



University of Bradford eThesis

This thesis is hosted in [Bradford Scholars](#) – The University of Bradford Open Access repository. Visit the repository for full metadata or to contact the repository team



© University of Bradford. This work is licenced for reuse under a [Creative Commons Licence](#).

DYNAMIC OPTIMISATION AND CONTROL OF BATCH REACTORS

Development of a general model for batch reactors, dynamic optimisation of batch reactors under a variety of objectives and constraints and on-line tracking of optimal policies using different types of advanced control strategies

Norashid AZIZ BEng (Hons)

submitted for the degree
of Doctor of Philosophy

Department of Chemical Engineering

University of Bradford

2001

Norashid Aziz

DYNAMIC OPTIMISATION AND CONTROL OF BATCH REACTORS

KEYWORDS: Batch reactors, Modelling, Dynamic Optimisation, On-line Tracking, Control Vector Parameterisation, Generic Model Control, Neural Network Techniques.

ABSTRACT

Batch reactor is an essential unit operation in almost all batch-processing industries. Different types of reaction schemes (such as series, parallel and complex) and different order of model complexity (short-cut, detailed, etc.) result in different sets of model equations and computer coding of all possible sets of model equations is cumbersome and time consuming. In this work, therefore, a general computer program (GBRM – General Batch Reactor Model) is developed to generate all possible sets of equations automatically and as required. GBRM is tested for different types of reaction schemes and for different order of model complexity and its flexibility is demonstrated. The above GBRM computer program is lodged with Dr. I.M. Mujtaba.

One of the challenges in batch reactors is to ensure desired performance of individual batch reactor operations. Depending on the requirement and the objective of the process, optimisation in batch reactors leads to different types of optimisation problems such as maximum conversion, minimum time and maximum profit problem. The reactor temperature, jacket temperature and jacket flow rate are the main control variables governing the process and these are optimised to ensure maximum benefit. In this work, an extensive study on mainly conventional batch reactor optimisation is carried out using GBRM coupled with efficient DAEs (Differential and Algebraic Equations) solver, CVP (Control Vector Parameterisation) technique and SQP (Successive Quadratic Programming) based optimisation technique. The safety,

environment and product quality issues are embedded in the optimisation problem formulations in terms of constraints. A new approach for solving optimisation problem with safety constraint is introduced. All types of optimisation problems mentioned above are solved off-line, which results to optimal operating policies.

The off-line optimal operating policies obtained above are then implemented as set points to be tracked on-line and various types of advanced controllers are designed for this purpose. Both constant and dynamic set points tracking are considered in designing the controllers. Here, neural networks are used in designing Direct Inverse and Inverse-Model-Based Control (IMBC) strategies. In addition, the Generic Model Control (GMC) coupled with on-line neural network heat release estimator (GMC-NN) is also designed to track the optimal set points. For comparison purpose, conventional Dual Mode (DM) strategy with PI and PID controllers is also designed. Robustness tests for all types of controllers are carried out to find the best controller. The results demonstrate the robustness of GMC-NN controller and promise neural controllers as potential robust controllers for future. Finally, an integrated framework (BATCH_REACT) for modelling, simulation, optimisation and control of batch reactors is proposed.

ACKNOWLEDGEMENTS

First and foremost, I would like to thank my supervisor Dr. I.M. Mujtaba for his valuable guidance, encouragement and support throughout this work.

I would also like to thank Dr. M.A. Hussain, University of Malaya, Malaysia for his inspiration and help in learning and using Neural Network Techniques in this work.

The financial support from University Sains Malaysia is gratefully acknowledged.

Special thanks to the staffs, students and friends that I have met during my research work especially those are in the Computational Process Engineering Group. A few to mention: Shebl, Austin, Michael, Adel, Apornchai and Abu Baker.

Lastly but most importantly, I would like to thank my parents for their unwavering love, continuous guidance, care and support. My thanks also to all my sisters and brothers for their continuous encouragements.

Table of Contents

<i>CONTENTS</i>	<i>PAGE</i>
Abstract	i
Acknowledgements	iii
List of Tables	viii
List of Figures	ix
Nomenclature	xii
Abbreviations	xv
1.0 Introduction	1
1.1 Aims of this Work	5
1.2 Outlines of the Thesis	6
2.0 Literature Review	8
2.1 Dynamic Optimisation of Batch Reactors	8
2.2 Solution Methods of Dynamic Optimisation Problems	15
2.3 On-line Tracking of Optimal Batch Reactors Profiles (Controller Design)	20
2.4 Modelling of Batch Reactors	28
2.5 Concluding Remarks	30
3.0 Dynamic Modelling	32
3.1 Introduction	32
3.2 General Batch Reactors Model (GBRM)	35
3.2.1 Mass Balance	36

3.2.2	Reaction Rate	37
3.2.3	Energy Balance	38
3.2.4	Model Equations and Degrees of Freedom Analysis	40
3.2.5	Examples	48
3.2.5.1	Consecutive reaction	48
3.2.5.2	Parallel reaction	50
3.2.5.3	Consecutive and reversible reaction	51
3.2.5.4	Complex reaction	53
3.2.5.5	Consecutive and parallel reaction	55
3.2.6	Simulation Results	58
3.3	Summary and Conclusions	60
4.0	Dynamic Optimisation (Optimal Control)	61
4.1	Introduction	61
4.2	Classical Formulation of Dynamic Optimisation (Optimal Control) Problems	64
4.3	Solution of Dynamic Optimisation Problems	65
4.3.1	Formulation of Optimal Control Problem as a Non-linear Programme	65
4.3.2	Control Vector Parameterisation (CVP) Technique	66
4.4	Optimisation Problem Formulation in Batch Reactors	67
4.5	Case Studies	71
4.5.1	Case Study 1	72
4.5.1.1	Model Equations	73
4.5.1.2	Degrees of Freedom Analysis	75
4.5.1.3	Results	75
4.5.2	Case Study 2	80
4.5.2.1	Models Equations	81
4.5.2.2	Degrees of Freedom Analysis	84
4.5.2.3	Results	85
4.5.3	Case Study 3	89
4.5.3.1	Optimisation Problem Formulation	92

4.5.3.2	Path Constraints in Batch Reactors	93
4.5.3.3	Models Equations	99
4.5.3.4	Degrees of Freedom Analysis	100
4.5.3.5	Results	101
4.5.4	Case Study 4	111
4.5.4.1	Optimisation Formulation of Semi-Batch Reactors	113
4.5.4.2	Models Equations	115
4.5.4.3	Degrees of Freedom Analysis	116
4.5.4.4	Results	117
4.5.4.5	Concluding Remark	121
4.5.5	Case Study 5	121
4.5.5.1	Profit Function	122
4.5.5.2	Descriptions of Different Problems	123
4.5.5.3	Results	124
4.5.6	Case Study 6	128
4.5.6.1	Model Equations	129
4.5.6.2	Degrees of Freedom Analysis	130
4.5.6.3	Results	131
4.6	Summary and Conclusions	132
5.0	Control (On-line Tracking of Optimal Profiles)	134
5.1	Introduction	134
5.2	Generic Model Control (GMC) Strategy	135
5.2.1	Theoretical Formulation	135
5.2.2	On-Line Estimation of the Heat-Release using Neural Network Techniques (GMC Strategy)	138
5.3	Neural Controller	145
5.3.1	Forward Model	145
5.3.2	Inverse Model	147
5.3.3	Direct Inverse Control Technique	148
5.3.4	Inverse-Model-Based Control Strategy (IMBC)	149
5.4	Dual-Mode (DM) Control Strategy	152

5.5	Applications (On-line tracking of optimal profiles)	153
5.5.1	Case Study 1	154
5.5.1.1	Models Equations	154
5.5.1.2	Results	157
5.5.2	Case Study 2	164
5.5.2.1	Model Equations	164
5.5.2.2	Results	166
5.5.3	Remarks from the Applications (Case Study 1 and 2)	178
5.6	Conclusions	182
6.0	BATCH_REACT: An Integrated Framework for Modelling, Simulation, Optimisation and Control of Batch Reactors	184
6.1	Overview	184
6.2	Four-Levels BATCH_REACT Framework	185
6.3	Steps to Follow by the User	187
6.4	Current Limitations of the Proposed BATCH_REACT	189
6.5	Future Development	189
7.0	Conclusions and Future Work	190
7.1	Conclusions and Contributions	190
7.2	Suggestions for future work	193
	References	195
	Appendices	
	Appendix A	207
	Appendix B	212

List of Tables

<i>TABLES</i>	<i>PAGE</i>
Table 2.1: Summary of the past work on batch reactor optimisation	14
Table 2.2: Summary of the past work on batch reactor control	27
Table 2.3: Review on modelling of conventional batch reactors	29
Table 3.1: Test results from GBRM	58
Table 3.2: Simulation results achieved from GBRM with different β_{ij} and ν_{ji} values	59
Table 4.1: Summary of the results (Problem P1)	76
Table 4.2: Summary of the results (Problem P2)	79
Table 4.3: Summary of the results	85
Table 4.4: Summary of the results	89
Table 4.5: Summary of the results – Maximum conversion problem	103
Table 4.6: Summary of the results – Minimum time problem	108
Table 4.7: Summary of the results for maximum conversion problem	118
Table 4.8: Summary of the results for minimum time problem	119
Table 4.9: Summary of the results for maximum profit problem	122
Table 4.10: Summary of results for Case Study 5	120
Table 4.11: Summary of the Results for Case Study 6	131
Table 5.1: The constant parameter values of the model and control equation	155
Table 5.2: Summary of the result for Case Study 1	156
Table 5.3: The constant parameter values of the model and control equation	165
Table 5.4: Summary of the results for Case Study 2	167

List of Figures

<i>FIGURES</i>	<i>PAGE</i>
Figure 3.1: Schematic diagram of jacketed batch reactor	35
Figure 4.1: Computational Sequence of Dynamic Optimisation Problem	71
Figure 4.2: Conversion and optimal temperature profiles for Run 2 (P1 problem)	77
Figure 4.3: Conversion and optimal temperature profiles for Run 4 (P1 problem)	78
Figure 4.4: Conversion and optimal temperature profiles for Run 4 (P2 problem)	80
Figure 4.5: Conversion profiles for Run 2a, 2b and 2c	87
Figure 4.6: Reactor temperature profiles for Run 2a, 2b and 2c	88
Figure 4.7: Typical reactor temperature profile	94
Figure 4.8: Violation during the reaction process	95
Figure 4.9: Reactor temperature and optimal coolant flow profiles for Run 1	104
Figure 4.10: Reactor temperature and optimal coolant flow profiles for Run 2 (P1)	104
Figure 4.11: Reactor temperature and optimal coolant flow profiles for Run 3 (P1)	105
Figure 4.12: Reactor temperature and optimal coolant flow profile for Run 4 (P1)	106
Figure 4.13: Reactor temperature and optimal coolant flow profiles for Run 5 (P1)	107
Figure 4.14: Conversion and temperature profiles for Runs 4 and 5 (P1)	107
Figure 4.15: Reactor temperature and optimal coolant flow profiles for Run 1 (P2)	109
Figure 4.16: Reactor temperature and optimal coolant flow profiles for Run 4 (P2)	109
Figure 4.17: Reactor temperature and optimal coolant flow profiles for Run 5 (P2)	110
Figure 4.18: Conversion and temperature profiles for Runs 4 and 5 (P2)	110
Figure 4.19: Schematic diagram of a jacketed semi-batch reactor	113
Figure 4.20: Percentage improvement in profit P_{3f} compared to P_{1f}	126

Figure 4.21: Percentage improvement in profit for various cases over the profit obtained in Case 2.	127
Figure 5.1: Neural network systems identification – Basic steps	139
Figure 5.2: Multi-layered feed forward neural network topology	140
Figure 5.3: Input/output map of neural network estimator	142
Figure 5.4: Forward modelling of heat-release by neural network	142
Figure 5.5: GMC strategy in controlling batch reactor	143
Figure 5.6: Input/output patterns for forward model in IMBC strategy	146
Figure 5.7: Method for the training of the inverse model for control	147
Figure 5.8: Input/output patterns for inverse model	148
Figure 5.9: Direct inverse control strategy	150
Figure 5.10: Inverse-model-based control (IMBC) strategy	151
Figure 5.11: GMC response (Case Study 1)	157
Figure 5.12: Performance of heat-release estimator (Case Study 1)	158
Figure 5.13: PID response (Case Study1)	159
Figure 5.14: Controllers responses for heat reaction change (Case Study 1)	160
Figure 5.15: Heat release estimator performance for heat reaction change (Case Study 1)	160
Figure 5.16: Controllers responses for rate constant change (Case Study 1)	161
Figure 5.17: Heat release estimator performance for rate constant change (Case Study 1)	161
Figure 5.18: Controllers responses for weight change (Case Study 1)	162
Figure 5.19: Heat release estimator performance for weight change (Case Study 1)	162
Figure 5.20: Controllers responses for heat transfer coefficient change (Case Study 1)	163
Figure 5.21: Heat release estimator performance for heat transfer coefficient change (Case Study 1)	163
Figure 5.22: GMC response for 1 Tsp (Case Study 2 - RUN1)	168
Figure 5.23: Performance of heat-release estimator (Case Study 2 - RUN1)	168
Figure 5.24: GMC response for dynamic set points (Case Study 2 - RUN2)	169
Figure 5.25: Performance of heat-release estimator (Case Study 2 - RUN2)	169
Figure 5.26: PI response for 1 Tsp (Case Study 2 - RUN1)	171

Figure 5.27: PI response for dynamic set points (Case Study 2 - RUN2)	171
Figure 5.28: PID response for 1 Tsp (Case Study 2 - RUN1)	172
Figure 5.29: PID response for dynamic set points (Case Study 2 - RUN2)	172
Figure 5.30: Direct Inverse response for 1 Tsp (Case Study 2 - RUN1)	173
Figure 5.31: Direct Inverse response for dynamic set points (Case Study 2 - RUN2)	174
Figure 5.32: IMBC response for 1 Tsp (Case Study 2 - RUN1)	174
Figure 5.33: IMBC response for dynamic set points (Case Study 2 - RUN2)	175
Figure 5.34: Controllers responses for heat reaction change (Case Study 2) - I	179
Figure 5.35: Controllers responses for heat reaction change (Case Study 2) - II	179
Figure 5.36: Controllers responses for heat transfer coefficient change (Case Study 2) - I	180
Figure 5.37: Controllers responses for heat transfer coefficient change (Case Study 2) - II	180
Figure 5.38: Controllers responses for molar/weight change (Case Study 2) - I	181
Figure 5.39: Controllers responses for molar/weight change (Case Study 2) - II	181
Figure 6.1: Four Level BATCH_REACT Framework	185
Figure 6.2: Flow diagram of the BATCH_REACT	188

Nomenclature

A	Heat transfer area
A_i	Inside heat transfer area
A_o	Outside heat transfer area
CC	Capital cost
C_i	Concentration of component i
C_{i0}	Inlet concentration of component i
c_p	Specific heat capacity of reactor contents
c_{pi}	Specific heat capacity of component i
c_{pJ}	Specific heat of coolant/heating element in jacket
c_{pm}	Specific heat of metal (wall)
E_i	Activation energy for reaction j
$E_m, TD-1, TD-2, PL$	Tuning parameters in dual mode (DM) control strategy
F_J	Jacket flow rate
k	Time step k
K_1, K_2	GMC controller constants
k_j^1	'Pre-exponential' rate constant for reaction j
k_j^2	Activation energy for reaction j
K_c, τ_I, τ_D	Tuning parameters in PID/PI controller
k_j	Reaction rate constant for reaction j
k_{j0}	Reaction rate constant for reaction j at reference temperature
M_i	Amount of component i in mole
O_pC	Operating cost
P	Profit
PF_J	Coolant cost
P_i	Price of component i
Q_J	Amount of heat transferred to jacket
Q_m	Amount of heat transferred to metal (wall)
Q_r	Heat released by reaction
R	Universal gas constant
R_i	Overall production rate of the component C_i

r_j	Reaction rate of reaction j
T	Reactor (reactant mixture) temperature
t	Time
t_f	Batch or final time
T_J	Jacket temperature
T_{J0}	Inlet jacket temperature
T_m	Temperature of the reactor metal (wall)
U	Overall heat transfer coefficient
$u(k)$	Control variable at time step k
$u(t)$	Control variable at time t
U_i	Inside heat transfer coefficient
U_o	Outside heat transfer coefficient
V	Volume of reactor contents
V_J	Volume of jacket
V_m	Volume of reactor wall
V_r	Volume of reactor
W_r	Total weight of reactant
x_i	Mole fraction of component i in the system
z^{-l}	Past value
z^{-k}	Value at past time step k

Greeks letter

ΔH_{rj}	Heat of reaction for reaction j
ρ	Density of reactor contents
ρ_m	Density of metal (wall)
ρ_J	Density of jacket element
ν_{ji}	Stoichiometric coefficient of component i in reaction j
Δt	Sample interval
β_{ij}	Reaction rate order of component i in reaction j
τ	Time constant
ν_i	Feeding rate of reactant i

Superscripts

*	Fixed value
'	Derivative value

Subscripts

<i>avg</i>	Average
<i>f</i>	Final
<i>i</i>	Component <i>i</i>
<i>J</i>	Jacket
<i>j</i>	Reaction <i>j</i>
<i>m</i>	Metal (wall)
<i>max</i>	Maximum
<i>min</i>	Minimum
<i>r</i>	Reactant
<i>sp</i>	Set point

Abbreviations

CP	Controller performance
CVP	Control vector parameterisation
D.F.	Degrees of freedom
DAEs	Differential algebraic equations
DM	Dual mode
DMP	Discrete maximum principle
DP	Desired product
GBRM	General batch reactors model
GMC	Generic model control
GMC-NN	Generic model control coupled with neural network
GRG	Generalised reduced gradient
GS	Golden search
IDP	Iterative dynamic programming
IMBC	Inverse-model-based control
IMC	Internal model control
MSE	Mean squared error
NLP	Non-linear programming
NN	Neural network
OC	Orthogonal collocation
ODEs	Ordinary differential equations
P1	Maximum conversion problem
P2	Minimum time problem
P3	Maximum profit problem
PARSEX	Partially simulated exothermic reactor
PF1	Profit function 1
PF2	Profit function 2
PI	Proportional-integral
PID	Proportional-integral-derivatives
PMP	Pontryagin's maximum principle
QP	Quadratic programming
RC1	Mettler reaction calorimeter

SLP	Successive linear programming
SQP	Successive quadratic programming
SRQPD	Successive reduced quadratic programming
SSE	Sum squared error
TNE	Total number of equations
TNV	Total number of variables

Chapter One

1.0 Introduction

Batch reactor is an essential unit operation in almost all batch-processing industries. It is used for small-scale operation, for testing new processes that have not been fully developed, for the manufacture of expensive products and for processes that are difficult to convert to continuous operations (Fogler, 1992). In batch reactor, there is no inflow or outflow of reactants or products while the reaction is being carried out. Here, the reactants are initially charged into a container, are well mixed and are left to react for a certain period. The resultant mixture is then discharged.

In the 1950s students of chemical reaction engineering might well have gained the impression that the ultimate mission of the chemical engineer was to transform old-fashioned batch processes into modern continuous ones (Rippin, 1983). With such a perspective it would be surprising to find that, today, fifty years later a significant proportion of the world's chemical production by volume and a much larger proportion by value is still made in batch plants and it does not seem likely that this proportion will decline. Moreover, many more products, which could be manufactured continuously, are in fact made in batch plants on economic grounds.

Batch production is usually carried out in relatively standardised types of equipment, which can easily be adapted and if necessary reconfigured to produce many other different products. This type of reactor can achieve high conversion with long batch time but may require high operating cost per unit production. Therefore, it is not feasible for large-scale production. It is particularly suitable for low volume, high value products such as pharmaceuticals, polymers, biotechnological or other fine chemicals

products for which annual requirement can be manufactured in a few days or a few batches in an existing plant. The flexibility of the production arrangements can also cope with the fluctuations or rapid changes in demand, which is often characteristic of products of this type. Batch reactors are also used when there are many processing steps in chemical process, when isolation is required for reasons of sterility or safety and when materials involved are hard to handle (Palanki et al., 1993).

Even though the optimisation and control of batch reactors has become the research of interest since a few decades but it still remains a real engineering challenge for process control engineers because of certain technical and operational conditions (Bonvin, 1998):

1. Time-varying characteristics. There is no steady state operation; chemical transformations proceed from initial state to a very different final state. Even at the constant reactor temperature, the concentrations and the reaction rates change significantly over the duration of the batch run. Therefore, there is no single operating point around which the control system can be designed.
2. Non-linear behaviour. It might originate from two main sources: (i) the non-linear dependency of the reaction rates on concentrations and temperature and (ii) the non-linear relationship between the heat transfer from the reactor to the jacket and the flow rate of the heating/cooling medium in jacket.
3. Model inaccuracies. The development of reliable models for industrial process is very time consuming. The completion of this task is often against the purpose of batch processing i.e. a short time-to-market.
4. Few specific measurements. Chemical composition is usually determined off-line. The on-line specific chemical sensors are still rare although reliable and

relatively inexpensive spectroscopic sensors have become available commercially in recent years.

5. **Constrained operation.** The presence of process or equipment constraints complicates the design of a simple operational strategy for at least two reasons:
(i) if the process is linear or has been linearised along a reference trajectory, constraint will make it non-linear and (ii) controllability might be lost when a manipulated variable hits a constraint.
6. **Presence of disturbances.** Operator mistakes (e.g. wrong stirrer or solvent choice) and processing problems (e.g. fouling of sensors and reactor walls, insufficient mixing, incorrect temperature of feeding profiles) represent major disturbances that cannot be totally excluded.

Due to these several characteristics, one of the challenges in batch reactors is to ensure the performance of individual batch reactor operations. The first step to rational design of a batch chemical reactor generally consists of guaranteeing safety and operability objectives. Once these goals are achieved, the next step is to make its operation profitable. The aim of fine chemical industries is to produce high quality and purity product, in general small amount of products with a control of polluting waste materials and losses of raw material. Therefore an optimisation of batch operating conditions such as temperature, operating time, etc. is the more efficient approach to obtain a maximum yield in a minimum time or minimum cost, as well as to reach the specific final conditions of the product in terms of quality and quantity. Therefore, operating batch reactors efficiently and economically is very important as far as the overall profitability is concerned. Because of the necessity to answer to strict constraints

and objectives, the optimisation problems encountered in fine chemical industries are very complex.

The main objective of dynamic optimisation (optimal control) of batch reactors in the past was to determine the optimum reactor temperature profiles, which would maximise or minimise the objective function or index of performance. The dynamic optimisation problem considered was either maximum conversion, minimum time or maximum profit problems (Rippin, 1983; Terwiesch et al., 1994; Bonvin, 1998). However, most of the work reported were very simple and implemented simple model to represent the system under consideration which might not be feasible for the real application.

The control of a batch reactor in a simple case consists of charging the reactor, controlling the reactor temperature to meet some processing criterion and shutting down and emptying the reactor. For an exothermic reaction, heat may be required to obtain the desired reaction temperature, and then cooling is used to maintain the proper reaction temperature. An excellent review on this subject can be found in Juba and Hamer (1986) and Berber (1996).

Although the number of the work on optimisation and control of batch reactors is large but the work on conventional general batch reactor still relatively low. Here conventional general batch reactor refers to all batch reactors excluding semi-batch, polymerisation and fermentation reactors. From now on, the 'batch reactor' will refer to conventional general batch reactor.

All the work reported previously have treated optimisation and control separately. Moreover, no attempt has been made to implement them on-line (implementing the off-line optimal profiles achieved to design a controller). On the

other hand, designing controller to implement the optimal control profiles or tracking the dynamic set points have become an important area of research for inherently dynamic batch processes in recent years.

1.1 Aims of this Work

The importance and the potential for further research on optimisation and control of batch reactors have been clearly shown in the previous section. Nevertheless, the listed characteristics of such system create a very challenging task to researchers especially to control engineers. The gap between the off-line dynamic optimisation, generating optimal operating conditions (set points) and the on-line tracking of these optimal set points needs to be filled.

Due to the reasons mentioned above, the aims of this work are to study the optimisation of conventional general batch reactors in an extensive manner and to build the bridge between the off-line optimisation and the on-line optimal tracking (designing controller). However, to tackle both optimisation and control issues, the modelling issue has also been taken into consideration.

In brief, the objectives of this work are as follows:

1. To develop a user friendly computer program which will automatically generate the required dynamic model (the mass and energy balances equations and the reaction rate equations) of batch reactor based on a very minimum information (such as reaction scheme and kinetic information) supplied by the user.
2. To formulate and solve all general types of off-line dynamic optimisation problems of batch reactors i.e. maximum conversion, minimum time and maximum profit problems using various types of reaction schemes and to

highlight the significance of all types of optimisation problems. Issues like product quality, waste minimisation and safety are embedded in the form of constraints into the optimisation problems.

