profiles, so that the final polymer product had a desired average chain length and polydispersity. The effect

of increased viscosity at high conversion levels on the heat transfer characteristics of the system was simulated by reducing the value of the overall heat transfer coefficient down to 50% of its initial value.

An approach based upon time-series and stochastic control methods (Dougherty et al., 1988) was developed to generate a simple PID controller to control reaction temperature. They used time-series identification techniques to generate a transfer function relating the control variable (reactor temperature) to the manipulated one (jacket coolant flow rate), with the manipulated variable subject to a Pseudo-random Binary Sequence (PRBS). Then, they developed their control scheme by minimizing an objective function similar to that of a constrained minimum variance control algorithm and recast the controller into a PID form.

Louie et al. (1985b) proposed a programmed oxygen injection method for curbing the gel effect and manipulating the system temperature in a batch MMA emulsion polymerization reactor. The primary application of oxygen control is to rapidly alter existing reaction conditions with respect to radical populations inside the reactor. To minimize possible detrimental effects on product molecular weights, they suggested pulse oxygen control to determine the effects of controller set point changes on molecular weights.

Choi and Ray (1985) studied the steady state behaviour of a fluidized bed reactor for ethylene polymerization as a function of catalyst injection and demonstrated that close temperature control is important for safety and reactor runaway problems.

Davidson (1987) described an intelligent microprocessor-based temperature control system for jacketed reactors. The intelligence was based on logic used by skilled operators and implemented using simple algorithms. This controller based on a logical model was advocated as easier to install and maintain than one using a mathematical model based on dynamics which may have modelling errors.

Kiparissides, Sidiropoulou, Voutetakis and Frousakis (1990) demonstrated the application of dynamic matrix control (DMC) and extended self-tuning regulator (ESTR) to bulk polymerization of MMA in a batch reactor under strong diffusion limitations of termination and propagation reactions. They controlled conversion and number average molecular weight (NAMW) along a state trajectory using temperature. In the face of impurities in the polymerization kinetics, product variations were considerably reduced by operating the reactor under closed-loop control. Performance was compared to a linear quadratic controller (LQC) and tuning parameters for DMC and ESTR were investigated in a simulation study.

Defaye, Renier, Chabanon, Caralp and Vidal (1993) showed that adaptive-predictive temperature control was workable on a moderately exothermic pilot plant copolymerization reactor. The system was the free radical, solution copolymerization of vinyl acetate and 2-ethyl-hexyl acrylate in ethyl acetate solvent. The initiator was benzoyl peroxide. Data filtering in the estimation model was important especially to deal with dynamic effects associated with reactant addition.

Chang, Hsu and Sung (1996) proposed a method for optimization and control of batch reactors. The system studied was for the synthesis of hexyl monoester of maleic acid reaction. A SISO modified globally linearizing control (MGLC) structure was proposed for temperature trajectory tracking. The heating/cooling system was shown to effectively control the highly exothermic reaction.

Polacco, Semino and Palla (1996) analyzed batch polymerization of MMA in an agarose suspension. Their focus was on the sensitivity of the system to operating conditions, the kinetic parameters, and the heat transfer properties. Maximum temperature was a key concern for this biomedical application. Initial initiator concentration and other parameters that increased the rate of polymerization did not affect much the maximum temperature but rather the time at which the maximum temperature appeared.

2.6 Continuous Reactor Control

This section discusses literature on continuous reactors with respect to the operating policies, the design of stable reactor systems, the estimation of unmeasured states, and the on-line closed-loop control of polymer or latex properties.

There has been an increasing number of publications in the literature on the control of continuous polymerization reactors. Increased competition and emphasis on product quality, together with improvements in control hardware and advances in control theory have provided many new tools to attack the continuous polymerization control problem. Our survey shows that most of the articles are from academia; only a few come from industry. Also, theoretical work greatly outnumbers experimental applications, and the continuous stirred tank reactor (CSTR) is the clear favourite for testing reactor control strategies at the simulation level.

MacGregor et al. (1984) divided continuous reactor control into the following viewpoints:

- steady-state optimization
- dynamic optimization
- reactor design optimization
- state estimation
- on-line control

Optimization papers have been reviewed in Sec. 2.2 and state estimation has been covered in Sec. 2.3.

Table 2.8 summarizes recent work on the on-line control of continuous polymerization reactors.

Table 2.8: Recent work on control of continuous polymer reactor systems

Reference	System	Work	Notes
Alvarez et al. (1993)		T	stabilization with saturated feedback
Alvarez (1996)	CSTR	S	output feedback control; free radical homopolymerization, example
Adebekun (1996)	CSTR	T	input-output linearization
Assala et al. (1997)	STY/CSTR	S	stabilization with input constraints
Brandolin et al. (1991)	PE	S	optimal policy comparison
Broadhead et al. (1996)	extrusion		
Chien & Penlidis (1994b,c)	MMA	S,X	conversion control, effect of impurities
De Souza et al. (1996)	VA	S	ANN (IMC) model predictive compared to PID; T control
Dittmar et al. (1991)	SAN/CSTR	S	compares T control by PID, GPC and HPC on open-loop unstable reactor
Dunia & Edgar (1996)		T	linear system; GMC
Gazi et al. (1996)	STY	T	evaluation of controllers
Han-Abedekun et al. (1997)	olefins, copolymer		design and control for kinetic studies
Inglis et al. (1991)	MMA/CSTR	S	long range GPC conversion control, also experimental T control in batch
Ishida et al. (1996)	STY/bulk	S	neural network; dynamic state prediction and control
Lee et al. (1996)		T	uncertain delay; robust stability, Smith predictor
Lie & Balchen (1992)	PP	S	comparison of controllers
Maner et al. (1996)		S	NL-MPC; 2nd order Volterra models
Maner & Doyle III (1997)	MMA/VAc	T,S	autoregressive+Volterra based MPC 2 polymerization reactor case studies
Maschio et al. (1994)	MMA	S,X	suspension polymerization
Jiang et al. (1997)	PE	X	heat removal in supercondensed mode
Mendoza-Bustos et al. (1990)	MMA	S	adaptive conversion control
Meziou et al. (1996)	EPDM	S	DMC control of composition & Mooney viscosity
Mrazek et al. (1991)	VC/suspension	X	gradual initiator dosage
Mutha (1996)	MMA	T,X	simulation NBR, experimental MMA; control nonaffine NL-MPC algorithm
Padilla & Alvarez (1997)	VA-MMA copolymer	T,S	control study - linear interaction compensator; MIMO reactor control
Qammar et al. (1996)		S	chaotic; linear and nonlinear controllers
Semino & Ray (1995a)		T	population balance; controllability study
Semino & Ray (1995b)	emulsion		constrained control action; control of oscillations
Soroush & Kravaris (1994)	MMA	X	nonlinear GLC, singular characteristic matrix
Soroush (1996)	NL-MIMO	T	evaluation of achievable control quality
Soroush & Kravaris (1996a)	MIMO	T,S	state feedback, non-linear, GLC
Soroush & Kravaris (1996b)			MPC formulation of GLC
Smith & Malone (1997)	PMMA	T	attainable regions for design of systems
Tan et al. (1996a)			analysis and design, Smith predictor

2.6.1 Steady State Optimization

Farber (1986) applied multiobjective optimization to study aspects of the steady state optimality of continuous methyl methacrylate-vinyl acetate and styrene-acrylonitrile copolymerization systems. He made use of the concept of Pareto optimality to enable the determination of non-inferior sets, which permits a clear decision on tradeoffs related to operating conditions and product specifications.

Das and Rodriguez (1990) studied the solution copolymerization of 2-ethylhexyl acrylate with vinyl chloroacetate in a CSTR. Based on physicochemical and diffusional considerations, they were able to model the steady state rates. However, their attempt to model the transient behaviour of the reactor was unsuccessful due to the rapid composition drift as the system approaches steady state.

2.6.2 Dynamic Optimization

Many papers have demonstrated that for certain polymerization processes forced periodic operation of continuous reactors is sometimes beneficial in improving the performance and/or the quality of the final product. Meira (1981) reviewed most of the papers that have appeared on this subject. There are only a few more recent papers which demonstrate the application of optimal periodic forcing of the feed flows on continuous "living" (nonterminated) anionic polymerization reactors to obtain a prespecified value of the MWD. These are summarized below:

Frontini et al. (1986, 1987) carried out simulations of a living anionic solution homopolymerization in an isothermal CSTR under optimal periodic forcing of the initiator and monomer flow rates in order to obtain prespecified number average chain length and polydispersity. When such a polymerization is operated at steady-state, two situations arise: the number-average chain length (or weight-average chain length) may be changed simply by adjusting the flow ratio between the monomer and initiator feed; the polymer produced exhibits a Schultz-Flory distribution (ie., polydispersity is close to 2). Alternatively, periodic operation of the CSTR can provide added flexibility in the quality of the average polymer produced. That is, by maximizing (minimizing) an objective functional, a range of the average polydispersity around the steady-state value of 2 can be obtained. It is possible then to produce polymers with any average polydispersity within this range.

The papers by Couso and Meira (1984) and Elicabe and Meira (1989) also examine the effects of periodic operation on several system variables, specifically those related to the MWD. Gugliotta et al. (1991) theoretically and experimentally investigated the effect of forced-feed oscillations of an intentionally added chain transfer agent on isothermal free-radical polymerizations in a CSTR. They found that under certain conditions it is possible to broaden the average MWD without affecting other properties.

Pinto and Ray (1995a,b) have published a numerous papers on the modelling, control and characteristics of the continuous solution copolymerization of vinyl acetate (VA) and methyl methacrylate (MMA) in tert-

butanol (TB). In 1995, they built a dynamic model to describe the system. They used experimental results to validate their kinetic parameters and proceeded to draw bifurcation diagrams for the dynamic behaviour of their system (Pinto, 1995a). Their next article (Pinto and Ray, 1995b) reported the dramatic effects of feed composition and studied the full bifurcation model. The following year, (Pinto and Ray, 1996), they showed theoretically and experimentally that small amounts of inhibitors in the feed stream may lead to unstable operation.

2.6.3 Start-Ups and Change of Grades

In many industrial free-radical polymerization processes, a reactor can be very sensitive to small changes in the concentration of the radical generator. Therefore, particular care should be paid to the effects of start-up procedures on reactor behaviour.

Accounts of experimental investigations, in which continuous-flow back-mixed reactors have been used, do not always describe the details of start-up procedures. However, some early studies have indicated that the dynamic behaviour upon start-up can be very interesting (Brooks, 1981). In the start-up regime the following problems are to be considered:

- The reaction can run away so rapidly that the safety of the operation is not guaranteed.
- As a consequence of the variation of concentrations and temperature, polymers produced in a non-optimum start-up regime are very heterogenous with respect to the molecular and chemical composition. These polymers possess bad properties.

The first problem can be solved either by starting with pure solvent in the reactor or by operating the reactor in the batch mode with an excess of solvent in the first period of the start-up regime. The second problem can be solved with a product-optimum start-up policy with temperature programming and feed ratio control. Based on the above strategies, Thiele (1986) developed an optimum start-up policy for a bulk styrene-acrylonitrile copolymerization reactor.

It was shown that changes in the start-up procedure during the continuous emulsion polymerization of styrene (Baddar and Brooks, 1984) and MMA (Brooks and Raman, 1987) affected both the transient and long-term behaviour. From their experiments, Brooks and co-workers found that reactor behaviour, particularly with respect to conversion, particle size distribution and particle nucleation rate, can be controlled by the choice of the initial conditions.

2.6.4 Reactor Design Optimization

Carrat, Sherwin and Soong (1984) developed a two-stage continuous reactor system for bulk methyl methacrylate polymerization. This process utilizes a CSTR as a first-stage prepolymerizer and a spray

tower as the second-stage finishing reactor. Use of a CSTR offers good temperature control and product uniformity during the early stage. Spraying the partially polymerized mixture into the tower as fine droplets prior to the onset of the gel effect eliminates the problems of transporting, agitating, and mixing a reacting system with a rapidly increasing viscosity. Based on their experimental work and simulation model, the authors came up with a practical design procedure for such systems.

Taylor and Reichert (1985) investigated the effects of micro- and macromixing and reactor type (batch reactor (BR), segregated CSTR (SCSTR), and homogeneous CSTR (HCSTR)) with respect to the breadth of the MWD in a vinyl acetate polymerization system. Their simulation results showed that the MWD in the three reactor types is not fixed, but instead is a strong function of reactant concentrations and predominantly of chain branching. As the monomer concentration is increased or polymer transfer and terminal double bond polymerization become more important, the MWD will become broader in the HCSTR than in the BR or SCSTR..

MacGregor (1986) and Penlidis et al. (1989b) have shown that by not considering reactor stability and control at the design stage, it is possible to have a situation in which the process behaviour can not be optimized by the application of any control theory. Redesigning the process, on the other hand, can lead to a well-behaved controllable process, and hence to a rather simplified control problem. Similar results have also been shown by Poehlein, Schork and co-workers (e.g., Temeng and Schork, 1987, 1989).

Ferrero and Chiovetta (1990) developed a short-cut method for the preliminary design of a tubular loop reactor for bulk propylene polymerization. A relationship between the average particle size in the reactor and the particle size at both inlet and outlet was obtained.

Kataoka, Ohmura, Kouzu, Shimamura and Okubo (1995) studied continuous emulsion polymerization of styrene in a continuous Taylor vortex flow reactor which has characteristics of a plug flow reactor (PFR). Steady state conversion, average molecular weight and size distribution of latex particles could be controlled by the flow condition or reactor temperature.

Russo and Bequette (1996b) studied the effect of assuming that the cooling jacket dynamics are negligible for a CSTR. They demonstrate areas of unfeasibility in reactor scale-up or operation.

2.6.5 Temperature Control

Problems often encountered in industrial polymerizations are associated with the heat released by the highly exothermic reactions which, combined with lowered thermal diffusivity through the increased viscosity of the reacting media over the course of polymerization, can cause thermal runaway. Onset of the gel (Trommsdorff) effect can cause excessive temperature rise, rapid increase in conversion, and plugging of equipment

For a typical addition polymerization, the heat of polymerization ranges from 10 to 20 Kcal/mole, which can result in an adiabatic temperature rise of 200 to 400°C. This large generation of heat, coupled with the low thermal diffusivity of the reacting system, often leads to thermal runway. Therefore, control of the process is difficult. A temperature rise generally causes a lowering of the degree of polymerization. Hence, large temperature variations during the course of the reaction broaden the product molecular-weight distribution, with accompanying deterioration of the mechanical properties of the polymer. Another serious problem occurring is the Trommsdorff or "gel" effect. The onset of the gel effect frequently causes uncontrollable reactions, resulting in excessive temperature rise, rapid conversion rise, and plugging of equipment.

Several publications tackled the temperature control problem. Henderson and Cornejo (1989) considered three different mechanisms of removing the heat of reaction (cooling coils and jacket, vaporization of monomer and solvent, and circulation of reaction mixture through an external heat exchanger) for temperature control of the continuous production of polystyrene by thermal polymerization. Their results are based on the simulation of a 5000 gal reactor. Using a model of the process, simulations were performed and the following process variables were estimated as a function of steady-state reaction temperature: conversion, weight-average molecular weight, zero-shear viscosity, Reynolds number, agitator shaft horsepower, and the heat generated due to the agitator only (ie., the heat of agitation). This work illustrates some of the problems encountered in the industrial operation of polymerization reactors.

Liptak (1986) discussed different temperature and pressure control strategies to maintain constant concentrations and safety. He pointed out that in large polymerization reactors having low heat-transfer coefficients and large changes in heat evolution, the conventional temperature cascade control loop is not fast enough. On the other hand, pressure measurements give an almost instantaneous indication of changes in temperature. Based on this observation, he proposed that a pressure-compensated temperature control system can be applied to a reactor with both jacket and overhead condenser cooling. The net result is the ability to operate the reactor at a much higher reaction rate, and thereby obtain higher productivity than is possible with temperature control alone.

In studying the dynamic behaviour of fluidized bed reactors for ethylene and propylene polymerization, Choi and Ray (1985) showed that even in closed-loop, temperature runaway in the reactor can occur in some situations in which the rate of heat removed from the recirculating gas becomes lower than the rate of heat generated in the reactor.

Kwalik and Schork (1985) applied the adaptive multivariable self-tuning regulator of Vogel and Edgar (1982) to a simulated continuous solution polymerization of MMA to control monomer concentration and temperature in the reactor. The effect of monomer impurities was simulated by a step change in the initiator efficiency factor. Manipulation of the reactor jacket temperature and initiator concentration in the feed proved successful in the regions where only unique steady-states occurred. Their results show that slight

variations in the manipulated variables produce unacceptable fluctuations in the controlled variables. Also, they found that in the region of multiple steady-states the interactions of the multivariable system are magnified and for this reason a multivariable controller without proper decoupling may not be suitable.

Whatley and Pott (1984) applied an adaptive gain strategy for temperature control by manipulating the circulation oil temperature. This technique was used to provide the necessary driving-force compensation in the oil-circulation loop temperature controller for the following situations: 1) many grades of polymer were made which required different sets of tuning constants for the controller; 2) if tuned without the adaptive gain strategy, the system response to upsets was very poor; and 3) set point changes must be made extremely slowly to preserve system stability.

Marini and Georgakis (1984) studied the temperature control of a continuous low density polyethylene reactor at the simulation stage. From a linearized model, they developed a reaction rate controller (a type of variable gain PI controller) which exhibited superior performance to that of constant gain classical PI controllers.

2.6.6 State Estimation / Filtering

State estimation techniques have been developed to provide acceptable estimates of state variables despite the fact that some of the polymer or latex properties of interest may not be directly measurable and/or may be subject to stochastic measurement error and process disturbances. The interest is in sequential estimates which can be used for on-line control schemes, ie., the estimates must be continuously updated by recursive algorithms. The best current (filtered) estimates may then be used in control applications.

Fundamentally, state estimation considers the problem of determining the values of states from the knowledge of the outputs (data) and the inputs (control, disturbances). For the estimation to be successful, the input-output information must provide a unique state estimation, which implies system observability and/or reachability (Ray, 1981).

Observability plays an important role in state estimation for polymerization processes. In general, the molecular weight states are non-observable from measurements on the material balance or the particle population balances states of the model. Since on-line measurements for the latter states are more readily available, this means that tracking the molecular weight development of a polymerization process is often very difficult.

Ray (1985) presents a table where the observability of monomer conversion, initiator conversion, and other variables or parameters related to the molecular structure is considered. Several typical measurements and combinations of measurements such as temperature, density, intrinsic viscosity, etc., are reviewed.

Several papers have been published dealing with state estimation via Kalman Filtering methods. MacGregor et al. (1986) tracked the material and population balance states in emulsion polymerization using information from on-line densitometers. They also stressed the importance of modelling the effect of impurities, and of incorporating nonstationary stochastic states into the model of the process in order to obtain realistic state estimators that are capable of tracking an industrial process. This work was later extended in the very interesting papers by Kozub and MacGregor (1989) and Gagnon and Macgregor (1991).

In other applications, Papadopoulou and Gilles (1986) applied the Kalman filtering method for continuous estimation of the chain length distribution in a polymerization reactor using on-line gel-permeation chromatographic measurements.

Adebekun and Schork (1989b) applied Kalman filter techniques to solution polymerization of MMA in a CSTR in combination with open-loop dynamic simulations. They showed that, even in the event of incomplete state observability, the open-loop performance of these estimators could still be acceptable. Then, they employed these estimates in closed-loop nonlinear model reference control schemes. In order to motivate realistic evaluations, an extended Kalman filter was incorporated for reconstruction of unavailable (and/or infrequently measurable) state variables (e.g., molecular weight distribution) from the available measurements (monomer, temperature, initiator, and solvent levels). They proposed that if a MWD measurement is available, then the measurement can be used to update the current estimates of MWD through a two-time scale filtering algorithm (Ellis et al., 1988). This is achieved by reinitializing the MWD estimates to the filtered estimates at the time the process sample was taken and then, resimulating the MWD subsystem to real time.

Daoutidis and Christofides (1995) tried to develop feedforward/static state feedback combined with state observers in a way similar to their pure output feedback system. The developed control methodology was applied to an exothermic CSTR in simulations.

Viel et al. (1995) showed global asymptotic stability of a polymerization reactor, high gain observer, and controller system after applying input/output linearization. They modelled solution polymerization of styrene in toluene using the standard free-radical kinetic mechanism and defined their system with a model consisting of three mass balances, one heat balance and the zeroth and second moments of dead polymer. Linearization was done for a MIMO controller with reactor temperature and feed monomer weight fraction as the control variables. The observer used measured temperature and density of the mixture.

2.6.7 Recent Efforts Towards On-Line Control

Although maintaining a stable process operation was possibly the only objective of control systems in the past, there is an increasing demand for modern control systems to satisfy one or more of the following performance criteria (Garcia et al., 1989):

- Economic - These can be associated with either maintaining process variables at the targets dictated by the optimization phase or dynamically minimizing an operating cost function.
- Safety and environmental - Some process variables must not violate specified bounds for reasons of personnel or equipment safety, or because of environment regulations.
- Equipment - The control system must not drive the process outside the physical limitations of the equipment.
- Product quality - Specifications on products must be satisfied. This is probably the most difficult performance criterion.
- Human preference - There exist levels of variable oscillations or jaggedness that the operator will not tolerate. There can also be preferred modes of operation.

The whole spectrum of process control methodologies is faced with the solution of the above problems. In an effort to satisfy all these integrated performance criteria, many recent pieces of work have shown promise in on-line control of polymerization reactor systems. The objective is to design a control system which can *on-line update the manipulated variables to satisfy multiple, changing performance criteria in the face of changing plant characteristics.*

In this section, different on-line control methodologies that have been published for control of continuous polymerization reactors are reviewed. Based on a rather loose classification of the techniques involved in the controller design, the schemes are reviewed in 7 parts, namely, adaptive control, inferential control, linear quadratic Gaussian control, model predictive control, model reference control, multivariable control and nonlinear model based control. Several of these schemes can be cast in the general internal model control (IMC) framework. One should note that, in some cases, there is no clear-cut distinction on the classification followed and hence, considerable overlapping is found in the design and implementation of these controllers, which makes the classification extremely difficult. This is also partly due to the fact that, up to now, there is still no "unified" classification of the many control theories and techniques published to date, which often may be slight variations on the same theme with a different name. Therefore, the purpose of our classification is basically to reflect the main structure of the control system and convenience in reviewing the papers found.

Adaptive Control

There has been lately a growing interest in the application of adaptive and self-optimizing control to polymer reactor systems. The reason for this trend is clear. Most commercial polymer reactors exhibit

complex, nonlinear dynamics, and the stability characteristics of the system may change drastically throughout the period of operation. These changes are especially large during start-up and set point changes. A poor control system may then lead to runaway situations and subsequent loss of production time and valuable raw materials.

Many different approaches to adaptive control of nonlinear systems have been proposed. Two schemes in particular have attracted most interest: model reference adaptive control (MRAC) and self-tuning regulators (STR). Both schemes actually turn out to be special cases of a more general design philosophy (Goodwin and Sin, 1984). A detailed review of the structure of the above controllers and their applications by earlier researchers has been provided by Elicabe and Meira (1988).

Takamatsu et al. (1986b) employ an adaptive/inferential control system for the control of weight-average molecular weight in the presence of deterministic disturbances. In particular, two polymerization systems are investigated at the simulation level; solution polymerization of isobutylene and styrene in a CSTR. In the first polymerization system either initiator concentration in the feed or total feedrate is used as the manipulated variable and the reactor temperature becomes a secondary output from which the unknown deterministic disturbances could be estimated. With the styrene polymerization the reactor temperature is kept constant and the weight-average molecular weight is controlled by manipulating the total feedrate. In this case the monomer concentration in the reactor is utilized as the secondary output.

Mendoza-Bustos et al. (1990a,b) present an adaptive controller to control monomer conversion in the presence of stochastic disturbances for the solution polymerization of MMA in a CSTR at the simulation level. The controller has the stability and robustness features that are essential in dealing with unmodelled dynamics and unmeasured disturbances. The disturbances are drifting concentration levels of reactive impurities in the feed to the reactor. The manipulated variable that is chosen to control the monomer conversion at its target value is the initiator concentration in the feed. Simulation results of this adaptive scheme are compared to those of the minimum variance and self-tuning controllers.

Inferential Control

A major difficulty with controlling product quality in industrial polymerization reactors is the lack of suitable on-line measurement technology for polymer quality variables (MacGregor et al., 1984; Elicabe and Meira, 1988). While temperature, pressure, flow rate and gas composition are routinely measured on-line, key quality variables such as molecular weight and copolymer composition must usually be measured off-line in quality control laboratories (Richards and Schnelle, 1988). These delayed and infrequent quality measurements provide essential feedback for both process control and model updating purposes. Combination of information from on-line and off-line measurements with process models can lead to improved estimates of quality variables between samples and to improved product property control.

McAuley and MacGregor (1991) developed a scheme to predict melt index and density in an industrial fluidized-bed ethylene copolymerization reactor. They used theoretically-based models to predict quality variables from on-line temperature and gas composition measurements. The adjustable parameters in these models were updated on-line using infrequent laboratory measurements and a recursive parameter estimation technique. It was shown that both melt index and density could be successfully predicted.

Linear Quadratic Gaussian (LQG) Control

In the past decade, several internal model-based control methods have been developed: Model Algorithmic Control (MAC); Dynamic Matrix Control (DMC); Quadratic Dynamic Matrix Control (QDMC); Internal Model Control (IMC); and Quadratic Programming Internal Model Control (QPIMC). These methods have been quite successful in industrial applications, especially from the petrochemical industry, because they provide a high degree of flexibility and allow for on-line tuning. Garcia and Morari (1982) gave an unified review of DMC, MAC and IMC, pointing out common features and noting analogies with classical forms of optimal control.

A shortcoming of the IMC-type methods is that an open-loop stable system is required; the nonminimum phase characteristics need to be "factored out" in advance in the IMC design procedures. The well-studied linear quadratic (LQ) feedback control method (e.g., MacGregor and Tidwell, 1980; Astrom and Wittenmark, 1984) can stabilize an open-loop unstable process and is applicable directly to processes with nonminimum-phase characteristics.

An excellent illustration of the empirical modelling approach was recently given in Kelly et al. (1987), concerning the linear-quadratic (LQ) control of a continuous industrial polymerization reactor train for the production of stereoregular butadiene rubber based on the use of quality control laboratory data. Their models were identified in the closed-loop mode and then used to design minimum variance type control strategies for monomer conversion and Mooney viscosity of the final product. To ensure identifiability under closed-loop conditions independent pseudo-random binary sequence (PRBS) perturbations were introduced into all inputs. These controllers manipulate the flowrates of the Ziegler-Natta catalyst components and a chemical transfer agent. The design of these controllers via spectral factorization, and their robustness/performance trade-off properties have been discussed thoroughly by Harris et al. (1982), Bergh and MacGregor (1987), and Harris and MacGregor (1987).