3. To implement the optimal operating parameters (such as reactor temperature and coolant flow rate) achieved from off-line optimisation study as the set points to be tracked by different types of controllers.
4. To design appropriate controllers using different types of advanced control strategies i.e. the generic model control coupled with neural network heat released estimator (GMC-NN), Direct Inverse and inverse-model-based control (IMBC) strategies to track on-line constant and dynamic set points. Other types of conventional controllers (dual mode (DM) control strategy with proportional-integral-derivatives (PID) and proportional-integral (PI) controllers) are also designed for comparison purpose.
5. To evaluate and compare the performances of different types of controllers used. Also to test the robustness of each controller toward a few process parameters changes.
6. To discuss BATCH_REACT, an integrated framework for modelling, simulation, optimisation and control of batch reactors.

1.2 Outlines of the Thesis

The structure of the thesis is outlined as follows:

Chapter 2 provides a literature review of earlier work on the dynamic optimisation and control of conventional general batch reactors. In the control part, three strategies are reviewed i.e. generic model control (GMC), neural controller in

Direct Inverse and IMBC strategies. The issues on modelling of batch reactors are also briefly addressed.

Chapter 3 is concerned with the modelling issue of batch reactors. It deals mainly with the first principle model for batch reactors and shows how general models of batch reactors can be developed to be used within optimisation framework. The degrees of freedom analysis have also been carried out for every types of model. Finally, examples on how the model can be generated for different types of reaction schemes are presented.

Chapter 4 considers the off-line dynamic optimisation of batch reactors. Initially, the method or approach used in this work is described. Then a few case studies with different reaction schemes and objectives are demonstrated.

Chapter 5 deals with the on-line tracking of constant and dynamic set points obtained in Chapter 4. Various types of advanced control strategies are implemented for two different reaction schemes. DM control strategy with PID and PI controllers are also implemented for comparison purposes. Their performance are evaluated and compared by consistent robustness tests.

Chapter 6 proposes the development of an integrated framework on the BATCH_REACT which will automatically generate model equations to describe the batch reactor operation, simulate the reactor, generate optimal operation policies off-line (set points) and finally design controllers to track the optimal set points. The work presented in Chapters 3-5 will be combined together in BATCH_REACT.

Finally, Chapter 7 summarises the contributions of this work and highlights future work to be undertaken.

Chapter Two

2.0 Literature Review

In this chapter two main areas are reviewed i.e. the dynamic optimisation (optimal control) and control of batch reactors. Dynamic optimisation of batch reactors refers to the reaction with known kinetics and the aim is to maximise or minimise a certain objective function such as maximum conversion, minimum batch time and maximum profit. On the other hand, control of batch reactors refers to the tracking of the set-point profile and disturbance rejection. Since both optimisation technique and model-based control rely on appropriate mathematical system representations, a modelling issue is also briefly addressed. This chapter only covers the conventional general batch reactor process and specific cases such as polymerisation and fermentation process are not reviewed as they are beyond the scope of this study. The study on semi-batch reactor is discussed separately in Chapter 4 in Case Study 4.

2.1 Dynamic Optimisation of Batch Reactors

The dynamic optimisation (optimal control) of chemical reactors has received major attention since the early works of Denbigh (1958) and Aris (1960). However, the number of work focusing on the conventional batch reactor is relatively low compared to other types of reactors such as semi-batch and tubular reactors.

Rippin (1983) has presented an excellent review for work carried out until 1982 on the optimal operation of several batch processes including batch reactors. However, again the work on conventional batch reactor is very limited. Moreover, most of the

publications gave only the general form of optimal operation policies without any numerical results.

To the best of the author's knowledge, the first work on the optimisation of conventional batch reactor was carried out by Siebenthal and Aris (1964). They presented two exothermic reactions with consecutive and competitive side reaction. The objective of the study was to minimise the side reaction and the problem was solved using Pontryagin's maximum principle (PMP). However, there was no constraint either on the state or on the control variable.

Later, Crestcitelli and Nicoletti (1973) obtained the optimum temperature profiles that maximised the product of an exothermic consecutive reaction. Here a constraint on the control variable was considered. The optimisation problem was solved using discrete maximum principle (DMP) method with time intervals of 3 and 6. In each case, the length of the time interval was optimised. The results achieved were compared with the optimal continuous solution (optimal continuous profile). They found the result achieved using greater number of time intervals is closer to the result achieved in the optimal continuous profile, as is normally expected.

Biegler (1984) optimised a batch reactor with parallel reaction scheme by using the orthogonal collocation (OC) with successive quadratic programming (SQP) technique. The aim was to find the temperature profiles that maximise the yield. The rate constant, k_1 (function of reactor temperature) was chosen as the optimisation variable. However, no constraint on the state variable was addressed. Later, Logsdon and Biegler (1989) solved the same problem with the same solution method but the reactor temperature was used as the control variable. However, they directly used the integration error information to construct accurate profiles. They also considered a

consecutive reaction scheme in their study. Here, the aim was to maximise the yield after a fixed reaction time and the control variable was the reactor temperature. Again, no state constraint was implemented in their study.

Logsdon (1990) considered optimisation of consecutive reaction in batch reactors. The objective was to find the optimum temperature (control variable) profiles that maximise the desired product in fixed reaction time. The problem was solved using two-point collocation and the null and range space code developed by Vasantharajan and Biegler (1988). Logsdon (1990) found that the method he used did not encounter any difficulty to solve this kind of problem. Later, Logsdon and Biegler (1993), considered the same reaction scheme but developed a relaxed simultaneous approach that leads to faster performance for the solution of optimisation problem. The optimal control problem remains the same as the previous study.

Eaton and Rawlings (1990) solved the optimisation of parallel batch reaction scheme using collocation method. The objective of the study was to find the optimal temperature profiles that maximise the concentration of the product at fixed final time. No state constraint was imposed in the system.

Luus (1994) used iterative dynamic programming (IDP) method in order to solve optimal control batch reactors with consecutive reaction scheme. The aim of his study was to maximise the yield at a fixed final time. Reactor temperature was chosen as the control variable and was optimised to achieve the objective function. Here, the switching time was fixed (was not optimised) and no constraint on the state variable was imposed on the system. He claimed that the results (amount of yield) obtained using the IDP method were better than those obtained using OC (Logsdon and Biegler, 1993) and could be obtained with reasonable computational effort.

Vassiliadis et al. (1994a) considered the formulation and solution of multistage dynamic optimisation problems in their work. They chose a system comprising two batch reactors as an example. In the first reactor, consecutive reaction took place in the presence of catalyst. The reactor was fitted with a heating coil, which can be used to manipulate the reactor temperature over time. After completion of the reaction, an amount of reactant is added instantaneously to the products of the first reactor, and the mixture is loaded into a second reactor where three parallel reactions took place under isothermal condition. The objective was to maximise the yield in the second reactor. The optimisation variables were the temperature profile in the first reactor, the batch times of the two reactors and the amount of reactant added at the mixing step. Reactor temperature was chosen as the control variable and final concentration of the product was imposed as the end-point constraint in the optimisation problem. Control vector parameterisation (CVP) technique employing piecewise polynomial function for control variables has been used to solve the above-mentioned problem. The initial conditions at the start of the first stage and junction conditions between stages can be embedded in the problem as interior point equality and inequality constraints at the end of each stage. They also stated that the CVP technique was able to handle large systems without the need to solve excessively large optimisation problems.

Garcia et al. (1995) studied a consecutive-competitive reaction scheme in their optimisation work. They converted the optimal control problems into a non-linear programming problem and then solved them by the generalised reduced gradient (GRG) procedure coupled with golden search (GS) method, to reach the final batch time. The objective was to find the temperature profiles with a fixed final batch time that will maximise the composition of the intermediate product. They were using 5 and 10

subintervals to discretise the profile and with the switching time equally spaced. They concluded that there is no significant change in the temperature profile when a more accurate level of discretisation is used.

Ruppen et al. (1995) considered a consecutive batch reactor in their study. They solved both maximum conversion and minimum time problems. The objective in the maximum conversion problem was to find the best temperature profiles that maximise the product at the fixed final time. In the minimum time problem, the product was specified and the object was to minimise the batch time. In both studies, OC method was implemented to transform the differential algebraic optimisation problem to the algebraic optimisation problem, which was solved, using the successive linear programming (SLP) method. Here, no constraint on the state variables was considered. They also carried out sensitivity analysis of reaction rate constant, k to the system.

Luus and Okongwu (1999) studied the optimisation of consecutive reaction schemes. The aim of the study was to maximise the production by manipulating the flow rate of hot and cold water (heating and cooling respectively). To prevent simultaneous flows of heating and cooling streams, they used a single heat flow term as the control variable. When positive, this heat flow term is equal to the heating stream flow rate; when negative, the magnitude of the heat flow term is equal to the coolant flow rate. They used an IDP method in order to solve the optimal control problem under consideration. They also studied the effect of heat transfer coefficient to the optimal policy.

All the work on the conventional batch reactors mentioned above are summarised in Table 2.1. From Table 2.1, it is found that most of the researchers only solved the maximum conversion problem except Ruppen et al. (1995). No one in the

past has considered the maximum profit problem for batch reactor system. Table 2.1 also shows that very few researchers considered the end-point constraint (e.g. amount of waste and final product quality and temperature) in their studies. Moreover, no one has considered the path constraint (e.g. reactor temperature should not go beyond a certain limit throughout the process) in their studies, which is very important from the safety point of view. In short, no attempt has been made to study the optimisation of batch reactors extensively. It might be due to the fact that the scope and the main interest of the previous researchers were to show the effectiveness and ability of the proposed numerical methods in solving the optimal control problem. Batch reactor system was just merely used as an example to fulfil the aim. Also, it might be due to the limitation of the numerical methods implemented.

In this work optimal operation of batch reactors is extensively studied. It is intended to apply suitable numerical methods from what are currently available to address some of the issues (shown in Table 2.1) not considered earlier. The purpose of this work clearly is not to develop further numerical methods. Here, we consider all three main types of optimisation problems i.e. maximum conversion, minimum time and maximum profit problems. Practical issues like safety, waste (by-product) minimisation and product quality are taken into consideration while formulating the optimisation problems.

Table 2.1: Summary of the past work on batch reactor optimisation

Author	Year	Reaction Scheme	O.F.	Method*	Control Variable	Interval Length	Constraint	On-line implementation
Sieenthal & Aris	1964	1. $A_1 \leftrightarrow A_2 \rightarrow A_3$ 2. $A_1 \leftrightarrow A_2$ $A_1 \rightarrow A_3$	Min side reaction	PMP	Temperature	NA	None	No
Crescitelle & Nicoletti	1973	$A \rightarrow B \rightarrow C$	Max B	DMP	Rate constant, k_1	Optimise	None	No
Biegler	1984	$A \rightarrow B, A \rightarrow C$	Max B	OC with SQP	Rate constant, k_1	NA	None	No
Logsdon & Biegler	1989	1. $A \rightarrow B, A \rightarrow C$ 2. $A \rightarrow B \rightarrow C$	Max B	OC	Temperature	Fixed	None	No
Eaton and Rawlings	1990	$A \rightarrow B, A \rightarrow C$	Max B	OC	Temperature	Fixed	None	No
Logsdon	1990	$A \rightarrow B \rightarrow C$	Max B	OC	Temperature	Fixed	None	No
Logsdon & Biegler	1993	$A \rightarrow B \rightarrow C$	Max B	OC with SQP	Temperature	Fixed	None	No
Luus	1994	$A \rightarrow B \rightarrow C$	Max B	IDP	Temperature	Fixed	None	No
Vassiliadis et al.	1994	2A $\rightarrow B \rightarrow C$ B $\rightarrow D, B \rightarrow E$ 2B $\rightarrow F$	Max D	CVP	Temperature	Optimise	End-point	No
Garcia et al.	1995	A + B $\rightarrow R$ R + B $\rightarrow S$	Max R	GRG with GS	Temperature	Fixed	None	No
Ruppen et al.	1995	A $\rightarrow B \rightarrow C$	Max B & Min time	OC with SLP	Temperature	Fixed	End-point	No
Luus & Okongwu	1999	A $\rightarrow B \rightarrow C$	Max B	IDP	Coolant/ Heating flow	Fixed	End-point	No
This work	2001	1. A $\rightarrow B \rightarrow C$ 2. A $\leftrightarrow B$ 3. 2A $\rightarrow B \rightarrow C$ B $\rightarrow D, B \rightarrow E$ 2B $\rightarrow F$ 4. A + B $\rightarrow C$ A + C $\rightarrow D$	Max Yield & Min time & Max profit	CVP with SQP	Temperature & Jacket flow rate	Optimise & Fixed	End-point & Path constraints	Yes

*Key: PMP, DMP etc. are in Abbreviations, page xv, O.F. = Objective Function, Min = Minimise, Max = Maximise, NA = Not Applicable

2.2 Solution Methods of Dynamic Optimisation Problems

The transient behaviour of many chemical engineering systems including batch reactor is described by mixed sets of Differential and Algebraic Equations (DAEs). A number of different solution approaches to dynamic optimisation problems for systems described by these Ordinary Differential Equations (ODEs) or DAEs have been proposed in the literature. Up to early 90's, most of the optimisation procedures or solutions were based on the Pontryagin's maximum principle (PMP) (Garcia et al., 1995). However, this approach is very difficult in most cases, especially when constraints on the state variables are present. This might be the reason for not considering the state constraint in most studies (Table 2.1) based on PMP method.

Since the last two decades, three main solution methods have been emerged and implemented (Carrasco and Banga, 1997). They are iterative dynamic programming technique (IDP) (Luus, 1990,1994; Luus and Okongwu, 1999; Luus and Hennessy, 1999; Rusnak et al., 2001), complete parameterisation approach or orthogonal collocation method (OC) (Biegler, 1984; Cuthrell and Biegler; 1989; Eaton and Rawlings, 1990; Logsdon, 1990; Ruppen et al., 1995, Loeblein et al., 1997) and control vector parameterisation approach (CVP) (Morison, 1984; Gritsis, 1990; Vassiliadis, 1993; Vassiliadis et al., 1994a,b; Barton et al., 1998; Sharif, 1999; Abel et al., 2000). For this reason, only these last three main approaches are reviewed in this work.

Luus (1990) developed iterative dynamic programming by implementing the Bellman's principle of optimality. The concept is based on the flow of information in a process and can be paraphrased as (Edgar and Himmelblau, 1989, p.398):

"If a decision forms an optimal solution at one stage in a process, then any remaining decisions must be optimal with respect to the outcome of the given

decision. This concept can be best implemented if the decision-making from stage to stage takes place in the direction of the information flow in the stages which may be in the opposite direction of the materials flow."

In brief, in IDP algorithm (Luus, 1990), the last subinterval (stage) is optimised first. After finding an approximate optimum there, the preceding stage is optimised regarding the control trajectories in the last stage as fixed and optimal. This procedure is repeated backwards to time $t = 0$ and for predefined number of iterations. To avoid optimisation on infinite space of all possible controls, at first N control trajectories are generated as uniform grid over the whole time interval. Then at the beginning of each stage, R_C constant control moves are generated for each of N trajectories and the optimum is searched among them (Fikar et al., 1998).

The complete parameterisation approach is an approach, which converts the dynamic optimisation problem to a finite dimensional non-linear program (NLP) through the discretisation of all variables. Initial work was based on finite difference approximation to the system constraint, but later global orthogonal collocation (Biegler, 1984) and orthogonal collocation on finite elements (Cuthrell and Biegler, 1989) were also used. In particular, the use of approximations over finite elements, the size and number of which can be determined automatically allows some control over the error discretisation (Logsdon and Biegler, 1989; Vasantharajan and Biegler, 1990).

The key characteristic of the complete discretisation approach outlined above is the fact that the optimisation is carried out in the full space of discretised variables, and the discretised constraints are in general, satisfied at the solution of the optimisation problem only. This is therefore often called an "infeasible path" approach.

The last approach is control vector parameterisation, which is an alternative approach to carry out the optimisation in the space of the decision variables only. In this case, it is necessary to discretise only the control variable $u(t)$. For given $u(t)$ and values of the other decision variables, it is then possible to integrate the underlying DAE system using standard integration algorithms so as to evaluate the objective function and other constraints that have to be satisfied by the solution. This technique corresponds to a “feasible path” approach since the DAEs are satisfied at each step of the optimisation algorithm.

All three main solutions mentioned above have their own advantages and disadvantages. The main advantage of IDP method is the ease of implementation. All that is needed is the calculation of the criterion at the final time and simulation of the system at one stage. It is suitable for solution of problems with small dimensionality. However, IDP is not directly suitable for handling equality or inequality terminal constraints (Fikar et al., 1998).

The complete discretisation approach has an advantage of not wasting computational effort in obtaining feasible DAE solutions away from the solution of optimisation. However, the size of NLP problem that is generated from this approach can be very large.

CVP on the other hand requires the integration of DAEs for constraint and objective values and the integration of variational equations (adjoint or sensitivity) in order to obtain the corresponding gradients, which can be very expensive. The complete parameterisation supporters claim this as its major drawback. On the other hand, the advantage of the CVP approach is that it decouples the DAEs from the optimisation stage. Each iteration is a feasible solution to the DAEs – hence functions are more likely

well behaved. Moreover, the stiffness of the DAEs is taken care of by the integrator, and no longer implies ill conditioning in the resulting NLPs (Vassiliadis, 1993). CVP has also been proved more superior and significantly faster compared to IDP (Fikar et al., 1998). After all, due to recent advances in numerical sensitivity technology, CVP is computationally more efficient than other approaches (Barton et al., 1998). In addition to the smaller size of the optimisation problem, the CVP approach has the advantage of efficiently controlling the discretisation error by adjusting the size and order of the integration steps using well-established ODE/DAE integration techniques. Also the number of such steps need not be known in advance, nor does it have to be constant during the course of the optimisation.

Due to the several advantages and arguments above, in this work we choose the CVP method to solve the batch reactor optimisation problems. In this approach, the process variables are partitioned into dependent and independent variables (decision variables), and for each choice of the decision variables, the simulator is used to converge the process equations (DAE's). Therefore the method includes two levels, the first level performs the simulation to converge all the equality constraints and to satisfy the inequality constraints and the second level performs the optimisation. The resulting optimisation problem is thus an unconstrained non-linear optimisation problem or a constrained optimisation problem with simple bounds for the associated decision variables plus any interior or terminal point constraints.

In general, there are four major approaches for solving non-linearly constrained optimisation (Chen, 1988):

1. Penalty function methods
2. Lagrange multiplier methods

3. Generalised reduced gradient methods
4. Successive quadratic programming methods

In this work, we use a successive quadratic programming (SQP) to solve the NLP problems as it is considered as one of the most effective method for NLP problems (Le Lann et al., 1999). SQP was also successfully used in other applications such as batch distillation (Mujtaba and Macchietto, 1998) and cogeneration plant (Rodriguez-Toral et al., 2000). This method generates at each step a search direction by solving a quadratic programming (QP) which is an approximation of the original problem. The Hessian of the quadratic programming subproblem is an approximation to the Hessian of the Lagrangian function. A procedure is taken to ensure convergence. The details of this method can be found in Chen (1988).

Successive quadratic programming (SQP) based optimisation codes that has been developed up by Chen (1988), known as SRQPD is used in conjunction with CVP technique to solve the optimal control problem. It implements a SQP method to find a local solution (local optimum) of a function subject to fixed bounds on the variables and to general equality and inequality constraints. It is suitable for optimisation problems where the number of variables and constraints is such that a dense (full matrix) representation can be stored in available computer. This code has been chosen because it permits an efficient solution of a sequence of similar optimisation problems (e.g. for parametric studies, sensitivity analysis, repeated online solution, etc.). SRQPD can also be used for the solution of systems of non-linear equations with/without constraints which is applicable to batch reactor. Besides that, it can also be used for maximisation problems.

2.3 On-line Tracking of Optimal Batch Reactors Profiles (Controller Design)

The term 'control' in batch operations can have widely differing objectives ranging from developing a complete operating strategy for productive and optimal use of available equipment i.e. plant scheduling to simple disturbance and trajectory tracking. Scheduling and sequencing of batch reactors is an important area of batch systems engineering that has been receiving an increasing level of interest in research and publications. However, our attention here will be confined to the control of individual batch operations, i.e. tracking the set point profile and disturbance rejection.

In terms of 'reactor control' in batch processing, one finds the concept of 'sequential control', 'logic control' and 'loop control'. Sequential control helps to step the reactor through its process sequence. Logic control is required for valve interlocks and alarm logic. Valve interlocks are used in addition to the sequence logic to prevent process upsets caused by a valve inadvertently opening or closing. Loop control is for maintaining operating parameters such as reactor temperature and pressure at their desired value. The integration of these three modes of control is required to effectively control the batch process (Berber, 1996). In this work, we will only concentrate with the loop control of batch reactors.

As we can see in Table 2.1, no attempt has been made to use the optimal operating profiles (set points) from optimisation study as a basis for controller design. The optimal profiles in the optimisation problem are basically achieved off-line and it is very essential to design controllers to track these set points on-line. In this study, three types of control strategies are considered. They are the generic model control (GMC), Direct Inverse and inverse-model-based control (IMBC) strategies. For comparison, a

dual mode (DM) with proportional-integral-derivatives (PID) and proportional-integral (PI) controllers are also implemented in this work.

Up to date, no work has been reported using the IMBC strategy to control the conventional batch reactor. Due to this reason, in this work, only two types of control strategies i.e. GMC and Direct Inverse implementation in conventional batch reactor will be reviewed. Readers are directed to consult the works of Juba and Hamer (1986) and Berber (1996) for more details on batch reactor control.

Generic model control (GMC) is a model-based control strategy developed by Lee and Sullivan (1988) and is one of several advanced process control algorithms implemented recently. This approach provides a rigorous and effective way of incorporating a non-linear energy balance model of the reactor and the heat exchange apparatus into the controller. It also allows the use of the same control algorithm for both heat-up and temperature maintenance, thereby eliminating the need to switch criterion between two separate control algorithms (as in the case of DM control strategy).

Cott and Macchietto (1989) have used the GMC algorithm as the controller for initial heat-up and subsequent temperature maintenance of exothermic batch reactors. They considered a well-mixed, liquid phase reaction system, in which two reactions occurred. The first reaction produced the desired product and the second reaction produced unwanted by-product (waste). The object was to maximize the conversion of the desired product and to minimise the by-product. Both reactions were strongly exothermic and the heating and cooling of the reactor contents were performed through the use of a single-pass jacket system. The on-line heat-release estimation method (Jutan and Uppal, 1984) using a three-term difference equation and exponential filters

was implemented in their GMC framework. The performance of GMC-based controller was also compared with commonly used DM controllers. Simulation studies showed that the new controller was as good as the DM controller for a nominal case for which both controllers were well tuned. However, the former was shown to be more robust with respect to changes in process parameters.

Later, Kershenbaum and Kittisupakorn (1994) considered the same reaction scheme of Cott and Macchietto (1989) and also used the GMC algorithm for the controller. However, the extended Kalman filter was used as the on-line heat-release estimator. They compared the simulation results with those carried out experimentally in a Partially Simulated Exothermic (PARSEX) reactor. The objective of these experiments was to observe the differences between results obtained from realistic simulation studies (with artificially imposed mismatches and uncertainties) and those from experiments in which the uncertainties are varied and poorly characterised. They found that the extended Kalman filter in experimental work was more sensitive to plant/model mismatch than would have been predicted from simulations alone. However, in both work mentioned earlier, the set points were not optimised beforehand.

Due to the robustness of the GMC reported as above, here we also consider in this work the same controller to track the optimal temperature profiles achieved in the off-line optimisation. However, we use neural network techniques as the on-line heat-release estimator because they have been proven to be an accurate and fast on-line dynamic estimator (Hussain, 1999). A multi-layered feed forward network is used which is trained using the back-propagation method. The back-propagation method has been chosen since it is the most well known and widely used algorithm associated with the training of a feed-forward neural network. A few tests have also been carried out in

order to test the robustness of this GMC based neural network controller (GMC-NN) with respect to changes in process parameters.

Since neural networks are able to model non-linear systems with some desired accuracy, it has high potential to be used for control task (as non-linear controller). Moreover, it is very versatile in nature that made it easily incorporated in various conventional and advanced control strategies for non-linear systems (Hussain and Kershenbaum, 2000). To date, the use of neural networks to control the batch reaction system is very few (Zaldivar et al., 1992; Dirion et al., 1996 and Galvan and Zaldivar, 1998). Extensive review on the implementation of neural networks in chemical process control can be found in Hussain (1999).