Chien (1992) are critically evaluated and experimentally verified the robustness/performance of LQ (e.g., minimum variance, constrained minimum variance and one-step optimal) controllers for controlling the conversion level of a continuous MMA polymerization reactor system. The results show that these LQ controllers provide better robustness and performance as compared to the conventional PID-class controllers.

Chien and Penlidis (1994c) studied the effect of impurities on the polymerization of MMA in toluene in a CSTR. The modelling and control objectives for conversion and molecular weight. In this second of two papers, they evaluate PID controllers: PID, Smith predictor, and Dahlin's control, and stochastic control strategies: minimum variance (MVC), constrained MVC, and one-step optimal. Simulations were used to identify promising scenarios and 2-3 day experimental runs were used to verify them. Conversion was controlled, impurity effects matched theoretical predictions, and controllers were implemented to work.

Model Predictive Control

Model predictive control (MPC) is a family of controllers in which there is a direct use of an explicit and separately identifiable model. Most of the design techniques emanating from MPC use some kind of a dynamic model to predict the effect of future control actions on the process output. Some example models are: step response (dynamic matrix control - DMC), impulse response (model algorithmic control - MAC), and transfer function (generalized predictive control - GPC). The future manipulations of the input are determined by an optimization step which minimizes a horizon of predicted errors between the process outputs and the desired outputs (set-points) subject to operating constraints.

Farber and Ydstie (1986) used a parameter-adaptive regulator algorithm similar to the dynamic matrix controller (DMC) (Cutler and Ramaker, 1978) in their simulation studies of the control of a continuous solution polymerization of styrene in ethylbenzene in the presence of AIBN initiator. To improve adaptation, they applied a variable, forgetting-factor estimator to enable adaptation. Also, an extended horizon controller, with a horizon larger than the expected delay time of the system, was used to enhance controller robustness in the presence of time-varying delays and non-minimum-phase system characteristics. In this way, they were able to trade off the difficulties in controlling non-minimum phase systems against the use of a higher order model.

Hidalgo and Brosilow (1990) used a combination of model predictive control (MPC) and coordinated control strategies to control a simulated, open-loop unstable, free-radical solution polymerization of styrene in a CSTR. The controls are the flowrate of cooling water supplied to the CSTR jacket and the flowrate of monomer to the reactor. They assumed that modelling errors are due to changes in the unmeasured initiator level in the reactor. By adjusting the initiator level, stable control and exact tracking of the set point at steady state can be accomplished. One practical problem of such a control strategy is the variation of monomer flowrate which will result in varying residence times or differing levels of monomer soluble impurities in the reactor. Also, their assumption that the process is a fourth-order nonlinear process was not justified.

The economic operation of a polymerization process usually requires that unreacted species be recovered and fed back into the process. Associated with this unreacted monomer recycle is also recycle of impurities.

Impurities are known to consume free radicals and therefore prevent polymer generation and growth. Only upon introduction of additional initiator to consume these impurities will the reaction proceed as desired. Polymer productivity is directly related to monomer conversion (concentration) in the reactor. Therefore, a reasonable control objective would be to minimize the variance of the predicted process outputs from the set-point trajectory. The optimal design may be implemented via long-range predictive controllers such as GPC (Inglis et al., 1991) by relating the observer polynomial to the disturbance dynamics. In practice, the observer polynomial can be used more effectively as a detuning tool to reduce the magnitude of the control action and improve robustness of the controller to modelling errors in the same way as the weighing parameter does in constrained minimum variance, or linear-quadratic output controllers.

Lawrence (1994) applied MPC and GPC to simulation and experimental studies of a continuous, solution polymerization of MMA in toluene. SISO MPC and GPC were applied to simulations and since GPC performed better, two SISO GPC controllers were chosen for experimental verification. The experimental work on the bench scale CSTR supported the simulation results.

Model Reference Control

In model reference control, one is interested in deriving a control law to enable a nonlinear process to follow some desired reference model. To do this, a mechanistic model is generally converted into dimensionless model equations. Thus, these control schemes are sufficiently general for most types of polymerization reactors. For example, it is possible for the dimensionless heat transfer coefficient to be a function of the reactor medium viscosity. Therefore, as long as such correlations have been properly identified, the control schemes are quite feasible.

Elicabe and Meira (1989) proposed a cascade model reference adaptive algorithm to control a continuous living anionic polymerization reactor, under forced oscillation of its feeds. According to their control scheme, the first two moments of the number chain length distribution were forced to follow predetermined periodic trajectories. The major limitation of this method was that a relatively simple model for a living anionic polymerization was chosen; and that the control method was strictly applicable to continuous systems without dead times. In real situations, a process size exclusion chromatograph would be required to measure monomer concentration and the MWD moments, and this would clearly imply discontinuous and lagged measurements.

Adebekun and Schork (1989a,b) applied a nonlinear reference model control algorithm on a simulation of a continuous methyl methacrylate polymerization reactor. Based on the full model of Tanner et al. (1987), they used bifurcation theory and "catastrophe" sets to demonstrate the importance of reactor "psychoanalysis" in design applications. Their simulation results showed that input multiplicity is a property of the system and not of the controller. The leading moments of the MWD were estimated using model reduction techniques as described in Schuler and Zhang (1985).

Multivariable Control

Internal model control (IMC) design techniques were discussed by Garcia and Morari (1982), but the philosophy of including process models in control system design has been used since the 1950s. The Smith Predictor is one of the first widely discussed applications of simple linear process models internal in a control system. It contains one element of the basic philosophy of IMC þ the inverse of the process model. Another characteristic of IMC is the form of the controller, which consists of an approximation to the process inverse and a filter to provide a desired process variable trajectory. Inverting the process forces the controller to follow the trajectory in an optimal fashion.

There are a number of publications (outside the scope of this review) on the successful application of IMC in the petrochemical (e.g., packed bed reactor (Kozub et al., 1989)) and pulp and paper industry (e.g., Matsko, 1985). Direct applications of IMC to controlling polymerization reactors are forthcoming.

Tanner et al. (1987) discussed a multivariable optimal Proportional-Integral control structure with local linearization for controlling the MWD in a continuous methyl methacrylate reactor. The polymerization model used in the simulations is that developed by Schmidt and Ray (1981). Monomer conversion, initiator concentration, and reactor temperature were among the controlled variables. The performance of the controller was evaluated for cases involving start-up, disturbance rejection, and set-point tracking.

Kwalik and Schork (1985) and Temeng and Schork (1987, 1989) proposed a similar multivariable control policy for the isothermal emulsion polymerization of MMA in a seeded CSTR. The multivariable pole-zero placement, dead-time compensator developed by Vogel (1982) was utilized to control monomer conversion and average particle diameter in the presence of unknown disturbances (inhibitor concentration in the feed). The manipulated variables were the flowrate of a concentrated initiator solution and the split of the water flow between the tubular reactor (seeding pre-reactor in which particle nucleation takes place) and the main CSTR. Their simulation results indicate good control of both conversion and particle size. The effectiveness of using the tubular reactor to eliminate the oscillations in the CSTR was verified in an open-loop experiment.

Richards and Congalidis (1987) proposed a control structure to regulate polymer production, copolymer composition, molecular weight and reactor temperature in a simulated methyl methacrylate/vinyl acetate continuous solution copolymerization system. Their strategy combines a feedforward ratio control of recycle streams to maintain constant composition and flow to the reactor, and multivariable feedback PI control for polymer rate, composition, molecular weight, and reactor temperature. Several combinations of control variables were analyzed and evaluated. Subsequently, Congalidis et al. (1989) applied an input/output pairing approach to identify the fundamental structure of the solution copolymerization problem. Their design procedure was based on ranking various candidate structures according to the

condition number, minimum singular value and relative gain array of the process transfer function for the specific structure. They used flow ratios as manipulated variables and implemented PI controllers to control conversion, molecular weights and composition. The attractive part of their control strategy is its ease of tuning. However, a more complex controller may be needed to improve robustness.

Ogunye (1994) developed multivariable control schemes for set-point tracking and disturbance rejection for the continuous solution MMA polymerization. These schemes were investigated experimentally in the control of both conversion and average molecular weight using the initiator flow rate and the reactor jacket cooling water flow rate (i.e., reactor temperature) as the manipulated variables. Both constrained minimum variance control and one-step optimal control strategies were studied, along with GPC and DMC.

Semino and Ray (1995b) continued their modelling of the emulsion polymerization of MMA as a population balance system. Oscillatory and unstable behaviour were previously identified as problems that could be controlled with feed rate. They proposed to control the polymerization in a CSTR by manipulating surfactant, initiator or inhibitor feed. They applied frequency domain tuning techniques to control the non-linear unstable process with SISO PI or PID controllers. Various combinations of manipulated and controlled variables were evaluated. When the dynamics were sufficiently slow, derivative action was unnecessary to guarantee stability. Seven combinations within the three manipulated and six output variables were successful control strategies, and conversion/initiator and total particle number/surfactant control were suggested as experimentally feasible. Constraints on the controller and measurements of outputs closely related to the onset of oscillations were identified as important issues.

Nonlinear Model Based Control

Cardner (1984), Fox et al. (1984), and Tatkar et al. (1985) considered the anionic polymerization of styrene in a simulated CSTR. The controlled variables were the number average molecular weight and the polymer production rate. The control scheme included the following: a) a state variable feedback to reduce the transient response, b) a decoupling first order lag to minimize interaction between controlled variables, and c) a PI controller for regulation against load changes.

Alvarez et al. (1990) attempted to apply a feedforward/feedback (FF/FB) control scheme on the MMA polymerization in a CSTR. Their simulation was based on the simplified model of Chiu et al. (1983). In their control scheme, conversion and temperature set points were regulated by manipulating initiator feed rate and heat removal rate. Upon measuring input disturbances, they were able to adopt the FF/FB control scheme by using single-loop tuning with conventional linear techniques. In a similar fashion, Daoutidis et al. (1990) also tried to apply multivariable FF/FB control schemes to reject step changes of two disturbances (inlet MMA concentration and inlet temperature) so as to control reactor temperature and number average molecular weights.

Alvarez et al. (1993) studied the saturated state-feedback stabilisation for a free-radical polymerization reactor operating around an open-loop unstable point. Their work was on numerical dynamic simulation models. Necessary and sufficient conditions were derived. Generally, this was a control study in non-linear feedback control.

Soroush and Kravaris (1994) applied nonlinear multivariable control to a pilot plant MMA solution polymerization reactor. The choice of residence time and jacket heat input as manipulated variables for controlling conversion and temperature created a singular characteristic matrix. An input/output lineaizing controller was calculated and successfully used to control reactor startup and setpoint tracking.

Mutha (1996) developed a new NL-MPC algorithm for control nonaffine (CNA) systems. The algorithm was tested in simulations, including a semi-batch nitrile rubber (NBR) polymerization reactor. The algorithm was further tested experimentally on an experimental, continuous solution MMA polymerization reactor. When compared to a control affine NLMPC algorithm from literature, the CNA NL-MPC algorithm was observed to have superior performance.

Maner and Doyle III (1997) studied autoregressive-plus Volterra-based MPC control of two simulated polymerization reactors. The nonlinear MPC outperformed both PI and linear MPC both in setpoint tracking and disturbance rejection. The stated advantages to their method were: (a) "plant-friendly" four-level input sequence that is less demanding than a continuously distributed disturbance, (b) computationally less intense nonlinear program to be solved at each sampling interval, and (c) semi-global stability in that global bounded-input bounded-output (BIBO) stability exists for the Volterra plus autoregressive model structure.

CHAPTER 3: PID CONVERSION CONTROL -- CONTROLLER TUNING AND SIMULATION STUDY

3.1 Introduction

This chapter presents the PID tuning method proposed by Wang, Barnes and Cluett (1995a) which will be referred to as the W-C method from now on. The PID tuning technique is based on a frequency domain design method that uses a least-squares fit of the actual to the desired open-loop Nyquist plot. The novel ideas lie in the closed-loop performance specification: the desired response of the control signal and two process frequency response points.

In terms of application of this to a CSTR, the polymer conversion was modelled as a single input single output (SISO) first order plus delay system with initiator feed rate as the manipulated variable and monomer to polymer conversion as the controlled variable.

Simulations show how the W-C tuning method was applied, and how its performance compares to other well known PID tuning methods.

3.2 Literature Survey of PID Controller Design

The classic, single-loop, proportional-integral-derivative (PID) controller is well documented in process control textbooks (Shinksey, 1979; Stephanopoulos, 1984; Seborg et al., 1989; Åström and Wittenmark, 1990; Ogunnaike and Ray, 1994).

Standard performance criteria, selection rules, and tuning methods, are well established, but some recent literature in these areas exists. Tuning is a major part of the literature. Ogunnaike and Ray's (1994) background, with tuning methods categorized by model type used has been adopted in Sec. 3.2.3 through Sec. 3.2.6.

Briefly summarizing the "approximation" models for processes that are either stable (proportional, self-regulating, approaching steady state) or unstable (integrating): 1- A common process model for stable processes is the first order plus dead time/delay model (FO+D) given in Eq. 3.1. 2 - Integrating processes, such as level control and some thermal and electromechanical processes, may be modelled as integrator plus some function, like integrator plus delay (I+D), or integrator plus first order (I+FO), even both I+FO+D. The I+D and I+FO models are given in Eq. 3.2a-b respectively.

$$G(s) = \frac{Ke^{-\theta s}}{\tau s + 1} \tag{3.1}$$

$$G(s) = \frac{1}{s} Ke^{-\theta s} \tag{3.2a}$$

$$G(s) = \frac{1}{s} \frac{K}{\tau s + 1} \tag{3.2b}$$

Here, the Laplace transfer function for the process, $G(s)$, is described by K, the process gain, τ the (dominant) time constant, and θ the dead time (time delay).

Ziegler-Nichols (Z-N), Cohen-Coon (C-C) and internal model control (IMC) based tuning methods are likely the best known.

Table 3.1 summarizes recent work on design and implementation of PID controllers. Many papers attempt to improve or extend on Åström & Hägglund (1984).

Table 3.1: Recent literature on the design and implementation of PID controllers

Reference	System	Work	Notes
Åström & Hägglund (1984)	general		automated Z-N tuning method "auto-tuning"
Åström & Hägglund (1988)			auto-tuning
Åström et al. (1992)	general	T,S	PID controller selection, tuning and evaluation
Barnes et al. (1993)	Laplace	T	frequency domain PID design method
Bobál (1995)	general	T,S	self-tuning Z-N PID
Cluett & Wang (1996)	Laplace	T	PID tuning rules
Cluett & Goberdhansingh (1990)			practical auto-tuning PID controller
Goberdhansingh et al. (1992)			robust controller design from frequency response
Hägglund & Åström (1991)	general	T	industrial adaptive FF and FB controllers; frequency response techniques
Horn et al. (1996)	general	T	filter design for IMC tuned PID systems
Huang et al. (1996)	Laplace	T	extension to auto-tuning to estimate FO+D parameters
Jacob & Chidambaram (1996)	FO+D	T	design for unstable systems
Jerome & Ray (1986)	delayed	S	multivariable control strategies
Loron (1997)	I+FO	T,X	nonsymmetrical optimum method (NSOM)
Luyben (1996)	I+D	T,S	tuning rule proposal
Park et al. (1997)	CSTH	T,S,X	on-line identification with Laguerre series, auto-tuning PID
Puleston & Mantz (1995)	MIMO	T,S	anti-wind-up for PI controller, tuning and dealing with saturation
Rivera & Gaikwad (1996)	FO+D I+D	T,S	digital PID design, uses Matlab ARX estimation
Tan et al. (1996a)			design of Smith Predictors
Tan et al. (1996b)	general	T,S,X	auto tuning procedure for PI/PID
Tham & Parr (1994)	general	T	filtering and data reconstruction
Tuffs & Clarke (1985)	general	T	eliminating offset in self-tuning control
Voda & Landau (1995a)	integrating		PID auto-calibration by symmetrical optimum method
Voda & Landau (1995b)	integrating	T,X	PI auto-calibration by 2 frequency measurements
Wang et al. (1995a)	general	T,S	frequency domain PID design method
Wang et al. (1997b)	general	T,S	frequency domain PID design method

3.2.1 Performance Measures

Performance criteria include (Ogunnaike and Ray, 1994):

1. Stability Criteria - closed loop process must be stable
2. Steady State Criteria - process must have close to no offset (zero error) at steady state
3. Dynamic Response Criteria

Although the first two criteria are fundamental, quantitatively evaluating the third one is not absolute. One way to evaluate dynamic controller performance is through some direct measures of maximum or minimum of the process output response curve:

- overshoot
- settling time
- rise time/slew rate
- decay ratio

Often an integrated function of the feedback error from desired setpoint can be used as well. Common error criteria are integrals of the absolute value of the error (IAE), squared error (ISE), time-weighted absolute error (ITAE), and time-weighted squared error (ITSE), given in Eq. 3.3a-d respectively.

$$\text{IAE} = \int_0^\infty |e(t)|\, dt \tag{3.3a}$$

$$\text{ISE} = \int_0^\infty e^2(t) dt \tag{3.3b}$$

$$\text{ITAE} = \int_0^\infty t|e(t)|\, dt \tag{3.3c}$$

$$\text{ITSE} = \int_0^\infty te^2(t) dt \tag{3.3d}$$

Alternatively, the *controller* output instead of the process output can be used in the above equations.

Chien (1992) evaluated controllers by summation of the input and output variance over equal time intervals. This corresponds to applying ISE to both the controller output, $u(t)$, (=process input) and the process output, $y(t)$ and adding them.

3.2.2 Controller Selection

Åström, Hang, Persson and Ho (1992) made recommendations on controller selection for stable and integrating processes (approximate models) with monotone step response. They suggest four dimensionless quantities, easily determined, that can be used to assess the achievable performance as well as selecting the correct form of PI or PID controllers. This also indicates if a more sophisticated control law would be suitable. The four values were normalised deadtime (divided by the dominant time constant), normalised process gain (multiply process gain and ultimate gain), process load disturbance error and normalised closed-loop rise time (divided by the dominant time constant).

Tan, Lee and Wang (1996b) applied normalised dead time as part of their rule-based automatic selection of PI or PID control in an auto-tuned PID controller.

3.2.3 Tuning with Fundamental Models

Given any process, the performance criteria given in Sec. 3.2.1 can be minimized as an optimisation problem and let the solution be the PID controller tuning parameters.

Analytical methods can be applied to low-order models with few parameters. This includes direct synthesis, pole placement and internal model control (IMC) design methods.

The Z-N continuous cycling (loop tuning, ultimate gain, or quarter decay) method applied to a process simulation is also tuning with fundamental models.

3.2.4 Tuning with Approximate Models

Åström et al. (1992) have studied these models (FO+D and I+FO+D) and made recommendations on selection and tuning methods. They recommended against using Z-N for a FO+D when the ratio of delay to time constant θ/τ is greater than 1. The CSTR process has a 0.7 ratio.

Z-N and C-C quarter decay methods, time integral tuning rules, direct synthesis and IMC tuning rules have all been applied to first order plus dead time models. Ogunnaike and Ray (1994) suggest ITAE and IMC/Direct synthesis settings as the best choice for the approximate model approach.

Voda and Landau (1995a) applied a "symmetrical optimum method" to auto-calibrate a PID controller on an integrating plant process using one frequency measurement. This was followed by auto-calibration of a PI controller using two frequency measurements (Voda and Landau, 1995b) to extend validity limits.

Loron (1997) presented the "non-symmetric optimum method" for tuning PID controllers on I+FO processes, as an extension to the work by Voda and Landau.

3.2.5 Tuning with Frequency Response Models

The majority of the industrial regulators are tuned using frequency response methods because modelling errors and application specifications can be expressed directly in the frequency domain (Tang and Ortega, 1993; Loron, 1997).

Tan et al. (1996b) enhanced auto-tuning by estimating the process frequency response at two specified phase lags on the Nyquist curve. They estimate the second point by placing a delay element on the relay controller. They then apply rules based on dynamics and normalised dead time to select PI or PID control. Simulation and experimental results show that this enhanced method has better performance than the standard auto-tuned PID.

Barnes, Wang and Cluett (1993) proposed an interesting new algorithm for designing PID controllers in the frequency domain. It was based on an optimisation problem using the frequency domain process response. The principle was to minimize the sum of squared errors between the actual and desired Nyquist plots of the open loop process. The problem was solved analytically. A more general non-linear control structure would require numerical solution.

Since the algorithm was solved on the open-loop Nyquist plot, the stability of the solution could be evaluated using the Nyquist stability criteria. Assessing robust stability was possible.

Wang, Barnes and Cluett (1995) applied this algorithm to formulate a novel PID tuning method. This method required minimal information about the process to be controlled. Specifically,

1. self-regulating (approaches steady state) or integrating process
2. frequency response at two frequencies related to the closed loop settling time of the process
3. overall process gain -- amount of change in the output with a given step in the input

The two advantages of this method were:

1. use of frequency domain data in the form of the process Nyquist plot at a limited number of points, required less structural information about the process
2. performance specification in time domain

The novelty in the new method was this closed loop performance specification of the desired controller output signal in time domain. A smooth control signal and process output is produced.

The key performance specification variable was α in Eq. 3.4.

$$u(t) = \frac{r}{K}\left[\alpha + (1-\alpha)(1-e^{-\frac{\alpha \cdot t}{T}})\right] \tag{3.4}$$

where $u(t)$ is the controller output with respect to time, t. The ratio r/K of setpoint change, r, to overall process gain, K gives the final required controller output for the new setpoint. The expression in the square brackets applies the smooth shape to the control signal and α is the performance parameter -- larger values making this controller more aggressive (typically in the range of 0.5 to 1.5). T is the process time constant.

Cluett and Wang (1996) also presented a simplified set of tuning rules using normalised parameters and PID tuning constants. The key measure of the difficulty in controlling a process was given as the ratio $L = T/d$, where T was the process time constant and d was the delay or dead time in the process. Small L with a long delay (large d) or fast process (small T) would indicate a process that is difficult to control.

PID controller tuning constants, K_c, τ_I and τ_D were normalised by multiplying the controller gain by the process gain, and dividing the controller time constants by the process dead time. Tables of normalized tuning constants for first order plus dead time and integrating processes, at various level of performance, were presented.

Wang, Hang and Bi (1997b) proposed a frequency domain controller design method that minimizes the closed loop response between the actual and desired process frequency response. They admit that their method is similar to Barnes et al. (1993), however they point to the lack of information on how to set the desired loop frequency responses, how to deal with unstable processes and how to obtain the multiple frequency response points. They propose using relay feedback auto-tuning data to generate an FFT-based process frequency response.

3.2.6 Tuning without Models

The Z-N continuous cycling method applied directly to the process by using a proportional controller falls into this category. However, applying this method is difficult in practice since oscillating (i.e. marginal stability) behaviour is required – not always applicable for the CSTR process.

Åström and Hägglund (1984) presented "auto-tuning" of PID controllers. Their technique relies on a relay controller to add small amplitude perturbation to the process instead approaching instability. The ultimate gain and ultimate period are used to tune the PID controller

Bobál (1995) described a self-tuning digital PID controller with an adaptive second order regression model. It used Z-N tuning rules but instead of oscillating the process output like Åström and Hägglund (1984), it specified placing the closed loop control response on the stability boundary using a proportional controller. Key parameters were critical gain and critical period of oscillation. Simulations showed that it could control proportional systems with or without minimum phase, and integrating systems.

Huang, Chen, Lai and Wang (1996) refined auto-tuning to better estimate FO+D models so that approximate model tuning methods could be applied to relay feedback results. They proposed that the magnitude of the perturbations could be made asymmetrical, that the relay could be automatically shut off once constant oscillations were observed, and that a test stoppage criterion could be applied. These changes allowed for model-based controllers more robust to error yet with better performance.

Park, Sung and Lee (1997) proposed using an under-damped, step or pulse change in setpoint with a normal P, PI or PID controller to perturb the process, and a Laguerre series to estimate a second order plus time delay model from the output. They use ITAE tuning based on this model to calculate the parameters for the PID controller. Simulation and experimental results are reported to be effective.

3.3 Theoretical Background

3.3.1 Process Description

The process is a solution, homopolymerization of methyl methacrylate (MMA) monomer in toluene solvent. The reaction is initiated by 2,2'-azobisisobutyronitrile (AIBN), a thermally decomposing free radical source. The continuously stirred tank reactor (CSTR) operates isothermally with a constant feed of monomer, solvent and initiator. A constant reactor volume is maintained by product overflow out of the vessel.

A PID controller was implemented to control percent conversion of MMA to poly-methyl methacrylate (PMMA) polymer by manipulating the AIBN initiator flow rate. Increasing the initiator flow rate into the reactor increases conversion.

A first order plus dead-time, discrete time, transfer function model for the CSTR in terms of initiator flow rate to percent conversion was previously identified by Chien (1992) as Eq. 3.5.

$$Y_t = \frac{0.116z^{-25}}{1-0.972z^{-1}}U_t \tag{3.5}$$

where U_t is the AIBN pump flow rate (ml/min) and Y_t is weight percent conversion of monomer to polymer in the reactor. Both variables are in deviation from steady state form. The time interval was one minute.

In order to implement PID controller tuning methods, this z-transform was converted to a continuous time Laplace transfer function, Eq. 3.6.

$$G_p(s) = \frac{-4.143e^{-25s}}{35.7s+1} \tag{3.6}$$

This transfer function has an overall process gain of -4.143, a dead time of 25 minutes, and a process time constant of 35.7 minutes. The process is stable, however, it has a long delay and large time constant -- indicating a relatively slow process.

3.3.2 Wang and Cluett (W-C) Tuning Method

The general first order plus time delay process transfer function (Eq. 3.1), for frequency response is given in Eq. 3.7.

$$G(j\omega) = \frac{Ke^{-dj\omega}}{\tau j\omega + 1} \tag{3.7}$$

If the desired closed loop transfer function is given as Eq. 3.8,

$$G'_{CL}(j\omega) = \frac{e^{-dj\omega}}{\tau_{CL} j\omega + 1} \tag{3.8}$$

then the real and imaginary parts of the process frequency response are Eq. 3.9a-b respectively.

$$\mathrm{Re}(Y) = \frac{1}{KQ}\left(\tau_{CL} + \frac{\sin(d\omega)}{\omega} - \tau(1-\cos(d\omega))\right) \tag{3.9a}$$

$$\mathrm{Im}(Y) = \frac{1}{KQ}\left(\tau\tau_{CL}\omega + \tau\sin(d\omega) + \frac{1-\cos(d\omega)}{\omega}\right) \tag{3.9b}$$

where

$$Q = \tau_{CL}^2 + 2\tau_{CL}\frac{\sin(d\omega)}{\omega} + 2\frac{1-\cos(d\omega)}{\omega^2} \tag{3.10}$$

The closed loop time constant is determined from the choice of α using Eq. 3.11.

$$\tau_{CL} = \tau / \alpha \tag{3.11}$$

The real and imaginary parts of the process frequency response, Y, are required at two frequencies, ω_1 and ω_2, and are calculated using Eq. 3.12a-b.

$$\omega_1 = 2\pi / T_s \tag{3.12a}$$

$$\omega_2 = 4\pi / T_s \tag{3.12b}$$

The closed loop settling time, T_s, was estimated as five process time constants and the delay, Eq. 3.13.

$$T_s = 5\tau + d \tag{3.13}$$

Actual tuning constants are given in terms of three intermediate variables (Eq. 3.14a-c):

$$c_2 = -\frac{Y_R(\omega_1) - Y_R(\omega_2)}{\omega_1^2 - \omega_2^2} \tag{3.14a}$$

$$c_1 = \frac{Y_I(\omega_1)}{\omega_1} \tag{3.14b}$$

$$c_0 = Y_R(\omega_1) + c_2\omega_1^2 \tag{3.14c}$$

So the final PID tuning constants are given by Eq. 3.15a-c.

$$K_c = c_1 \tag{3.15a}$$

$$\tau_I = c_1 / c_0 \tag{3.15b}$$

$$\tau_D = c_2 / c_1 \tag{3.15c}$$

Wang et al. (1995a) used α values of 0.5, 1.0 and 1.5 in their example of tuning a PID controller for a FO+D process using their method as compared to IMC, Z-N and another method. Tuning of the PID controller with α values in the range of 0.5 to 1.5, as well as more aggressive values yields tuning parameters as given in Table 3.2.

Table 3.2: Tuning constants for PID controller tuned using W-C rules

Parameter	W-C $\alpha = 0.5$	W-C $\alpha = 1.0$	W-C $\alpha = 1.5$	W-C $\alpha = 2.0$	W-C $\alpha = 5.0$	W-C $\alpha = 10.0$
K_c	-0.099	-0.165	-0.211	-0.245	-0.343	-0.395
τ_I	38.7	40.6	41.8	42.6	44.9	46.0
τ_D	2.49	4.13	5.24	6.0	8.1	9.1

3.3.3 Controller Tuning

For comparison, the conversion controller will also be tuned using Z-N, C-C, ITAE minimized, and IMC based tuning methods on the same first order plus dead time process model. Numerical results are given in Table 3.3.

Ziegler-Nichols (Z-N) Tuning

Z-N approximate model tuning rules for PID controllers are given in Eq. 3.16a-c.

$$K_c = \frac{1.2}{K}\frac{\tau}{\theta} \tag{3.16a}$$

$$\tau_I = 2.0\theta \tag{3.16b}$$

$$\tau_D = 0.5\theta \tag{3.16c}$$

Cohen-Coon (C-C) Tuning

C-C tuning rules are given in Eq. 3.17a-c.

$$K_c = \frac{1}{K}\frac{\tau}{\theta}\left[\frac{16\tau + 3\theta}{12\tau}\right] \tag{3.17a}$$

$$\tau_I = \frac{\theta[32 + 6(\theta/\tau)]}{13 + 8(\theta/\tau)} \tag{3.17b}$$

$$\tau_D = \frac{4\theta}{11 + 2(\theta/\tau)} \tag{3.17c}$$

ITAE Tuning

Minimum ITAE tuning rules for PID controllers using approximate models are optimised for setpoint tracking and disturbance rejection, given by Eq. 3.18a-c and Eq. 3.19a-c, respectively.

$$K_c = \frac{0.956}{K}\left(\frac{\tau}{\theta}\right)^{0.855} \tag{3.18a}$$

$$\tau_I = \frac{\tau}{\left[0.796 - 0.147\left(\frac{\theta}{\tau}\right)\right]} \tag{3.18b}$$

$$\tau_D = 0.308\tau\left(\frac{\theta}{\tau}\right)^{0.929} \tag{3.18c}$$

$$K_c = \frac{1.357}{K}\left(\frac{\tau}{\theta}\right)^{0.947} \tag{3.19a}$$

$$\tau_I = \frac{\tau}{0.842}\left(\frac{\theta}{\tau}\right)^{0.738} \tag{3.19b}$$

$$\tau_D = 0.381\tau\left(\frac{\theta}{\tau}\right)^{0.995} \tag{3.19c}$$

IMC Tuning

Tuning rules based on IMC are given by Eq. 3.20a-c.

$$K_c = \frac{2\tau + \theta}{2K(\lambda + \theta)} \tag{3.20a}$$

$$\tau_I = \tau + \frac{\theta}{2} \tag{3.20b}$$

$$\tau_D = \frac{\tau\theta}{2\tau + \theta} \tag{3.20c}$$

The filter parameter, λ, is the time constant for the desired closed loop first order response. It must satisfy $\lambda > 0.2\tau$ and the recommended choice is given by Eq. 3.21.

$$\frac{\lambda}{\theta} > 0.25 \tag{3.21}$$

Table 3.3: Tuning constants for PID controller tuned by various tuning rules

Parameter	Z-N	C-C	ITAE setpoint	ITAE disturbance	IMC $\lambda/\theta = 0.25$	IMC $\lambda/\theta = 0.9$
K_c	-0.4136	-0.5199	-0.3129	0.4590	-0.3723	-0.2448
τ_I	50.	48.66	51.51	32.59	48.2	48.2
τ_D	12.5	6.048	0.2211	9.538	9.2583	9.2583

3.3.4 PID Simulation

The classical PID controller is described by Eq. 3.22, where the controller output, $u(t)$, is calculated as a function of the error, $e(t)$. The error is defined as the difference between the desired process output, $r(t)$, and the measured output, $y(t)$, in Eq. 3.23.

$$u(t) = K_c\left[e(t) + \frac{1}{\tau_I}\int^t e(s)ds + \tau_D\frac{de(t)}{dt}\right] \tag{3.22}$$

$$e(t) = r(t) - y(t) \tag{3.23}$$

Velocity Form

A simple digital implementation using the velocity (or deviation, or incremental) form of this equation is discretized using finite differences as in Eq. 3.24. The subscript, k, indicates the sample number in time, and Δt is the sampling interval.

$$\Delta u_k = K_c\left[\left(e_k - e_{k-1}\right) + \frac{\Delta t}{\tau_I}e_k + \frac{\tau_D}{\Delta t}\left(e_k - 2e_{k-1} + e_{k-2}\right)\right] \tag{3.24}$$

Using the velocity form avoids problems related to the integrator term:

- integrator windup
- bumps for parameter changes when offset is zero
- bumps switching from manual to automatic with zero offset

Åström and Wittenmark (1990) point out that a good PID controller is not a simple digital implementation of the "textbook" analog PID controller given above. They explain that the derivative action should be limited to avoid amplifying measurement noise, and separated for setpoint and measurement effects on $e(t)$.

Integral or reset windup can occur to a PID controller in position form. For the velocity form, a saturated signal can cause problems when the proportional and derivative terms negate the integral term as the controller output leaves the saturated region back to normal operating regions.

When a large control signal output saturates the actuator action, integral or reset windup results. The error signal will continue to accumulate.

Ogunnaike and Ray (1994) presented a simple scheme to prevent reset windup using feedback from the actuator to the controller. The difference between the actuator input (requested by the controller) and

output (achieved by the actuator) is fed back to the integrator at a rate determined by a tracking time constant.

Integral Action

For incremental improvement, the integral term of the digital controller can be calculated using a trapezoidal instead of a rectangular approximation as in Eq. 3.25.

$$\int_0^{n\Delta t} e(t)dt \approx \sum_{k=1}^{n} \frac{\left(e_k + e_{k-1}\right)}{2}\Delta t \tag{3.25}$$

Åström and Wittenmark (1990) relate how using 24 bits of precision in internal computation is frequently used to avoid integration offset due to quantization error.

Derivative Action

Derivative kick is the large change in the controller output that results when a step change is made in the setpoint. This be can be avoided by applying derivative action only to the measurement error using Eq. 3.26, or by ramping any setpoint changes over time.

$$\frac{de(t)}{dt} \approx \frac{e_k - e_{k-1}}{\Delta t} \rightarrow \frac{y_k - y_{k-1}}{\Delta t} \tag{3.26}$$

Sampling Interval

Åström and Wittenmark (1990) give a rule of thumb for choosing sampling frequency based on continuous time arguments. If the phase margin can be decreased by 5° to 15°, then, Eq. 3.27 can be used to choose the sampling frequency, h, based on the crossover frequency, ω_c, in radians per second.

$$h\omega_c \approx 0.15 - 0.5 \tag{3.27}$$

Filtering

A first order or low-pass filter (also exponential or alpha) can be implemented in digital form as Eq. 3.28, where y_k is the k th filtered measurement and r_k is the k th raw reading. α is the filtering factor between 0 and 1, where 0 applies no filtering and 1 uses only historical values.

$$y_k = \alpha \cdot y_{k-1} + (1-\alpha) \cdot r_k \tag{3.28}$$

Using a first order filter delays dynamics and will tend to make the control system sluggish.

Tham and Parr (1994) outlined a simple procedure based on statistical theory to validate and reconstruct data using a combination of low pass filters, spike filters and heuristics. This filters out noise and spikes without masking the dynamics or introducing significant delays characteristic of simple first order filters.

3.4 Simulation Results

3.4.1 Open Loop Results

Figure 3.1 shows the open loop process response to a step change in the process input. The control variable is conversion, $y(t)$ in percent, in the top plot. The manipulated AIBN flow rate, $u(t)$ in ml/min, is on the bottom plot. Both variables are given in deviation form from initial steady state values.

At time 50 minutes, the AIBN flow rate was increased from the initial steady-state value by 2 ml/min. The conversion rose by about 8%. Although not shown on the plot, when the AIBN flow rate was decreased, the conversion decreased back to the original value. The time required to complete 95% of the step in the process was about 132 minutes, including 25 minutes of delay.

At time 300 minutes, a large, unmeasured, first order disturbance in the conversion was introduced for 20 minutes, after which, the process was allowed to return to steady state. The conversion dropped about 6 % and then recovered back to the higher level.

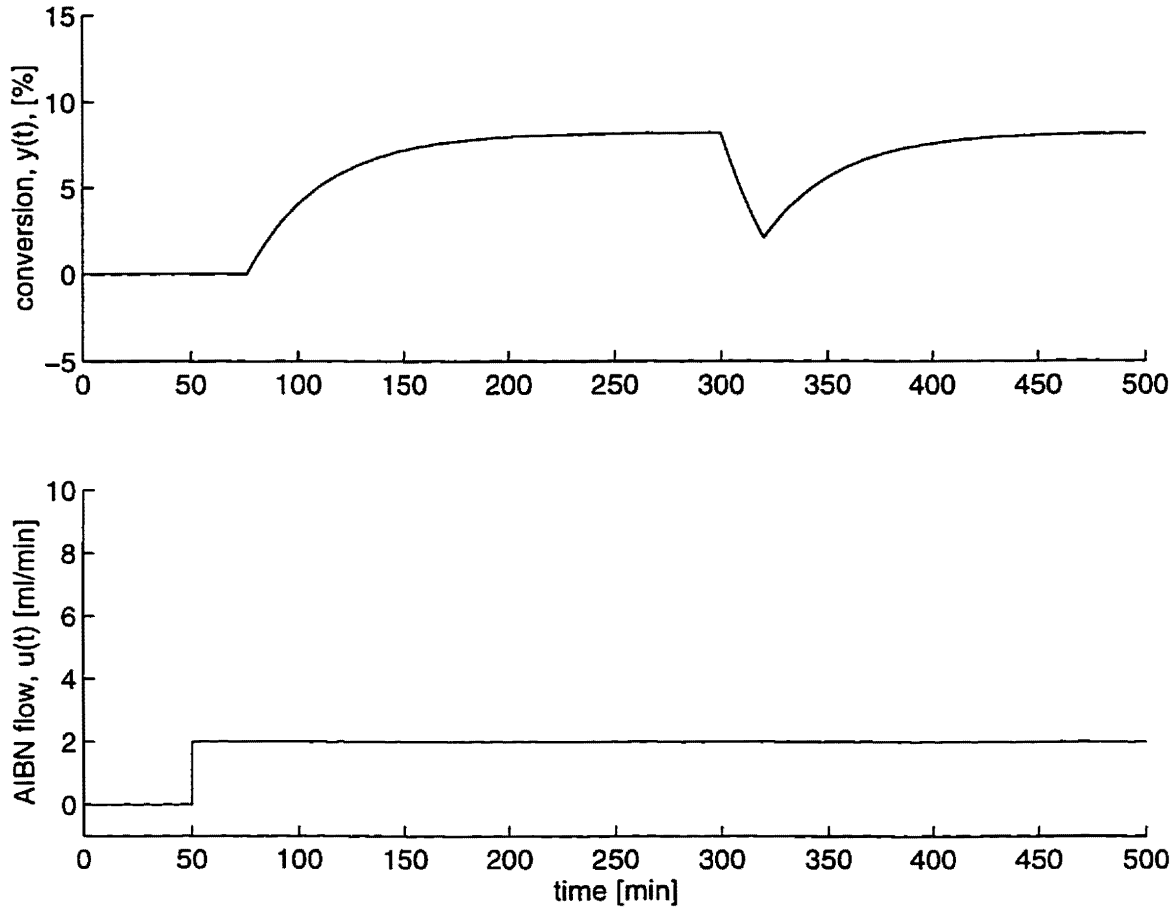


Figure 3.1: Simulated open loop process response to a 2 ml/min increase in AIBN flow rate

3.4.2 Closed Loop Results

Figures 3.2 through 3.4 show representative plots of the simulated, closed loop process response with the controller tuned with W-C tuning rules at $\alpha = 0.5$, 1.5 and 10.0. Figures 3.5 and 3.6 show the same loop with Z-N and C-C tuning rules for approximate first order and delay models. Figures 3.7 and 3.8 show simulated closed loop response for the controller tuned with ITAE minimized for setpoint change and disturbance rejection. Figures 3.9 and 3.10 show the closed loop response with the controller tuned by IMC tuning rules with λ/θ values of 0.25 and 0.9. The top plots show the percent conversion, $y(t)$, and the bottom plots show the manipulated AIBN flow rate, $u(t)$ in ml/min. Both conversion and AIBN flow rate are in deviation form, so that the initial steady state is reference zero.

All the closed loop simulations were started in closed loop control with the PID controller tuned according to the selected method. The process was started at steady state on the first conversion setpoint. At time 50 minutes, the setpoint was increased from the steady-state value by 10 %. At time 300 minutes, a large, unmeasured, first order disturbance was introduced into the conversion for 20 minutes, after which, the process was allowed to return to its previous state.

All the controllers were stable and able to change the conversion to the new setpoint, as well as recover from the disturbances.

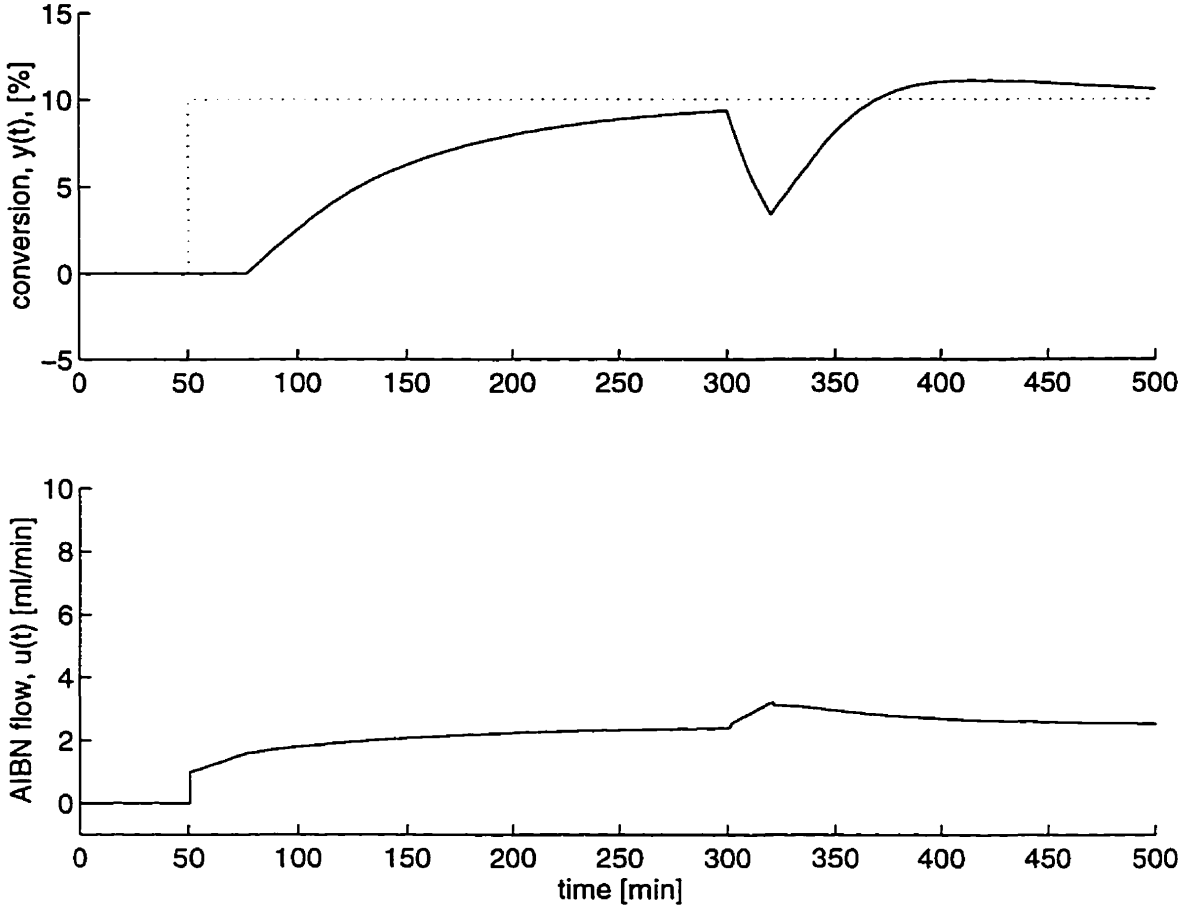


Figure 3.2: Simulated closed loop process response with W-C tuned (α = 0.5) controller

(— simulation, ··· setpoint)

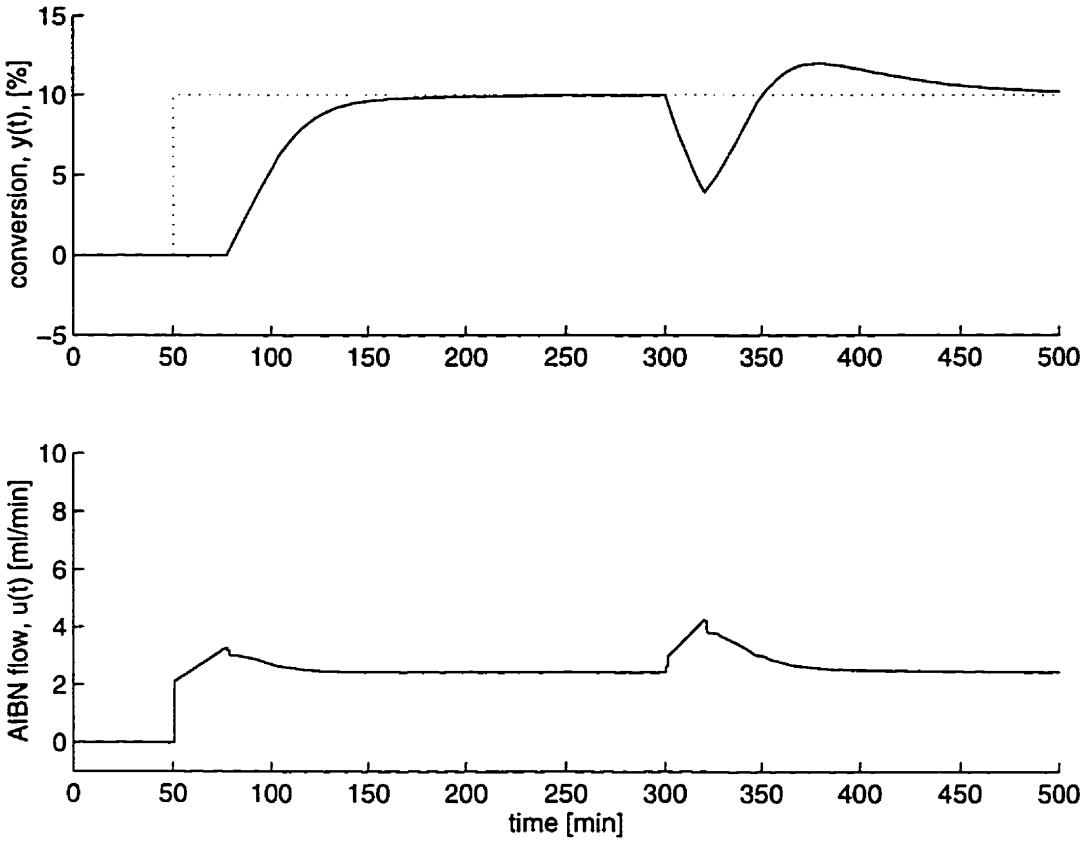


Figure 3.3: Simulated closed loop process response with W-C tuned (α = 1.5) controller

(— simulation, ··· setpoint)

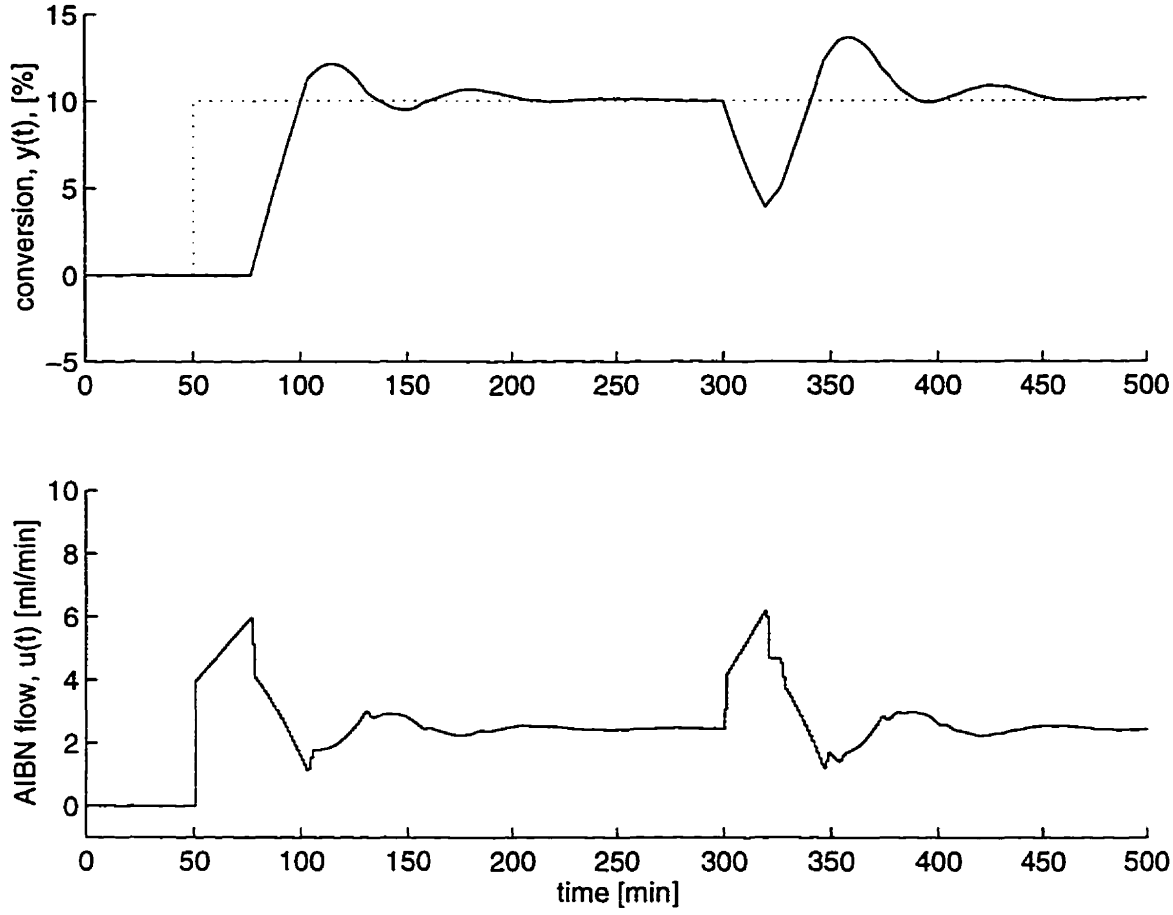


Figure 3.4: Simulated closed loop process response with W-C tuned (α = 10.0) controller

(— simulation, ··· setpoint)

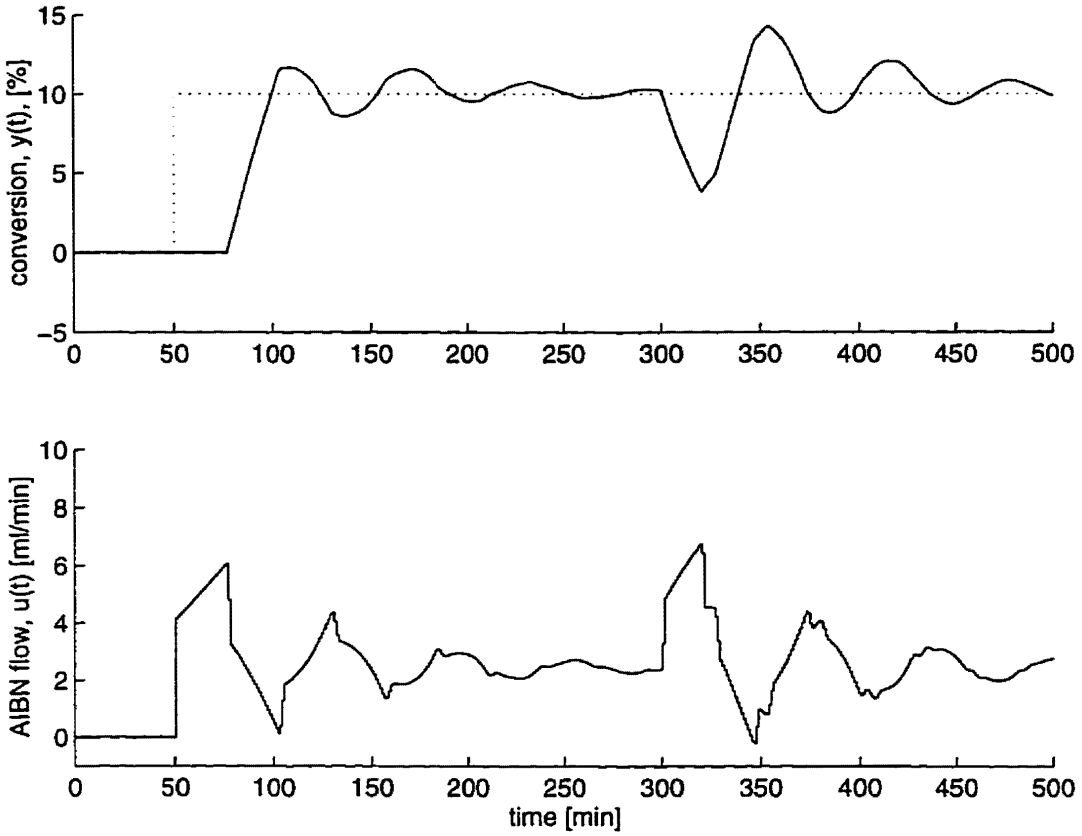


Figure 3.5: Simulated closed loop process response with Z-N tuned controller

(— simulation, ··· setpoint)