Zaldivar et al. (1992) used the neural networks for the adaptive control of temperature in a jacketed vessel in which an exothermic reaction took place. They have compared the conventional adaptive controllers and adaptive neural network based controllers with respect to closed-loop system stability, speed of adaptation, noise rejection and the number of required calculations through simulation. Feed-forward networks, which are trained using the back-propagation learning algorithm, have been implemented. They used the Mettler reaction calorimeter RC1 as an example of a typical reactor. RC1 is a computer-controlled batch reactor system able to carry out isothermal, adiabatic, isoperibolic and temperature programmed experiments. Learning data for neural network was taken from a simple neutralisation reaction between hydrochloric acid and sodium hydroxide reaction (in semi-batch mode) and the validation tests were carried out with the data from esterification of 2-butanol by propionic anhydride (in conventional batch mode). The results achieved showed that the

neural network approach functions well in the presence of noise, works for linear and non-linear systems and can be implemented very efficiently.

Dirion et al. (1996) applied neural networks to control batch reactor temperature. Again, the feed-forward multi-layered networks with back-propagation algorithm have been used here. The objective of the implementation of the neural controller (Direct Inverse approach) was to provide the appropriate process input in order to obtain the desired output. In other words, they used neural networks to calculate the adequate valve opening, giving the required inlet jacket temperature in order to make the mixture temperature follow the desired temperature profile. Here, the learning data set was obtained through heating and cooling solvent (water) with no chemical reaction which represented the dynamics range corresponding to the temperature domain in which the reactor is supposed to be operated. The learning database was composed of dynamic set point, the reactor temperature and control value computed by the adaptive generalised predictive controller. In the generalisation test, the trained neural network was used to control the reactor with exothermic chemical reaction (reaction between sodium thiosulphate and hydrogen peroxide) as a thermal disturbance for the neural controller. They reported that neural controllers might exhibit some limitations for extrapolation or for new operating conditions. This is obvious since the neural networks are being designed to learn from experience. They cannot predict or identify the parameter/condition beyond the training range. Dirion et al. also implemented an adaptive neural controller in order to improve on-line temperature control. However, the results achieved were equivalent to the Direct Inverse approach.

Galvan and Zaldivar (1998) applied the neural inverse and predictive control systems in the real-time control of heat transfer fluid temperature in a pilot chemical

reactor. The process was carried out in isothermal conditions (reactor temperature was maintained at its desired value – set point) by adjusting the jacket temperature (mixture of glycol-water). There was no reaction taking place in the reactor. The predictive control system used the neural networks to calculate the control action, which reduced the computational effort. They compared the performance of the neural-based controller and the self-tuning PID controller performance currently installed in the plant. It was found that the former was able to improve the performance of the real plant.

In this work, we implement a Direct Inverse control strategy and an IMBC strategy (Internal Model Control strategy) using neural networks to control the optimal temperature profiles of an exothermic reaction. Unlike previous work, our Direct Inverse approach also incorporates the on-line neural networks based heat-released estimator. The same estimator is also implemented in IMBC. In all the applied control strategies, the reactor temperature has been chosen as the control variable and jacket reactor temperature as the manipulated variable. Here, the neural network is trained based on the data collected while the batch reactor is in operation and the robustness tests have been carried out by changing process parameters of the system.

Internal Model Control (IMC) strategy is one of the many control strategies that can be applied to various chemical process plants. It promises to offer better control and reliability compared to other control strategies (Hussain, 1999). In this scheme, both the forward and inverse models are used directly as elements within the feedback loop and is known as IMBC strategy here. The network inverse model is utilised in control strategy by simply cascading it with the controlled system or plant. In this case the neural network acting as the controller has to learn to supply at its output the appropriate control parameters u , for the desired targets y_{sp} , at its input. In addition, the

neural network forward model is placed in parallel with the plant, to cater for plant or model mismatches and the error between the plant output and neural network forward model is subtracted from the set point before being feedback into the inverse model.

The neural network inverse model and the neural network forward model obtained a prior will be incorporated in this IMBC strategy.

Finally, we also implement the DM control strategy with PID and PI controllers and the performances (based on conversion of the desired product and on response of each controller) of all the various control strategies applied are compared. All reviewed works on the control of batch reactors are summarised in Table 2.2.

Table 2.2: Summary of the past work on batch reactor control

Author	Year	Reaction Scheme	Controller	Control Variable	Manipulated Variable	Robustness test	Set points optimisation (Off-line)
Cott & Macchietto	1989	A + B → C A + C → D	GMC with three-term difference equation and exponential filter	Temperature	Jacket temperature set point	Yes	No
Kershenbaum & Kittisupakorn	1994	A + B → C A + C → D	GMC with Kalman filter	Temperature	Jacket temperature set point	Yes	No
Zaldivar et al.	1992	Neutralisation & esterification	Adaptive NN	Temperature	Cooling flow & electric heating power	Yes	No
Dirion et al.	1996	No reaction	Direct Inverse & adaptive NN	Temperature	Valve opening	Yes	No
Galvan & Zaldivar	1998	No reaction	Direct Inverse & predictive NN	Heat transfer fluid temperature	Jacket temperature set point	No	No
This work	2001	1. A → B → C 2. A + B → C A + C → D	GMC with NN & Direct Inverse & IMBC	Temperature	Jacket temperature set point	Yes	Yes

GMC = Generic Model Control, NN = Neural Network, IMBC = Inverse-Model-Based Control

2.4 Modelling of Batch Reactors

The aim of this section is to highlight two types of batch reactor models (simple and detailed models). The simple model is based on mass balance equations and the reaction kinetic information and the detailed model is based on mass balance equations, reaction kinetic information and energy balance equations.

All information related to the model used for the work mentioned in Table 2.1 and Table 2.2. are summarised in Table 2.3. Table 2.3 shows that most of the work on optimisation of batch reactors implemented the simple model and only the work on the control implemented the detailed model. Also, most of previous work used rate of reaction as a function of concentration and the Arrhenius' reaction rate constant equation

The aim of the optimisation study, which used a simple model, was normally to find the best reactor temperature policy that maximise or minimise a certain performance index (maximum conversion, minimum time or maximum profit problems). For this reason, simple model was enough to correlate between the reactor temperature and the concentration (which is a measure of conversion) or time. The results obtained could then be applied as the basis to design batch reactors. The detailed model was normally used for optimising the existing plants with fixed cooling or heating capabilities. The energy balances in the detailed model provides the necessary information related to heating/cooling of the reactor and therefore the work on control has used the detailed model.

Table 2.3: Review on modelling of conventional batch reactors

Author	Year	Type of study	Model	Reaction Schemes
Siebenthal & Aris	1964	Optimisation	Simple	1. Consecutive 2. Parallel Both with reversible
Crescitelle & Nicoletti	1973	Optimisation	Simple	Consecutive
Biegler	1984	Optimisation	Simple	Parallel
Cott & Macchietto	1989	Control	Detailed	Complex
Logsdon & Biegler	1989	Optimisation	Simple	1. Consecutive 2. Parallel
Eaton & Rawlings	1990	Optimisation	Simple	Parallel
Logsdon	1990	Optimisation	Simple	Consecutive
Zaldivar et al.	1992	Control	Temperature controller model	Neutralisation & esterification
Logsdon & Bigler	1993	Optimisation	Simple	Consecutive
Kershenbaum & Kittisupakorn	1994	Control	Detailed	Complex
Luus	1994	Optimisation	Simple	Consecutive
Vassiliadis et al.	1994	Optimisation	Simple	Complex
Garcia et al.	1995	Optimisation	Simple	Complex
Ruppen et al.	1995	Optimisation	Simple	Consecutive
Dirion et al.	1996	Control	NA	NA
Galvan & Zaldivar	1998	Control	NA	NA
Luus and Okongwu	1999	Optimisation	Detailed	Consecutive
This Work	2001	Optimisation & Control	Simple & Detailed	Consecutive & Complex & Reversible

*NA = Not applicable

The number of model equations varies with the reaction schemes. It will take considerable amount of time to develop computer program for each set of the model to embed within optimisation and control problem formulation. Therefore, in this work, we develop a computer program, which can generate the mass and energy balance equations, the reaction rate and reaction rate constant equations automatically based on very basic information available. The user just has to supply basic information such as the total number of component, the type of reactions taking place in the reactor and the value of process parameters and constants. The non-zero matrix and the gradient information related to model equations also will be automatically generated. Non-zero

matrix is a matrix indicates only non-zero elements. This type of matrix is normally used to represent a sparse matrix (one in which there are many more zero elements than non-zero ones), which will save memory for storage and expense of computer time. The gradient information of the model equations is needed to solve the gradient-based optimisation technique such as CVP coupled with SQP.

2.5 Concluding Remarks

The review of this chapter has highlighted the work done on the optimisation and control of conventional general batch reactors. It also highlights the types of model used for optimisation and control. The review on control application mainly considered two control strategies i.e. GMC and Direct Inverse approach. Past work has revealed several relevant points, which have given impetus and direction to our work, as summarised below:

1. There can be several reaction schemes in batch reactors, which will end up with different number of model equations. It is cumbersome and time consuming to develop problem specific model each time for variety of possible reaction scheme that may be encountered. Therefore, a computer program is developed to generate the required model equations automatically.
2. There are only limited work on optimisation of conventional batch reactors and most of them focussed on the development of numerical methods. In this work, we carried out an extensive study on batch reactor optimisation including several practical constraints. All common types of optimisation problems (maximum conversion, minimum time and maximum profit problems) are studied for a variety of reaction schemes.

3. Work in the past has not implemented the optimal profiles obtained by off-line optimisation to design the controllers. In this work, we implement the off-line optimal profiles as set points to be tracked on-line and various types of controllers are designed for the purpose. Both constant and dynamic set points tracking are considered.
4. Neural networks are versatile in nature and are capable of being in various well-known non-linear control methods and strategies. In this work, we demonstrate this capability further by employing it in the Direct Inverse and IMBC strategies for controlling the optimal profiles of batch reactor. Since the multi-layered feed forward with sigmoidal activation function is widely used (Hussain, 1999), this is implemented in all neural network applications.
5. GMC has been approved to be a robust controller in batch reactor system but no attempt has been made to compare its performance with that of neural network based controller. Here, we first implemented GMC coupled with on-line neural network heat release estimator (GMC-NN) to control a batch reactor. The robustness of this algorithm is further tested in the event of process parameter changes. Then we implement neural controller (in Direct Inverse and IMBC strategies) to track the same set points (as implemented in GMC-NN). Similar robustness tests (as those in GMC-NN) are carried out toward these controllers and their performances are evaluated and compared.

Chapter Three

3.0 Dynamic Modelling

3.1 Introduction

The development of faster computers and sophisticated numerical methods has enabled modelling and solution of complete system (process), while in the past one had to separate the system to its constituent parts. 'Mathematical Modelling' of a system (or process) concerns with quantitative rather than a qualitative treatment of the process. This mathematical model is a collection (set) of equations that described some aspects of the behaviour of the system under investigation. It can also be defined as a set of relationship which links the inputs, outputs and other state variables of any system together. However, for many complex chemical processes, the models result to a set of non-linear equations requiring numerical solution.

'Mathematical Modelling' has a very wide field of application: from economics to engineering and science. They are essential for understanding or controlling the system. Generally, there are three types of models (Bonvin, 1998):

1. Data-driven black-box models
2. Knowledge-driven white-box models
3. Hybrid grey-box models

The black-box models are developed through observing the input-output open loop or closed-loop behaviour of the system. This method is simple and relatively easy to obtain but it is not accurate in extrapolation. Moreover, it represents the dynamic relationship only between variables that are manipulated or measured. In batch reactors,

the important variables such as heat of reaction and concentration are still unmeasured. This approach is commonly known as systems identification.

White-box models are referred to as mathematical modelling and the models are called as mechanistic first-principles-model. It is developed based on stoichiometric and kinetic knowledge and on material and energy balances of the reactor. This approach is preferred for batch reactor modelling. In this model, the effect of temperature and concentration to the rate of each reaction is described by the kinetic model and the reactor model relates the states (concentrations, temperature and volume) to the inlet streams, reaction terms and possible disturbances. However, they may not easily be obtainable in many cases and can be time consuming. The grey-box model is a combination of the two extreme cases mentioned above.

In this study, only the white-box model is considered and it will be used for all case studies. The model of a chemical process (individual unit or complete process) can be described in terms of a set of equations consisting of:

1. Mass and energy balances
2. Thermo-physical properties
3. Geometrical constraints and any other process constraint

From a gross kinetic point of view, chemical reactions are mainly divided into two classes i.e. single and multiple reactions (Laguerie and Delmas, 1989).

The single reactions comprise:

1. Uni-directional reactions:



2. Reversible reactions:



3. Auto-catalytic reactions:



where the presence of the product P is necessary to provoke the conversion of the reactant A

The multiple reactions comprise a variety of possible kinds of reaction, among which three basic types can be given:

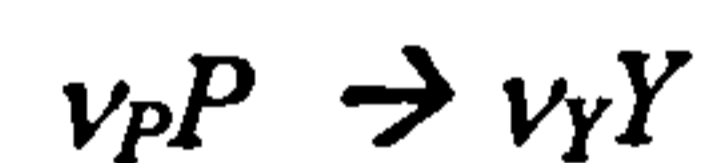
1. Parallel reactions



2. Consecutive reactions



3. Complex reaction which combinations of parallel and consecutive reactions



Single reactions are described by a single reaction balance. Multiple reactions are described by a system of reaction balances, the number of which is equal to the number of the reactions involved.

In a conventional batch reactor, a given amount of material is introduced first, but no material is fed or withdrawn as the reaction proceeds. The total mass of the reaction mixture remains constant, while the composition is a time function.

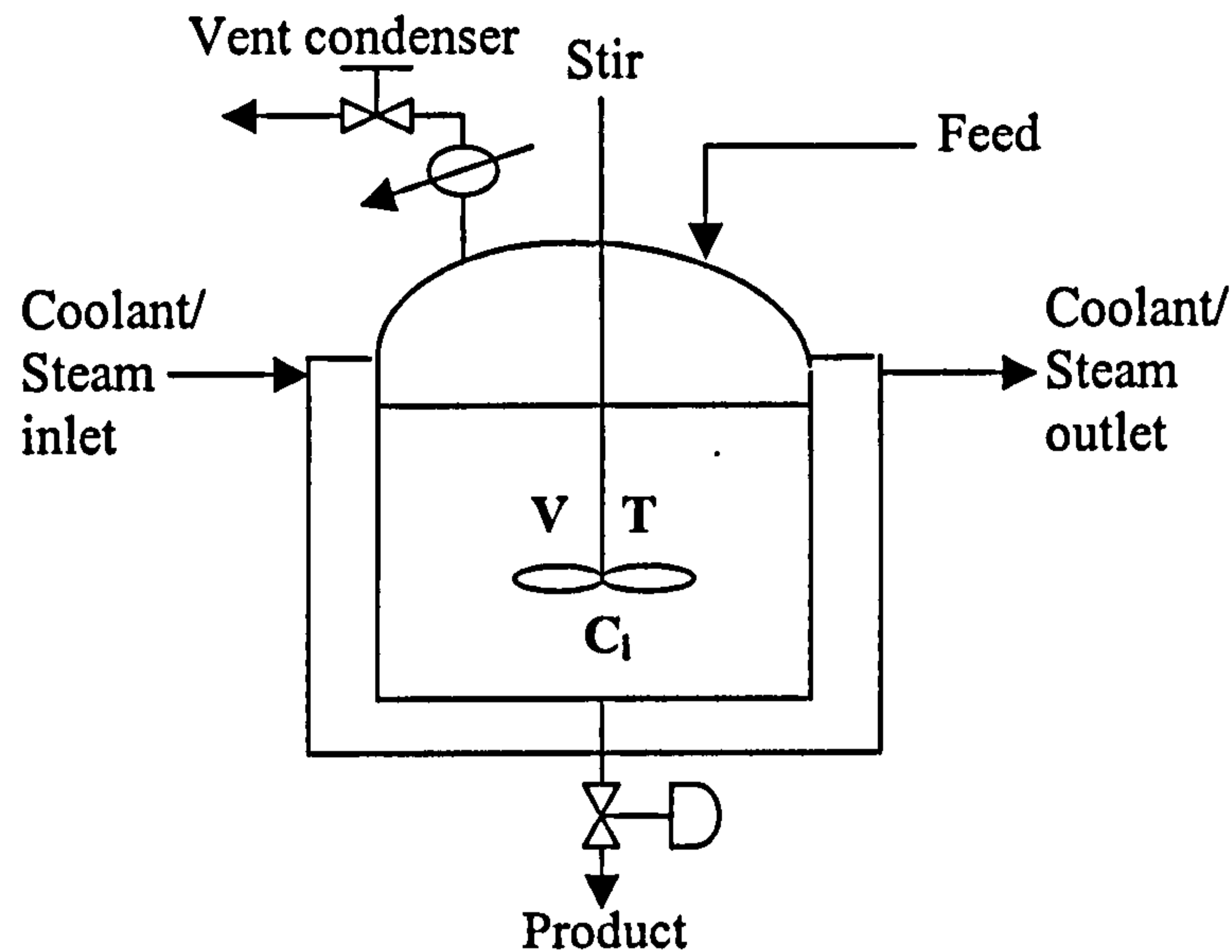


Figure 3.1: Schematic diagram of jacketed batch reactor

3.2 General Batch Reactors Model (GBRM)

The development of dynamic model of batch reactors is an essential task before the dynamic optimisation and control studies can be carried out. Different types of reactions will end up with different types of dynamic model. Rather than building up the model for different cases separately, a generic computer program is developed which will automatically build up the required dynamic model (the mass and energy balances, the reaction rate and reaction rate constant equations, etc.) of batch reactors based on the information supplied by the user. Jacobian (gradient information) matrix of the model will also be automatically generated, together with the locations of the non-zero elements in the Jacobian matrix.

Efficient solution of model equations requires the evaluation of gradients (Jacobian Matrix) of each equation with respect to all the variables of the system. As

only a subset of the total variables appear in each equation, the Jacobian will have more zero elements than non-zero elements. In such cases, one must confront the problem of the abundance of zeros contributing to the expense of computer time and memory. This can be overcome with a special 'sparse matrix' technique in such a way that only non-zero matrix elements are stored and processed.

The information that must be supplied by the user are:

1. Choose the option to include or exclude the energy balance and choose control variable of the process.
2. Give the total number of reactant and product, m .
3. Give the total number of reaction, n .
4. Provide all the values of the specified parameters (e.g. ν_{ji} , k_{j0} , E_j , R , β_{ij})

3.2.1 Mass Balance

In this work, only mass balance in term of concentration, C_i and reaction rate, r_j is used and one of the parameters (reactor temperature, jacket temperature or jacket flow rate) is chosen as the control variable. It can be written as:

$$\frac{dC_i}{dt} = R_i \quad \forall i \quad (3.1)$$

where R_i the overall production rate of the component C_i :

$$R_i = \sum_{j=1}^n \nu_{ji} r_j \quad \forall i \quad (3.2)$$

Combine Equations 3.1 and 3.2:

$$\frac{dC_i}{dt} = \sum_{j=1}^n v_{ji} r_j \quad \forall i \quad (3.3)$$

where v_{ji} is the stoichiometric coefficient.

$v_{ji} > 0$ if element or component i is a product of reaction j

$v_{ji} < 0$ if element or component i is a reactant of reaction j

$v_{ji} = 0$ if element or component i is not concerned with reaction j

3.2.2 Reaction Rate

Chemical reaction rates are expressed as functions of the concentrations or partial pressure. In the case where two reactants A and B are involved, the kinetic rate is often expressed by:

$$r = k C_A^a C_B^b \quad (3.4)$$

where a and b are the order of the reaction with respect to A and B respectively. The total order of the reactions is $a+b$.

For reversible reactions, one has:

$$r = k_1 C_A^a C_B^b - k_2 C_P^p C_Q^q \quad (3.5)$$

and for autocatalytic reactions, this becomes:

$$r = k C_A^a C_P^p \quad (3.6)$$

In general, the chemical reaction rate equations can be written as:

$$r_j = k_j \prod_{i=1}^m C_i^{\beta_{ij}} \quad \forall j \quad (3.7)$$

where β_{ij} is the order of reaction rate with respect to component C_i for reaction j .

In reaction rate equations, the influence of temperature is taken into account in the reaction rate constant, k_j which is expressed by an Arrhenius equation:

$$k_j = k_{j0} \exp\left(-\frac{E_j}{RT}\right) \quad \forall j \quad (3.8)$$

where k_{j0} is the reaction rate constant at reference temperature, E_j is the activation energy, R is the universal gas constant and T is the reactor temperature.

3.2.3 Energy Balance

The energy balance equation depends on the control variable chosen.

Option 1

Plain reactor – Energy balance is not considered and the reactor temperature (T) is used as control variable

Option 2

Jacketed reactor – Energy balance with negligible heat retention in the wall is considered and the jacket temperature (T_j) is used as control variable.

The energy balance equations will then be:

$$\frac{dT}{dt} = \frac{Q_r - Q_j}{c_p V \rho} \quad (3.9)$$

$$Q_r = \sum_{j=1}^n r_j (-\Delta H_{rj}) V \quad (3.10)$$

$$Q_j = UA (T - T_j) \quad (3.11)$$

Option 3

Jacketed reactor – Energy balance with negligible heat retention in the wall is considered and the jacket flow rate (F_J) is used as control variable.

The energy balance equations used are:

$$\frac{dT}{dt} = \frac{Q_r - Q_J}{c_p V \rho} \quad (3.12)$$

$$Q_r = \sum_{j=1}^n r_j (-\Delta H_{rj}) V \quad (3.13)$$

$$Q_J = UA (T - T_J) \quad (3.14)$$

$$\frac{dT_J}{dt} = \frac{F_J}{V_J} (T_{J0} - T_J) + \frac{Q_J}{c_{pJ} V_J \rho_J} \quad (3.15)$$

Option 4

Jacketed reactor – Energy balance with heat retention in the wall is considered and the jacket temperature (T_J) is used as control variable

The energy balance equations used are:

$$\frac{dT}{dt} = \frac{Q_r - Q_m}{c_p V \rho} \quad (3.16)$$

$$Q_r = \sum_{j=1}^n r_j (-\Delta H_{rj}) V \quad (3.17)$$

$$Q_m = U_i A_i (T - T_m) \quad (3.18)$$

$$\frac{dT_m}{dt} = \frac{Q_m - Q_J}{c_{pm} V_m \rho_m} \quad (3.19)$$

$$Q_J = U_o A_o (T_m - T_J) \quad (3.20)$$

Option 5

Jacketed reactor – Energy balance with heat retention in the wall is considered and the jacket flow rate (F_J) is used as control variable

Energy balance equations used are:

$$\frac{dT}{dt} = \frac{Q_r - Q_m}{c_p V \rho} \quad (3.21)$$

$$Q_r = \sum_{j=1}^n r_j (-\Delta H_{rj}) V \quad (3.22)$$

$$Q_m = U_i A_i (T - T_m) \quad (3.23)$$

$$\frac{dT_m}{dt} = \frac{Q_m - Q_J}{c_{pm} V_m \rho_m} \quad (3.24)$$

$$Q_J = U_o A_o (T_m - T_J) \quad (3.25)$$

$$\frac{dT_J}{dt} = \frac{F_J}{V_J} (T_{J0} - T_J) + \frac{Q_J}{c_{pJ} V_J \rho_J} \quad (3.26)$$

3.2.4 Model Equations and Degrees of Freedom Analysis

For all the options mentioned in section 3.2.3 the model equations are written below and the degrees of freedom analysis is provided.

Option 1

Model equations:

$$\frac{dC_i}{dt} = \sum_{j=1}^n \nu_{ji} r_j \quad \forall i \quad (3.27)$$

$$r_j = k_j \prod_{i=1}^m C_i^{\beta_{ij}} \quad \forall j \quad (3.28)$$

$$k_j = k_{j0} \exp\left(-\frac{E_j}{RT}\right) \quad \forall j \quad (3.29)$$

Total number of variables (TNV):

$$\begin{aligned} & \frac{dC_i}{dt} C_i, v_{ji}, r_j, k_j, \beta_{ij}, k_{j0}, E_j, R, T, t \\ & = 4n + 2m + 2mn + 3 \end{aligned}$$

Total number of equations (TNE):

$$= 2n + m$$

Degrees of freedom (D.F.),

$$\begin{aligned} \text{D.F} &= \text{TNV} - \text{TNE} \\ &= 4n + 2m + 2mn + 3 - (2n + m) \\ &= 2n + m + 2mn + 3 \end{aligned}$$

Specifications:

$$\begin{aligned} \text{Parameters} &= v_{ji}, k_{j0}, E_j, R, \beta_{ij} \\ &= 2n + 2mn + 1 \end{aligned}$$

Initial values of:

$$C_i \text{ at } (t=0) = m$$

Control variable, $T = 1$

Independent variable, $t = 1$

Note C_i is differential variable of the model equations. So initial value at $t = 0$ must be given. t is the independent variable of the model equations. T is used as the control variable and therefore it is relaxed from the specification set and optimised.