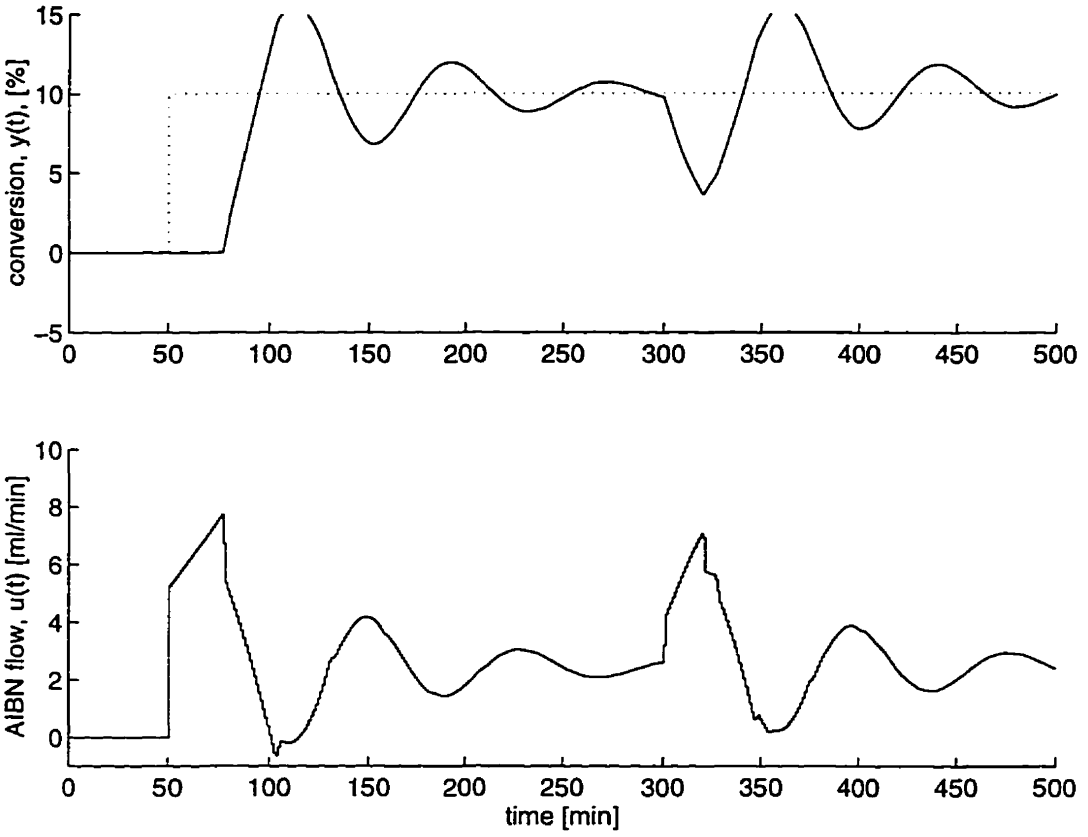


Figure 3.6: Simulated closed loop process response with C-C tuned controller

(— simulation, ··· setpoint)

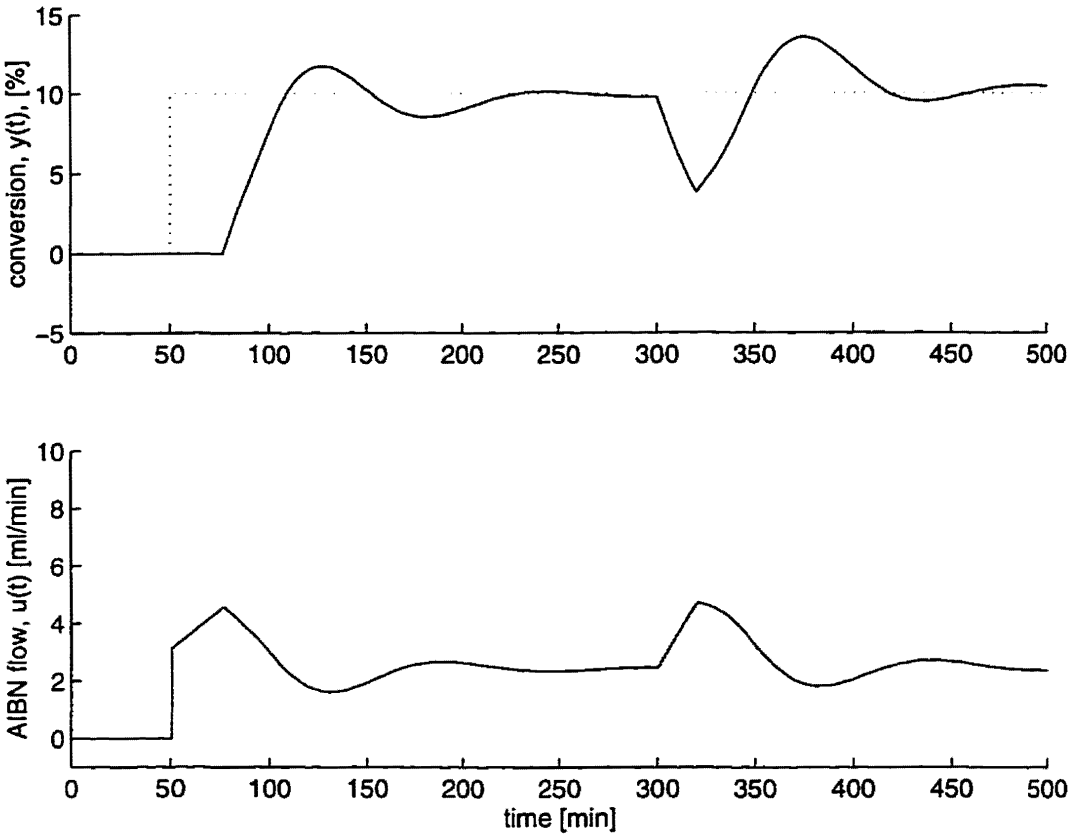


Figure 3.7: Simulated closed loop process response with ITAE tuned (setpoint change) controller

(— simulation, ··· setpoint)

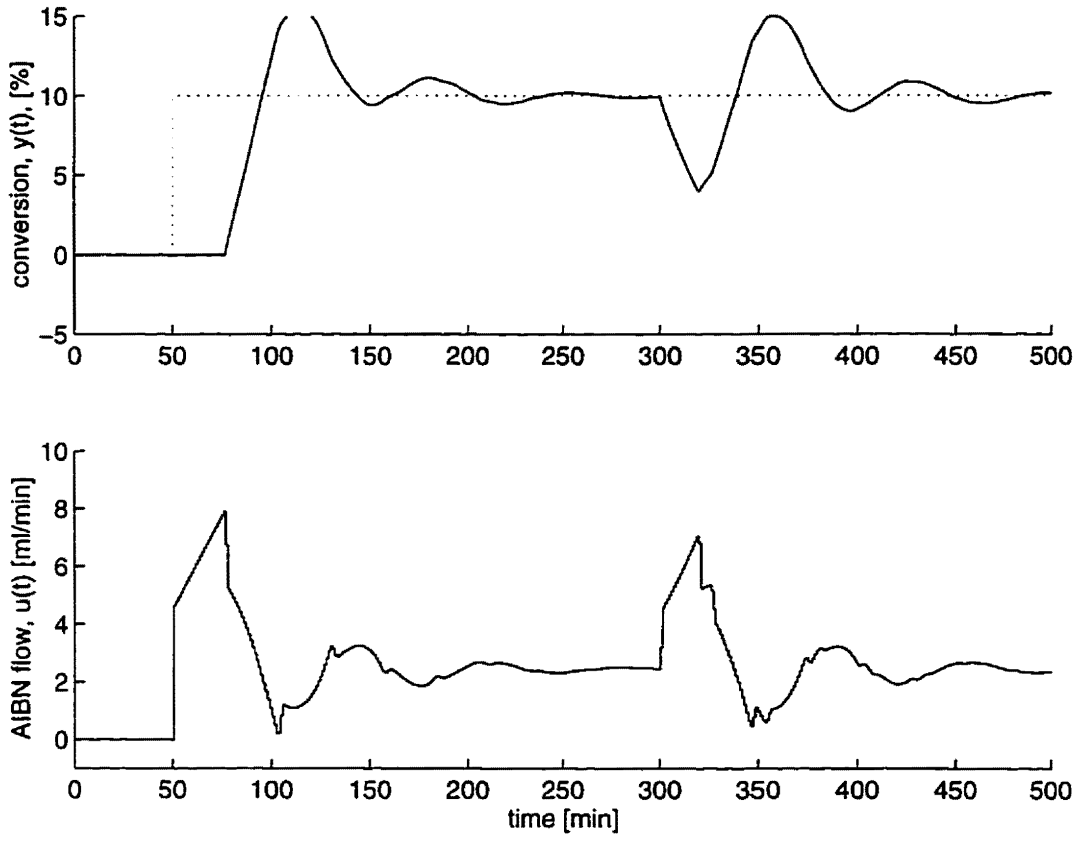


Figure 3.8: Simulated closed loop process response with ITAE tuned (disturbance rejection) controller

(— simulation, ··· setpoint)

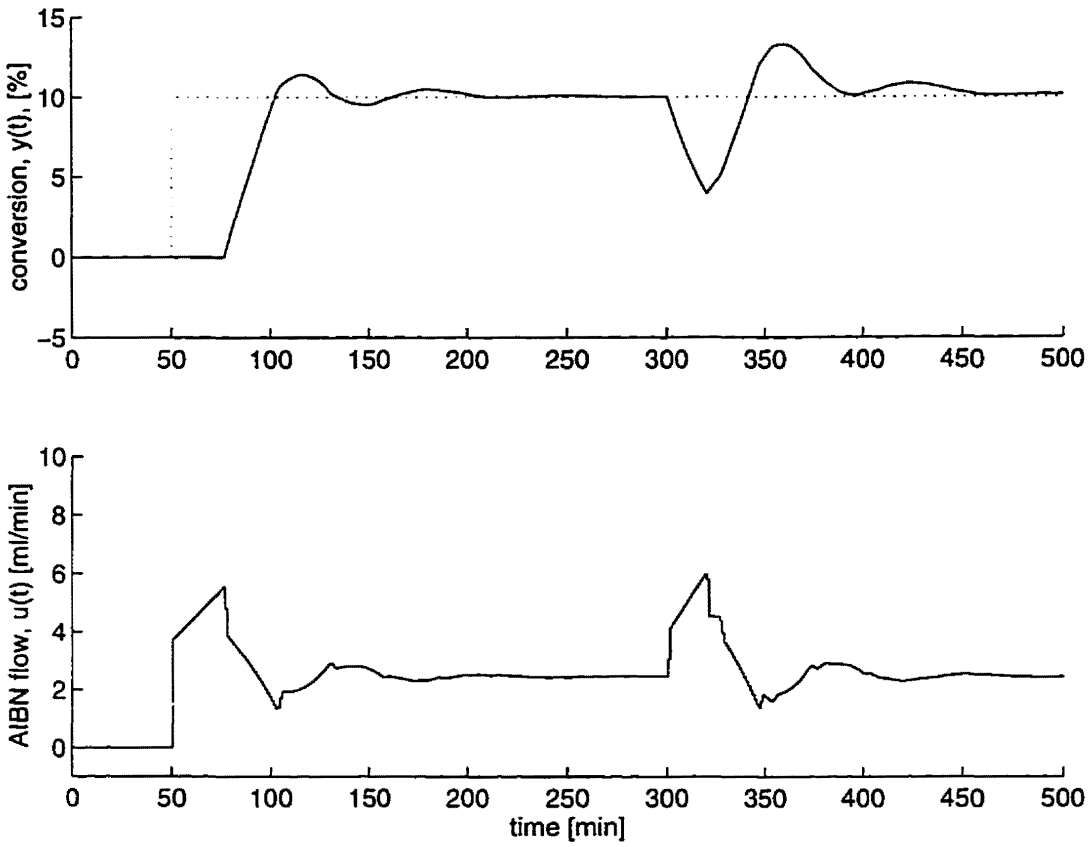


Figure 3.9: Simulated closed loop process response with IMC ($\lambda/\theta = 0.25$) tuned controller

(— simulation, ··· setpoint)

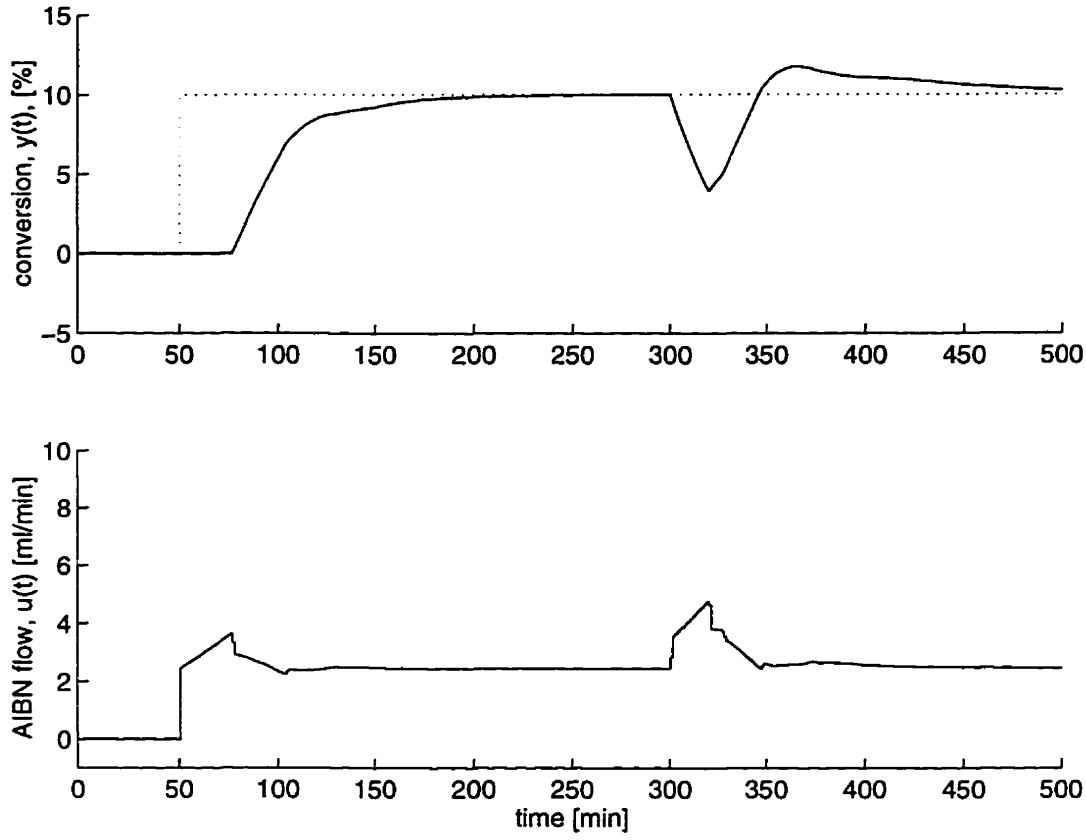


Figure 3.10: Simulated closed loop process response with IMC tuned ($\lambda/\theta = 0.9$) controller

(— simulation, ··· setpoint)

3.4.3 Evaluation

All the controllers were closed loop stable, satisfying the stability criteria.

Although W-C (α=0.5) tuned response in Figure 3.2 did not reach steady state in 250 minutes, there is clear indication that it is approaching the setpoint and further simulations showed that it achieved the 9.9% of the setpoint change after 465 minutes. Therefore, all the controllers satisfied the steady state criteria.

Integral error performance measures given in Eq. 3.3a-d were calculated for all the controllers. Table 3.4 summarizes numerical results of IAE, ISE, ITAE and ITSE for the closed loop response with the different controller tuning methods. The four functions are subdivided into whether the test was a setpoint step change or a load disturbance

Table 3.4: Time-integral performance measures of the simulated closed loop responses to setpoint change and load disturbances

Controller	IAE		ISE		ITAE (thousands)		ITSE (thousands)	
	setpoint	distur-bance	setpoint	distur-bance	setpoint	distur-bance	setpoint	distur-bance
W-C α = 0.5	995	329	6479	1061	703	208	2815	367
W-C α = 1.0	659	324	4698	920	294	215	1276	366
W-C α = 1.5	534	314	4160	899	173	196	941	348
W-C α = 2.0	470	307	3917	891	121	182	815	335
W-C α = 5.0	446	287	3569	892	128	150	673	313
W-C α = 10.0	462	278	3515	928	150	139	673	331
Z-N	503	328	3519	1039	221	203	722	437
C-C	671	423	4171	1523	401	282	1310	752
ITAE - setpoint	539	344	3783	1151	236	201	844	494
ITAE - disturbance	567	325	3978	1177	245	175	1025	486
IMC λ/θ = 0.25	445	278	3511	884	133	143	653	306
IMC λ/θ = 0.9	531	289	4007	806	180	176	879	277

3.5 Discussion of Simulation Results

Figure 3.11 compares the various controllers by their ISE after setpoint changes and load disturbances. Since the ISE does not weigh errors over time, if a controller takes a long time to reach steady state at a new setpoint, it would not be so heavily penalized. However, since error is squared, large deviations from setpoint are penalized. This would indicate which controllers are better at minimizing offset.

IMC appears to be the best controller for setpoint changes and disturbance rejection by ISE performance measures, with the W-C (1.5) and W-C (2.0) being very close.

A clear trend can be seen as the α parameter was increased from 0.5 to 5.0 for the W-C tuned controller -- it performed better, and ISE decreased. The improvement in performance using more aggressive control (higher α) was more pronounced for changes in setpoint than for disturbance rejection (about half as much, percentage-wise as for setpoint changes). As α increased to 10.0, ISE for disturbance rejections increased. This was visible in Figure 3.4 with agressive tuning, the controller output overshot the setpoint after the disturbance, and oscillated before returning to the desired setpoint.

Z-N tuned controllers and those tuned to minimize ITAE performed poorer than W-C controllers. Their aggressive, oscillatory process response was penalized by the squared error in ISE. Z-N and ITAE minimized for setpoint changes both performed better than W-C for setpoint changes for the same reasons.

C-C tuned control did not perform well.

Comparison of the controllers by ITAE is shown in Figure 3.12. Since ITAE penalizes more than ISE for slow approaches to setpoint, slow reacting and oscillatory controllers performed poorly with this test.

IMC was again the best controller by ITAE criteria with W-C being close.

For the W-C tuned controller, increasing values of α from 0.5 through 10.0 improved performance. The trend was very clear. IMC with a time filter constant equal to the process time constant ($\lambda/\theta = \tau/\theta$ so ~1.428) having performance somewhere between W-C with α between 1.0 and 1.5 is evident and in agreement with comments by Wang et al. (1995a).

Z-N tuning appeared to have performance somewhere between W-C with α of 1.0 and 1.5. However, comparison of the simulation plots for α=1.0 and α=1.5 (Figure 3.3) shows that Z-N has faster response but a larger overshoot that does poorly when the error is squared in ISE (Figure 3.11).

ITAE tuned controllers appeared to perform better than W-C methods in disturbance rejection, especially the one tuned for such. However, they suffered from oscillatory response and fared poorly in the ISE test.

C-C tuned control did not perform well by ITAE measures either.

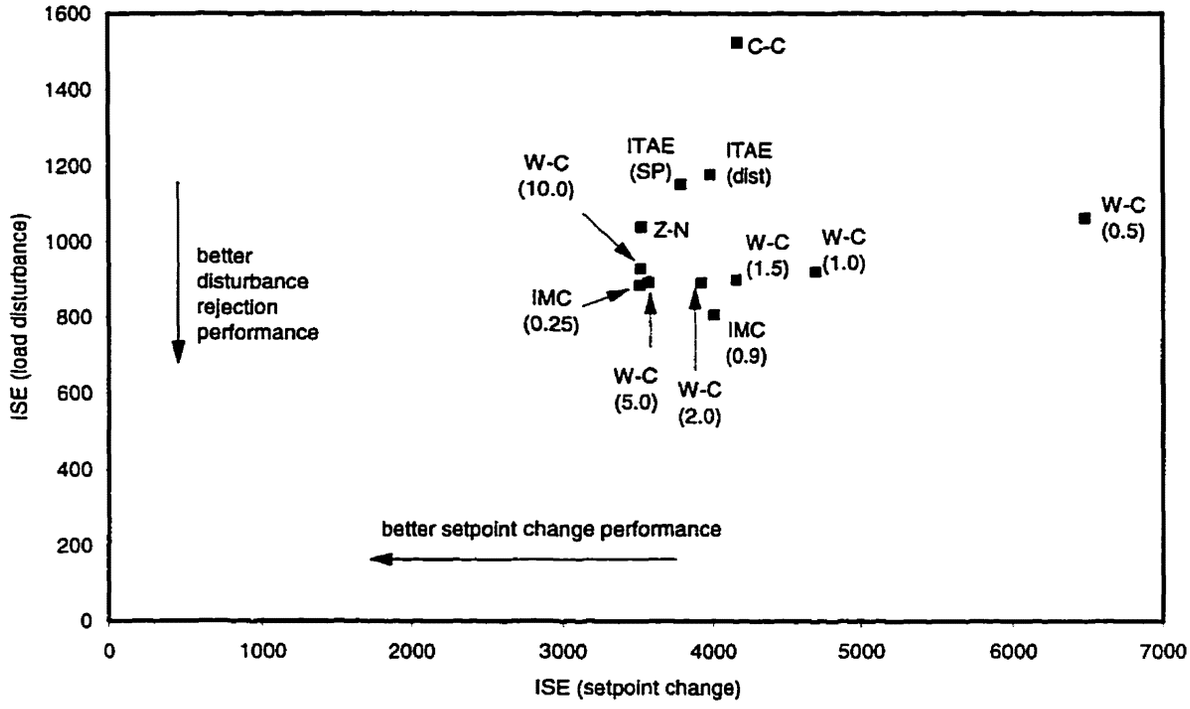


Figure 3.11: Comparison of closed loop controller performance by ISE

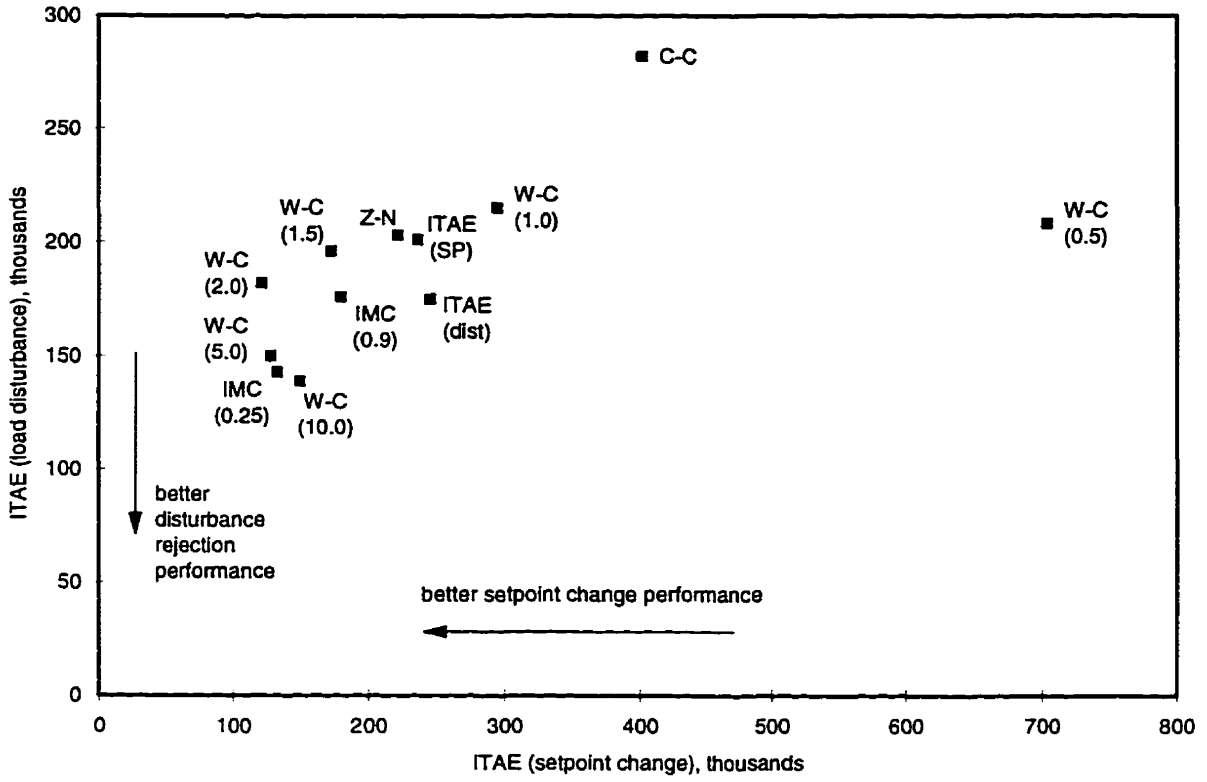


Figure 3.12: Comparison of closed loop controller performance by ITAE

3.6 Concluding Remarks

IMC appears to be the best type of controller tuning method for tuning the first order plus dead time model of the CSTR process when compared with ISE or ITAE performance criteria.

Using ISE and ITAE performance criteria was a good compromise for evaluating controllers. ISE weighter the comparison against controllers with large deviations from setpoint, such as with overshoot. ITAE weighted the comparison against controllers with long term offset. The results could be compared in a quick, visual, plot using the two measures as axes.

IMC could be expected to give good performance for the control of the experimental CSTR process. This would be a good choice for comparison with W-C tuning.

If W-C tuning with α values of 0.5 are to be used, a long waiting period will be required for the process to reach steady state.

CHAPTER 4: EXPERIMENTAL VERIFICATION OF A PID CONTROL TUNING METHOD FOR CONVERSION IN A SOLUTION MMA CSTR

4.1 Introduction

The experimental system consists of a heated 1 L reactor in which MMA was polymerized in solution. Monomer, toluene solvent and AIBN initiator dissolved in toluene were fed in at controlled rates. Conversion was controlled in the range of 20 to 35%. Setpoints were changed both up and down. Disturbances were introduced by a pulse injection of 2,2-diphenyl-1-picrylhydrazyl hydrate (DPPH) into the feed stream, that consumed free radicals and slowed down the reaction.