Option 2

Model equations:

$$\frac{dC_i}{dt} = \sum_{j=1}^n \nu_{ji} r_j \quad \forall i \quad (3.30)$$

$$r_j = k_j \prod_{i=1}^m C_i^{\beta_{ij}} \quad \forall j \quad (3.31)$$

$$k_j = k_{j0} \exp\left(-\frac{E_j}{RT}\right) \quad \forall j \quad (3.32)$$

$$\frac{dT}{dt} = \frac{Q_r - Q_J}{c_p V \rho} \quad (3.33)$$

$$Q_r = \sum_{j=1}^n r_j (-\Delta H_{rj}) V \quad (3.34)$$

$$Q_J = UA (T - T_J) \quad (3.35)$$

Total number of variables (TNV):

$$\begin{aligned} & \frac{dC_i}{dt}, \frac{dT}{dt}, C_i, \nu_{ji}, r_j, k_j, \beta_{ij}, k_{j0}, E_j, R, T, Q_r, c_p, V, \rho, \Delta H_{rj}, Q_J, U, A, T_J, t \\ & = 5n + 2m + 2mn + 12 \end{aligned}$$

Total number of equations (TNE):

$$= 2n + m + 3$$

$$\begin{aligned}
 \text{D.F} &= \text{TNV} - \text{TNE} \\
 &= 5n + 2m + 2mn + 12 - (2n + m + 3) \\
 &= 3n + m + 2mn + 9
 \end{aligned}$$

Specifications:

$$\begin{aligned}
 \text{Parameters} &= v_{ji}, k_{j0}, E_j, R, c_p, V, \rho, AH_{rj}, U, A, \beta_{ij} \\
 &= 3n + 2mn + 6
 \end{aligned}$$

Initial values of:

$$C_i \text{ at } (t=0) = m$$

$$T \text{ at } (t=0) = 1$$

Control variable, $T_j = 1$

Independent variable, $t = 1$

Option 3

Model equations:

$$\frac{dC_i}{dt} = \sum_{j=1}^n v_{ji} r_j \quad \forall i \quad (3.36)$$

$$r_j = k_j \prod_{i=1}^m C_i^{\beta_{ij}} \quad \forall j \quad (3.37)$$

$$k_j = k_{j0} \exp\left(-\frac{E_j}{RT}\right) \quad \forall j \quad (3.38)$$

$$\frac{dT}{dt} = \frac{Q_r - Q_j}{c_p V \rho} \quad (3.39)$$

$$Q_r = \sum_{j=1}^n r_j (-\Delta H_{rj}) V \quad (3.40)$$

$$Q_J = UA (T - T_J) \quad (3.41)$$

$$\frac{dT_J}{dt} = \frac{F_J}{V_J} (T_{J0} - T_J) + \frac{Q_J}{c_{pJ} V_J \rho_J} \quad (3.42)$$

Total number of variables (TNV):

$$\frac{dC_i}{dt}, \frac{dT}{dt}, \frac{dT_J}{dt}, C_i, v_{ji}, r_j, k_j, \beta_{ij} k_{j0}, E_j, R, T, Q_r, c_p, V, \rho, \Delta H_{rj}, Q_J, U, A, T_J, F_J,$$

$$V_J, T_{J0}, c_{pJ}, \rho_J, t$$

$$= 5n + 2m + 2mn + 18$$

Total number of equations (TNE):

$$= 2n + m + 4$$

$$\text{D.F} = \text{TNV} - \text{TNE}$$

$$= 5n + 2m + 2mn + 18 - (2n + m + 4)$$

$$= 3n + m + 2mn + 14$$

Specifications:

$$\text{Parameters} = v_{ji}, \beta_{ij} k_{j0}, E_j, R, c_p, V, \rho, \Delta H_{rj}, U, A, V_J, T_{J0}, c_{pJ}, \rho_J$$

$$= 3n + 2mn + 10$$

Initial values of:

$$C_i \text{ at } (t=0) = m$$

$$T \text{ at } (t=0) = 1$$

$$T_J \text{ at } (t=0) = 1$$

Control variable, $F_J = 1$

Independent variable, $t = 1$

Option 4

Model equations:

$$\frac{dC_i}{dt} = \sum_{j=1}^n v_{ji} r_j \quad \forall j \quad (3.43)$$

$$r_j = k_j \prod_{i=1}^m C_i^{\beta_{ij}} \quad \forall j \quad (3.44)$$

$$k_j = k_{j0} \exp\left(-\frac{E_j}{RT}\right) \quad \forall j \quad (3.45)$$

$$\frac{dT}{dt} = \frac{Q_r - Q_m}{c_p V \rho} \quad (3.46)$$

$$Q_r = \sum_{j=1}^n r_j (-\Delta H_{rj}) V \quad (3.47)$$

$$Q_m = U_i A_i (T - T_m) \quad (3.48)$$

$$\frac{dT_m}{dt} = \frac{Q_m - Q_J}{c_{pm} V_m \rho_m} \quad (3.49)$$

$$Q_J = U_o A_o (T_m - T_J) \quad (3.50)$$

Total number of variables (TNV):

$$\frac{dC_i}{dt}, \frac{dT}{dt}, \frac{dT_m}{dt}, C_i, v_{ji}, r_j, k_j, \beta_{ij}, k_{j0}, E_j, R, T, Q_r, Q_m, c_p, V, \rho, \Delta H_{rj}, U_i, A_i, T_m, Q_J,$$

$$c_{pm}, V_m, \rho_m, U_o, A_o, T_J, t$$

$$= 5n + 2m + 2mn + 20$$

Total number of equations (TNE):

$$= 2n + m + 5$$

D.F = TNV - TNE

$$= 5n + 2m + 2mn + 20 - (2n + m + 5)$$

$$= 3n + m + 2mn + 15$$

Specifications:

Parameters = $v_{ji}, k_{j0}, E_j, R, c_p, V, \rho, AH_{rj}, U_i, A_i, c_{pm}, V_m, \rho_m, U_o, A_o, \beta_{ij}$

$$= 3n + 2mn + 11$$

Initial values of:

$$C_i \text{ at } (t=0) = m$$

$$T \text{ at } (t=0) = 1$$

$$T_m \text{ at } (t=0) = 1$$

Control variable, $T_j = 1$

Independent variable, $t = 1$

Option 5

Model equations:

$$\frac{dC_i}{dt} = \sum_{j=1}^n v_{ji} r_j \quad \forall j \quad (3.51)$$

$$r_j = k_j \prod_{i=1}^m C_i^{\beta_{ij}} \quad \forall j \quad (3.52)$$

$$k_j = k_{j0} \exp\left(-\frac{E_j}{RT}\right) \quad \forall j \quad (3.53)$$

$$\frac{dT}{dt} = \frac{Q_r - Q_m}{c_p V \rho} \quad (3.54)$$

$$Q_r = \sum_{j=1}^n r_j (-\Delta H_{rj}) V \quad (3.55)$$

$$Q_m = U_i A_i (T - T_m) \quad (3.56)$$

$$\frac{dT_m}{dt} = \frac{Q_m - Q_J}{c_{pm} V_m \rho_m} \quad (3.57)$$

$$Q_J = U_o A_o (T_m - T_J) \quad (3.58)$$

$$\frac{dT_J}{dt} = \frac{F_J}{V_J} (T_{J0} - T_J) + \frac{Q_J}{c_{pJ} V_J \rho_J} \quad (3.59)$$

Total number of variables (TNV):

$$\frac{dC_i}{dt}, \frac{dT}{dt}, \frac{dT_m}{dt}, \frac{dT_J}{dt}, C_i, v_{ji}, r_j, k_j, \beta_{ij}, k_{j0}, E_j, R, T, Q_r, Q_m, c_p, V, \rho, \Delta H_{rj}, U_i, A_i,$$

$$T_m, Q_J, c_{pm}, V_m, \rho_m, U_o, A_o, T_J, F_J, V_J, T_{J0}, c_{pJ}, V_J, \rho_J, t$$

$$= 5n + 2m + 2mn + 27$$

Total number of equations (TNE):

$$= 2n + m + 6$$

$$\text{D.F} = \text{TNV} - \text{TNE}$$

$$= 5n + 2m + 2mn + 27 - (2n + m + 6)$$

$$= 3n + m + 2mn + 21$$

Specifications:

Parameters = $v_{ji}, k_{j0}, E_j, R, c_p, V, \rho, AH_{rj}, U_i, A_i, c_{pm}, V_m, \rho_m, U_o, A_o, V_J, T_{J0}, c_{pJ}, V_J, \rho_J,$

$$\beta_{ij}$$

$$= 3n + 2mn + 16$$

Initial values of:

$$C_i \text{ at } (t=0) = m$$

$$T \text{ at } (t=0) = 1$$

$$T_m \text{ at } (t=0) = 1$$

$$T_J \text{ at } (t=0) = 1$$

Control variable, $F_J = 1$

Independent variable, $t = 1$

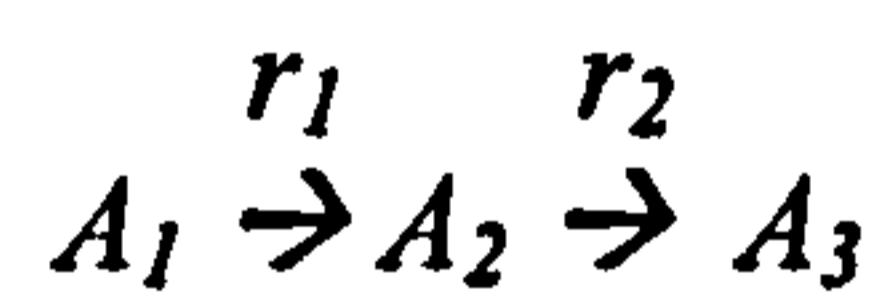
3.2.5 Examples

3.2.5.1 Consecutive reaction

Option 1

$$m = 3$$

$$n = 2$$



Model equations that will be generated ($2n + m = 7$):

$$\frac{dC_1}{dt} = v_{11}r_1 + v_{21}r_2 \quad (3.60)$$

$$\frac{dC_2}{dt} = v_{12}r_1 + v_{22}r_2 \quad (3.61)$$

$$\frac{dC_3}{dt} = v_{13}r_1 + v_{23}r_2 \quad (3.62)$$

$$r_1 = k_1 C_1^{\beta_{11}} C_2^{\beta_{21}} C_3^{\beta_{31}} \quad (3.63)$$

$$r_2 = k_2 C_1^{\beta_{12}} C_2^{\beta_{22}} C_3^{\beta_{32}} \quad (3.64)$$

$$k_1 = k_{10} \exp\left(-\frac{E_1}{RT}\right) \quad (3.65)$$

$$k_2 = k_{20} \exp\left(-\frac{E_2}{RT}\right) \quad (3.66)$$

Accordingly, the values of parameters that must be supplied are

(2j + 2ij + 1 = 17):

$v_{11}, v_{21}, v_{12}, v_{22}, v_{13}, v_{23}, \beta_{11}, \beta_{21}, \beta_{31}, \beta_{12}, \beta_{22}, \beta_{32}, E_1, E_2, R, k_{10}, k_{20}$

Here the values of v_{11} and v_{22} are equal to -1 . The values of v_{12} and v_{23} are equal to 1 and the rest of stoichiometric coefficients values are zero.

So Equations 3.60 to 3.62 will become:

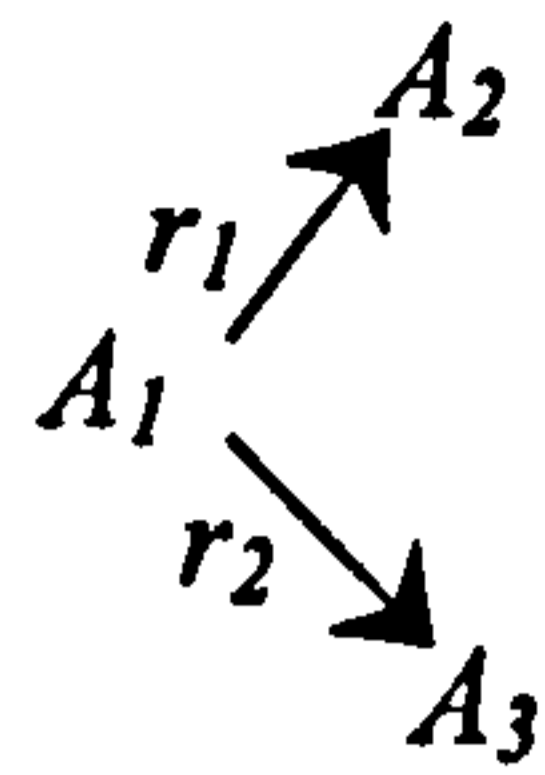
$$\frac{dC_1}{dt} = -r_1 \quad (3.67)$$

$$\frac{dC_2}{dt} = r_1 - r_2 \quad (3.68)$$

$$\frac{dC_3}{dt} = r_2 \quad (3.69)$$

3.2.5.2 Parallel reaction

Option 2

 $m = 3$ $n = 2$ Model equations that will be generated ($2n + m + 3 = 10$):

$$\frac{dC_1}{dt} = v_{11}r_1 + v_{21}r_2 \quad (3.70)$$

$$\frac{dC_2}{dt} = v_{12}r_1 + v_{22}r_2 \quad (3.71)$$

$$\frac{dC_3}{dt} = v_{13}r_1 + v_{23}r_2 \quad (3.72)$$

$$r_1 = k_1 C_1^{\beta_{11}} C_2^{\beta_{21}} C_3^{\beta_{31}} \quad (3.73)$$

$$r_2 = k_2 C_1^{\beta_{12}} C_2^{\beta_{22}} C_3^{\beta_{32}} \quad (3.74)$$

$$k_1 = k_{10} \exp\left(-\frac{E_1}{RT}\right) \quad (3.75)$$

$$k_2 = k_{20} \exp\left(-\frac{E_2}{RT}\right) \quad (3.76)$$

$$\frac{dT}{dt} = \frac{Q_r - Q_J}{c_p V \rho} \quad (3.77)$$

$$Q_r = -r_1(AH_{r1})V - r_2(AH_{r2})V \quad (3.78)$$

$$Q_J = UA(T - T_J) \quad (3.79)$$

Accordingly, the values of parameters that must be supplied are

$(3n + 2mn + 6 = 24)$:

$v_{11}, v_{21}, v_{12}, v_{22}, v_{13}, v_{23}, \beta_{11}, \beta_{21}, \beta_{31}, \beta_{12}, \beta_{22}, \beta_{32}, E_1, E_2, R, k_{10}, k_{20}, c_p, V, \rho, AH_{r1},$
 AH_{r2}, U, A

Here the values of v_{11} and v_{21} are equal to -1 . The values of v_{12} and v_{23} are equal to 1 and the rest of stoichiometric coefficients values are zero.

So Equations 3.70 to 3.72 will become:

$$\frac{dC_1}{dt} = -r_1 - r_2 \quad (3.80)$$

$$\frac{dC_2}{dt} = r_1 \quad (3.81)$$

$$\frac{dC_3}{dt} = r_2 \quad (3.82)$$

3.2.5.3 Consecutive and reversible reaction

Option 3



Model equations that will be generated $(2n + m + 4 = 13)$:

$$\frac{dC_1}{dt} = v_{11}r_1 + v_{21}r_2 + v_{31}r_3 \quad (3.83)$$

$$\frac{dC_2}{dt} = v_{12}r_1 + v_{22}r_2 + v_{32}r_3 \quad (3.84)$$

$$\frac{dC_1}{dt} = v_{13}r_1 + v_{31}r_2 + v_{33}r_3 \quad (3.85)$$

$$r_1 = k_1 C_1^{\beta_{11}} C_2^{\beta_{21}} C_3^{\beta_{31}} \quad (3.86)$$

$$r_2 = k_2 C_1^{\beta_{12}} C_2^{\beta_{22}} C_3^{\beta_{32}} \quad (3.87)$$

$$r_3 = k_3 C_1^{\beta_{13}} C_2^{\beta_{23}} C_3^{\beta_{33}} \quad (3.88)$$

$$k_1 = k_{10} \exp\left(-\frac{E_1}{RT}\right) \quad (3.89)$$

$$k_2 = k_{20} \exp\left(-\frac{E_2}{RT}\right) \quad (3.90)$$

$$k_3 = k_{30} \exp\left(-\frac{E_3}{RT}\right) \quad (3.91)$$

$$\frac{dT}{dt} = \frac{Q_r - Q_J}{c_p V \rho} \quad (3.92)$$

$$Q_r = -r_1 (AH_{r1})V - r_2 (AH_{r2})V - r_3 (AH_{r3})V \quad (3.93)$$

$$Q_J = UA (T - T_J) \quad (3.94)$$

$$\frac{dT_J}{dt} = \frac{F_J}{V_J} (T_{J0} - T_J) + \frac{Q_J}{c_{pJ} V_J \rho_J} \quad (3.95)$$

Accordingly, the values of parameters that must be supplied are

$(3n + 2mn + 10 = 37)$:

$v_{11}, v_{21}, v_{31}, v_{12}, v_{22}, v_{32}, v_{13}, v_{23}, v_{33}, \beta_{11}, \beta_{21}, \beta_{31}, \beta_{12}, \beta_{22}, \beta_{32}, \beta_{13}, \beta_{23}, \beta_{33}, E_1, E_2, E_3,$

$R, k_{10}, k_{20}, k_{30}, c_p, V, \rho, AH_{r1}, AH_{r2}, AH_{r3}, U, A, V_J, T_{J0}, c_{pJ}, \rho_J$

Here the values of v_{11}, v_{22} and v_{33} are equal to -1 . The values of v_{12}, v_{21} and v_{33} are equal to 1 and the rest of stoichiometric coefficients values are zero.

So Equations 3.83 to 3.85 will become:

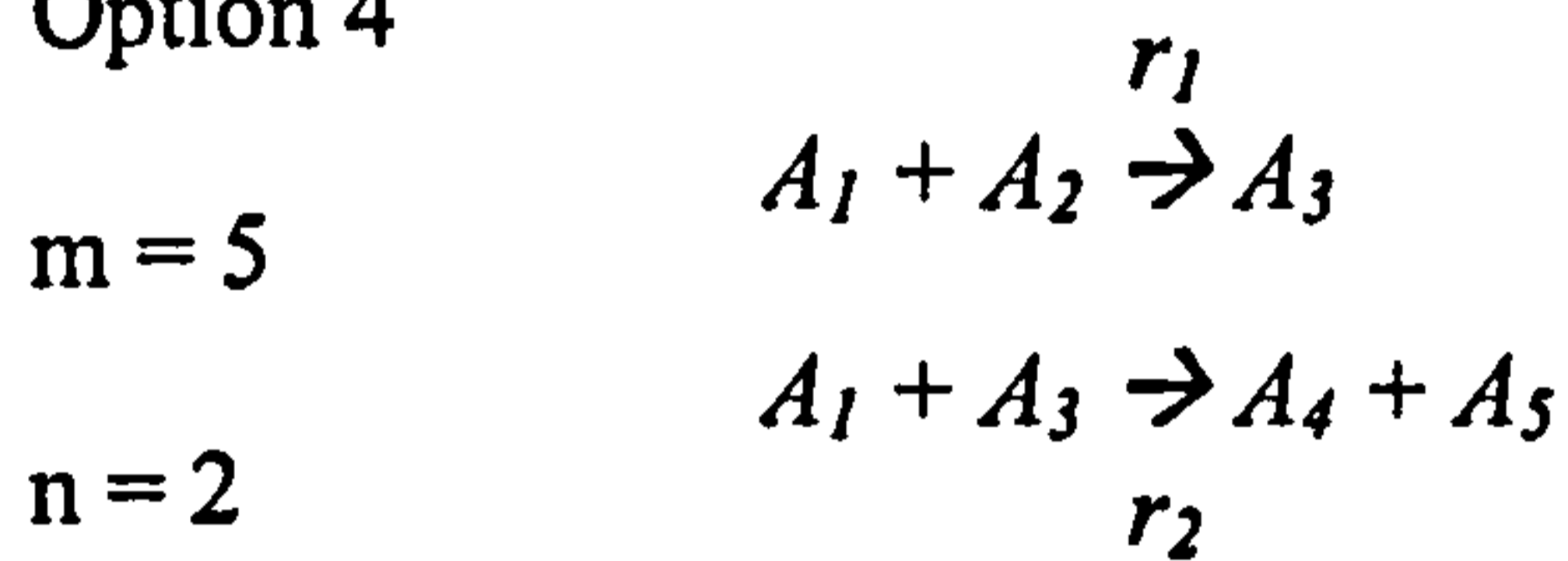
$$\frac{dC_1}{dt} = -r_1 + r_2 \quad (3.96)$$

$$\frac{dC_2}{dt} = r_1 - r_2 - r_3 \quad (3.97)$$

$$\frac{dC_3}{dt} = r_3 \quad (3.98)$$

3.2.5.4 Complex reaction

Option 4



Model equations that will be generated ($2n + m + 5 = 14$):

$$\frac{dC_1}{dt} = v_{11}r_1 + v_{21}r_2 \quad (3.99)$$

$$\frac{dC_2}{dt} = v_{12}r_1 + v_{22}r_2 \quad (3.100)$$

$$\frac{dC_3}{dt} = v_{13}r_1 + v_{23}r_2 \quad (3.101)$$

$$\frac{dC_4}{dt} = v_{14}r_1 + v_{24}r_2 \quad (3.102)$$

$$\frac{dC_5}{dt} = v_{15}r_1 + v_{25}r_2 \quad (3.103)$$

$$r_1 = k_1 C_1^{\beta_{11}} C_2^{\beta_{21}} C_3^{\beta_{31}} C_4^{\beta_{41}} C_5^{\beta_{51}} \quad (3.104)$$

$$r_2 = k_2 C_1^{\beta_{12}} C_2^{\beta_{22}} C_3^{\beta_{32}} C_4^{\beta_{42}} C_5^{\beta_{52}} \quad (3.105)$$

$$k_1 = k_{10} \exp\left(-\frac{E_1}{RT}\right) \quad (3.106)$$

$$k_2 = k_{20} \exp\left(-\frac{E_2}{RT}\right) \quad (3.107)$$

$$\frac{dT}{dt} = \frac{Q_r - Q_m}{c_p V \rho} \quad (3.108)$$

$$Q_r = -r_1 (AH_{r1})V - r_2 (AH_{r2})V \quad (3.109)$$

$$Q_m = U_i A_i (T - T_m) \quad (3.110)$$

$$\frac{dT_m}{dt} = \frac{Q_m - Q_J}{c_{pm} V_m \rho_m} \quad (3.111)$$

$$Q_J = U_o A_o (T_m - T_J) \quad (3.112)$$

Accordingly, the values of parameters that must be supplied are

($3n + 2mn + 11 = 37$):

$v_{11}, v_{21}, v_{12}, v_{22}, v_{13}, v_{23}, v_{14}, v_{24}, v_{15}, v_{25}, \beta_{11}, \beta_{21}, \beta_{31}, \beta_{41}, \beta_{51}, \beta_{12}, \beta_{22}, \beta_{32}, \beta_{42}, \beta_{52}, E_1,$
 $E_2, R, k_{10}, k_{20}, c_p, V, \rho, AH_{r1}, AH_{r2}, U_i, A_i, U_o, A_o, c_{pm}, V_m, \rho_m$

Here the values of v_{11}, v_{12}, v_{21} and v_{23} are equal to -1 . The values of v_{13}, v_{24} and v_{25} are equal to 1 and the rest of stoichiometric coefficients values are zero.

So Equations 3.99 to 3.103 will become:

$$\frac{dC_1}{dt} = -r_1 - r_2 \quad (3.113)$$

$$\frac{dC_2}{dt} = -r_1 \quad (3.114)$$

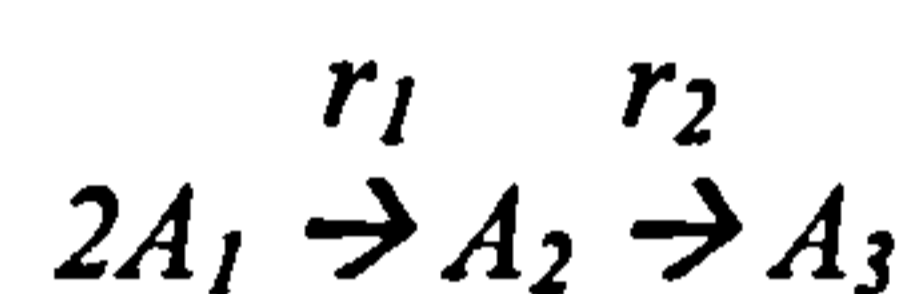
$$\frac{dC_3}{dt} = r_1 - r_2 \quad (3.115)$$

$$\frac{dC_4}{dt} = r_2 \quad (3.116)$$

$$\frac{dC_5}{dt} = r_2 \quad (3.117)$$

3.2.5.5 Consecutive and parallel reaction

Option 5



m = 6



Model equations that will be generated ($2n + m + 6 = 22$):

$$\frac{dC_1}{dt} = v_{11}r_1 + v_{21}r_2 + v_{31}r_3 + v_{41}r_4 + v_{51}r_5 \quad (3.118)$$

$$\frac{dC_2}{dt} = v_{12}r_1 + v_{22}r_2 + v_{32}r_3 + v_{42}r_4 + v_{52}r_5 \quad (3.119)$$

$$\frac{dC_3}{dt} = v_{13}r_1 + v_{23}r_2 + v_{33}r_3 + v_{43}r_4 + v_{53}r_5 \quad (3.120)$$

$$\frac{dC_4}{dt} = v_{14}r_1 + v_{24}r_2 + v_{34}r_3 + v_{44}r_4 + v_{54}r_5 \quad (3.121)$$

$$\frac{dC_5}{dt} = v_{15}r_1 + v_{25}r_2 + v_{35}r_3 + v_{45}r_4 + v_{55}r_5 \quad (3.122)$$

$$\frac{dC_6}{dt} = v_{16}r_1 + v_{26}r_2 + v_{36}r_3 + v_{46}r_4 + v_{56}r_5 \quad (3.123)$$

$$r_1 = k_1 C_1^{\beta_{11}} C_2^{\beta_{21}} C_3^{\beta_{31}} C_4^{\beta_{41}} C_5^{\beta_{51}} C_6^{\beta_{61}} \quad (3.124)$$

$$r_2 = k_2 C_1^{\beta_{12}} C_2^{\beta_{22}} C_3^{\beta_{32}} C_4^{\beta_{42}} C_5^{\beta_{52}} C_6^{\beta_{62}} \quad (3.125)$$

$$r_3 = k_3 C_1^{\beta_{13}} C_2^{\beta_{23}} C_3^{\beta_{33}} C_4^{\beta_{43}} C_5^{\beta_{53}} C_6^{\beta_{63}} \quad (3.126)$$

$$r_4 = k_4 C_1^{\beta_{14}} C_2^{\beta_{24}} C_3^{\beta_{34}} C_4^{\beta_{44}} C_5^{\beta_{54}} C_6^{\beta_{64}} \quad (3.127)$$

$$r_5 = k_5 C_1^{\beta_{15}} C_2^{\beta_{25}} C_3^{\beta_{35}} C_4^{\beta_{45}} C_5^{\beta_{55}} C_6^{\beta_{65}} \quad (3.128)$$

$$k_1 = k_{10} \exp\left(-\frac{E_1}{RT}\right) \quad (3.129)$$

$$k_2 = k_{20} \exp\left(-\frac{E_2}{RT}\right) \quad (3.130)$$

$$k_3 = k_{30} \exp\left(-\frac{E_3}{RT}\right) \quad (3.131)$$

$$k_4 = k_{40} \exp\left(-\frac{E_4}{RT}\right) \quad (3.132)$$

$$k_5 = k_{50} \exp\left(-\frac{E_5}{RT}\right) \quad (3.133)$$

$$\frac{dT}{dt} = \frac{Q_r - Q_m}{c_p V \rho} \quad (3.134)$$

$$Q_r = -r_1(AH_{r1})V - r_2(AH_{r2})V - r_3(AH_{r3})V - r_4(AH_{r4})V - r_5(AH_{r5})V \quad (3.135)$$

$$Q_m = U_i A_i (T - T_m) \quad (3.136)$$

$$\frac{dT_m}{dt} = \frac{Q_m - Q_J}{c_{pm} V_m \rho_m} \quad (3.137)$$

$$Q_J = U_o A_o (T_m - T_J) \quad (3.138)$$

$$\frac{dT_J}{dt} = \frac{F_J}{V_J} (T_{J0} - T_J) + \frac{Q_J}{c_{pJ} V_J \rho_J} \quad (3.139)$$

Accordingly, the values of parameters that must be supplied are

($3n + 2mn + 16 = 91$):

$v_{11}, v_{21}, v_{31}, v_{41}, v_{51}, v_{12}, v_{22}, v_{32}, v_{42}, v_{52}, v_{13}, v_{23}, v_{33}, v_{43}, v_{53}, v_{14}, v_{24}, v_{34}, v_{44}, v_{54}, v_{15},$
 $v_{25}, v_{35}, v_{45}, v_{55}, v_{16}, v_{26}, v_{36}, v_{46}, v_{56}, \beta_{11}, \beta_{21}, \beta_{31}, \beta_{41}, \beta_{51}, \beta_{61}, \beta_{12}, \beta_{22}, \beta_{32}, \beta_{42}, \beta_{52},$
 $\beta_{62}, \beta_{13}, \beta_{23}, \beta_{33}, \beta_{43}, \beta_{53}, \beta_{63}, \beta_{14}, \beta_{24}, \beta_{34}, \beta_{44}, \beta_{54}, \beta_{64}, \beta_{15}, \beta_{25}, \beta_{35}, \beta_{45}, \beta_{55}, \beta_{65}, E_1, E_2,$
 $E_3, E_4, E_5, R, k_{10}, k_{20}, k_{30}, k_{40}, k_{50}, c_p, V, \rho, AH_{r1}, AH_{r2}, AH_{r3}, AH_{r4}, AH_{r5} U_i, A_i, c_{pm}, V_m,$
 $\rho_m, U_o, A_o, V_J, T_{J0}, c_{pJ}, V_J, \rho_J$

Here the values of v_{11} and v_{52} are equal to -2. The values of v_{22}, v_{32} and v_{42} are equal to -1, the values of $v_{12}, v_{23}, v_{34}, v_{45}$ and v_{56} are equal to 1 and the rest of stoichiometric coefficients values are zero.

So Equations 3.118 to 3.123 will become:

$$\frac{dC_1}{dt} = -2r_1 \quad (3.140)$$

$$\frac{dC_2}{dt} = r_1 - r_2 - r_3 - r_4 - 2r_5 \quad (3.141)$$

$$\frac{dC_3}{dt} = r_2 \quad (3.142)$$

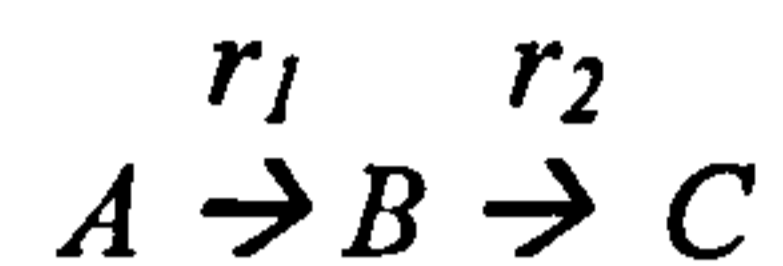
$$\frac{dC_4}{dt} = r_3 \quad (3.143)$$

$$\frac{dC_5}{dt} = r_4 \quad (3.144)$$

$$\frac{dC_6}{dt} = r_5 \quad (3.145)$$

3.2.6 Simulation Results

In this section, some simulation results using the GBRM are presented. The reaction scheme is:



where A is the reactant, B is the product and C is the by-product. This reaction is same as that presented in section 3.2.5.1 i.e. consecutive reaction with number of components, $m=3$ and number of reactions, $n=2$. Here the values of β_{11} and β_{22} are equal to 1. The remaining values of β_{ij} are equal to zero. The initial values of A, B and C are 0.975, 0.025 and 0.0 respectively.

The simulation of batch reactor is carried out using the GBRM with all the available options (1-5). The batch time is 3.5 hours for all cases and the following variables were bounded to a certain values.

Option 1 – reactor temperature

Option 2 – jacket temperature

Option 3 – jacket flow rate

Option 4 – jacket temperature

Option 5 – jacket flow rate

Table 3.1: Test results from GBRM

Option	Simulation Results (conversion to product B)	
	GBRM	Problem specific model
1	0.6631	0.6631
2	0.6643	0.6643
3	0.6409	0.6409
4	0.6651	0.6651
5	0.6521	0.6521

The results achieved are then verified through simulation using the problem specific model. The results are shown in Table 3.1.

Table 3.1 shows that in all cases the results achieved from GBRM is as accurate as the results achieved from the specific case model. This shows the reliability of the developed GBRM.

Further tests have been carried out to demonstrate the flexibility of GBRM for different values of β_{ij} and ν_{ji} . The simulation results obtained are presented in Table 3.2.

Table 3.2: Simulation results achieved from GBRM with different β_{ij} and ν_{ji} values

Case	Values of β_{ij}			Values of ν_{ji}						Resulting Reaction scheme	Product B
	β_{11}	β_{12}	β_{22}	ν_{11}	ν_{21}	ν_{12}	ν_{22}	ν_{13}	ν_{23}		
1	1.0	0.0	1.0	-1.0	0.0	1.0	-1.0	0.0	1.0	$A \rightarrow B \rightarrow C$	0.6631
2	3.0	0.0	2.0	-1.0	0.0	1.0	-1.0	0.0	1.0	$A \rightarrow B \rightarrow C$	0.4911
3	1.0	0.5	1.5	-1.0	0.0	1.0	-1.0	0.0	1.0	$A \rightarrow B \rightarrow C$	0.5752
4	1.0	0.0	1.0	-2.0	0.0	1.0	-1.0	0.0	1.0	$2A \rightarrow B \rightarrow C$	0.4042
5	1.0	0.0	1.0	-1.0	-1.0	1.0	0.0	0.0	1.0	$A \rightarrow B$ $A \rightarrow C$	0.7542
6	1.0	0.0	1.0	-1.0	-2.0	1.5	0.0	0.0	1.0	$A \rightarrow 1.5B$ $2A \rightarrow C$	0.9342

In Table 3.2, Case 1 is similar to option 1 of Table 3.1. Cases 2 – 3 refer to the case where the value of β_{ij} changes meaning different order of reaction rate. Cases 4 – 6 refer to the case where the value of ν_{ji} changes (generating different reaction scheme). These tests have been carried out just to show how this GBRM can easily be used to solve different types of reaction schemes with different kinetic information.

3.3 Summary and Conclusions

This chapter mainly deals with the dynamic modelling of batch reactors based on the mechanistic first-principle-model. To summarise, the items presented in this chapter are:

1. Modelling issues, its basic principle and classifications.
2. The development of the dynamic modelling of batch reactor based on the white-boxed-driven-model.
3. The development of a general computer program which will generate the dynamic model of batch reactor and Jacobian matrix in sparse form (using the information of the locations of non-zero elements in the Jacobian matrix).
4. Examples to show how the general equations of batch reactor can be generated.
5. Simulation results using the GBRM and problem specific model. In addition, tests are presented to show the flexibility of the GBRM.

From Chapter 2, it was found that it was essential to develop a computer program, which can develop the required dynamic models of batch reactors. It is because different types of reactions will end up with different types of models. Tasks to develop these models for different cases is time consuming. For this reason, here we developed a computer program (GBRM) which will generate the required dynamic model of batch reactors using limited information supplied by the user.

Chapter Four

4.0 Dynamic Optimisation (Optimal Control)

4.1 Introduction

Typical problems in chemical engineering process design or plant operation can be represented by some equations and they have many, and possibly an infinite number of solutions. An optimisation is concerned with selecting the best among the entire set by efficient quantitative methods. The goal of the optimisation is to find the values of the variables in the process that yields the best value of the performance criterion.

Engineers work to improve the initial design of equipment, and strive for enhancements in the operation of the equipment once it is installed in order to maximise the production, minimise the operation time, maximise the profit, minimise cost, minimise the energy usage and so on. The use of monetary value provides a convenient measure of the compatible objectives, however not all problems have to be considered in monetary (cost versus revenue) terms.

In plant operations, benefits arise from improved plant performance, such as improved yields of valuable products (or reduced yields of contaminants), reduced energy consumption, higher processing rates, and reduced period between shutdowns. Optimisation can also lead to reduced maintenance costs, less equipment wear, and better staff utilisation. In addition, intangible benefits arise from the interactions among plant operators, engineers and management. It is extremely helpful to systematically identify the objective, constraints, and degrees of freedom in a process or plant, leading to benefits such as improved quality of design, faster and more reliable trouble-shooting and faster decision-making.

Optimisation can be applied in numerous ways to chemical processes and plants. Typical projects where optimisation has been used include (Edgar and Himmelblau, 1989):

1. Determination of the best sites for plant location
2. Routing of tankers for distribution of crude and refined products
3. Pipeline sizing and layout
4. Equipment and entire plant design
5. Maintenance and equipment replacement scheduling
6. Operation of equipment such as reactors, columns, absorbers etc.
7. Evaluation of plant data to construct model of a process
8. Minimisation of inventory charges
9. Allocation of resources or services among several processes
10. Planning and scheduling of construction

Every optimisation problem contains three essential categories:

1. At least one objective function to be optimised (e.g. profit function, cost function etc.) often called the economic model
2. Equality constraints (e.g. process model equations)
3. Inequality constraints (e.g. lower and upper bounds of operating variables, such as temperature in a reactor)

By a feasible solution of the optimisation problem we mean a set of variables that satisfy categories 2 and 3 to the desired degree of precision. An optimal solution is a set of values of the variables that satisfy the components of categories 2 and 3 and also provides an optimal value for the function in category 1.

No single method or algorithm of optimisation exists that can be applied efficiently to all problems. The method chosen for any particular case will depend primarily on:

1. The character of the objective function and whether it is known explicitly
2. The nature of the constraints
3. The number of independent and dependent variables

The general objective of optimisation is to choose a set of values of the variables subject to the various constraints that will produce the desired optimum response for the chosen objective function.

In batch reactor, the optimisation problem can be classified into three general categories:

- P1. Maximum Conversion Problem – The operation time is fixed a priori.
- P2. Minimum Time Problem – The conversion is fixed a priori.
- P3. Maximum Profit or Productivity Problem – None of the batch time and conversion are fixed but both are optimised.

4.2 Classical Formulation of Dynamic Optimisation (Optimal Control)

Problems

The classical optimal control problem is well stated by Bryson and Ho (1975). It has the basic unconstrained form:

Find $u(t)$ which minimises an objective, J

$$J = F_0(x(t_f), t_f) + \int_{t_0}^{t_f} L(x(t), u(t), t) dt \quad (4.2.1)$$

where $x(t)$ is determined by a system of ODE's

$$x'(t) = f(x(t), u(t), t), \quad x(t_0) = x_0, \quad t \in [t_0, t_f] \quad (4.2.2)$$

and $x(t)$ is an n -dimensional vector, $u(t)$ is an m -dimensional vector and $f(x, u, t)$ is an n -dimensional vector function (or more concisely: $x(t) \in R^n$, $u(t) \in R^m$, $f: R^n \times R^m \times R \rightarrow R^n$) and $x'(t)$ is the derivative of $x(t)$ with respect to t .

This basic problem can be constrained in a number of ways:

Terminal constraints (end-point or final time constraints)

$$F(x(t_f), t_f) \geq 0 \quad (4.2.3)$$

Interior-point constraints

$$F(x(t_1), t_1) \geq 0, \quad t_0 \leq t_1 \leq t_f \quad (4.2.4)$$

Control constraints, which are usually simple bounds

$$C(u(t), t) \geq 0 \quad (4.2.5)$$

Path constraints (state or phase constraints)

$$g(x(t), u(t), t) \geq 0 \quad (4.2.6)$$

where $u(t)$ may or may not appear explicitly.

4.3 Solution of Dynamic Optimisation Problems

It has been mentioned in Chapter 2 that there are a large variety of computational methods available for solving dynamic optimisation problem presented in section 4.2. Some of the methods are based on numerically satisfying the necessary conditions for optimality of (4.2.1) subject to (4.2.2), while others involve more direct search algorithms. However, due to merits that have been discussed in the literature review, we choose the control vector parameterisation (CVP) technique to be applied in this work.

4.3.1 Formulation of Optimal Control Problem as A Non-linear Programme

Due to the inherent dynamic nature in batch reactors system, their modelling will result to differential and algebraic equations (DAEs) as shown in the previous chapter. The DAEs can be represented in the form of:

$$f(t, \underline{x}'(t), \underline{x}(t), \underline{u}(t), \underline{v}) = 0, \quad [t_0, t_f] \quad (4.3.1)$$

where t is the independent variable, $\underline{x}(t) \in R^n$ is the set of all state variables, $\underline{x}'(t)$ denotes the derivatives of $\underline{x}(t)$ with respect to time, $\underline{u}(t) \in R^m$ is a vector of control variables such as reactor temperature and jacket flow rate, and \underline{v} is a vector of time invariant parameters (design variables) such as volume of the reactor. Suitable initial

conditions $x(t)$ are defined at time $t = t_0$. The time interval of interest is $[t_0, t_f]$ and the function $f: R \times R^n \times R^n \times R^m \times R^p \rightarrow R^n$ is assumed to be continuously differentiable with respect to all its arguments.

The system is subject to bounds on the controls:

$$a^u(t) \leq u(t) \leq b^u(t), \quad t \in [t_0, t_f] \quad (4.3.2)$$

where $a^u(t)$ and $b^u(t)$ are given continuous function of time on $[t_0, t_f]$ and interior point or terminal constraints at time t_p of the form:

$$a^F \leq F(t_p, \underline{x}'(t_p), \underline{x}(t_p), \underline{u}(t_p), \underline{v}) \leq b^F \quad (4.3.3)$$

where $F(t, \underline{x}', \underline{x}, \underline{u}, \underline{v}) \in R^n$ and is continuously differentiable with respect to its arguments. At the terminal point $t_p = t_f$. The system performance is measured in terms of a scalar objective function to be minimised:

$$J = F(t_f, \underline{x}'(t_f), \underline{x}(t_f), \underline{u}(t_f), \underline{v}) \quad (4.3.4)$$

The optimal control problem is to choose an admissible set of controls $u(t)$, and final time, t_f , to minimise the objective function, J , subject to the bounds on the controls and constraints.

4.3.2 Control Vector Parameterisation (CVP) Technique

In order to pose the optimal control problem as a non-linear programming problem, this approach (Morison, 1984; Vassiliadis, 1993; Mujtaba and Macchietto, 1998; Sharif, 1999) applies parameterisation to the control variables $u(t)$ only. They are approximated by a finite dimensional representation. The time interval $[t_0, t_f]$ is divided

into a finite number of subintervals, each with a set of basis functions involving a finite number of parameters:

$$u(t) = \Phi^j(t, z_j), \quad t \in (t_{j-1}, t_j), \quad j = 1, 2, \dots, J \quad (4.3.5)$$

where $t_J = t_f$. The function $\Phi^j(t, z_j)$ is assumed to be continuously differentiable with respect to t and z_j , and the derivatives are uniformly bounded. The control is thus defined by the parameters, z_j and the switching time $t_j, j = 1, 2, \dots, J$.

The control constraints now become:

$$a^u(t) \leq \Phi^j(t, z_j) \leq b^u(t), \quad t \in (t_{j-1}, t_j), j = 1, 2, \dots, J \quad (4.3.6)$$

The set of decision variables for the non-linear program can now be written as:

$$y = \{z_1, z_2, \dots, z_J, t_1, t_2, \dots, t_J\} \quad (4.3.7)$$

4.4 Optimisation Problem Formulation in Batch Reactors

In general, the optimisation problem presented earlier can be written as:

Find the set of decision variables, y (Equation 4.3.7) which

minimise $J(y)$

subject to Equality constraints [Equation (4.3.1)]

Inequalities constraints [Equations (4.3.3) and (4.3.6)]

The various optimisation problems for batch reactors mentioned earlier can be detailed as follows:

P1 – Maximum Conversion Problem

The problem can be described as:

Given	the fixed volume of the reactor and the batch time
Optimise	the jacket flow or jacket temperature profile or reactor temperature profile
So as to maximise	the conversion of the desired product
Subject to	any constraints bounds on the jacket flow, bounds on the jacket temperature, bounds on the reactor temperature

Mathematically the optimisation problem can be written as:

$$\begin{array}{ll}
 \text{Max} & X \\
 F_J(t) \text{ or } T_J(t) \text{ or } T(t) & \\
 \text{s.t} & f(t, \underline{x}'(t), \underline{x}(t), \underline{u}(t), \underline{v}) = 0 \text{ (process model)} \\
 & t_f = t_f^* \\
 & F_{JL} \leq F_J \leq F_{JU} \text{ or } T_{JL} \leq T_J \leq T_{JU} \text{ or } T_L \leq T \leq T_U
 \end{array}$$

where:

X is the conversion of the limiting reactant to the desired product

T is the reactor temperature

F_J is the jacket flow

t_f is the batch time

F_{JL} and F_{JU} are the lower and upper bounds of the jacket flow

T_{JL} and T_{JU} are the lower and upper bounds of the jacket temperature

T_L and T_U are the lower and upper bounds of the reactor temperature

t_f^* is the fixed batch time

P2 – Minimum Time Problem

The problem can be described as:

Given the fixed volume of the reactor and the conversion to the desired product

Optimise the jacket flow or jacket temperature profile or reactor temperature profile

So as to minimise the batch time

Subject to any constraints

 bounds on the jacket flow, bounds on the jacket temperature, bounds on the reactor temperature

Mathematically the optimisation problem can be written as:

$$\begin{aligned} \text{Min} & \quad t_f \\ & F_J(t) \text{ or } T_J(t) \text{ or } T(t) \\ \text{s.t} & \quad f(t, \underline{x}'(t), \underline{x}(t), \underline{u}(t), \underline{v}) = 0 \text{ (process model)} \\ & \quad X = X^* \\ & \quad F_{JL} \leq F_J \leq F_{JU} \text{ or } T_{JL} \leq T_J \leq T_{JU} \text{ or } T_L \leq T \leq T_U \end{aligned}$$

where:

X^* is the fixed conversion

t_f is the batch time

P3 – Maximum Profit or Productivity Problem

The problem can be described as:

Given the fixed volume of the reactor
 Optimise the jacket flow or jacket temperature profile or reactor temperature profile
 So as to maximise the profit or productivity (can be expressed in terms of conversion to the desired product and batch time)
 Subject to any constraints
 bounds on the jacket flow, bounds on the jacket temperature, bounds on the reactor temperature

Mathematically the optimisation problem can be written as:

$$\begin{array}{ll}
 \text{Max} & P \\
 F_J(t) \text{ or } T_J(t) \text{ or } T(t) & \\
 \text{s.t} & f(t, \underline{x}'(t), \underline{x}(t), \underline{u}(t), \underline{v}) = 0 \text{ (process model)} \\
 & F_{JL} \leq F_J \leq F_{JU} \text{ or } T_{JL} \leq T_J \leq T_{JU} \text{ or } T_L \leq T \leq T_U
 \end{array}$$

where P is the profit or productivity function.

These constrained non-linear optimisation problems can be solved using efficient successive quadratic programming (SQP) algorithm (Chen, 1988). The details of this algorithm can be found in Appendix A.

Each 'function evaluation' of the optimiser requires a full integration of DAE system [Equation (4.3.1)] and this is achieved using an efficient Gear's-type integrator. Solution of the optimisation problem requires the derivatives of the objective function and constraints with respect to all the arguments of y [Equation (4.3.7)]. These are evaluated in an efficient way using adjoint variables (Morison, 1984). Figure 4.1

illustrates a typical computational sequence for the solution of the optimisation problem used in this work.