This system exhibits significant time delays of about half an hour. Based on the feed rate, the average residence time is 25 minutes. Product flowing out of the reactor, through a cooling system, and through the on-line instruments also delays the measurements by about 10 minutes.

Conversion is measured using an on-line densitometer. The increase in density as monomer is converted to polymer is converted to a weight conversion that is fed back to the PID controller running on a PC. This was a significant source of measurement noise. Air bubbles entrained in the product stream passed through the densitometer causing occasionally a momentary drop in the density reading.

Results show that the PID controller tuned using the W-C tuning technique and an approximate process transfer function controlled the conversion to desired changes in the setpoint and recovered from process disturbances.

An IMC tuning based PID controller was also tested. This controller was more aggressive, and tended to saturate the controller signal. Because of its aggressive tuning, setpoint changes were effected more quickly, but at the cost of an oscillating controller signal.

4.2 Experimental System

The CSTR laboratory holds the 1 litre continuous stirred tank reactor (1L-CSTR), and the control system composed of a desktop PC, and OPTOMUX OPTO-22 digital and analogue input/output controller.

The CSTR is used for control studies on a continuous free radical solution polymerization process. The monomer is MMA, the solvent is toluene, and the initiator is AIBN. Typical reaction conditions are:

- temperature around 50 to 90°C
- atmospheric pressure
- 60 % toluene, 40 % MMA in the feed

Poly(methyl methacrylate) of between about 20,000 and 40,000 weight-average molecular weight (MW) is produced at approximately 20 to 40 % conversion.

Small quantities of DPPH can be used to "poison" the reaction.

The apparatus and procedures for the 1L-CSTR are described in detail in "Operating Procedure for 1-Litre Continuous Stirred Tank Reactor" (Chien and Penlidis, 1994a).

A review of the apparatus, chemicals, and operational procedures was written in the "Health and Safety Assessment Report: Polymer CSTR Laboratory E1-2550A" (Tanaka, 1995).

A demonstration run was completed by R.K. Mutha on November 1, 1995, where several control strategies were evaluated.

Figure 4.1 shows a schematic diagram of the experimental apparatus. The system consists of three feed lines, the reactor, heating and cooling equipment, on-line sensors, data collection system, and a process control computer. The system was developed and described in detail in Chien (1992). A brief overview of the equipment and procedures follows.

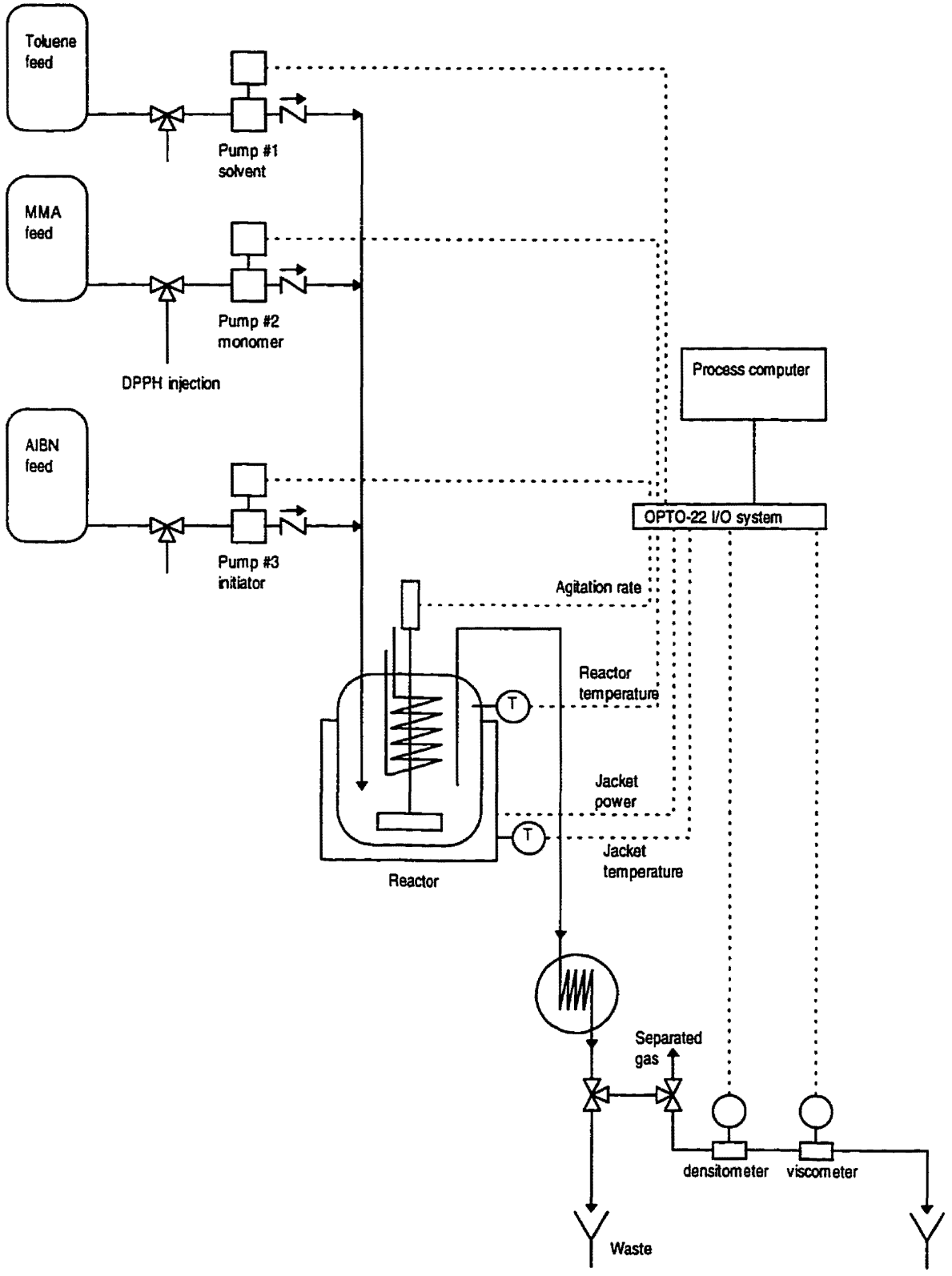


Figure 4.1: Schematic of experimental apparatus

4.2.1 Apparatus and Materials

Feed

The three feed lines are MMA monomer, toluene solvent, and AIBN initiator dissolved in toluene. Each species is pumped from a drum or flask by a computer controlled, ProMinent EXtronic (model BX2A1006SB for pump #1 and pump #3, externally pulsed model BX2E1006SB for AIBN pump #2) diaphragm-type metering pump, through stainless steel tubing, and into the reactor. The whole feed system operates at room temperature.

Toluene (Aldrich Chemical, commercial grade 99% toluene) is delivered in 18 L drums. It is not purified before use. Toluene is fed directly from the drums, through 1/8" tubing into metering pump #3. The solvent pulses out, through a Nupro 20 lb. check valve and along 1/8" tubing into a feed header on the reactor.

Methyl methacrylate inhibited with 10 ppm MEHQ (Aldrich Chemical, commercial grade 99% MMA) is delivered in 17 L drums. It is not purified before use. MMA is fed by a small, variable speed positive displacement pump (FMI Lab pump, 1/4" piston) through 1/8" stainless steel tubing into a magnetically stirred 2 L aspirator bottle. Nitrogen gas is bubbled slowly through monomer in the bottle to remove dissolved oxygen. Metering pump #3 pulses MMA through a Nupro 3-50 lb. relief valve (manually adjusted to 15 lb.), along 1/8" tubing and into the reactor feed header.

AIBN (Dupont, VAZO-64) is delivered in 10 lb fibre drums. 43.794 g (47.892g/L in Chien, 1992) of the white crystals are dissolved in 1 L of toluene at room temperature to make 0.032 mol/L initiator solution. It is held in a magnetically stirred, 2 L aspirator bottle. Nitrogen gas is bubbled slowly through the solution to remove dissolved oxygen. 1/4" tubing connects the bottle to the pump. Metering pump #2 pumps solution, through a Nupro 20 lb. check valve, along 1/4" tubing, and into the reactor feed header.

DPPH (Aldrich Chemical) is delivered in 2 g vials. It is dissolved in 200 ml of MMA and injected into the monomer feed line in place of the regular MMA feed.

Reactor

The reactor is a Chemineer mini-reactor series 1000 ml model. The 316 stainless steel vessel has an inner diameter of 4.25" and approximate 1309.5 ml volume. A sparge/dip tube, a finned thermowell-baffle, 1/4" coil and an agitator shaft all descend through the vessel head.

The agitator consists of a six blade, ~2.25" diameter turbine attached 4.25" below the top of the reactor and driven by a 1/2 hp air motor. Agitator speed is set by manually adjusting the air pressure, measured by a tachometer, and is recorded on the process computer. Normal operating speed is about 300 rpm.

Cooling water flows through the coil. The coil is made of 1/4" O.D. tubing with a 2.25" I.D. and 37 $in.^2$ surface area. Cooling water flow from utility into the coil is controlled by a needle valve on computer

control. Water temperature as it leaves the vessel is measured by a transducer and is recorded by the process computer. The thermowell is empty.

An electric heating mantle heats the reactor. The heater is controlled by proportionating the on/off cycle time with a signal from the process computer. For all the experiments, the duty cycle was maintained at 65% on, except during startup and shutdown. For the beginning and the end of the run, the heater duty was changed to 100% and 0%, respectively.

Product

Product from the reactor overflows through 1/4" tubing through a cooling water bath, bypass valve, deaerator, on-line viscometer, on-line densitometer/flow meter, and into a waste drum.

The cooling water bath is filled with utility cold water at approximately 25 °C. The bypass valve can send reactor effluent directly to the waste without passing through the sensitive on-line instruments. The deaerator removes bubbles of gas: most likely nitrogen formed from the decomposition of AIBN as well as entrained nitrogen gas and toluene and MMA vapours, which can cause problems in the densitometer.

Viscosity is measured by a Contraves Covimat (CVM 105-DC40E) in-line rotational viscometer. It measures braking torque between a rotating measuring bob (cylinder) and drive motor which is converted to viscosity.

The densitometer consists of a Micromotion Mass Flow Meter (Model D012S100) and support equipment (Remote Flow Transmitter RFT-9712; Rosemount 268 Smart Family interface) that measures fluid mass flow rate as it passes through an oscillating "U" shaped tube. Rated accuracy is about +/- 0.2% of rate and the measurement range was configured to 0.256 kg/min. Temperature is also measured as part of the information required to calculate density/flow.

Measurements taken from on-line instruments are converted and read through the OPTOMUX Opto-22 interface. Communication is through an RS-485. The Opto-22 is interfaced to a 486DX33 Intel CPU based personal computer running the MS-DOS 5.0 operating system and a custom process control program.

4.2.2 I/O Interface and Process Computer

The OPTOMUX Opto-22 data acquisition system connected to the reactor consists of one 16 position analog mounting rack (PB16AH) with a "brain" board (B2) and another digital 16 channel "quad-pak" mounting rack (PB16HQ), RS-485 communication link to the PC, and some support circuitry (power supply, wiring, etc.). The input and output modules plugged into the boards are summarized in Table 4.1.

The original reactor control program was written in Microsoft QuickBASIC version 4.5 (Chien, 1992). The control program was updated and revised for multivariate control, as well as adding new controllers (Ogunye, 1994; Lawrence, 1994). Conversion control manipulating AIBN flowrate was reprogrammed and the whole program was cleaned up in QuickBASIC version 7.0. The program flow diagram for the updated program is shown Figure 4.2.

Table 4.1: Opto-22 I/O module summary

Position	Module	Description	Function
0	AD5T (type J)	thermocouple input	reactor temperature
1	AD5T (type J)	thermocouple input	jacket/mantle temperature
2	AD5T (type J)	thermocouple input	cooling water outlet temperature
3	AD3T	4 ~ 20 mA input	cooling water valve position
4	DA4	0 ~ +5 V output	4.858 V supply to read CW pot.
5	-		
6	AD7	0 ~ 10 V input	product temperature
7	AD3T	4 ~ 20 mA input	density
8	AD20		mass flow
9	-		
10	DA5	0 ~ 10 V input	jacket voltage
11	AD7	0 ~ 10 V input	agitator RPM
12	AD6T	0 ~ 5 V input	viscometer
13	DA3	4 ~ 20 mA ouput	MMA pump flow
14	DA3	4 ~ 20 mA ouput	AIBN pump flow
15	DA3	4 ~ 20 mA ouput	toluene pump flow

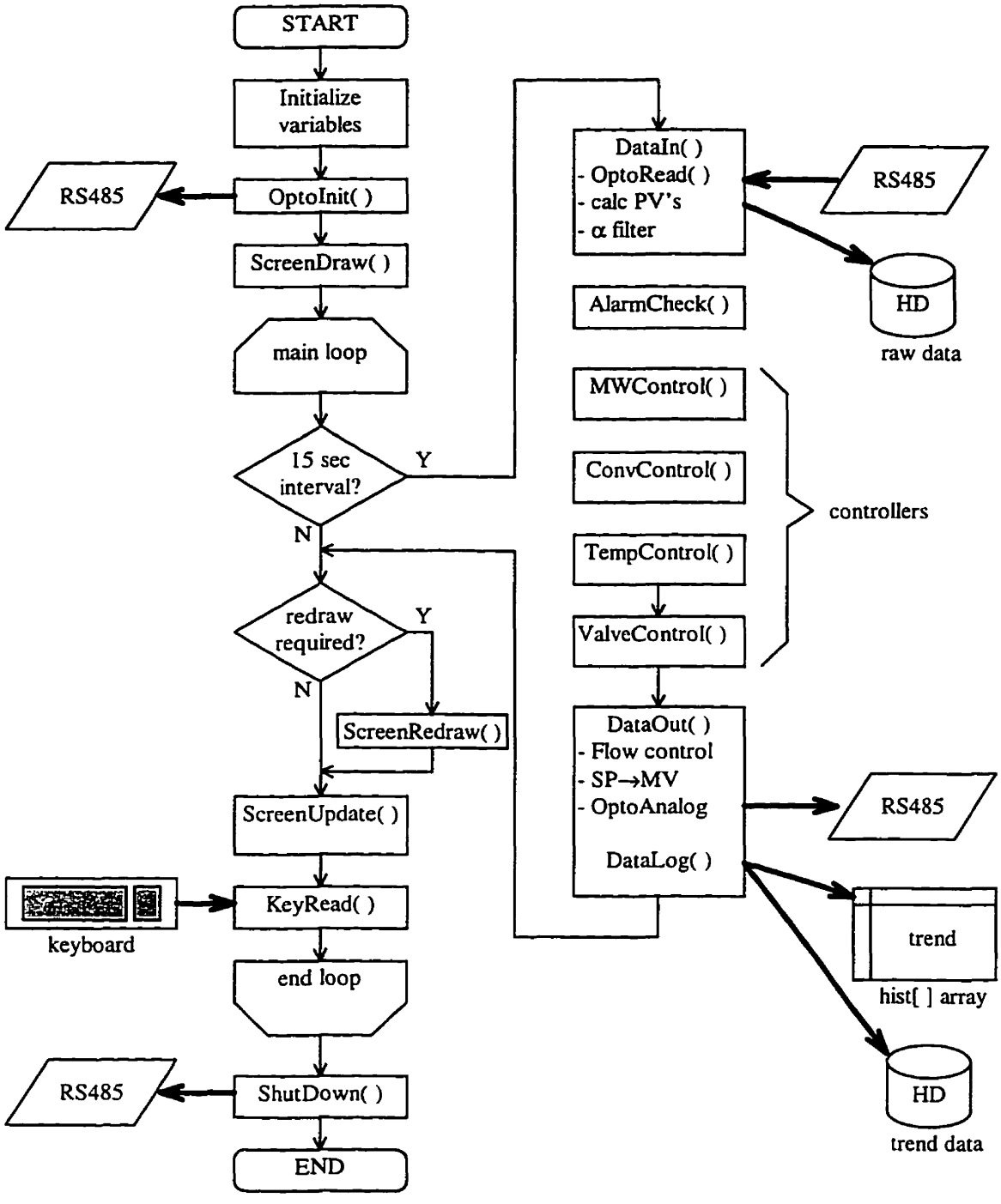


Figure 4.2: CSTR control program flow sheet

4.2.3 Operating Conditions and Procedures

Detailed descriptions of equipment, operating conditions and procedures are given in Chien (1992) and the 1-litre CSTR Operating Manual (Chien and Penlidis, 1994a). Nominal operating conditions are tabulated in Table 4.2.

Level control is by overflow out of the reactor into the product lines. Pressure is at atmospheric level plus the back pressure in the product lines and on-line instrumentation.

Table 4.2: Nominal operating conditions

State	Nominal Value	Normal Operating Range
reactor mixture temperature	80 °C	isothermal 70-90 °C
feed monomer:solvent ratio	40:60 by vol.	constant
monomer feed rate	13.33 ml/min	constant
solvent feed rate	16.00 ml/min	5-20 ml/min
initiator solution feed rate	4.00 ml/min	0-11 ml/min
residence time	39.3 min	constant
mass conversion monomer to polymer	30 %	20-40 %

4.2.4 Temperature Controller

One problem with temperature control is that the reactor is heated and cooled by multiple sources. This is a multiple input, single output (MISO) control problem. Heat sources are the heating jacket, and heat released by the exothermic polymerization reaction. Heat is removed by the cold water (CW) flowing through the internal coil, and via heat losses from the reactor and jacket, and through the product.

Mutha (1996) studied the heat balance around the reactor. He implemented a thermocouple on the CW outlet to better predict the heat load.

The current temperature controller varies the CW flow rate while maintaining a constant heating jacket power input. The constrained minimum variance control (CMVC) temperature controller implemented by Ogunye (1994) was used with a small modification. It calculates a CW valve position in order to maintain a reactor temperature. The percentage is converted to a CW valve position which is implemented separately by turning the needle valve clockwise or counter-clockwise a number a seconds. Previously, the fraction of maximum flow rate was given in instrument units (mA). It was converted to a percentage of maximum.

The duration, dur, to turn the valve is given by the following method. It is recalculated at every iteration at 15 second intervals. The present and setpoint values (PV and SP) are valve position in mA as measured by the potentiometer.

$$v_2 = v_1$$
$$v_1 = v_0$$
$$v_0 = PV - SP$$
$$K_v = \left(\frac{36}{SP}\right)^2$$
$$T_{iv} = 360$$
$$T_{dv} = 0$$
$$T_s = \Delta t_{valve}$$
$$sum = sum + v_0$$
$$dur = K_v \times v_0 + \frac{T_s}{T_{iv}} \times sum$$

Minimum and maximum valve position measurements are hard-coded into the control program and used as the 0% and 100% limits of the manipulated (MV) controller output.

4.2.5 Conversion Controller

The conversion controller is a digital PID controller in velocity form. It manipulates AIBN flow into the reactor to achieve a desired conversion setpoint. Tuning and implementation of this type of single input single output (SISO) controller was discussed in detail in Chapter 3.

Figure 4.3 is block diagram for this controller.

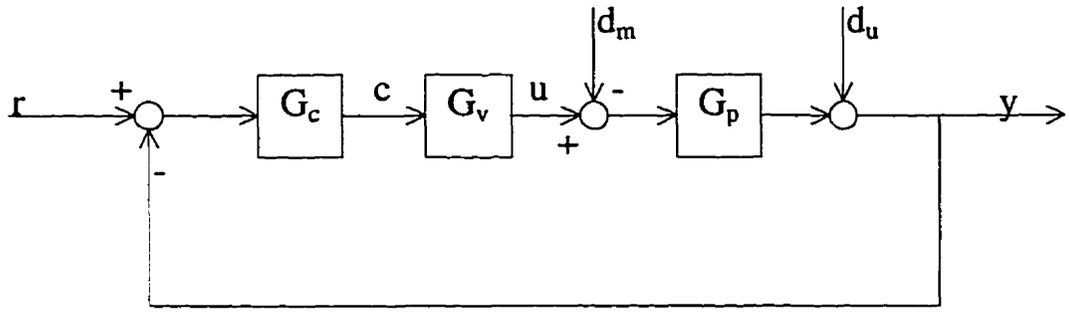


Figure 4.3: Conversion control block diagram

The setpoint, r, is compared with the measured output, y, and input into the controller, G_c. The controller calculates a required output, c, that is clamped within the physical limits of the pump output, G_v, and a physically achievable output, u, is sent to the process. A measured disturbance, d_m, inhibitor injection (DPPH), can be introduced at this point. The process, G_p, and any unmeasured disturbances, d_u, are measured together and form the perceived output, y.

4.3 Methodology

4.3.1 Experimental Run Objectives

One experimental run was completed to verify the performance of the W-C tuned PID conversion controller. The run consisted of three parts: an open loop segment without the controller, a closed loop segment with the controller tuned using W-C tuning rules, and a final closed loop segment with the controller tuned using IMC tuning rules.

The open loop segment verified that the process model previously identified and used in the simulation studies was a good representation of the actual process. The segment had three tests, consisting of a step change in the initiator flow rate from a high value to a low value, and then back up to the high value, followed by a pulse disturbance.

The first closed loop segment verified that the W-C tuned PID controller performed according to the specification and as predicted by the simulations. The three tests were: a controlled conversion change from a high setpoint to a low setpoint, a move back up to the high setpoint, and recovery from a pulse disturbance. A conservative α=0.5 case was chosen as a safe example since the behaviour of the PID controller had not been recently tested.

The second closed loop segment compared the W-C tuned PID controller with a PID controller tuned using IMC tuning rules, a well known and industrially accepted tuning method. This segment also consisted of three tests: a controlled conversion setpoint step down, a step up, as well as recovering from a pulse disturbance. The controller was tuned with a more aggressive λ/θ=0.9 since this would be the second closed loop test and would make a good contrast to the conservative W-C tuned controller.

4.3.2 Experimental Run Procedures

The proposed experimental run procedure steps are summarized in Table 4.3.

The run was divided into three segments: open loop, closed loop with W-C tuning, and closed loop with IMC tuning. Each segment consisted of three tests: a conversion step down, another step up, and recovery from a pulse disturbance.

The run started with the reactor initially charged with a 40:60 monomer:solvent solution to match the constant feed mix ratio. There was no initiator in the reactor at the beginning of the run. The reactor temperature was raised to 80 °C and maintained within 0.5 °C of the target temperature for the duration of the run. All three feed pumps were started. Once at steady state with 4 ml/min initiator mix feed rate, the first, open loop, segment tests were started.

For open loop mode, the AIBN flow rate was manually set to either a high (4 ml/min) or low (2 ml/min) rate. For closed loop mode, the AIBN flow rate was on computer control, and automatically adjusted by the computer. For closed loop mode, the conversion setpoint was manually set to the high or low value.

DPPH was manually added into the MMA feed stream as a pulse injection. Flow was either on (1) or off (0).

Table 4.3: Run procedure summary

Time [min]	Conversion Controller	Manipulation	Target (wait for)
0	open loop	start heating, and initiator flow	steady state
180		AIBN flow rate step down 4 → 2 ml/min	steady state at low conversion
300		AIBN flow rate step down 2 → 4 ml/min	steady state at high conversion
420		DPPH injection	recovery to high conversion
660	closed loop W-C tuning	conversion setpoint step down 40 → 30 %	steady state at low conversion
780		conversion setpoint step up 30 → 40 %	steady state at high conversion
900		DPPH injection	recovery to high conversion and steady state
1140	closed loop IMC tuning	conversion setpoint step down 40 → 30 %	steady state at low conversion
1260		conversion setpoint step up 30 → 40 %	steady state at high conversion
1380		DPPH injection	recovery to high conversion and steady state
1560	open loop	shut down heating and AIBN flow	ambient conditions

4.3.3 On-line Data Collection

Temperature readings are taken every 15 seconds, and trended on the screen. Every minute, they are used by the temperature controller to control reactor temperature.

Flow rates for the metering pumps are set every minute.

Density measurements are taken every 15 seconds and converted to conversion based on Eq. 4.1. These values are trended on the screen. Every minute, the conversion controller uses this measurement as the input. Readings are stored to disk every minute as well.

$$conversion = \frac{\frac{\rho_{in}}{\rho} - 1}{1 - \frac{\rho_p}{\rho_m}} \tag{4.1}$$

ρ_{in} is the density of the feed (constant 40% monomer, 60% solvent), ρ is the measured product density, ρ_m is the pure monomer density and ρ_p is the pure MMA polymer density.

Viscosity measurements are taken every 15 seconds. They are saved to disk every minute. Lawrence (1994) obtained parameter estimates for the viscosity to molecular weight correlation given in Eq. 4.2.

$$\ln \eta = \left(\alpha_1 + \frac{\alpha_2}{T} \right) + \alpha_3 X \overline{M_w}^{\frac{1}{2}} \tag{4.2}$$

where η is the solution viscosity in cP, T is the product temperature in °C, X is the conversion, and $\overline{M_w}$ is the weight average molecular weight in thousands. The 95% confidence intervals for the parameters are (Lawrence, 1994):

$$\alpha_1 = -1.048 \pm 3.84 \times 10^{-3}$$
$$\alpha_2 = 19.137 \pm 0.924$$
$$\alpha_3 = -9.095 \times 10^{-3} \pm 2.819 \times 10^{-5}$$

Samples of the product were collected every 30 minutes, inhibited with hydroquinone, and stored in a freezer.

4.3.4 Off-line Measurements

Gravimetry as described by Chien (1992) and Lawrence (1994) was used to verify the measurement of conversion off-line using the samples that were collected during the run.

Briefly, part of each sample was weighed ($mass_{initial}$), vacuum oven dried, and weighed again ($mass_{dry}$). Conversion was calculated using Eq. 4.3.

$$conversion = \frac{\left(\frac{mass_{dry}}{mass_{initial}}\right)}{\left(\frac{x_m \rho_m}{x_m \rho_m + x_s \rho_s}\right)} \tag{4.3}$$

where x_m and x_s are the mass fraction monomer and solvent in the feed, and ρ_m and ρ_s are the densities of the pure monomer and solvent.

Gel permeation chromatography (GPC) was used to determine molecular weight characteristics of some of the samples that were collected during the run.

4.4 Experimental Results

Experimental data was divided into the three segments of the experimental run. The results from the first segment, operation in open loop mode, is described in Section 4.4.1.

Section 4.4.2 describes the results from the second and third segments. The second segment was closed loop operation with the W-C (α=0.5) tuned controller. The third and final segment was closed loop operation with the IMC (λ/θ=0.9) tuned controller.

Off-line conversion and molecular weight measurement results are presented in Section 4.4.3.

4.4.1 Open Loop Results

Figure 4.4 shows the results from the open loop mode of the run. The three plots, from top to bottom, are the conversion (%), AIBN flow rate (ml/min) and DPPH flow (on/off) trends from the 153rd to 523rd minutes of data collection. DPPH flow was either "on" (1) or "off" (0).