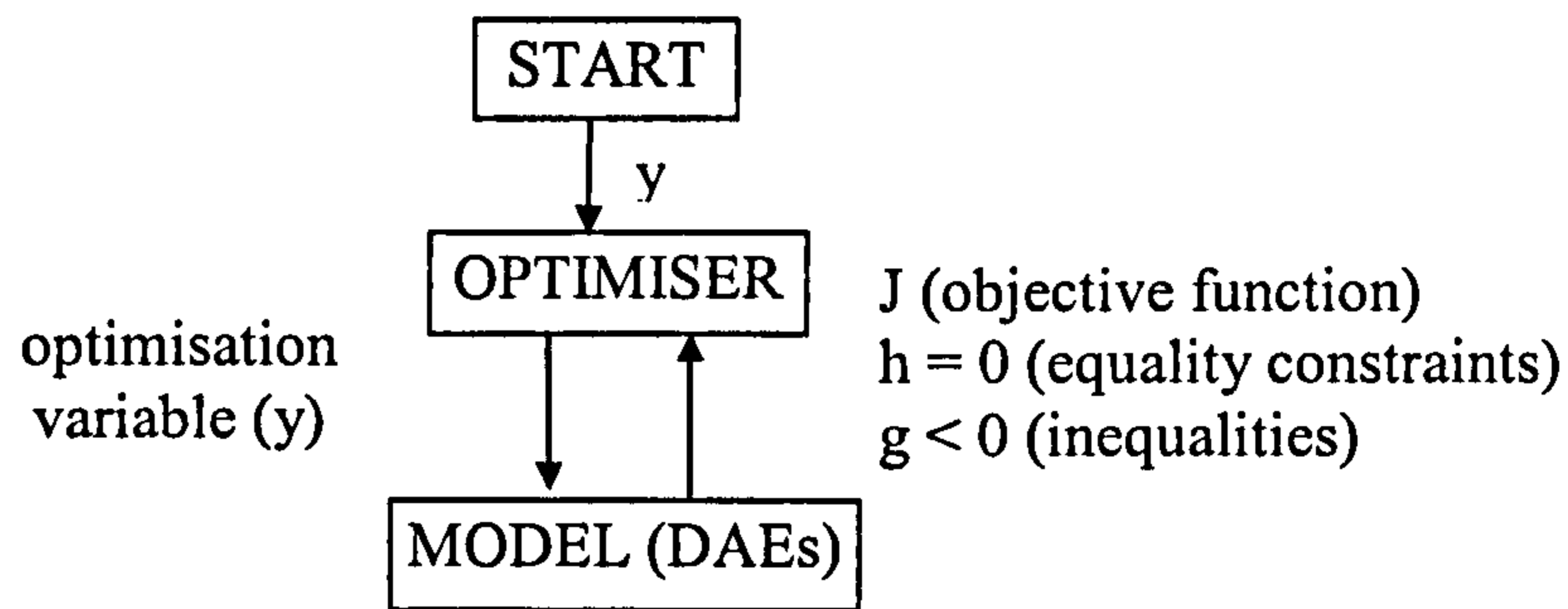


Figure 4.1: Computational Sequence of Dynamic Optimisation Problem

4.5 Case Studies

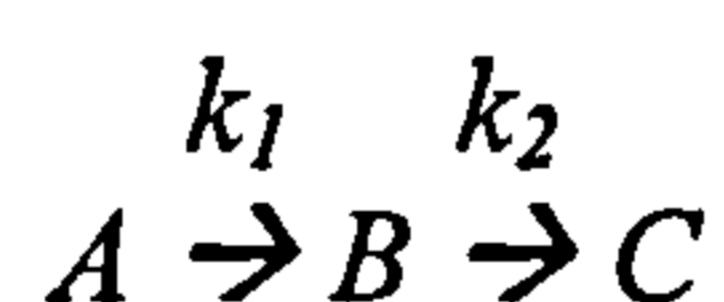
Six case studies are presented in this section with different aims and objectives for each case. The aim of Case Studies 1 and 2 are to solve the maximum conversion problem (P1) and the minimum time problem (P2) for two different reaction schemes. Another aim of these studies is to compare the results achieved with those from the literature (solved by iterative dynamic programming (IDP), Luus, 1994). This will demonstrate the efficiency of the CVP technique compared to IDP technique. Case Study 3 is carried out to tackle path constraint (i.e. batch reactor should not go beyond a safety limit, see section 4.5.3.1) problem. Here the method of Vassiliadis et al. (1994b) is implemented but is solved with new solution approach (the problem is solved sequentially). In Case Study 4, an exothermic semi-batch reaction is presented. Here, we solve all types of optimisation problems (P1, P2, P3) mentioned in section 4.4. The aim of this study is to analyse and evaluate each type of optimisation problems and to find the situations when each of these problems results to maximum benefit. In Case Study 5, a sensitivity analysis of the profit function in maximum profit problem in batch

reactor is studied. How the prices of reactant, product, by-product, operating cost and reactor volume (capital cost) influence the overall profit for a given reaction scheme is analysed in this case study. Finally (Case Study 6), a maximum conversion problem with fixed time interval for a complex reaction is carried out. The aim of this case study is to provide dynamic set points in terms of reactor temperature for its on-line tracking by various types of controllers in Chapter 5. In other word, how the results of the off-line optimisation (in Case Study 6) can be implemented in the on-line operation will be shown in the next chapter.

4.5.1 Case Study 1

(Part of this work was presented at the IChemE Symposium on Advances in Process Control 5, University of Wales, Swansea, 2-3 September 1998, Aziz and Mujtaba, 1988)

The non-linear batch reaction (Ray, 1981) used for optimisation studies by Logsdon and Biegler (1993) and Luus (1994) is chosen in this study. The reaction scheme is:



where B is the desired product. This is one of the classical batch reactor control problems. Both temperature and batch time will influence the selectivity and conversion (yield) of the desired product. The general reaction scheme shown above is of a considerable practical importance in a number of chemical processing operations, such as the oxidation of HCs or the chlorination of aromatics.

In their optimisation studies, Logsdon and Biegler (1993) used orthogonal collocation method with a relaxed reduced space SQP method and Luus (1994) used an

iterative dynamic programming (IDP) method in order to obtain optimal temperature profiles to maximise the yield (they solved the maximum conversion problem only). In this work, both maximum conversion and minimum time problems are solved and the results achieved are compared with those obtained by above-mentioned researchers.

4.5.1.1 Model Equations

We use the simple model involving only mass balance and reaction kinetics (as was used by the previous researchers). The conversion to B from A follows a second order reaction rate while conversion to C from B follows a first order reaction rate. The reactor volume is assumed constant.

Based on the general model for batch reactor (GBRM) that has been discussed in Chapter 3, this type of reaction is of option 1 (plain reactor - energy balance is not considered). Here, number of component, $m = 3$ and number of reaction, $n = 2$. So the sets of equation generated are:

Component A

Refer to the Equations 3.27 – 3.29 in Chapter 3 (Option 1 model):

The values of ν_{11} and ν_{22} are equal to -1 ; the values of ν_{12} and ν_{23} are equal to 1 and the rest of ν_{ji} are equal to zero. The values of β_{11} and β_{22} are equal to 2 and 1 respectively.

The rest of β_{ij} are equal to zero. Equations 3.27 – 3.29 result to:

Mass Balance:

$$\frac{dC_A}{dt} = -r_A \quad (4.5.1.1)$$

Reaction rate:

$$r_A = k_1 C_A^2 \quad (4.5.1.2)$$

Combining Equations (4.5.1.1 – 4.5.1.2):

$$\frac{dC_A}{dt} = -k_1 C_A^2 \quad (4.5.1.3)$$

Component B

Similar to component A

$$\frac{dC_B}{dt} = k_1 C_A^2 - k_2 C_B \quad (4.5.1.4)$$

where the reaction rate constants are:

$$k_1 = 4000 \exp\left(-\frac{2500}{T}\right) \quad (4.5.1.5)$$

$$k_2 = 6.2 \times 10^5 \exp\left(-\frac{5000}{T}\right) \quad (4.5.1.6)$$

The final set of model equations is from Equations (4.5.1.3 – 4.5.1.6), which does not contain any design parameters (e.g. reactor volume, reactor jacket cooling/heating configuration etc.). This type of model can be used at the design stage to find optimal reactor temperature profile to achieve the target performance of the reactor. This temperature profile can then be used for detailed design of the reactor including controller design.

4.5.1.2 Degrees of Freedom Analysis

There are 8 variables in Equations (4.5.1.3 – 4.5.1.6) and they are:

$$\frac{dC_A}{dt}, \frac{dC_B}{dt}, k_1, k_2, C_A, C_B, T, t$$

Therefore the degrees of freedom,

$$\begin{aligned} \text{D.F} &= \text{Total number of variables} - \text{total number of equations} \\ &= 8 - 4 = 4 \end{aligned}$$

Note C_A and C_B are differential variables of the model equations. So initial value at $t = 0$ must be given. For this case study $C_A(0) = 1.0$ and $C_B(0) = 0$. t is the independent variable of the model equations. T is used as the control (optimisation) variable and therefore it is relaxed from the specification set and optimised.

4.5.1.3 Results

Maximum Conversion Problem (Problem P1)

The objective is to maximise the conversion of the desired product B while optimising the reactor temperature profile within the specified safe bounds on the temperature.

A fixed batch time of 1 hr, as used by Luus (1994) is used here to compare the results. The reactor temperature is optimised within the bounds: $298 \leq T \leq 398\text{K}$ (same as used by Luus). It is assumed that the optimum temperature profile is obtainable by controlling external heating/cooling (not shown in the reactor description or in the model). A number of cases are presented. Up to 8 time intervals is considered within the total batch operation time. In each interval, the temperature as well as the length of the

interval are optimised. The total number of optimisation variables varies from case to case (15 variables for Run 4). The results are summarised in Table 4.1.

Table 4.1: Summary of the results (Problem P1)

Run	Optimum Temperature Profile	Max Conv, C_B
1	Temperature, K 335.34	0.60595
	Switching time, hr $t=0$ 1.0	
2	Temperature, K 338.58 331.59	0.60748
	Switching time, hr $t=0$ 0.46 1.0	
3	Temperature, K 356.59 334.44 327.00	0.60948
	Switching time, hr $t=0$ 0.15 0.58 1.0	
4	Temperature, K 352.2 342.5 339.8 337.7 335.7 333.5 332.5 328.2	0.60960
	Switching time, hr $t=0$ 0.14 0.24 0.26 0.30 0.36 0.51 0.68 1.0	

Table 4.1 shows that by using one time interval, the conversion achieved after one hour ($t_f = 1$ hr) of reaction is $C_B = 0.60595$. This value is about 1% lower than the value $C_B = 0.61079$ reported by Luus (1994). When three time intervals are used a conversion of $C_B = 0.60948$ is achieved which is much closer to the value reported by Luus (the difference is only about 0.2%). However, the important point is that the minimum length of the time interval is of the order of 10 minutes in our case, which will allow even a manual control. Luus had used 100 piecewise constant time intervals to approximate a continuous temperature profile meaning that every 36s the temperature has to be switched to the next optimum value by the controller. This will require a very sophisticated controller and may not be possible to implement practically (will demand capital investment to change to this type of controller).

However it is also shown in Table 4.1 that when the number of intervals is increased (to 8), the optimum results achieved are better but manual control can be difficult to achieve. Beginning with a larger value the optimal reactor temperature gradually decreases with batch time. Luus (1994) also has reported similar trend in the optimal temperature profile. Conversion and optimal temperature profiles for Run 2 and Run 4 are shown in Figures 4.2 and 4.3.

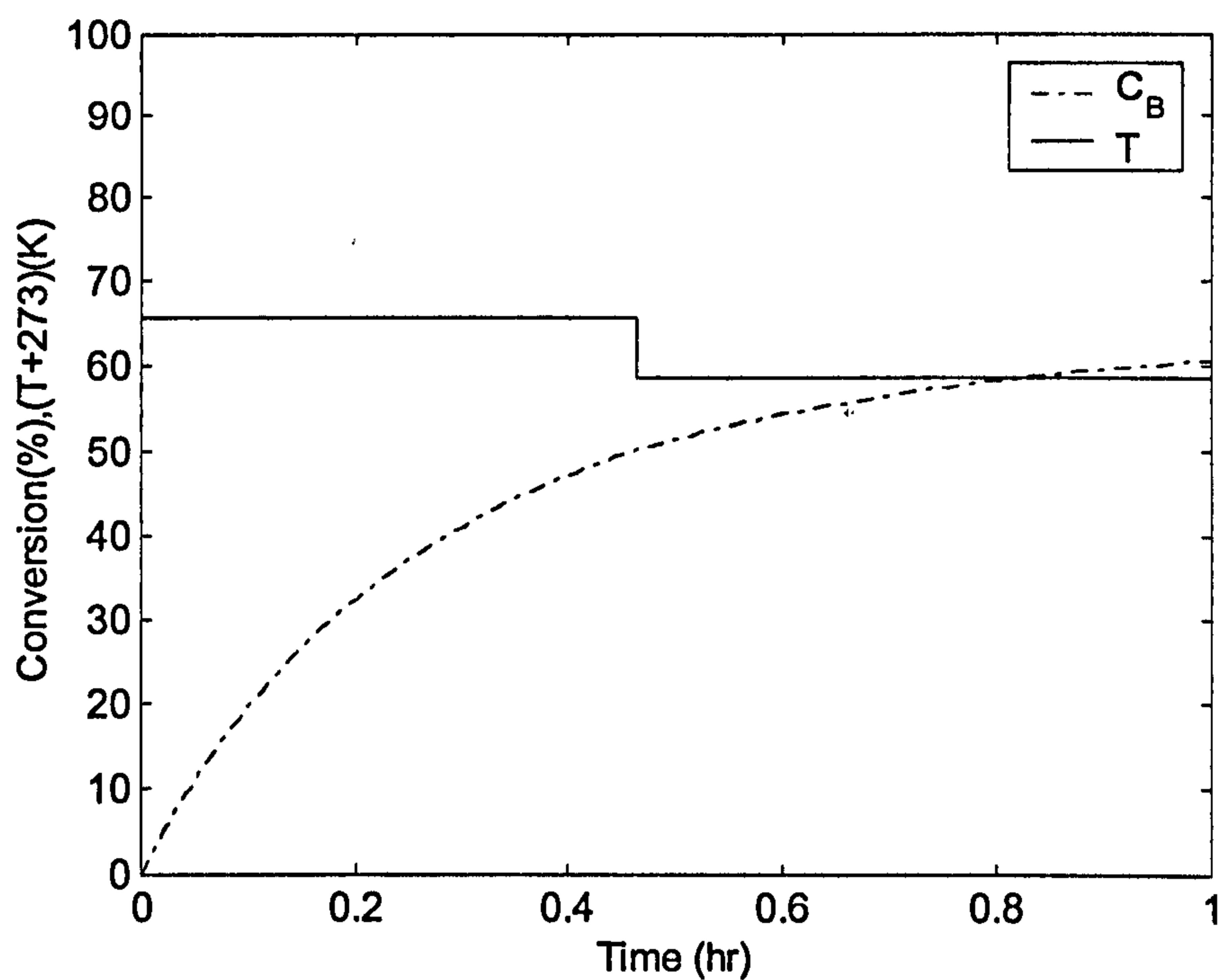


Figure 4.2: Conversion and optimal temperature profiles for Run 2 (P1 problem)

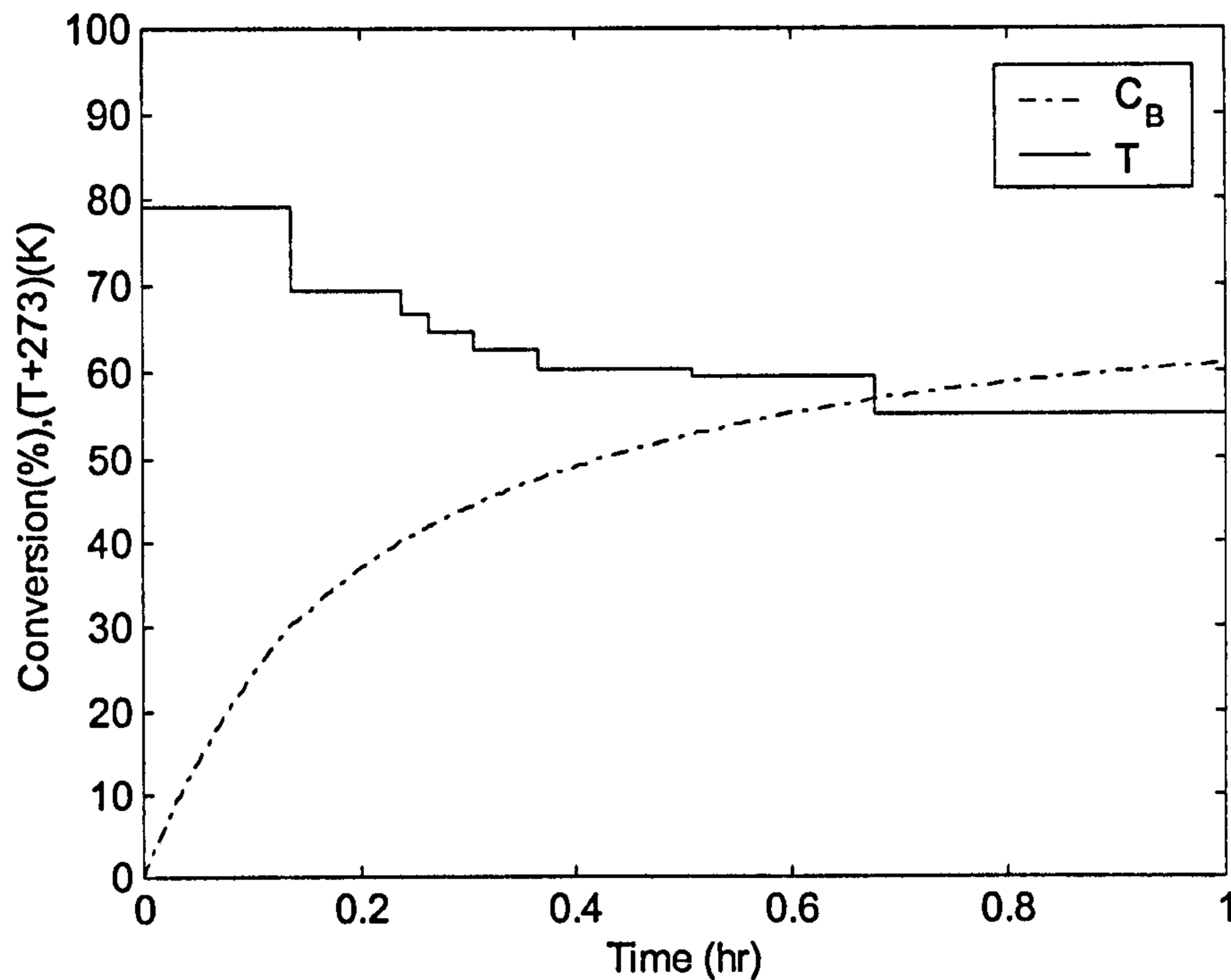


Figure 4.3: Conversion and optimal temperature profiles for Run 4 (P1 problem)

Minimum Time Problem (Problem P2)

The objective is to minimise the batch time for a given conversion to B while optimising the reactor temperature profile within a physical and safe bounds on the temperature. Several cases were run but with different fixed conversion. The bounds on the temperature are the same as before. For Runs 1-3 we considered one time interval which is minimised. Run 4 is presented with three intervals when switching times are optimised while minimising the final time. The optimal temperature profiles, conversion and the minimum batch time are presented in Table 4.2 for all cases.

Table 4.2: Summary of the results (Problem P2)

Run	Optimum Temperature Profile	Conv to B (Fixed)	Min Time	Conv to C
1	Temperature, K 332.10 ----- Switching time, hr $t=0$ 1.19	0.62	1.19	0.0987
2	Temperature, K 320.25 ----- Switching time, hr $t=0$ 1.71	0.65	1.71	0.0856
3	Temperature, K 328.23 ----- Switching time, hr $t=0$ 3.28	0.70	3.28	0.0782
4	Temperature, K 316.19 321.13 298.03 ----- ----- Switching time, hr $t=0$ 0.13 0.26 4.30	0.72	4.30	0.0832

Table 4.2 shows that the minimum batch time increases with the increase in conversion to the desired product, which is obvious. This type of operation may become unavoidable if the reactant is very expensive or if we need to fulfil a strict requirement of conversion to the desired product in the downstream process. Conversion and optimal temperature profiles for Run 4 are shown in Figure 4.4.

At low desired conversion to B (Run 1), there is more room for some B to convert to C. So operating at higher temperature although increases the rate constants (k_1, k_2), desired conversion to B is achieved quicker although produces more C. At high desired conversion to B (compare Runs 2,3,4 to Run 1), there are little rooms for B to convert to C. Therefore, to keep the rate constants lower, the reactor needs to be operated at lower temperature (Run 2,3,4) compared to Run 1. This consequently requires higher minimum batch time.

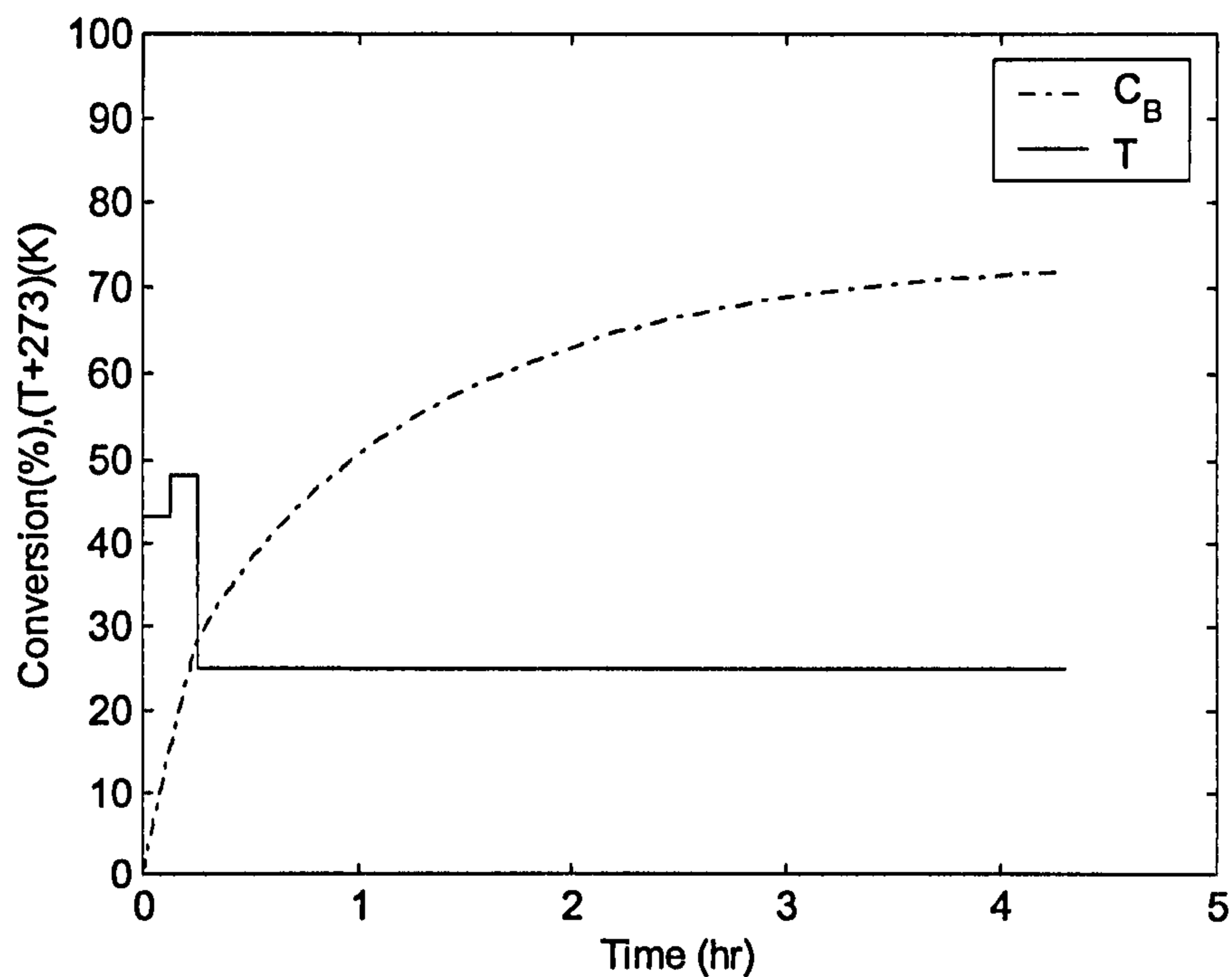
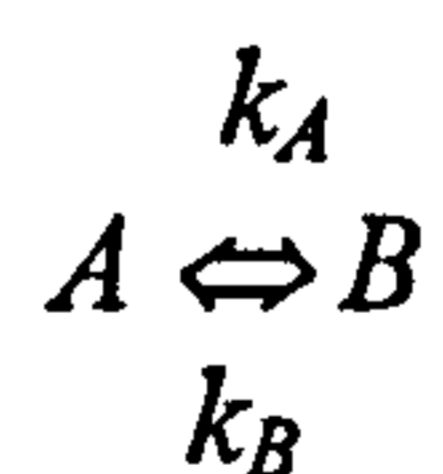


Figure 4.4: Conversion and optimal temperature profiles for Run 4 (P2 problem)

4.5.2 Case Study 2

A first order, reversible and exothermic chemical reaction (Ko and Stevens, 1971; Reddy and Hussain, 1981 and Luus, 1994) is considered. The reaction can be written as:



where A is the reactant and B is the product. k_A and k_B represents the forward and backward reaction rate respectively. This type of reaction is applied in the problem of disposing of exhaust gases and other ore-processing operation.