Steady state conversion with the initial 4 ml/min AIBN flow rate was approximately 31%. When the initiator flow rate was dropped to 2 ml/min, the conversion dropped to about 24%. It took about 2 to 3 hours to achieve steady state. The AIBN flow was increased to 4 ml/min. When the system had stabilized back to about 32% conversion, DPPH dissolved in MMA was injected into the monomer feed line. Conversion dropped about 8% before recovering to its original value.

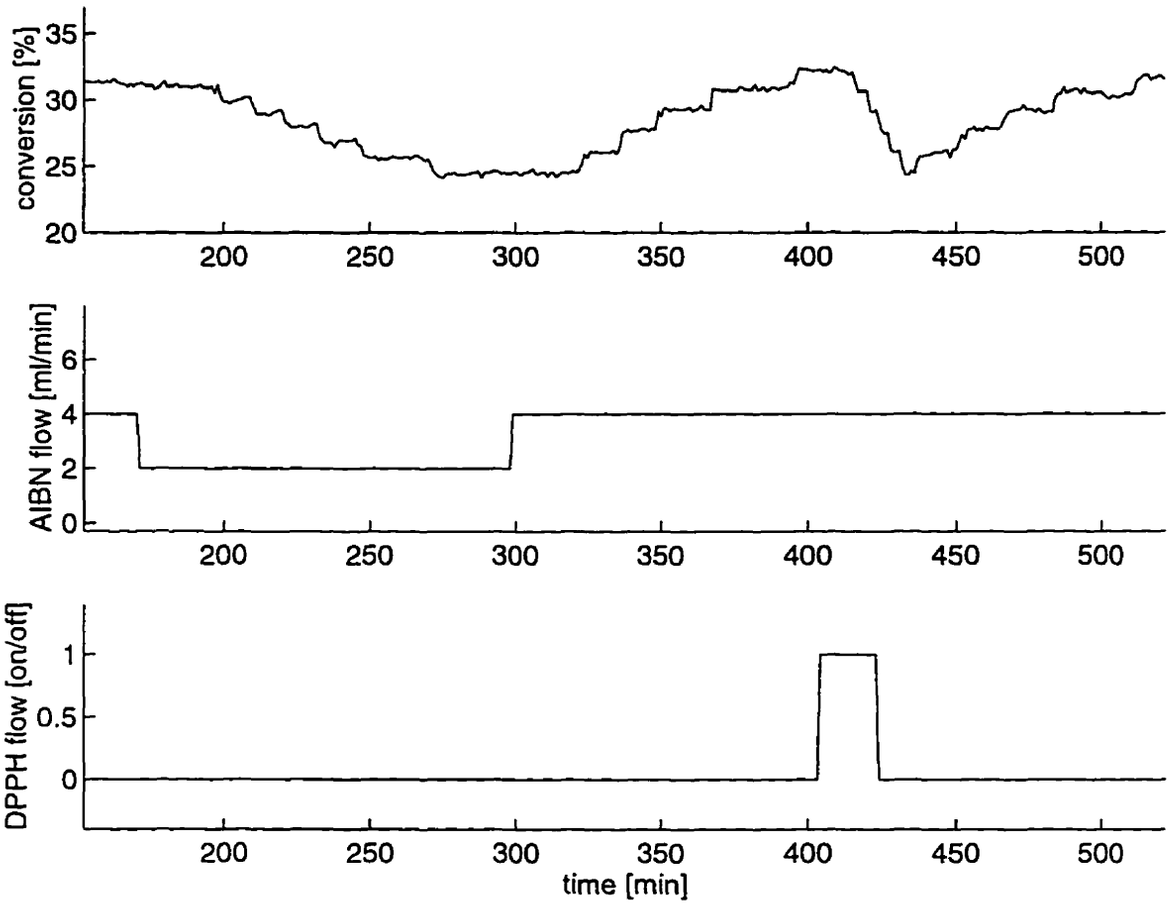


Figure 4.4: Open loop response of conversion process

4.4.2 Closed Loop Results

Figure 4.5 shows the results from the second segment, closed loop mode with the W-C (α=0.5) tuned controller. The three plots, from top to bottom, are the conversion (%), AIBN flow rate (ml/min) and DPPH flow (on/off) trends from the 636th to 1028th minutes of data collection. DPPH flow was either "on" (1) or "off" (0).

The process was first allowed to reach steady state at about 32% conversion with 4 ml/min AIBN flow at the open loop mode. Then, the controller was turned on with the W-C tuned parameters and a setpoint at the steady state value (32%).

The conversion setpoint was decreased by 10% to 22%. The controller decreased the AIBN flow rate and was able to control conversion down to about 22% after approximately two hours.

Once more at steady state, the conversion setpoint was increased 10% to 32% and the conversion was controlled back up to 30%. The reluctance to rise to 32% even though the process response curve might suggest that it should, and the sudden jump to 32% just before the disturbance injection suggest plugging or small pockets of poor mixing in the reactor apparatus.

At the high 32% conversion level, under closed loop control, DPPH dissolved in MMA was injected into the feed. The conversion dropped approximately 7% before being controlled back up to the setpoint.

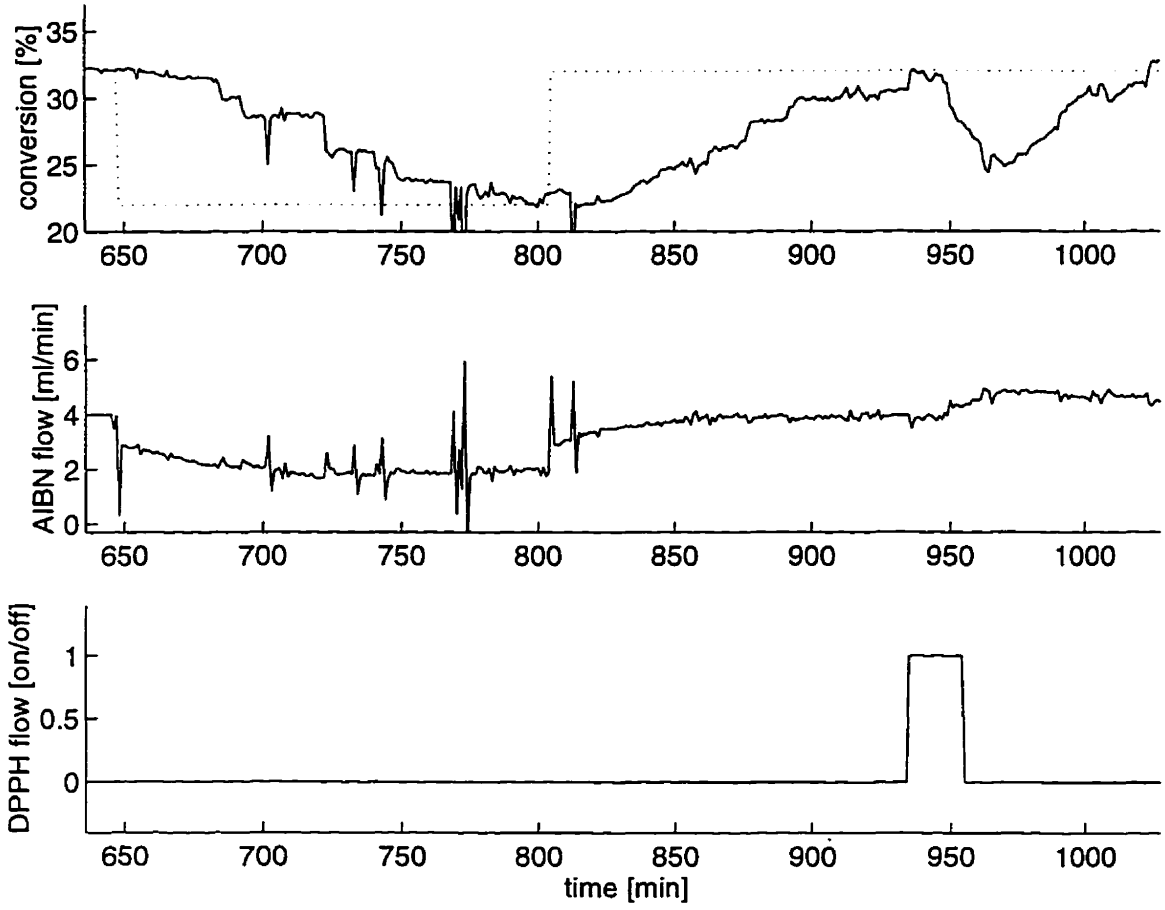


Figure 4.5: Conversion control with W-C tuned controller

(— experimental, ... setpoint)

Figure 4.6 shows the conversion control with the IMC (λ/θ=0.9) tuned controller. The three plots, from top to bottom, are the conversion (%), AIBN flow rate (ml/min) and DPPH flow (on/off) trends from the 1156th to 1525th minutes of data collection. DPPH flow was either "on" (1) or "off" (0).

The procedures was similar to the W-C tuned controller tests but the results were quite different. The initial steady-state was higher, so the high and low setpoints of 35% and 25% were used for this controller.

The controller was able to manipulate the AIBN flow rate to control conversion to the new setpoints faster than with the W-C controller (about 1.5 hours versus 3 hours). There were less problems with what may have been hysteresis in the conversion measurement, although another explanation may be that the system response at 22% conversion is different from that at 32% (ie. nonlinear).

The AIBN flow rate adjustments were very noisy, probably accentuated by the noisy conversion measurement. On a number of occasions, considerable spikes up appear in the AIBN flow rate settings when the conversion measurement spiked down and the controller tried to compensate. These low conversion spikes were caused by gas bubbles entrained in the product stream that give falsely low density reading in the densitometer (Chien, 1992). Saturation of the flow rate was then due to the combined measurement noise and aggressive IMC tuning.

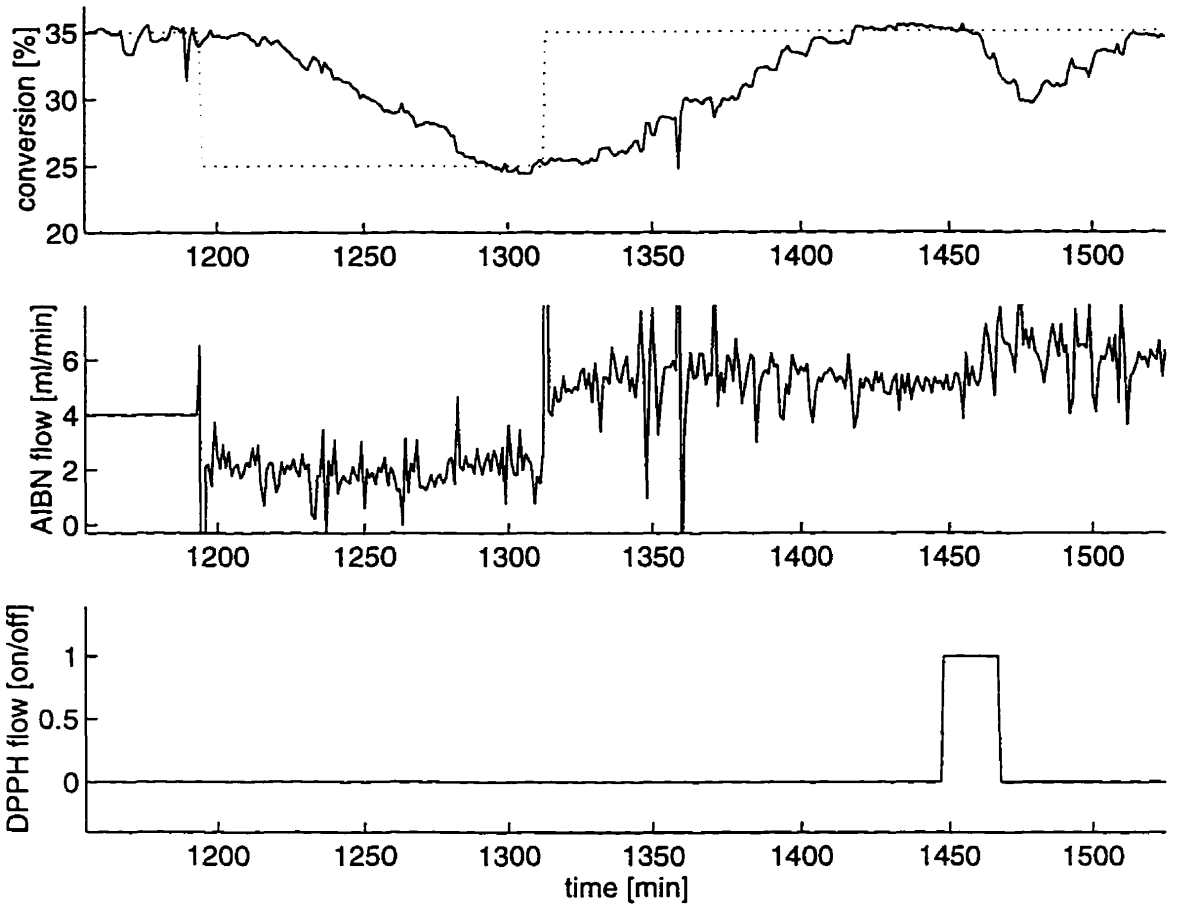


Figure 4.6: Conversion control with IMC tuned controller

(— experimental, ... setpoint)

4.4.3 Off-line Results

Figure 4.7 shows the gravimetry results superimposed on the conversion versus time graph of the whole run. Since conversion is calculated directly from density, even with temperature compensation in the densities, the two show exactly the same information.

We compared on-line conversion predictions with off-line gravimetry analysis. Although the agreement in trends is good, the off-line measurements are systematically higher than the on-line predictions. This may have been caused by a number of factors. The most likely is that unreacted monomer, solvent, or some impurities were still unevaporated and trapped in the dried polymer samples and in weighing the samples, the excess was incorrectly measured as PMMA. Although Lawrence did report that *not* first precipitating the polymer before drying still gave accurate results, it may have been a factor here.

Figure 4.8 shows number and weight average molecular weights of the samples as measured by GPC. The correlation used (Eq. 4.2) was reported to be systematically off with molecular weights below 35,000 (Mutha, 1996). However, Mutha's correction would not correct the measurements enough to give good agreement.

Another possible explanation is a high molecular weight tail, caused by poor local mixing in areas of the reactor and lines. Since the molecular weight correlation uses weight average molecular weight and yet even the number average molecular weights are higher than predicted, there may be a systematic error elsewhere.

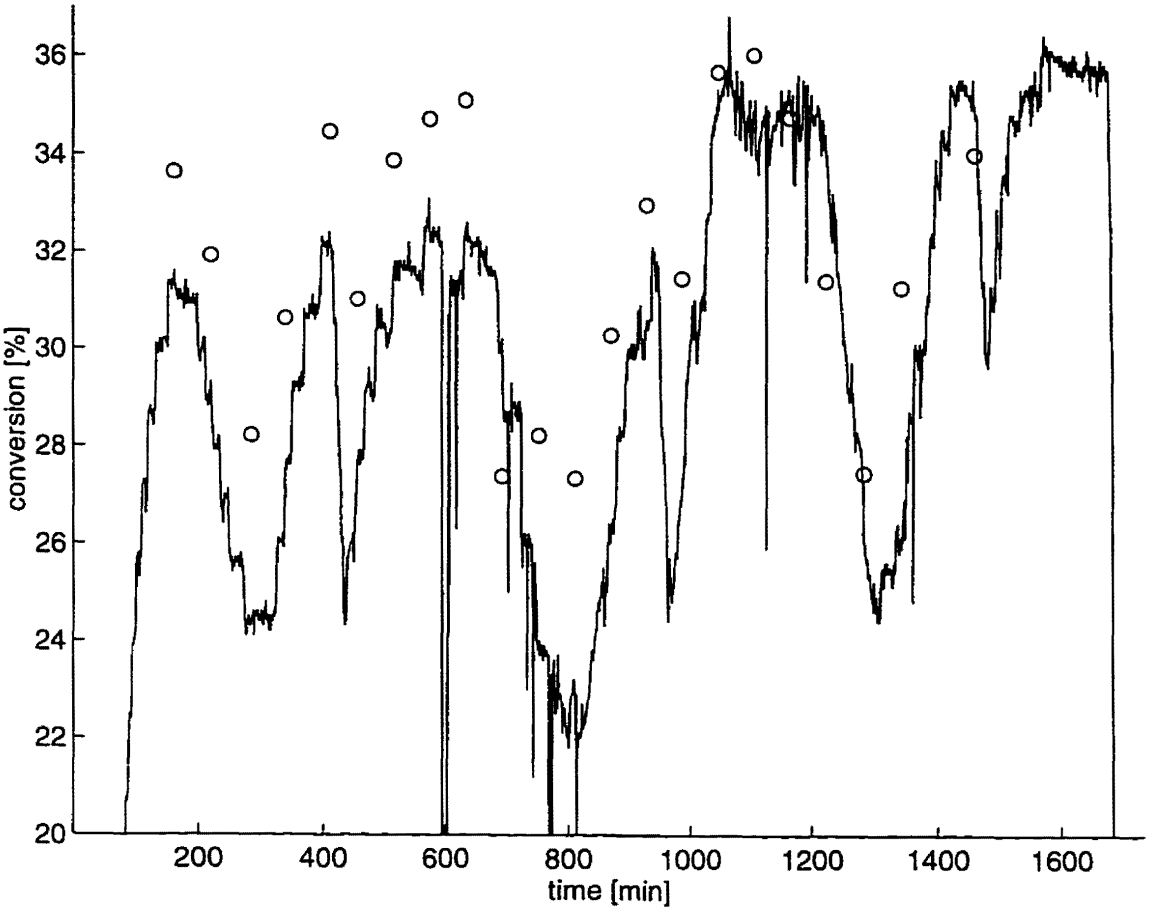


Figure 4.7: Off-line conversion measurement

(— on-line estimation from density, O off-line gravimetry measurement)

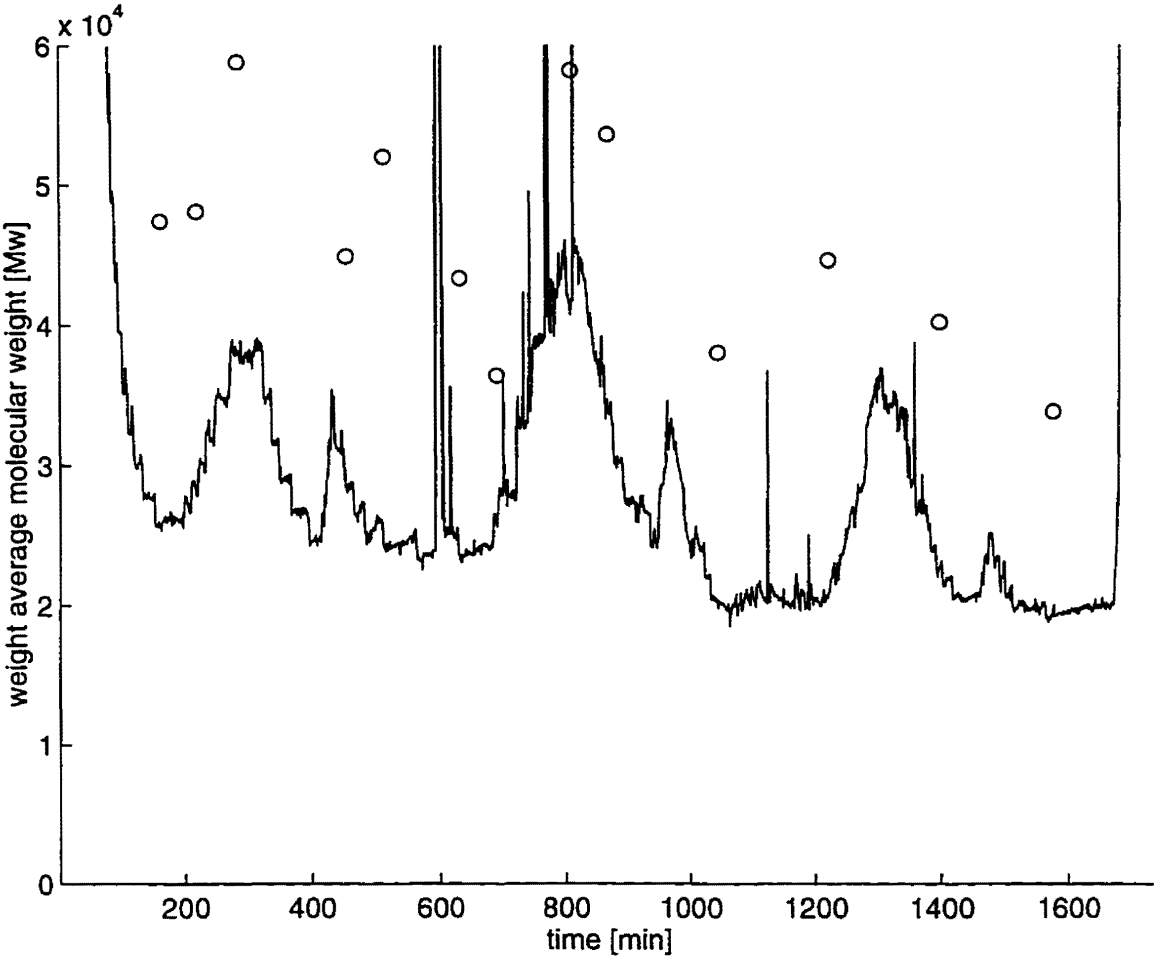


Figure 4.8: Off-line molecular weight measurement

(— on-line estimation from viscosity, O off-line GPC measurement)

4.5 Discussion of Results

4.5.1 Comparison of Chien (1992) Model with Experimental Results

Chien (1992) identified a discrete time transfer function for the open loop process relating the initiator flow rate to conversion. This model was described in Section 3.3.1 and the transfer function was given in Eq. 3.5. The model was compared to the open loop experimental results. Conversion offset was calculated from a steady state conversion of 32 % and AIBN flow rate was deviation from 4 ml/min.

Figure 4.9 is a plot of the model prediction and the open loop experimental results. The three graphs show the simultaneous trends of conversion level, AIBN flow and DPPH injection. There is good agreement between simulation and experimental data, indicating this model is a good process model and that there was little change in the process between 1992 and 1997. The sum of squared errors between simulation and experimental data was 177.7 over 371 minutes or 0.479/min.

Figure 4.10 is a plot of the model prediction and the W-C tuned segment of the closed loop experimental results. The three graphs show the simultaneous trends of conversion level, AIBN flow and DPPH injection. There is good agreement between simulation and experimental data. The sum of squared errors between simulation and experimental data was 652.3 over 392 minutes or 1.664/min.

Figure 4.11 is a plot of the model prediction and the IMC tuned segment of the closed loop experimental results. The three graphs show the simultaneous trends of conversion level, AIBN flow and DPPH injection. There is good agreement between simulation and experimental data. There is a visibly larger deviation of the model prediction from the experimental results than for the two previous figures. The sum of squared errors between simulation and experimental data was 2635.2 over 369 minutes or 7.14/min.