Generally there are two types of model i.e. shortcut model and detailed model. The former allows determination of optimal reactor temperature profile (to achieve the

desired performance) which can be used for detailed design of the reactor and the latter allows optimising operating conditions for an already designed batch reactors. Here the detailed model consists of mass and energy balances and rate equations are considered. Ko and Stevens (1971) and Reddy and Hussain (1981) applied modified gradient method to reach to the vicinity of the optimum temperature profile. Again, Luus (1994) used an iterative dynamic programming (IDP) method in order to obtain optimal temperature profiles to maximise the yield (they solved the maximum conversion problem only). Here, both maximum conversion and minimum time problems are solved with the CVP technique.

4.5.2.1 Models Equations

Here, the set of equations and the control variable used by the previous researchers are kept similar for the reference purpose. However, they can be derived from the GBRM presented in Chapter 3. This type of reaction is of option 2 (jacketed reactor - energy balance with negligible heat retention is considered). Here, number of component, $m = 2$ and number of reaction, $n = 2$.

The value of ν_{11} and ν_{22} are equal to -1 ; the values of ν_{12} and ν_{21} are equal to 1 and the rest of ν_{ji} are equal to zero. The values of β_{11} and β_{22} are equal to 1. The rest of the β_{ij} are equal to zero. Equations 3.30 – 3.35 then result to:

Mass balance:

$$\frac{dC_A}{dt} = -k_A C_A + k_B C_B$$

$$\frac{dC_B}{dt} = k_A C_A - k_B C_B$$

Given in Ko and Stevens (1971) that $C_{A0} + C_{B0} = 1$ and since the stoichiometric coefficients are equal to 1, it is true that at any time the value of $C_A + C_B = 1$.

Substitute, $C_A = 1 - C_B$ then:

$$\frac{dC_B}{dt} = k_A(1 - C_B) - k_B C_B$$

Since it is given that $C_{A0} + C_{B0} = 1$ (Ko and Stevens, 1971), above equation will

become:

$$\frac{dC_B}{dt} = R(C_B, T) = k_A(C_{A0} + C_{B0} - C_B) - k_B C_B \quad (4.5.2.1)$$

Energy balance (equations 3.33 – 3.35):

$$\frac{dT}{dt} = \frac{Q_r - Q_J}{c_p V \rho}$$

$$Q_r = -r_1(\Delta H_{r1})V - r_2(-\Delta H_{r2})V$$

$$Q_J = UA(T - T_J)$$

Combining all three equations results to:

$$\frac{dT}{dt} = \frac{-r_1(\Delta H_{r1})V - r_2(\Delta H_{r2})V - UA(T - T_J)}{c_p V \rho}$$

Here, the $T_J = T_C$, $U = h$, $A=V=1$ and $-r_1(\Delta H_{r1})V - r_2(\Delta H_{r2})V$ is same as

$-\Delta H \times R(C_B, T)$ [as $\Delta H_{r1} = \Delta H_{r2}$]. After simple manipulation the above equation will

result to:

$$c_p \rho \frac{dT}{dt} = -\Delta H \times R(C_B, T) - h(T - T_C) \quad (4.5.2.2)$$

Let:

$$\frac{h}{c_p \rho} = u$$

Given (Ko and Stevens, 1971):

$$C_{A0} + C_{B0} = 1; \quad \frac{-\Delta H}{c_p \rho} = 300; \quad T_c = 290K$$

Equation (4.5.2.1) becomes:

$$\frac{dC_B}{dt} = k_A(1 - C_B) - k_B C_B \quad (4.5.2.3)$$

and Equation (4.5.2.2) becomes:

$$\frac{dT}{dt} = 300[k_A(1 - C_B) - k_B C_B] - u(T - 290) \quad (4.5.2.4)$$

where

$$k_A = k_{A0} \exp\left(-\frac{E_A}{RT}\right); \quad k_B = k_{B0} \exp\left(-\frac{E_B}{RT}\right)$$

$$k_{A0} = 1.7536 \times 10^5; \quad k_{B0} = 2.4885 \times 10^{10}$$

$$E_A = 1.1374 \times 10^4 \text{ cal/mole}; \quad E_B = 2.2748 \times 10^4 \text{ cal/mole}$$

So:

$$k_A = 1.7536 \times 10^5 \exp\left(-\frac{1.1374 \times 10^4}{1.9872T}\right) \quad (4.5.2.5)$$

and $k_B = 2.4885 \times 10^{10} \exp\left(-\frac{2.2748 \times 10^4}{1.9872T}\right) \quad (4.5.2.6)$

4.5.2.2 Degrees of Freedom Analysis

There are 8 variables in the Equations (4.5.2.3 – 4.5.2.6) and they are:

$$\frac{dC_B}{dt}, \frac{dT}{dt}, k_A, k_B, C_B, T, u, t$$

Therefore the degrees of freedom,

$$D.F = 8 - 4 = 4.$$

Hence we need 4 specifications to satisfy the D.F. Note C_B (conversion to $B = x_I$) and T are differential variables. So initial values at $t = 0$ must be given. The specification are: t is independent variable; $C_B(0) = 0.0$; $T(0) = 380.0$ K. We use u as control variable and therefore this variable is relaxed and optimised.

All optimisations are carried out with:

$$u_{min} \leq u \leq u_{max} \quad (4.5.2.7)$$

where u_{min} and u_{max} are 0 and 0.36 respectively (the lower and upper bounds of u). In this case, u is proportional to the heat transfer coefficient, h which can be controlled by

manipulating the flow rate of coolant. $u_{min} = 0$ corresponds to no cooling while u_{max} corresponds to the maximum allowable coolant flow rate.

4.5.2.3 Results

Maximise Product B in Fixed Batch Time

Here we considered two different batch times of 5 and 25 minutes respectively. These durations are same as those used by the previous researchers. For each batch time, we divided the total time into 2,3 and 4 intervals and we optimise the interval lengths together with u . The results are presented in Table 4.3.

Table 4.3: Summary of the results

Run	Optimal Control Profile	Max Conv, x_f
1a	Control, u ----- ----- 0.0000 0.32650	0.66882
	Switching time, min $t=0$ 1.78 5.0	
1b	Control, u ----- ----- ----- 0.0000 0.36000 0.26277	0.67398
	Switching time, min $t=0$ 1.80 3.29 5.0	
1c	Control, u ----- ----- ----- ----- 0.0000 0.17230 0.36000 0.19082	0.67522
	Switching time, min $t=0$ 1.77 1.87 3.80 5.0	
2a	Control, u ----- ----- 0.12730 0.037091	0.89132
	Switching time, min $t=0$ 12.49 25.0	
2b	Control, u ----- ----- ----- 0.12828 0.07702 0.03467	0.89155
	Switching time, min $t=0$ 11.24 14.42 25.0	
2c	Control, u ----- ----- ----- ----- 0.0000 0.3600 0.09511 0.03099	0.90414
	Switching time, min $t=0$ 1.83 4.29 10.38 25.0	

For batch time of 5 minutes, it is found that the optimum value of conversion achieved with four time intervals is 0.67522 (Run 1c) which is almost the same as those (0.67532, 0.6722) reported by Luus (1994) and Reddy and Hussien (1981) respectively. Again in this case, Luus had used a non-practical time interval, $P = 44$ meaning every 6.82 seconds, the coolant flow rate must be switched to the next optimum value. In our run we need to have four short time intervals with different coolant load in each time interval and it does not seem very practical for only a small improvement in the conversion. Use of two time intervals seems practical with a yield of 0.66882.

Similarly for a batch time of 25 minutes, we can further improve the value of the conversion. With only four time intervals, the value achieved is 0.90414 (Luus achieved a conversion of 0.90460 using 27 time intervals).

Table 4.3 shows that for cases with 5 minutes batch time, u takes a zero value in the first time interval and then higher values in the following intervals. This observation can be explained as follows. With a short period of operation, the reactor requires to produce B as quickly as possible. For the first interval, the coolant is off so that the reaction can be initiated. However, since the production of B is favoured at lower temperature based on the given kinetics, the value of u is kept high towards the end of the operation so that the coolant can absorb more heat to maintain a low reactor temperature.

On the other hand, for Cases 2a and 2b with 25 minutes reaction time, the high value of u (imposed on the system for almost the first half of the batch time) maintains low reactor temperature and produces most of the product B (see Figure 4.6). In the second half of the batch time, as the conversion rate is low (Figure 4.5), the amount of heat produced is also low. So a small value of u is needed to maintain the reactor

temperature for a further conversion to B (although small). For Case 2c, low value of u for the short first interval builds up the temperature quickly to initiate forward reaction. After the first interval, u follows the same trend as in Cases 2a and 2b. Case 2c produces the most B compared to Cases 2a and 2b. It shows increasing the number of control intervals to some extent gives more flexibility in operation.

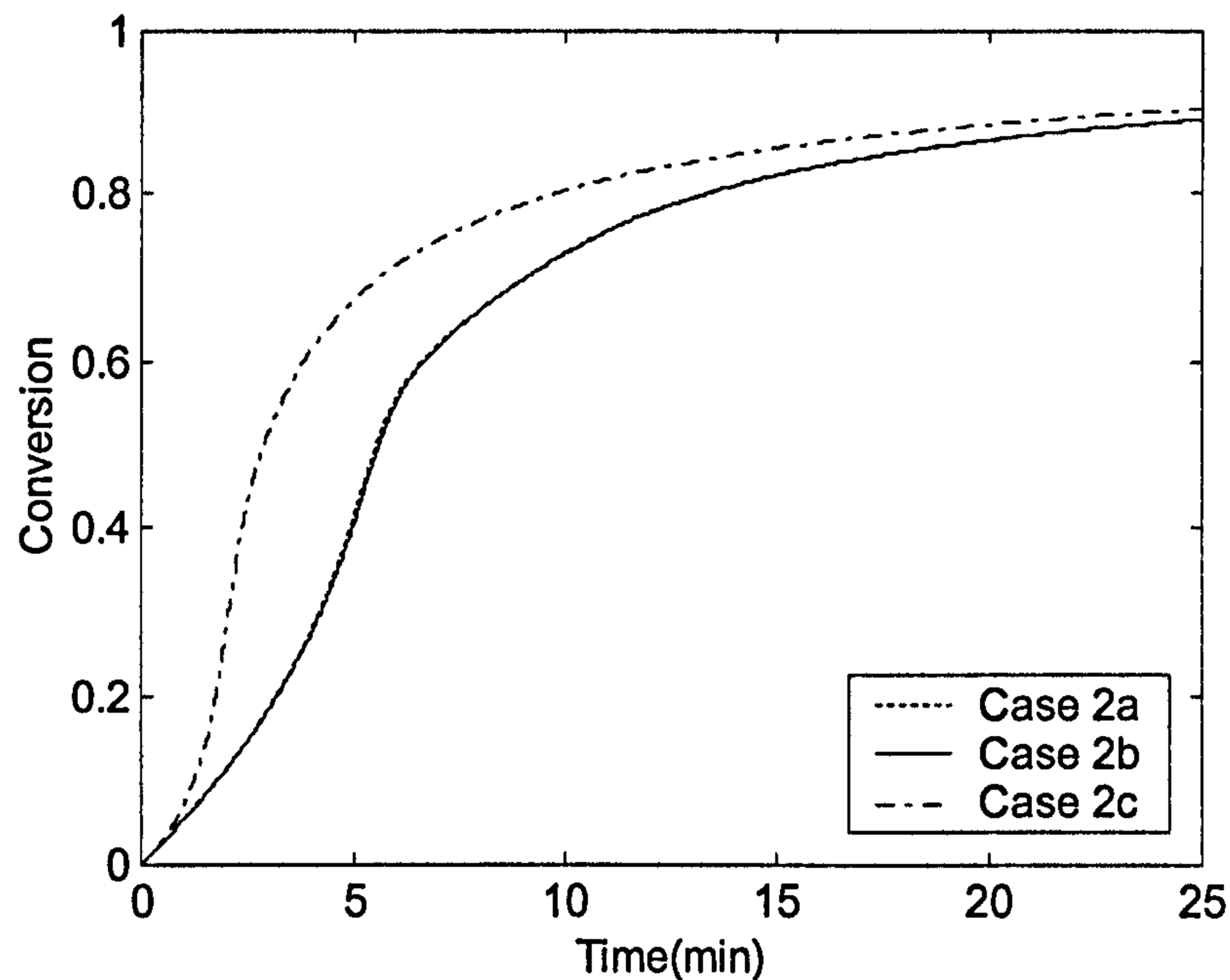


Figure 4.5: Conversion profiles for Run 2a, 2b and 2c

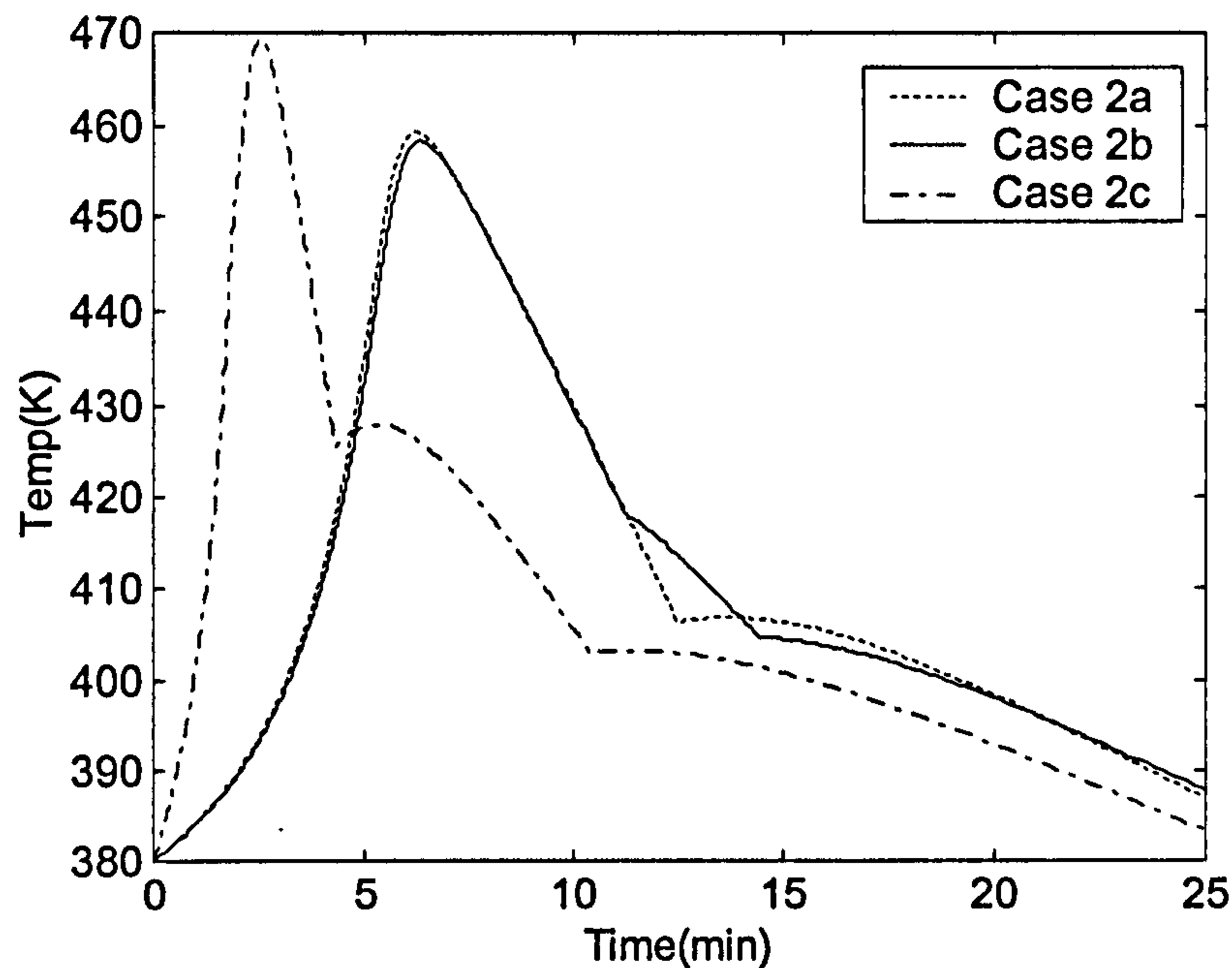


Figure 4.6: Reactor temperature profiles for Run 2a, 2b and 2c

Minimum Time Problem

Here we considered different but fixed conversion to product B and would like to minimise the batch time required to achieve the conversion. We used three time intervals and the length of the time interval together with the control u are optimised. For each run a total of 6 values were optimised. The results are summarised in Table 4.4.

From the results, it is observed that the conversion can be increased to 0.97 but the batch time needed will increase up to 100 minutes, which may not be profitable. Here, two types of control profiles are observed. In the first type of profile (Runs 1 and 3), the coolant is fully off for the first but short interval then takes a high value followed by low value. In the second type of profile, the coolant takes a high value at the beginning followed by low value towards the end (Runs 2 and 4). The reasons of these

different types profiles achieved can be explained in the same way as was done for the maximum conversion problem.

It is to be noted that initial values of optimisation variables (interval lengths and u values in each interval) can lead to a number of feasible solutions although the same objectives (maximum conversion, minimum time) are achieved.

Table 4.4: Summary of the results

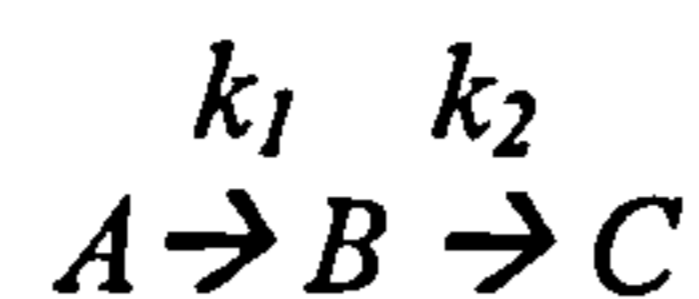
Run	Optimal Control Profile				Conv, x_f (Fixed)	Min Time
1	Control, u	0.0000	0.256902	0.035834	0.91	28.14
	Switching time, min	$t = 0$	1.56	6.51		
2	Control, u	0.125633	0.130696	0.02665	0.92	34.91
	Switching time, min	$t = 0$	6.97	12.97		
3	Control, u	0.0000	0.157914	0.018783	0.95	58.63
	Switching time, min	$t = 0$	2.34	11.58		
4	Control, u	0.126816	0.032960	0.080448	0.97	99.64
	Switching time, min	$t = 0$	12.80	30.28		

4.5.3 Case Study 3

(Part of this work has been accepted for the Chemical Engineering Journal, Aziz and Mujtaba, 2001b)

In this example a consecutive reaction scheme (same as Case Study 1) is considered. However the batch reactor system is described using a detailed model consisting mass and energy balances and detailed design parameters of the reactor. The reaction order also differs from Case Study 1.

The reaction type is:



where A is the raw material, B is the desired product and C is the waste or by-product.

The conversion to B from A and conversion C from B follow a first order reaction rate.

In order to operate the reactor safely, we impose a path constraint in the system to make sure that the reactor temperature throughout the batch does not go beyond a certain temperature. The path constraint is translated into an equivalent end point constraint in the optimisation problem using an extra differential equation in the model equations. The total batch time is divided into a finite number of intervals (*multi-path-interval*) and for each interval, constrained optimisation problem formulation is proposed which should be solved sequentially. This approach allows reactor operation at high temperature (more close to the safe temperature limit) for a longer period compared to the traditional *one-path-interval* method and results in better performance of the reactor.

Each waste or by-product of the reaction has a certain value (threshold value) before it can be discharged to the environment. So in this work, rather than producing the waste and treating it at the end, we put a limit on the waste production. The resulting optimisation problem thus not only has path constraint but also has end point constraints, which result in complex optimisation problems.

The reaction temperature is the key variable, which governs (controls) the batch reactor operation. Most of the previous researchers considered the reactor temperature directly as the control variable. They assumed that the optimal temperature profiles obtained from the solution of the optimisation problems can always be achieved. This assumption does not require consideration of some of the design parameters (such as the

reactor volume), operating parameters (such as the reactor jacket temperature) and energy balance in the model. Therefore in most cases the models used by previous researchers were simple (one such model is presented Case Study 1 and required only mass balance and kinetic information).

However, in practice, the reactor temperature is affected by the external heating or cooling depending whether the reaction is endothermic or exothermic. Therefore for optimising the operation of existing batch reactors (with fixed design in terms of reactor volume, jacket configuration, maximum limits to coolant (or heating media) flow), it is more desirable to manipulate coolant flow (or heating) to maintain a safe reactor temperature. The reactor model should therefore include both mass and energy balances together with the design parameters. It is to be noted that the simple model is useful at the preliminary design stage, where the required optimal temperature profile can be used as a guide to determine the size of the reactor, jacket and to configure the cooling or heating requirement. Here, cooling or heating rate is chosen as the control (optimisation) variable of the process.

4.5.3.1 Optimisation Problem Formulation

Two optimisation problems are considered (Problems P1 and P2). The optimisation problems in this case study are similar with those shown in section 4.6.

However, they are subject to specific conditions mentioned below:

P1 – Maximum Conversion Problem

$$\begin{array}{ll}
 \text{Max} & X \\
 & F_J(t) \\
 \text{s.t} & f(t, \underline{x}'(t), \underline{x}(t), \underline{u}(t), \underline{v}) = 0 \text{ (process model)} \\
 & t_f = t_f^* \\
 & F_{JL} \leq F_J \leq F_{JU} \\
 & W \leq W^*
 \end{array}$$

where X is the conversion of the limiting reactant to the desired product; F_J is the coolant flow; t_f is the batch time and W is the waste, F_{JL} and F_{JU} are the lower and upper bounds of the coolant flow. t_f^* is the fixed batch time and W^* is the threshold limit for the waste.

P2 – Minimum Time Problem

$$\begin{array}{ll}
 \text{Min} & t_f \\
 & F_J(t) \\
 \text{s.t} & f(t, \underline{x}'(t), \underline{x}(t), \underline{u}(t), \underline{v}) = 0 \text{ (process model)} \\
 & X = X^* \\
 & F_{JL} \leq F_J \leq F_{JU} \\
 & W \leq W^*
 \end{array}$$

where X^* is the fixed conversion, t_f is the batch time.

4.5.3.2 Path Constraints in Batch Reactors

When a certain process variable (e.g. the reactor temperature) cannot exceed a given limit for the entire processing period, the process is said to have a path constraint. There are two kinds of path constraints: equality and inequality path constraints. When the concerned process variable is always on the limit, the process is said to have an equality path constraint. In all other situation, the process is said to have an inequality path constraint.

Bryson and Ho (1975) introduced an integral penalty terms in objective function to be minimised in order to treat the equality path constraint. To avoid the numerical difficulties, which may be caused by the use of the penalty term, an alternative approach that involves the conversion of the path constraint to an equivalent end-point constraint was introduced by Sargent and Sullivan (1977). A common characteristic of all these techniques is that the penalty terms or end-point constraints introduced have zero gradients with respect to the optimisation parameters at the solution. This, in turn may result in a reduced convergence rate near the solution, and as noted by Goh and Teo (1988), the success of such techniques depends very strongly on the line-search merit function used by the optimisation algorithm.

Most of the established approaches for dealing with inequality path constraints are similar to the techniques for handling equality path constraints. Thus they rely on defining a measure of the constraint violation over the entire horizon and then penalising it in the objective function, or forcing it directly to zero through an end-point constraint. For example, Vassiliadis et al. (1994b) considered an integral expression to provide a single measure of the violation of an inequality path constraint over the entire time horizon of interest, permitting, for instance, the replacement of each such

constraint by a single end-point constraint. To illustrate this idea, consider a typical reactor temperature profile in Figure 4.7.