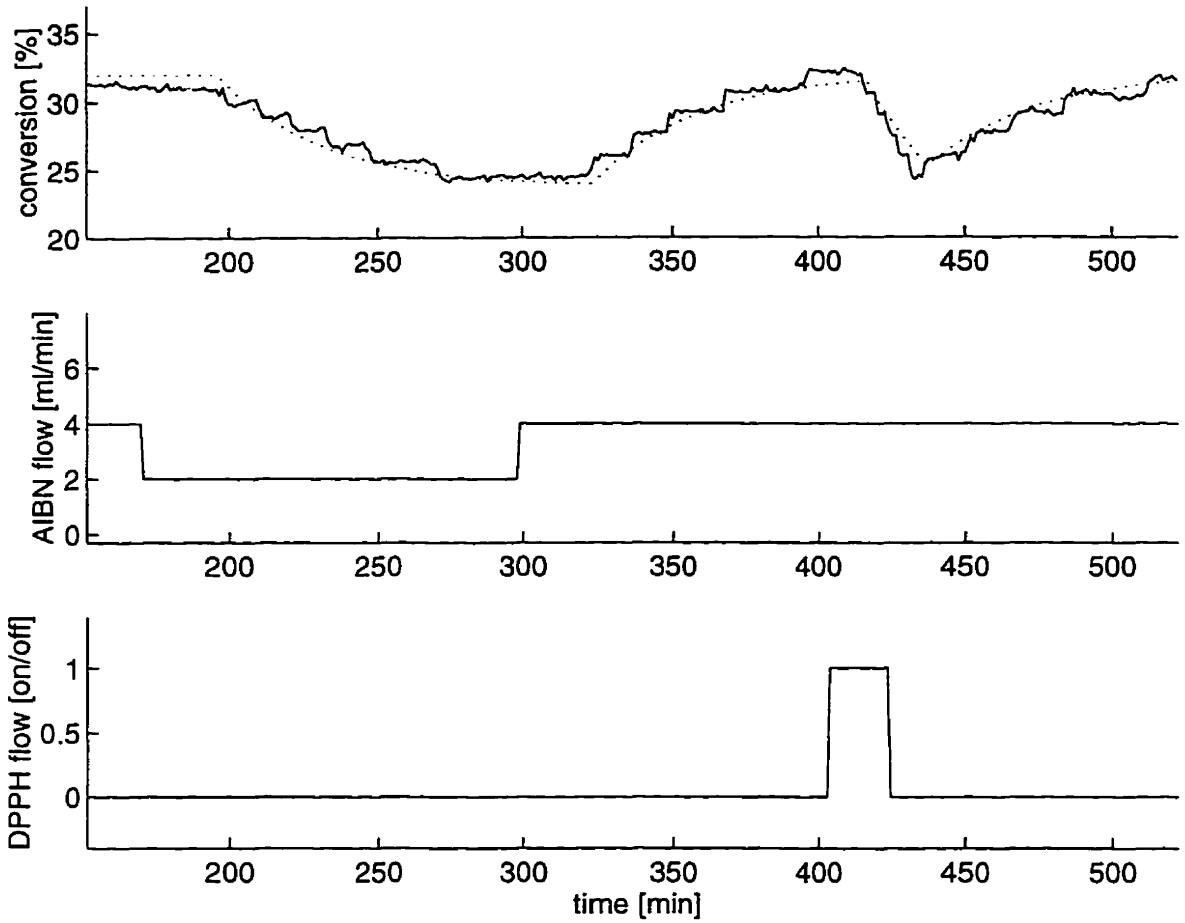


Figure 4.9: Comparison of simulation with Chien (1992) model with open loop experimental results

(— experimental, ... simulation)

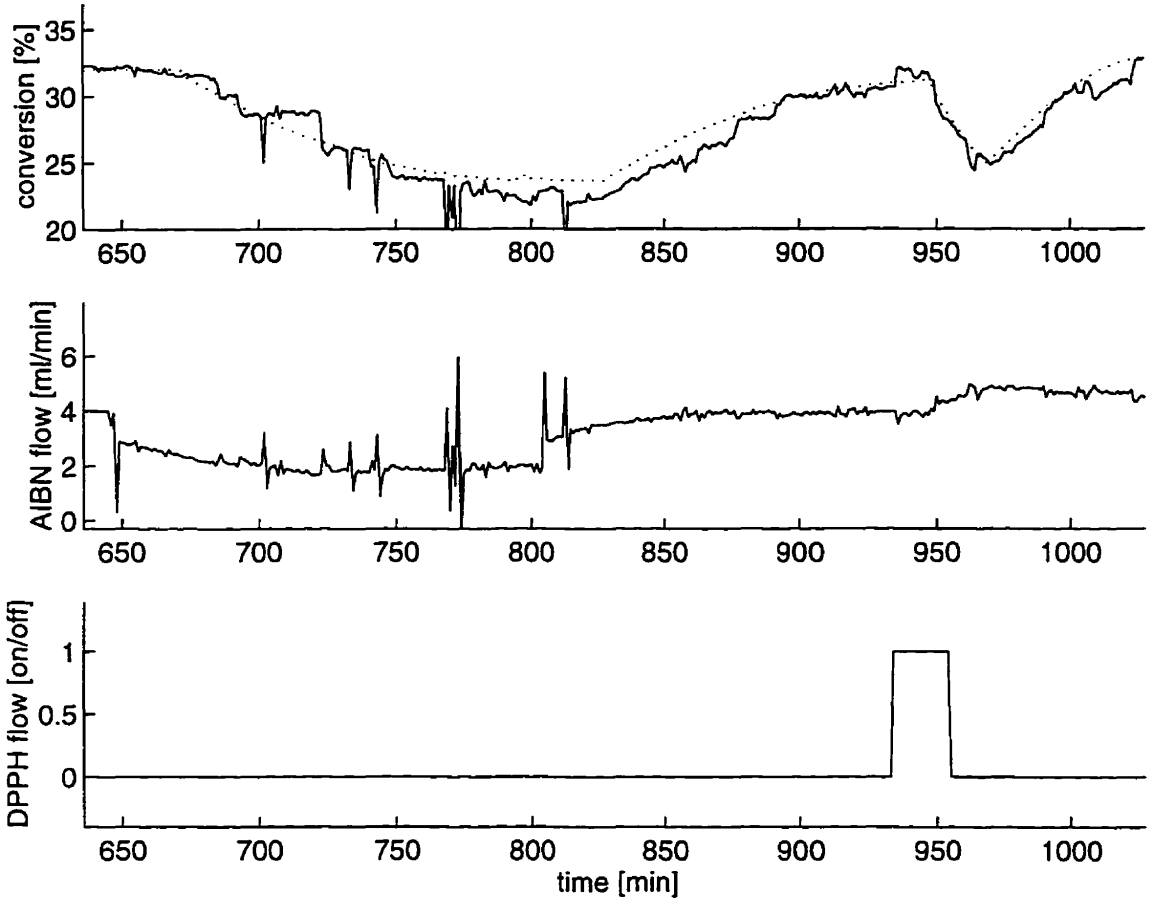


Figure 4.10: Comparison of simulation with Chien (1992) model with W-C segment of closed loop experimental results

(— experimental, ... simulation)

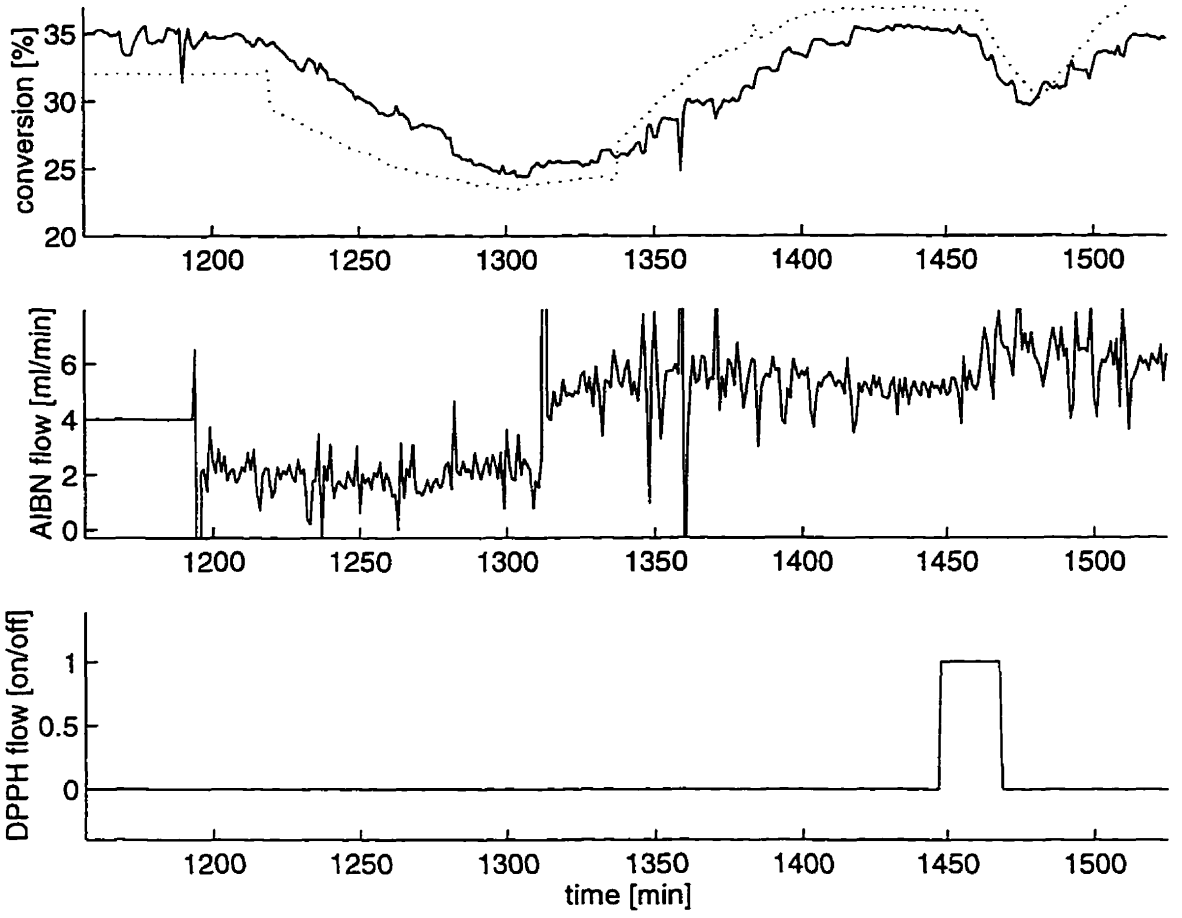


Figure 4.11: Comparison of simulation with Chien (1992) model with IMC segment of closed loop experimental results

(— experimental, ... simulation)

4.5.2 Transfer Function Model Parameters

Each of the parameters in the model originally estimated by Chien (1992) were recalculated base on the new experimental data. Simulations based on the new parameter values were compared with the experimental results.

Chien (1992) Model

Figure 4.12 combines the conversion trends for the Chien (1992) model predictions and experimental results for all three segments. Sum of the difference between the simulation predictions and experimental results are tabulated in Table 4.4.

Open Loop Estimated Model

A FO+D model was estimated using the open loop experimental results. Matlab was used to estimate the parameters for the general linear model. The estimated model is described by Eq. 4.4.

$$Y_t = \frac{0.111z^{-25}}{1-0.971z^{-1}}U_t + \frac{-0.461z^{-14}}{1-0.972}D_t \qquad (4.4)$$

where Y_t is the percent conversion, U_t is the AIBN flow rate (ml/min), and D_t is whether the DPPH injection is on (1) or off (0). Steady state conversion was assumed to be 32%, dead time 25 minutes, and AIBN flow rate was calculated as deviation from 4 ml/min. Figure 4.13 compares the simulation based on this open loop fitted model with the same set of experimental results as Figure 4.12. The sums of squared errors between simulation and experimental data are included in Table 4.4.

When the sums of squared errors between simulation and experimental results are compared for the open loop and two closed loop segments we note the following:

- open loop and IMC tuned closed loop segments, this model gives better agreement then Chien's model
- W-C tuned closed loop segment, this model does not predict behaviour as well as Chien's model

Mean Conversion Estimate

The steady state conversion level was estimated from the conversion in open loop mode with constant 4 ml/min AIBN flow rate. Since process drift may have caused this value to change, the mean conversion was varied from 30 to 35% for each segment to estimate the mean for minimal discrepancy between model predictions and experimental data. Figure 4.14 shows the effect of estimated mean conversion on the simulation predictions. The mean conversion was varied from 30 to 36%. The lowest band on the smooth lines is 30%, and the upper smooth line is 36%.

The best estimates for mean conversion were chosen based on minimizing the sums of squared errors for each of the segments. The minima for the open loop, W-C tuned closed loop and IMC tuned closed loop

segments were with 32.0, 31.4 and 32.3% mean conversion respectively.. The minimum sums of squared errors between simulation and experimental data are summarized in Table 4.4.

Gain Estimate

A FO+D model was fitted separately to each segment of the experimental results. Matlab was used to estimate the parameters for the general linear model. The estimated model for the open loop segment was given earlier in Eq. 4.4. For the W-C and IMC tuned closed loop segments, the models could not be accurately identified so the gains were estimated from the low and high steady state values.

For all segments, steady state conversion was assumed to be 32%, dead time 25 minutes, and AIBN flow rate was calculated as deviation from 4 ml/min. Figure 4.15 shows the simulation and experimental data for the three segments.

Steady state process gains for the AIBN flow to conversion rate are 3.83, 4.77 and 4.06 for three segments respectively. Chien (1992) estimated 4.14, and Lawrence (1994) estimated 2.39 for the same process.

Dead Time Estimate

The dead time for the AIBN to conversion process transfer function in Chien's model was varied from 23 through 37 minutes. For all segments, the steady state conversion was assumed to be 32%, and AIBN flow rate was calculated as deviation from 4 ml/min.

The minimum sum of squared errors for the open loop and W-C tuned closed loop segments were with 24 min and 32 min dead times. For the IMC tuned closed loop segment, the dead time could not be minimized since the model prediction was too far removed from the experimental data and sum of squared errors were an order of magnitude greater than the other test. Dead time estimates for open loop and W-C tuned closed loop segments also depended on the range of data points chosen, process gain and the choice of steady state conversion. Model predictions using 24 min, 32 min, and 37 min are given in Figure 4.16

The DPPH disturbance had best fit with a 14 minute dead time.

Table 4.4: Sum of squared errors for different parameter estimates for the model

	Open Loop	W-C Tuned Closed Loop	IMC Tuned Closed Loop
Chien (1992)	178	652	2635
open loop estimated model	123	852	2235
mean conversion adjusted model	178	524	2606
gain adjusted model	123	398	885
dead time adjusted model	175	570	1584

The results in Table 4.4 show how variability in the process model gain may have a significant effect on the prediction errors. The gain is related to the efficiency in increasing conversion by increasing the AIBN flow rate. The variability of the efficiency with which the AIBN is initiating the reaction may not be constant over time or at different conversion levels. This is problematic since this is directly related to the manipulated AIBN flow rate.

Factors that may be affecting the efficiency of initiation are: batch-to-batch variability as the AIBN initiator solution is being made, non-uniform mixing of initiator and reactants, decomposition rate varying with temperature. A possible solution is adaptively modifying the gain.

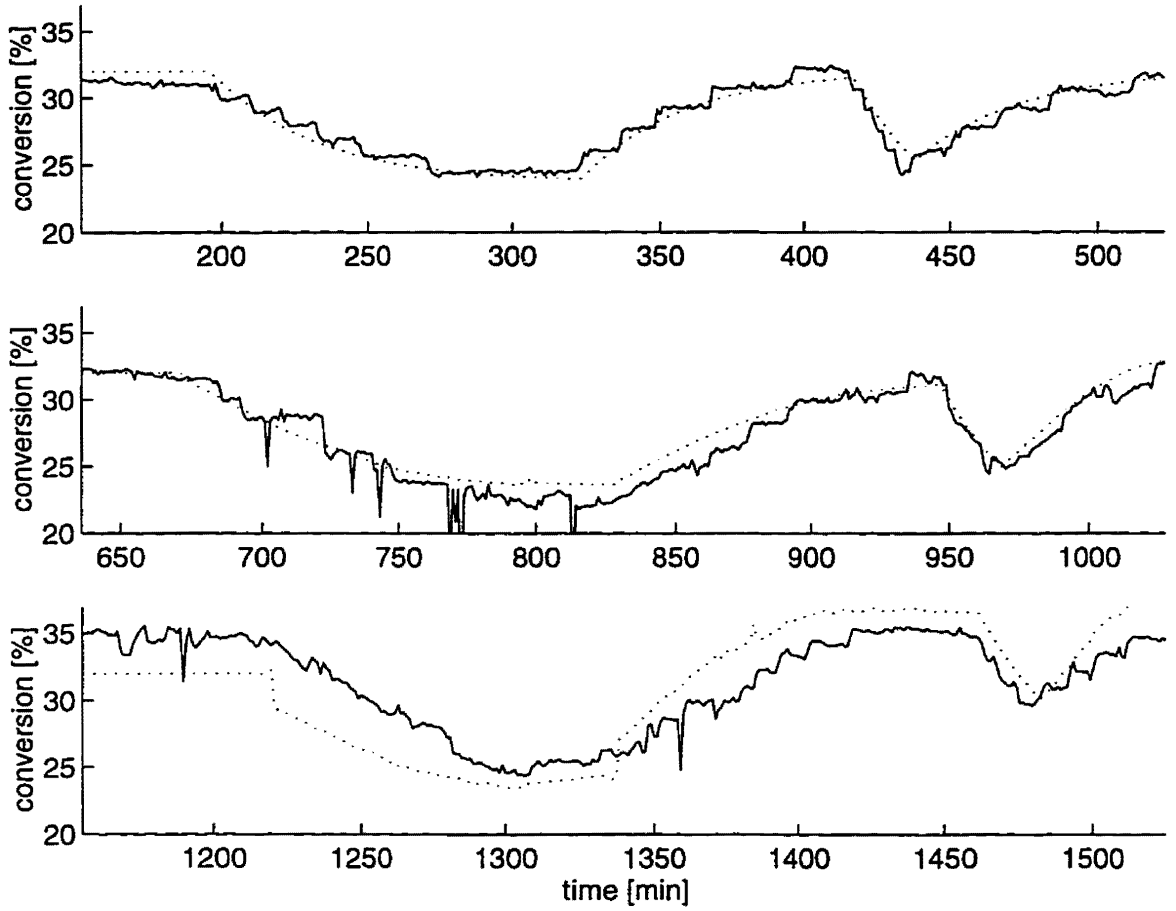


Figure 4.12: Comparison of the Chien (1992) model with experimental results

(— experimental, ... simulation)

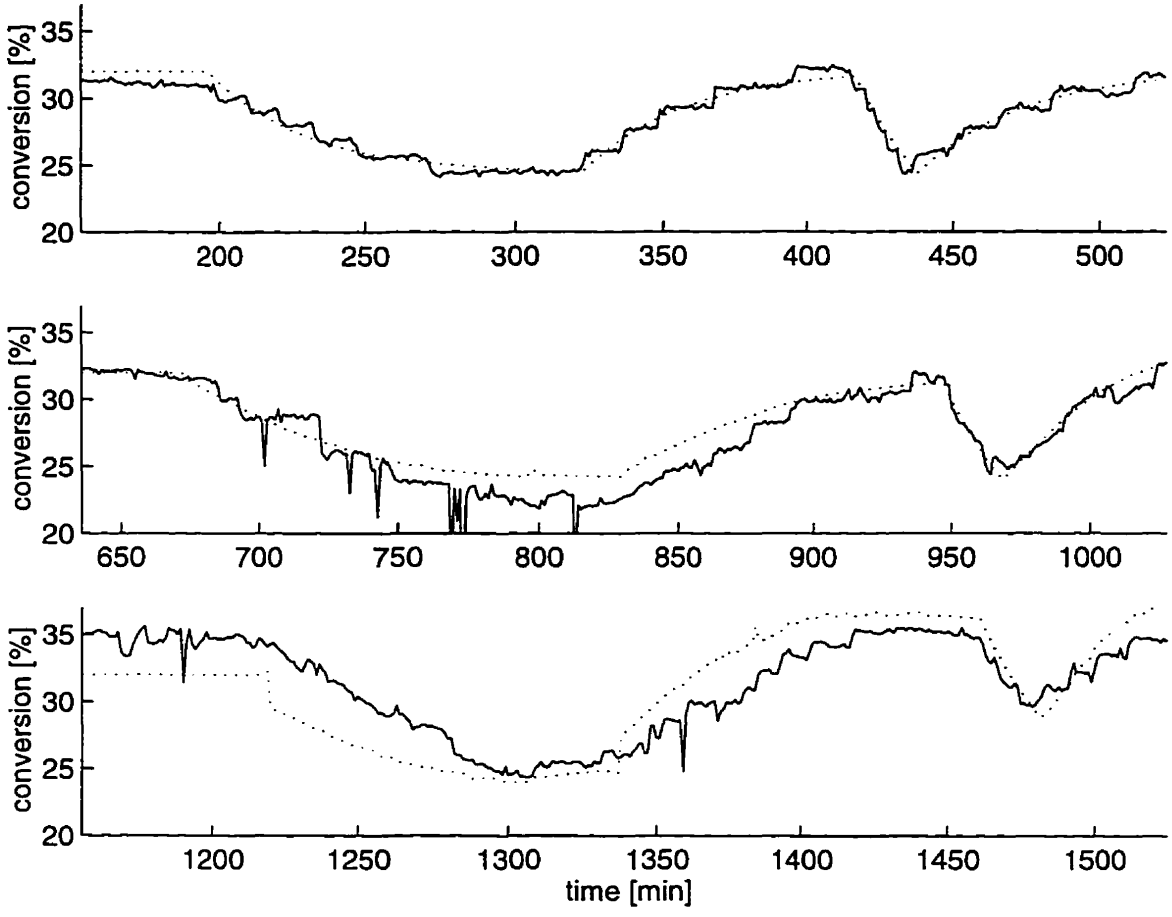


Figure 4.13: Comparison of the open loop estimated model with experimental results

(— experimental, ... simulation)

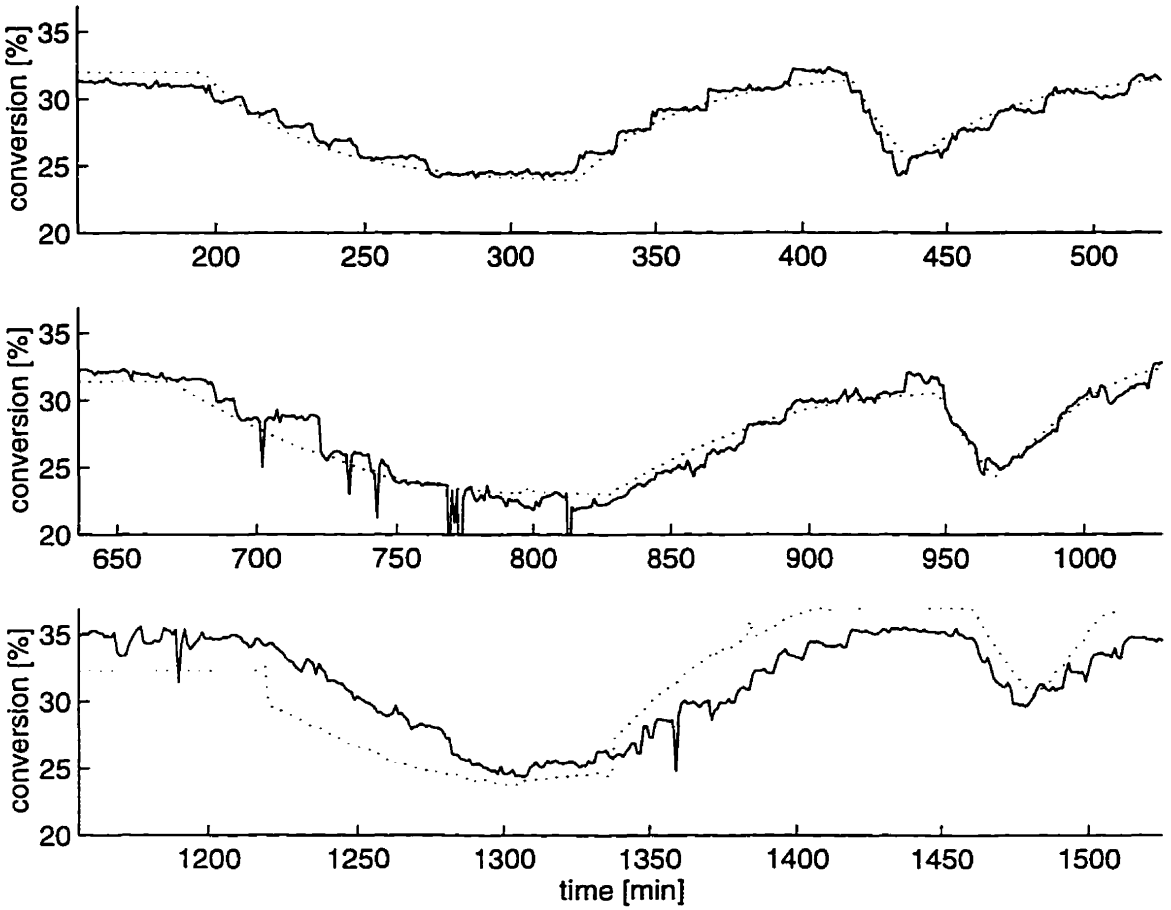


Figure 4.14: Comparison of the mean conversion adjusted model with experimental results

(— experimental, ... simulation)

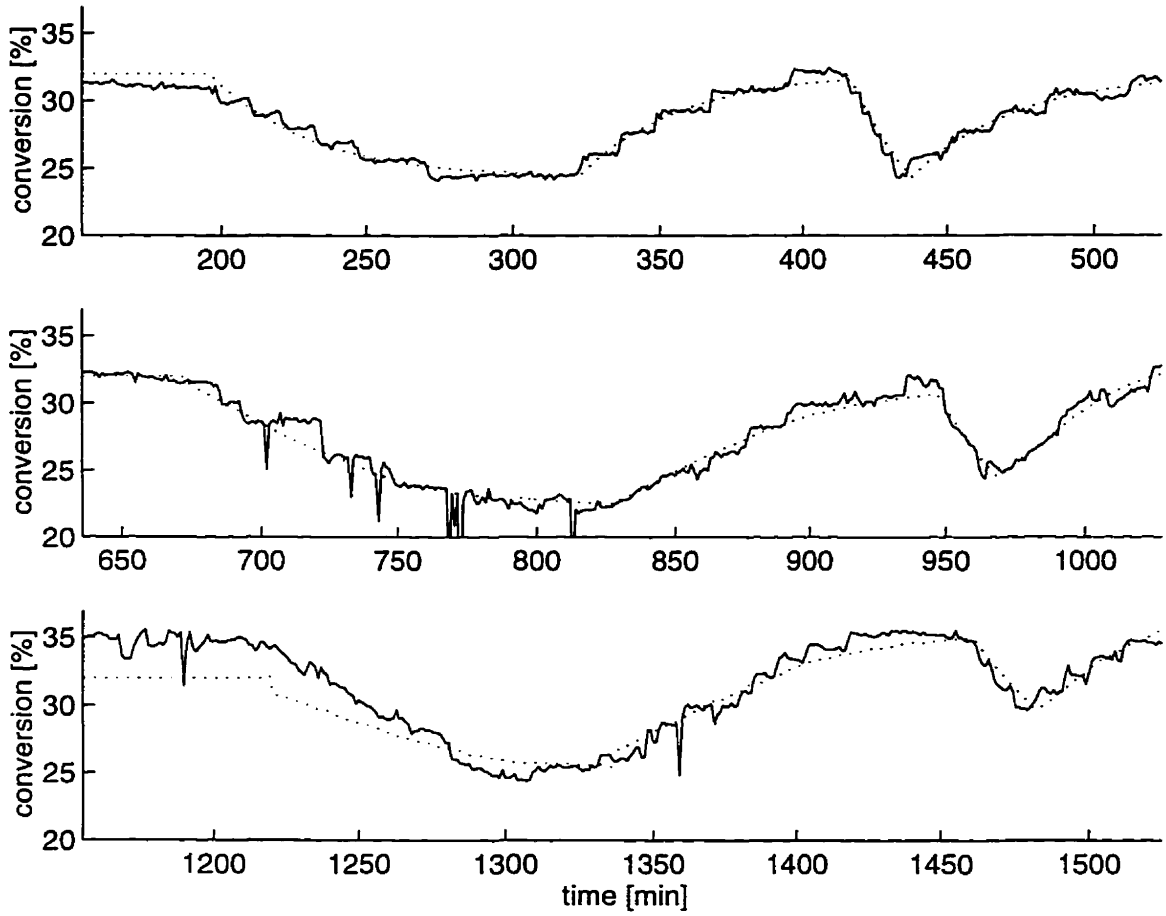


Figure 4.15: Comparison of gain adjusted model with experimental results

(— experimental, ... simulation)

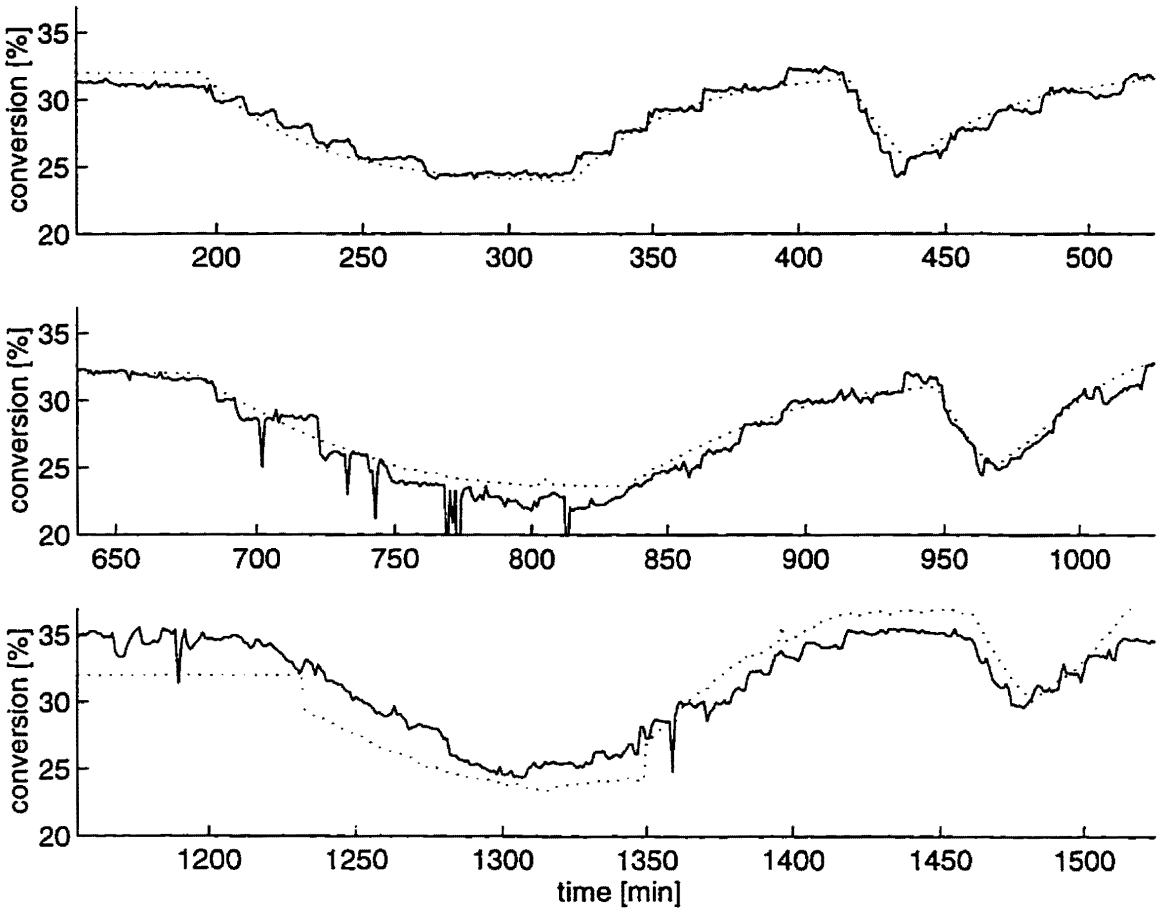


Figure 4.16: Comparison of dead time adjusted model with experimental results

(— experimental, ... simulation)

4.5.3 Quality of Control

As discussed earlier in Section 3.2.1, the controller performance includes stability, steady state, and dynamic response criteria. For the two closed loop mode segments, the process was visibly stable. The steady state criteria, that there should be zero offset from setpoint, was achieved with both the W-C tuned and IMC tuned controller. With W-C tuning, there appears to be a small offset from setpoint, but this can be attributed to conversion measurement error, a slow responding controller, and not allowing the system to settle (for 4~5 time constant intervals) – as was previously identified in Chapter 3 as a possible problem if W-C (α=0.5) is not allowed sufficient time to reach steady state.

For dynamic response performance measures, the same ISE and ITAE measures used in Chapter 3 were applied. However, since there is noise in the measured conversion, this must be taken into consideration when directly comparing with simulation results.

Figure 4.17 shows the closed loop data plots of conversion versus time. The top plot is for W-C tuned control and the bottom is for IMC tuned control. The solid line trends are experimental data, the dotted lines are setpoints, and the vertical lines are demarcation lines between, in order, steady state, setpoint down, setpoint down, and disturbance rejection regions.

Table 4.5 summarizes the ISE and ITAE results.

Integral error for setpoint down and up were averaged, and both controllers were compared with simulations. Figures 4.18 and 4.19 are scatter-plots of the experimental points along with the simulation predictions from Chapter 3. W-C(exp't) is the point for segment two and IMC(exp't) is the point for segment three.

The controllers evaluated by ISE measures had good agreement with simulation predictions. The W-C tuned controller was in the vicinity of W-C (0.5). The IMC tuned controller was close to the IMC (0.9) predicted point.

With ITAE measures, the controllers did not have as good agreement with simulation predictions. Both W-C and IMC tuned controllers had poor performance by ITAE measures.

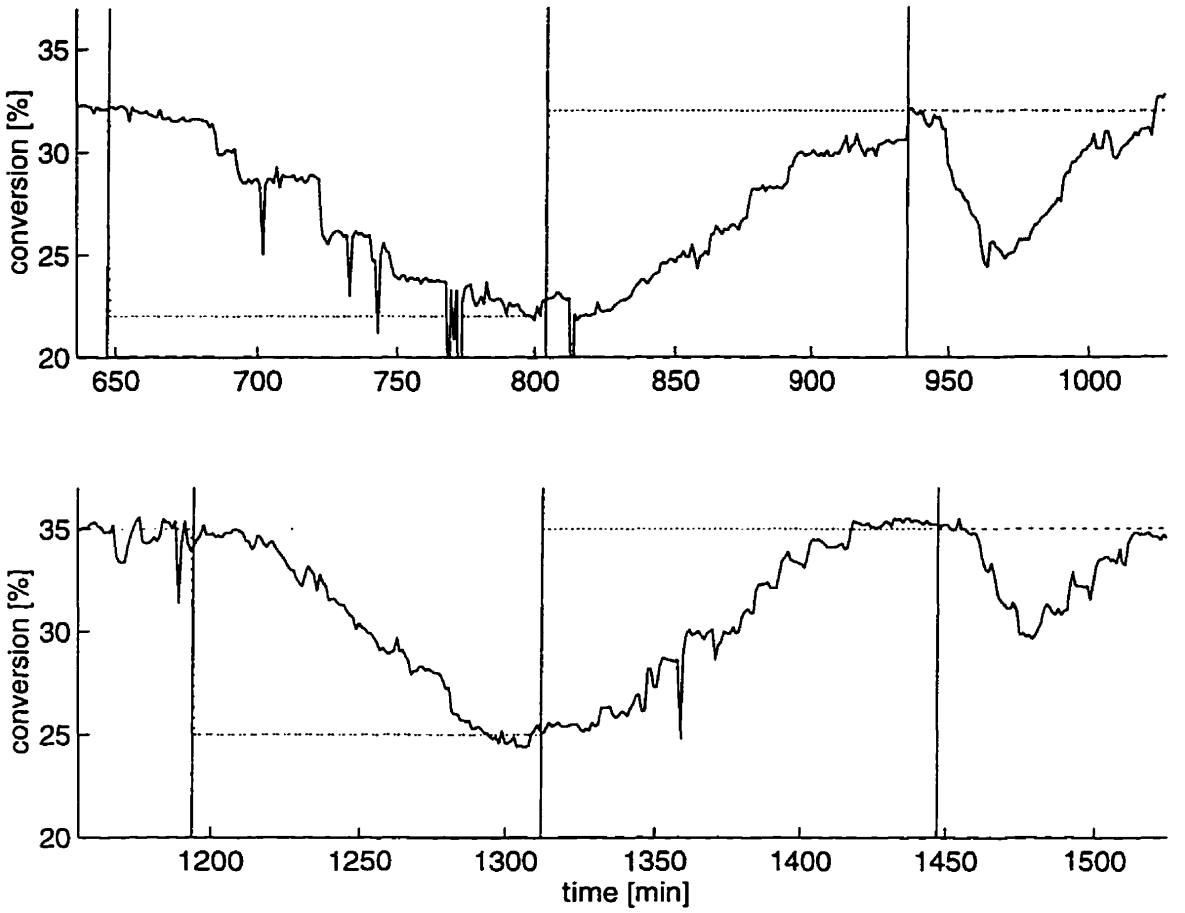


Figure 4.17: Closed loop data regions for performance measure

(— experimental, ... setpoint, | demarcation)

Table 4.5: Closed loop mode experimental performance measures

Controller	Test	minutes	ISE	ITAE
W-C	steady state	12	<1	1
	setpoint down	157	5936	790
	setpoint up	131	5254	718
	disturbance rejection	93	1411	289
	total	393	12600	1799
IMC	steady state	39	29	22
	setpoint up	118	4489	597
	setpoint down	135	4319	591
	disturbance rejection	78	569	163
	total	370	9407	1372

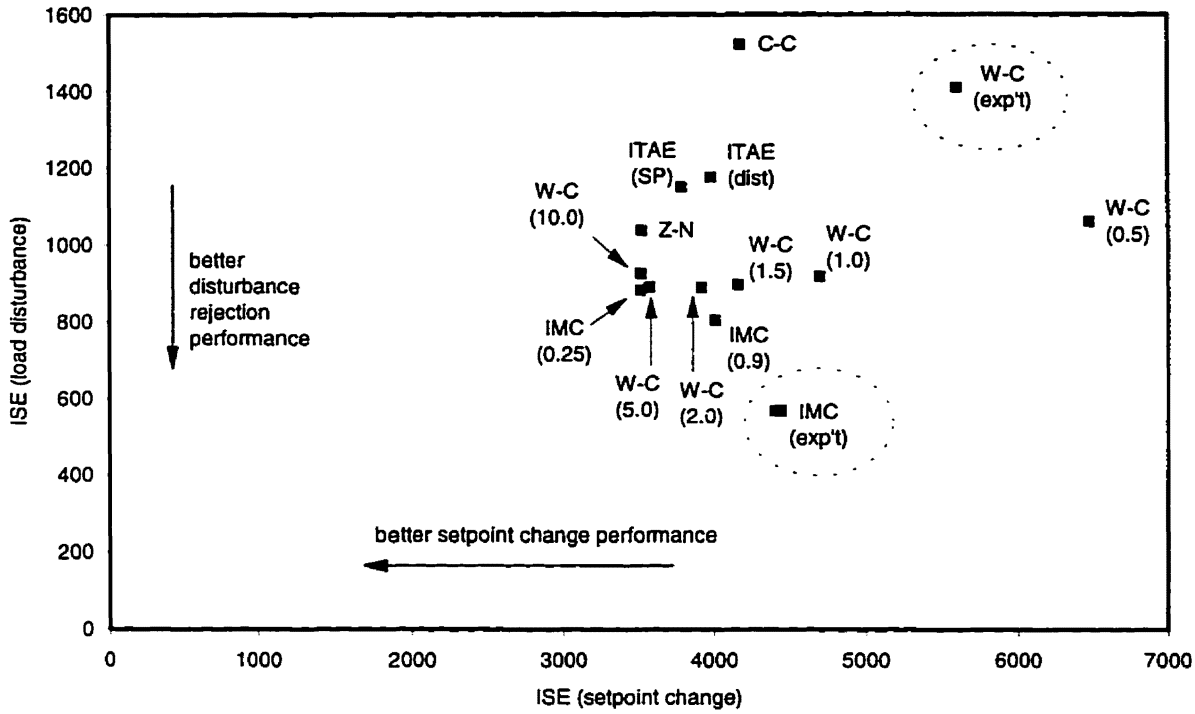


Figure 4.18: Comparison of closed loop controller performance by ISE

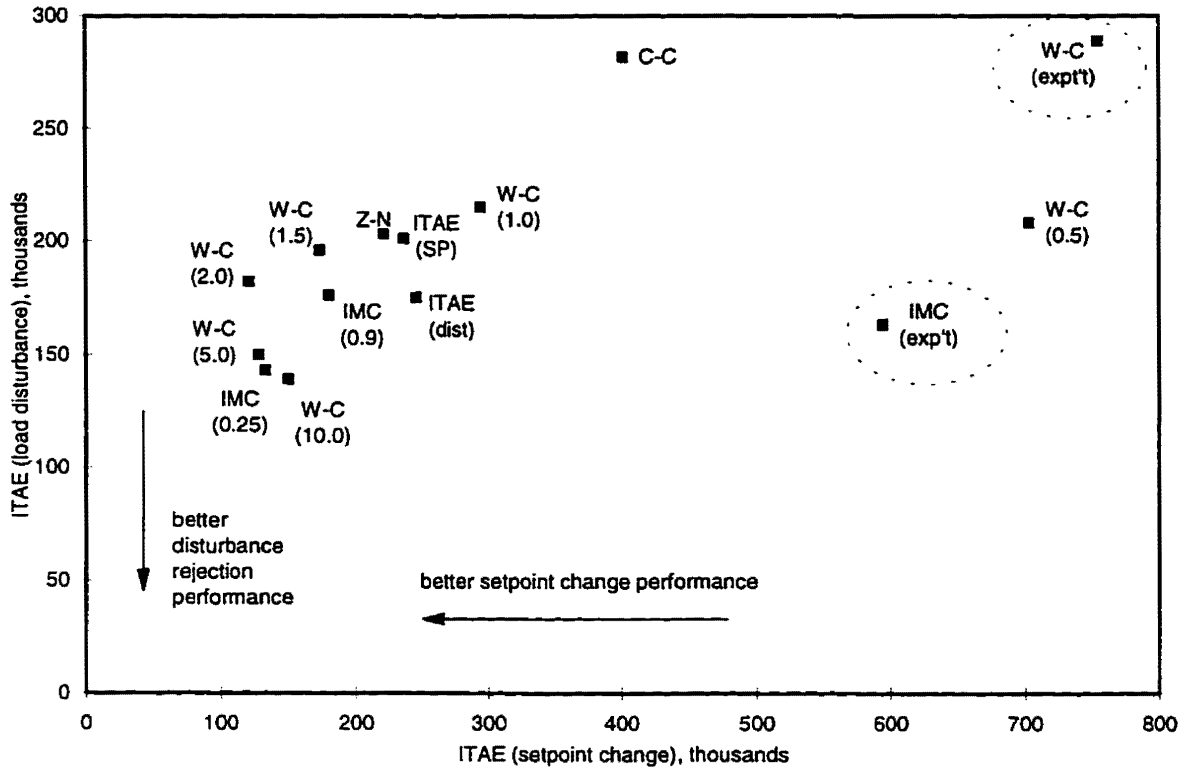


Figure 4.19: Comparison of closed loop controller performance by ITAE

4.5.4 Control Signal Specification

Figure 4.20 compares the simulation prediction for the control signal and the actual experimental AIBN flow rate signal. The top plot is the conversion setpoint (%) and the bottom plot is the AIBN flow rate (ml/min). The simulation for AIBN flow rate was based the W-C (α=0.5) controller regulating the FO+D z-transform model given in Chapter 3. The control signal necessary for the setpoint changes in the top plot were calculated and superimposed on the experimental AIBN flow rate settings. The sudden rise in the experimental AIBN flow rate at about 935 minutes was caused by the experimental system being disturbed by the DPPH injection.

There is very good agreement with the predicted controller output.

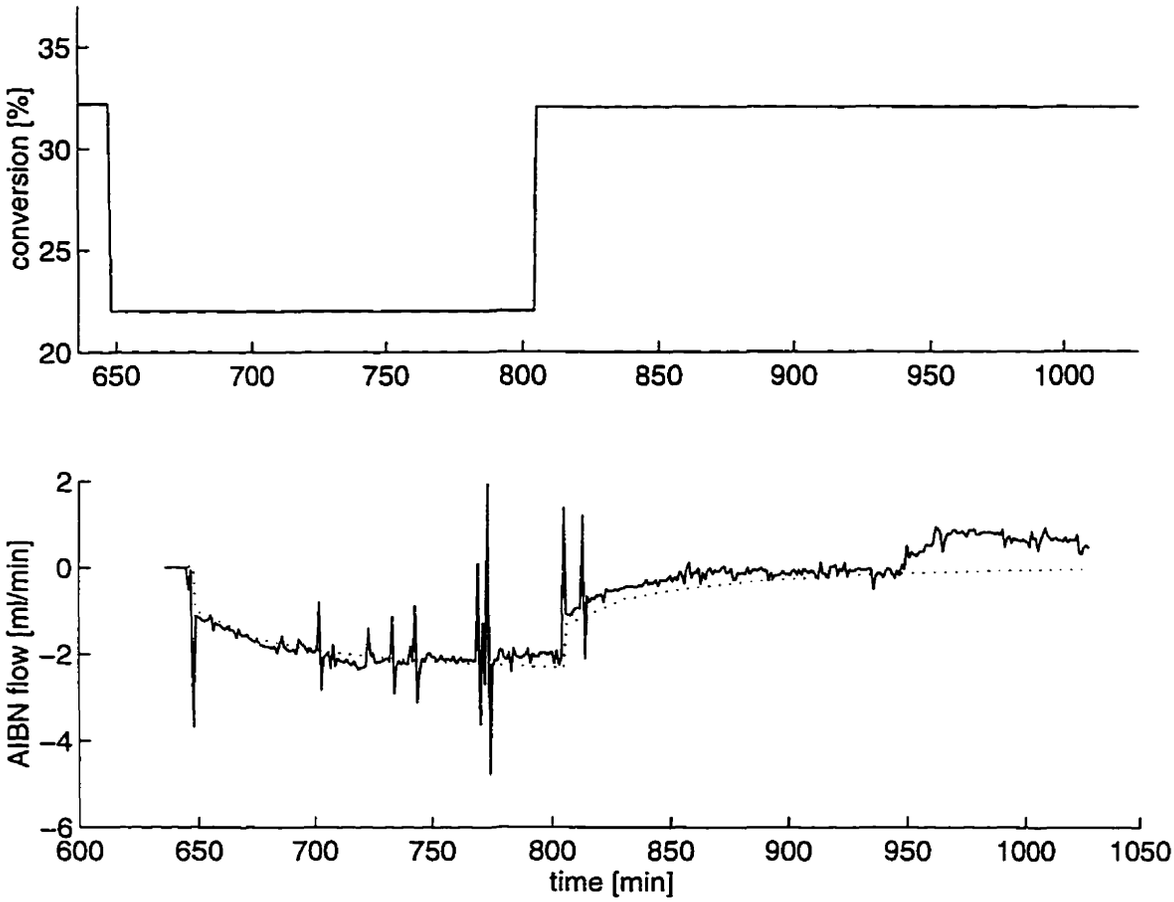


Figure 4.20: Comparison of predicted control signal with experimental results

(— experimental, ... simulation)

4.5.5 Off-line Gravimetry

Off-line gravimetry showed that the conversion estimates from on-line densitometer readings were systematically lower than by gravimetry. Although the accuracy of both the conversion measurements may be in question, the trends are the same and for the objective of controlling conversion to an arbitrary state, had good precision.

4.5.6 Molecular Weight Measurement

Molecular weight measurements from off-line GPC showed that the high/low molecular weight trends were correctly detected by the on-line viscometer. Further work may be required to yield better accuracy in the molecular weight estimates from viscosity measurements. Although Lawrence (1992) conducted extensive system identification in this area, the results had a large variability. Mutha (1996) used an empirical correlation to correct for lower molecular product because his results differed so much from Lawrence. However, his correlation does not work with this data set in improving the agreement between on-line and off-line measurements.

4.6 Concluding Remarks

There was very good agreement between the open loop experimental results with the previously identified z-transform transfer function model of the system by Chien (1992) that related initiator flow rate to monomer conversion in the reactor.

With closed loop control, the model was not as closely matched with the simulations. Refitting a first order plus delay model with the current open loop experimental results, the new model still was unable to predict the closed loop behaviour better than the previous model.

Varying three parameters of the model, mean conversion, overall process gain, and dead time, showed that the reaction rate may have changed during the course of the experimental run, causing a change in the overall process gain and steady state mean conversion. Several factors may have contributed, including variance between initiator batch make-ups and initiator efficiency. Since the experimental run was so long, there were many opportunities for the process to change or the operator to have erred.

Quantifying the of quality of control by integral error performance measures showed that the experimental results were not as good as the simulations. This was expected since the experimental results included noise which would lower the integral error performance.

The W-C tuned controller was easy to tune and behaved as predicted by simulations.

The IMC tuned controller was similarly easy to tune and behaved as predicted by simulations.

CHAPTER 5: CONCLUSIONS AND RECOMMENDATIONS

Some conclusions were drawn from the results of the simulation runs.

1. All the PID controllers tuning methods, W-C (α=0.5~10.0), Z-N, C-C, ITAE (setpoint, disturbance), and IMC (λ/θ=0.25,0.9) produced controllers that met minimum performance criteria for closed loop stability and steady state offset.
2. IMC tuned PID appeared to have better closed loop performance for conversion control than Z-N, C-C, or W-C (α<1.0) tuned versions when compared by ISE and ITAE measures. However, the W-C method had close to the same level of performance as IMC when tuned with α values greater than 1.0.
3. C-C and W-C (α=0.5) tuned controllers stood out as performing worse than most of the other controllers; C-C because it was more aggressive and oscillated the output signal, and W-C (α=0.5) because it was slower approaching a new setpoint or returning to a setpoint after a disturbance.

From the on-line experimental results, a number of conclusions were drawn.

1. Predictions using the z-transform model of CSTR process and open loop experimental results were in good agreement. The assumption that the process had not significantly changed since Chien (1992) was validated. The significance was that Chien's experimental runs testing other PID controllers (Smith predictor, Dahlin's control) could be directly compared with data from this experimental run.
2. The W-C (α=0.5) tuned controller performed as expected during the experimental run, with good agreement with simulation predictions. This included the prediction that the system would not be at steady state, even after 250 minutes, still exhibiting a slight offset from the desired setpoint as the controller was slow to react.
3. The controller output signal for the W-C (α=0.5) tuned controller was almost identical to that predicted by simulations based solely on the setpoint changes and first order plus delay (FO+D) transfer function model. This result verifies that, with the W-C tuning method using a FO+D model, the controller output signal could be used accurately for the specification for the desired level of performance.
4. IMC tuned PID control did not perform as closely to its simulation predictions as W-C (α=0.5) had done to its own. The process output was not as accurately predicted by the transfer function model, and the controller output had many, regular spikes. An aggressively tuned controller was expected to

perform with a noisy controller output as previously seen in Chien (1992). However, performance was worse than expected when compared using ITAE measures. Process drift may have been a cause.

5. On-line conversion measurements were systematically lower than off-line gravimetry measurements taken after the experimental run. Samples had been regularly collected during the experimental run, and analyzed days later.

6. On-line molecular weight estimates were about half of the measured weight average molecular weights calculated off-line using GPC. Samples collected during the experimental run were inhibited and analyzed days later.

Recommendations for future work include the following.

1. Since the W-C tuning method has been verified to work as specified, it could be used to tuned the controller for better performance. The experimental work was not to compare W-C against IMC tuned controllers' performance. However, that would be a logical next step since the experimental verification was successful. Some simulations had shown much better performance. Further runs could show what is the best possible performance given the noisy conditions.

2. Extending the control study to higher levels of conversion, where diffusional limitations become more significant, has been recommended by previous researchers. However, in order to deal with the non-linearities and ensure safe operation, tighter, guaranteed control would be desirable. Having a well understood PID controller with predictable behaviour and reliability would be required.

3. The IMC and W-C tuned controllers were run at different steady state conversions. The process may have different gains at these levels and caused the IMC tuned controller to behave noisily. More runs at the other conversions could verify this.

The following are suggestions for consideration before future work, based on experience gained during this thesis work.

1. The review of polymer reactor control had a large number of references, including many articles that had relevance in several areas of study. This made it difficult to categorize them. Organization into a general database format, with searchable indices and continual updates would be useful. Since many search indices are already available, having general interfaces to convert information from others would be advantageous.

2. Although Mutha (1996) developed NL-MPC control program that runs in a "real-time" UNIX environment, the use of a proprietary operating system is not necessarily better. Problems coding control programs and interfaces (man-machine and PC-process) were often a major problem encountered here. The NL-MPC algorithm could be included in the current, easier to understand, QuickBASIC program or in a similarly simple development environment.

APPENDIX A: ABBREVIATIONS AND NOMENCLATURE

A.1 General Abbreviations and Acronyms

Abbreviation	Full Name
ANN	artificial neural network
C-C	Cohen-Coon (tuning method)
CCD	chemical composition distribution
CLD	chain length distribution
CSTH	continuous stirred tank heater
CSTR	continuous stirred tank reactor
DAE	differential algebraic equations
DLS	dynamic light scattering
DMC	dynamic matrix control
EKF	extended Kalman filter
ESTR	extended self-tuning regulator
FB	feed back
FF	feed forward
FFT	fast Fourier transform
FO+D	first order plus delay or deadtime (process)
GC	gas chromatography
GLC	global linearizing control
GPC	gel permeation chromatography
HDC	hydrodynamic chromatography
I+FO	integrating plus first order (process)
IAE	integral absolute error
IMA	integrated moving average (process)
IMC	internal model control (tuning method)
ISE	integral square error
ITAE	integral time-weighted absolute error
ITSE	integral time-weighted squared error
KF	Kalman filter
LALLS	low angle laser light scattering
LCB	long chain branching
LO	Luenberger observer
LQC	linear quadratic controller
MHE	moving horizon estimator
MI	melt index
MIMO	multiple input multiple output
MW	molecular weight
MWD	molecular weight distribution
M_n	number average molecular weight
M_w	weight average molecular weight
NACL	number average chain length
NCLD	number chain length distribution
NIR	near-infared reflectance spectroscopy
NL	nonlinear
NL-PCA	nonlinear principal component analysis
NN	neural network
NMR	nuclear magnetic resonance
ODE	ordinary differential equations

Abbreviation	Full Name
PCA	principal component analysis
PDI	polydispersity index
PFR	plug flow reactor
PI	proportional integral (controller)
PID	proportional integral derivative (controller)
PRBS	pseudo-random binary sequence
PSD	particle size distribution
QC	quality control
RNN	recurrent neural net
SEC	size exclusion chromatography
SISO	single input single output
SPC	statistical process control
SS	steady-state
T_g	glass transition temperature
UNIPOL	polyethylene production process (Union Carbide)
W-C	Wang and Cluett tuning method
WCLD	weight chain length distribution
Z-N	Ziegler-Nichols tuning method

A.2 Initiator, Monomer, Polymer, and Other Chemical Species' Abbreviations

Abbreviation	Full Name
ACN	acrylonitrile
AIBN	2,2'-azobisisobutyronitrile
AVN	2,2',4,4'-tetramethyl-2,2'-azovaleronitrile
BDN	butadiene
BPO	benzoyl peroxide
BuA	butyl-acrylate
BUAc	n-butyl acrylate
DPPH	2,2-diphenyl-1-picrylhydrazyl hydrate
EHA	2-ethylhexyl acrylate
EPM	ethylene/propylene monomer
EPDM	ethylene/propylene/butadiene monomer
HIPS	high impact polystyrene
LDPE	low density polyethylene
LLDPE	linear low density polyethylene
LPO	dilauroylperoxide
MEHQ	mono methyl ether hydroquinone
MMA	methyl methacrylate
MSTY	p-methylstyrene
NBR	(acrylonitrile) nitrile-butadiene rubber
NMR	nuclear magnetic resonance (imaging)
PE	polyethylene
PET	poly(ethylene terephthalate)
PMMA	polymethyl methacrylate
PP	polypropylene
PS	polystyrene
PVC	polyvinyl chloride
SAN	styrene-acrylonitrile copolymer
SB	styrene-butadiene latex
SBR	styrene/butadiene rubber
STY	styrene
VA	vinyl acetate
VC	vinyl chloride
VCLAC	vinyl chloroacetate
Z-N	Ziegler-Natta (catalyst)

APPENDIX B: PROGRAM LISTINGS

A copy of the QuickBASIC control program can be obtained from:

Professor A. Penlidis
Department of Chemical Engineering
University of Waterloo
Waterloo, Ontario
Canada N2L 3G1

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