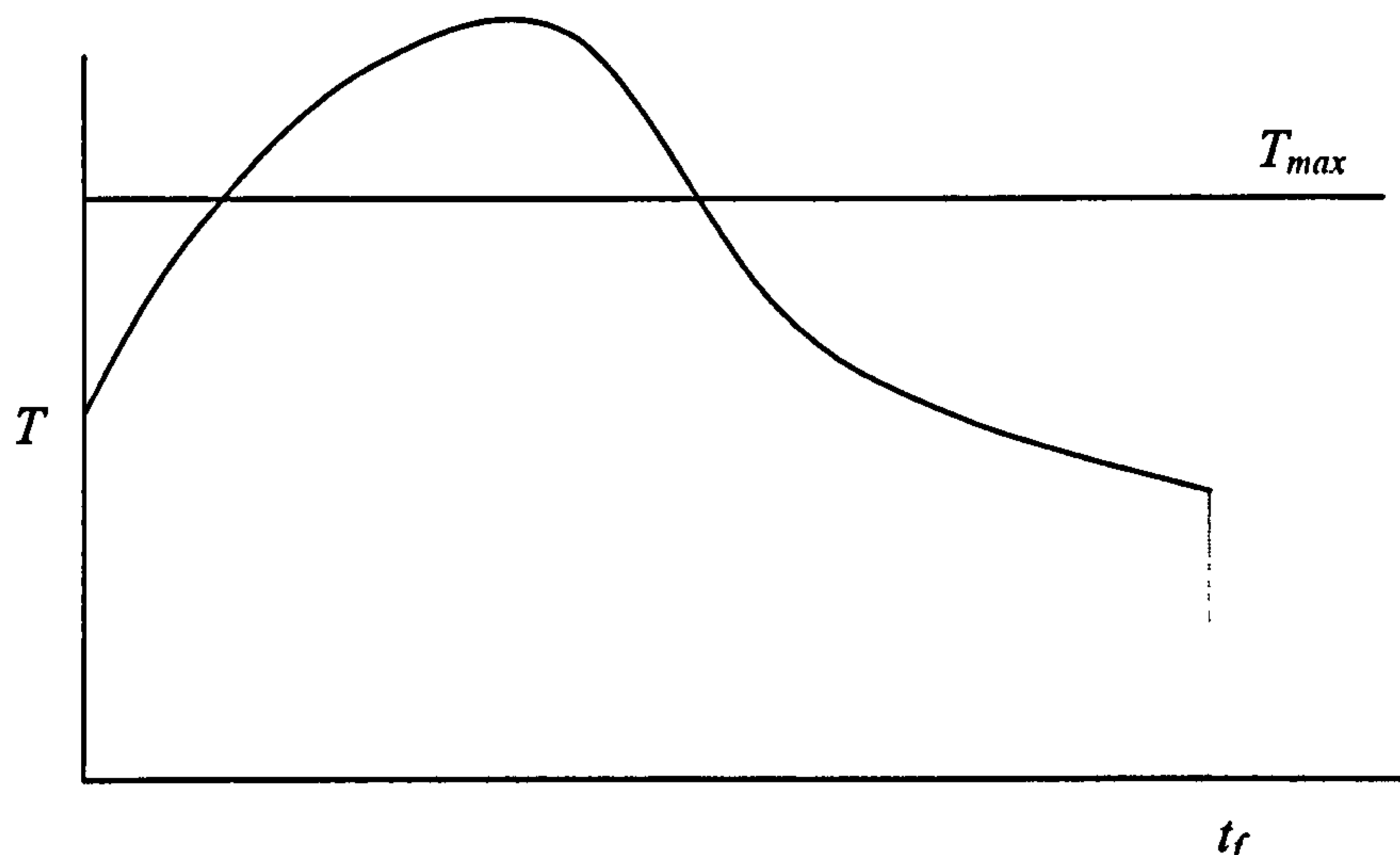


Figure 4.7: Typical reactor temperature profile

T_{max} shows the maximum allowable reactor temperature for a safe operation. In order to make sure that the reactor temperature, T does not go above T_{max} at any time, a path constraint on T is imposed in the optimisation problem.

In Figure 4.7, the reactor operates without any control on the reactor temperature, and therefore T goes above the limit T_{max} during the batch. At any time during the batch this violation can be defined as:

$$V = \text{violation of safe operation} = (T(t) - T_{max})^2 \text{ if } T > T_{max}$$

$$\text{and } V = 0 \text{ if } T < T_{max}$$

A typical plot of V versus batch time, t is shown in Figure 4.8.

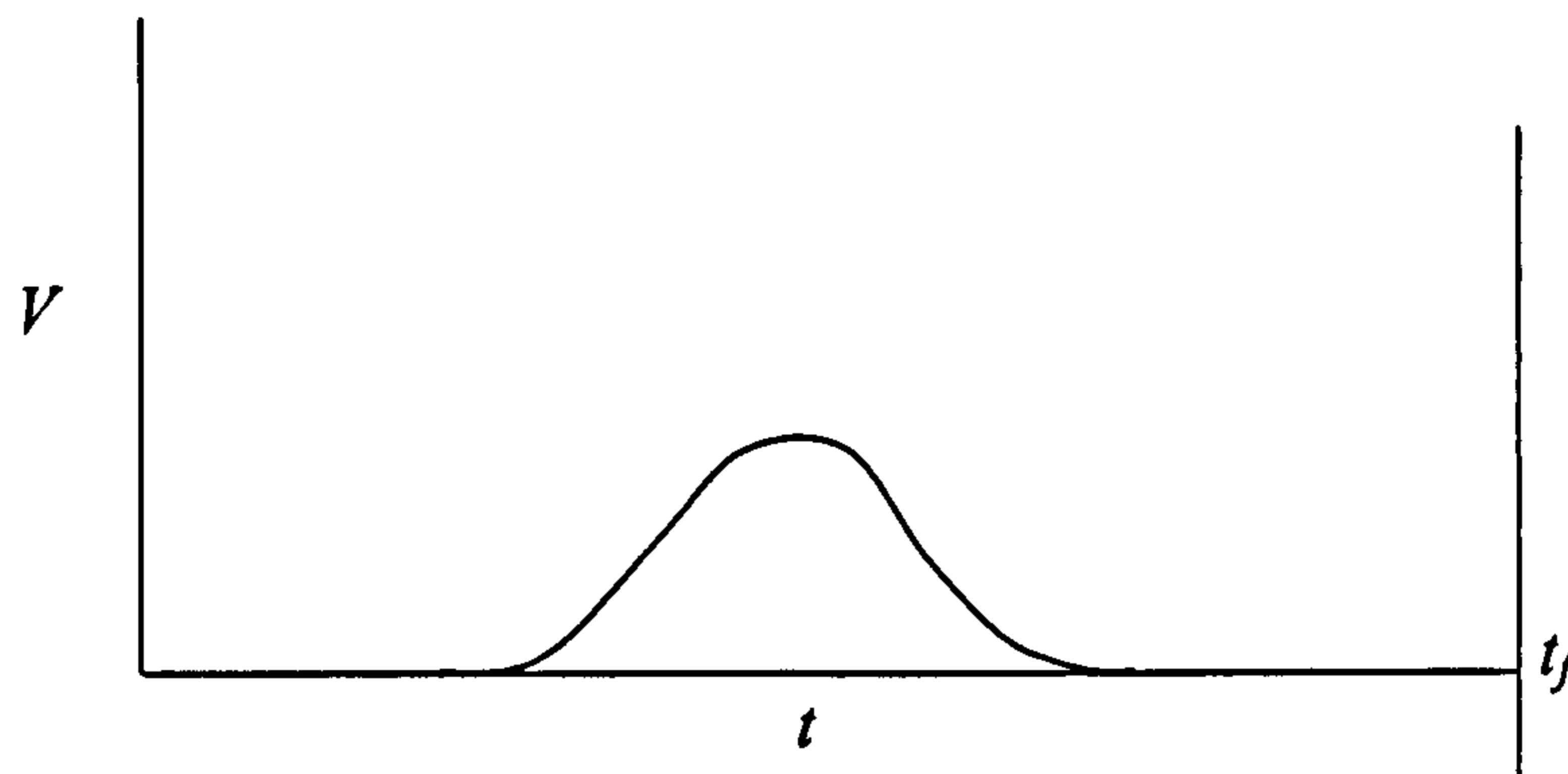


Figure 4.8: Violation during the reaction process

The total accumulated violation over the entire period can be written as:

$$V_T = \int_{t=0}^{t_f} V(t) dt$$

Differentiation of this equation leads to:

$$\therefore \frac{dV_T}{dt} = V(t) = (T(t) - T_{\max})^2 \quad (4.5.3.1)$$

In this work, Equation (4.5.3.1) is added to the process model equations. Also the following additional terminal constraint is added in each of the optimisation formulation presented earlier.

$$0 \leq V_T(\text{at } t_f) \leq \varepsilon \quad (4.5.3.2)$$

where ε is a very small finite positive number. The above constraint will ensure that $T(t)$ will always be $\leq T_{\max}$.

However, it is to be noted that for a given bounds on the optimisation variables (e.g. the coolant flow rate), the numerical value of the end-point constraint V_T at the final time t_f can be very large (can be as high as 10^5) or very low (close to zero) compared to other constraints (e.g. environmental, conversion, etc.) (usually less than 10) in the optimisation problem (P1, P2 or P3). During the solution of the optimisation problem this can result to a high overall constraint violation and can lead to an oscillatory behaviour in the optimisation variables (e.g. coolant flow rate), hitting either the lower or the upper bounds. This may result to a failure of the optimisation problem or to a sub-optimal solution or a very large number of iteration. The use of narrow bounds in the optimisation variables in many cases may not satisfy $V_T \leq \varepsilon$ at t_f and other constraint bounds. Therefore scaling of the optimisation variables as well as constraints may be necessary.

In the past, V_T has always been evaluated at the final time t_f (at the end of the process). However, in this work (instead of using narrow bounds or scaling the variables or constraints) we propose that the total batch time t_f be divided into a finite number of intervals 'n' with t_j as the length of each interval (as is done in CVP method). At the junction of each interval (called '*path-interval*') Equation (4.5.3.2) is satisfied. This results to a series of 'n' dynamic optimisation problems solved sequentially. The final state of any optimisation problem $j-1$ becomes the initial state of the optimisation problem j .

The optimisation problem P1 for any time interval j can be written as:

$$\begin{aligned}
 \text{P1 (j):} \quad & \text{Max} && \text{Conversion (X}_j\text{)} \\
 & && F_J(t) \\
 & \text{s.t} && f(t, \underline{x}'(t), \underline{x}(t), \underline{u}(t), \underline{v}) = 0 \text{ (process model)} \\
 & && F_{JL} \leq F_J \leq F_{JU} \\
 & && T \leq T_U \text{ (path constraint)} \\
 & && x_0^j = x_f^{j-1} \\
 & && t_j = t_j^* = \left(\frac{t_f^*}{n} \right) \\
 & && V_T \leq 0 \text{ (at } t = t_j\text{)}
 \end{aligned}$$

Note each *path-interval* t_j can be subdivided and CVP technique can be applied for the control (optimisation) variables. Maximisation of conversion in each *path-interval* will ensure maximum conversion at the final time ($X_{Total} = \sum_{j=1}^n X_j$). The above *multi-path-interval* method is conceptually similar to multistage dynamic optimisation problem formulation of Vassiliadis et al. (1994b) but solved sequentially. The case study presented later shows the merits of this approach over the *single-path-interval* method.

The optimisation problem P2 for any time interval j can be written as:

$$\begin{aligned}
 \text{P2 (j):} \quad & \text{Min} && t_j \\
 & && F_J(t) \\
 & \text{s.t} && f(t, \underline{x}'(t), \underline{x}(t), \underline{u}(t), \underline{v}) = 0 \text{ (process model)} \\
 & && F_{JL} \leq F_J \leq F_{JU} \\
 & && T \leq T_U \text{ (path constraint)} \\
 & && x_0^j = x_f^{j-1} \\
 & && X_j = X_j^* = f(X^*, n) \\
 & && V_T \leq 0 \text{ (at } t = t_j\text{)}
 \end{aligned}$$

As shown earlier, the minimum time formulation (optimisation problem P2) requires fixed product conversion, say X^* . We can assume that in *multi-path-interval* optimisation formulation, the desired conversion at the end of each *path-interval* t_j is X_j^*
 $= \left(\frac{X^*}{n} \right)$ (assuming linear conversion profile over time). Alternatively we can choose

$$X_j^* \text{ so that } X^* = \sum_{j=1}^n X_j^*.$$

The total minimum batch time, $t_{\min} = \sum_{j=1}^n t_j$

The optimisation problem P3 for any time interval j can be written as:

$$\begin{array}{ll} \text{P3 (j):} & \text{Max} & \text{Profit (} P_j \text{)} \\ & & F_j(t) \\ & \text{s.t} & f(t, \underline{x}'(t), \underline{x}(t), \underline{u}(t), \underline{v}) = 0 \text{ (process model)} \\ & & F_{JL} \leq F_j \leq F_{JU} \\ & & T \leq T_U \text{ (path constraint)} \\ & & x_0^j = x_f^{j-1} \\ & & V_T \leq 0 \text{ (at } t = t_j \text{)} \end{array}$$

In maximum profit problem (P3), both conversion and time are optimised. The total

$$\text{profit will be } P_{\max} = \frac{\sum_{j=1}^n P_j \times t_j}{t_f}$$

4.5.3.3 Models Equations

Here the Option 5 model of Chapter 3 with $m = 3$ and $n = 2$ is considered.

Mass balances:

The values of ν_{11} and ν_{22} are equal to -1 ; the values of ν_{12} and ν_{23} are equal to 1 and the rest of ν_{ji} are equal to zero. The values of β_{11} and β_{22} are equal to 1 . The rest of β_{ij} are equal to zero. The equations 3.51 and 3.52 in Chapter 3 will then result to:

$$\frac{dC_A}{dt} = -k_1 C_A \quad (4.5.3.3)$$

$$\frac{dC_B}{dt} = k_1 C_A - k_2 C_B \quad (4.5.3.4)$$

$$\frac{dC_C}{dt} = k_2 C_B \quad (4.5.3.5)$$

Energy balance (refer to equations 3.54 – 3.59):

$$\frac{dT}{dt} = \frac{Q_r - Q_m}{c_p V \rho} \quad (4.5.3.6)$$

$$\frac{dT_m}{dt} = \frac{Q_m - Q_J}{c_{pm} V_m \rho_m} \quad (4.5.3.7)$$

$$\frac{dT_J}{dt} = \frac{F_J}{V_J} (T_{J0} - T_J) + \frac{Q_J}{c_{pJ} V_J \rho_J} \quad (4.5.3.8)$$

$$Q_r = -\Delta H_1 (k_1 C_A V) - \Delta H_2 (k_2 C_B V) \quad (4.5.3.9)$$

$$Q_m = U_i A_i (T - T_m) \quad (4.5.3.10)$$

$$Q_J = U_o A_o (T_m - T_J) \quad (4.5.3.11)$$

Rate equations:

The equation 3.53 in Chapter 3 will result to:

$$k_1 = k_{10} \exp\left(-\frac{E_1}{RT}\right) \quad (4.5.3.12)$$

$$k_2 = k_{20} \exp\left(-\frac{E_2}{RT}\right) \quad (4.5.3.13)$$

4.5.3.4 Degrees of Freedom Analysis

Total number of variables in Equations (4.5.3.3 – 4.5.3.13) = 40:

$$\frac{dC_A}{dt}, \frac{dC_B}{dt}, \frac{dC_C}{dt}, \frac{dT}{dt}, \frac{dT_m}{dt}, \frac{dT_J}{dt}, k_1, k_2, Q_r, Q_m, Q_J, C_A, C_B, C_C, T, T_m, T_J, \Delta H_1,$$

$$\Delta H_2, \rho, \rho_m, \rho_J, c_p, c_{pm}, c_{pJ}, V, V_M, V_J, E_1, E_2, F_J, T_{J0}, R, k_{10}, k_{20}, U_i, U_o, A_i, A_o, t$$

Degrees of Freedom,

$$\begin{aligned} \text{DF} &= \text{Total number of variables} - \text{Total number of equations} \\ &= 40 - 11 \\ &= 29 \end{aligned}$$

Specifications:

Parameters = 21:

$$\Delta H_1, \Delta H_2, \rho, \rho_m, \rho_J, c_p, c_{pm}, c_{pJ}, V, V_M, V_J, E_1, E_2, T_{J0}, R, k_{10}, k_{20}, h_i, h_o, A_i, A_o$$

Independent variable = 1 = t

Initial value at time $t = 0$ for all the differential variables ($C_A, C_B, C_C, T, T_M, T_J$) to be specified = 6.

The above specifications count to 28 variables. In this work F_J (coolant flow) is relaxed from the specifications and is optimised.

It is assumed that the system is preheated to 350 K before the actual process starts and the jacket is full with water at room temperature. The initial value of C_A , C_B and C_C are 0.975, 0.025 and 0.0 respectively. The initial value of T , T_m and T_J are 350K, 373K and 300K respectively.

The constant parameters of the process model are:

$$\begin{aligned} \Delta H_1 &= -6.50E8 \text{ J/kmol}; & \Delta H_2 &= -1.20E8 \text{ J/kmol} \\ \rho &= 800.0 \text{ kg/m}^3; & \rho_m &= 8200.0 \text{ kg/m}^3 \\ \rho_J &= 1000.0 \text{ kg/m}^3; & c_p &= 4200.0 \text{ J/kg.K} \\ c_{pm} &= 500.0 \text{ J/kg.K}; & c_{pJ} &= 4200.0 \text{ J/kg.K} \\ E_1 &= 3.49E7 \text{ J/kmol}; & E_2 &= 4.65E7 \text{ J/kmol} \\ V &= 1.23 \text{ m}^3; & V_m &= 0.27 \text{ m}^3 \\ V_J &= 0.53 \text{ m}^3; & k_{10} &= 4.38E4 \text{ hr}^{-1} \\ k_{20} &= 3.94E5 \text{ hr}^{-1}; & R &= 8314.0 \text{ J/kmol.K} \\ U_i &= 3.27E6 \text{ J/hr.K.m}^2; & U_o &= 8.18E6 \text{ J/hr.K.m}^2 \\ A_i &= A_o = 5.25 \text{ m}^2; & T_{J0} &= 298 \text{ K} \end{aligned}$$

4.5.3.5 Results

Maximum Conversion Problem (Problem P1 or P1(j))

In this reaction, an end-point (final time) constraint for the reaction product temperature ($T \leq 320\text{K}$) is imposed to guarantee that the products are at or below certain desired temperature. The total batch time is 3.5 hours and the number of control

intervals is 4. Coolant flow is bounded between 0 (valve fully closed) and 9 m³/hr (valve fully open). The end point constraint for the waste (product C) is set at 0.10.

Five different runs have been presented for the maximum conversion problem (Problem P1 or P1(j)). Run 1 is with an end point constraint on the reactor temperature only. Run 2 is with end point constraints on the reactor temperature and on the waste (product C). Run 3 has been solved with path (*one-path-interval*) and end point constraints on the reactor temperature and an end point constraint on the waste. Runs 4 and 5 are solved with path and end point constraints on the reactor temperature. However, Run 4 deals with *one-path-interval* on T and Run 5 deals with *two-path-interval* on T. For Runs (3-5) the reactor temperature must not exceed 370K at any time (path constraint). The results for all runs are summarised in Table 4.5 in terms of conversion to B (desired product) and C (waste).

Table 4.5 shows that, a higher conversion to B is achieved in Run 1 compared to that of Run 2. This can be explained as: the rate of production of C depends on both the kinetic constant k_2 and the amount of B present at any time. Also since k_2 is much larger than k_1 , near the end of the reaction process, the rate of production of B is slower than the rate of production of C. Therefore the only way to satisfy the constraint on the waste production is by producing less B. Production of more B will produce more C for the type of reaction considered here (Run1, Table 4.5).

Table 4.5: Summary of the results – Maximum conversion problem

Run	Type of problem	Conv to B	Conv to C (waste)
1	With end point constraint on T	0.6462	0.1659
2	With end point constraint on T and waste	0.6249	0.1000
3	With path and end point constraint on T and end point constraint on waste	0.6253	0.0991
4	With path and end point constraint on T (<i>one-path-interval</i>)	0.6253	0.0991
5	With path and end point constraint on T (<i>two-path-interval</i>)	0.6401	0.1161

Run 3 results to a further reduction in conversion of B due to added path constraint on the reactor temperature. For Runs 1-3 the optimal coolant flow profile and the reactor temperature profile are shown in Figures 4.9 – 4.11. Figures 4.9 and 4.10 clearly show that although in both runs end point constraints on the reactor temperature are satisfied, the maximum reactor temperature goes beyond the safe limit (370K), to produce more B (as it is favoured at higher temperature). For Run 3, path constraint is satisfied (Figure 4.11) and therefore results to low conversion of B compared to Runs 1 and 2. Also it is interesting to note that Run 1 demands higher reactor temperature as the batch proceeds to favour more conversion to B while Run 2 demands lower reactor temperature to produce less waste (C). These temperature demands are fulfilled by lower and higher coolant flow rate (Figures 4.9 and 4.10) respectively.

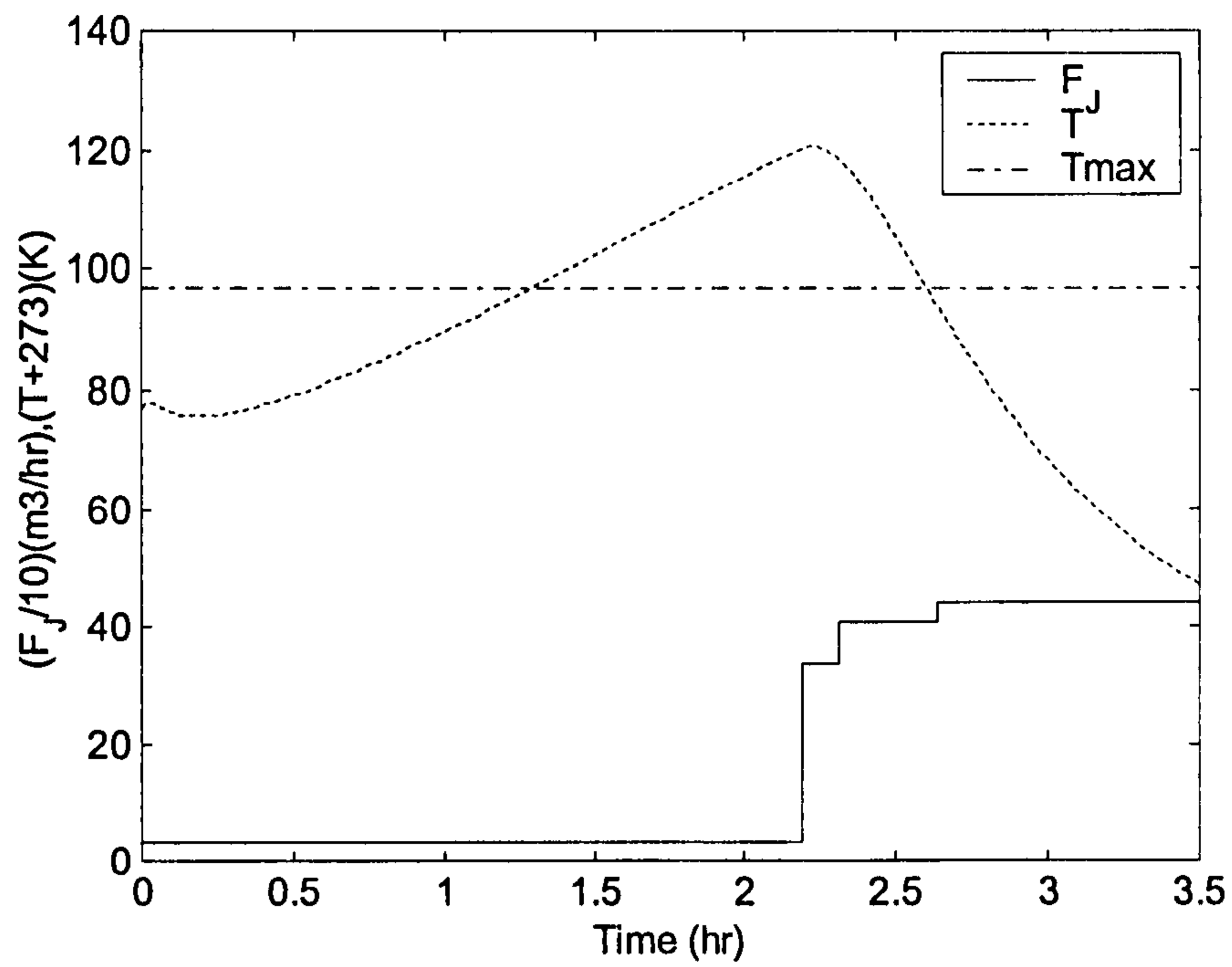


Figure 4.9: Reactor temperature and optimal coolant flow profiles for Run 1

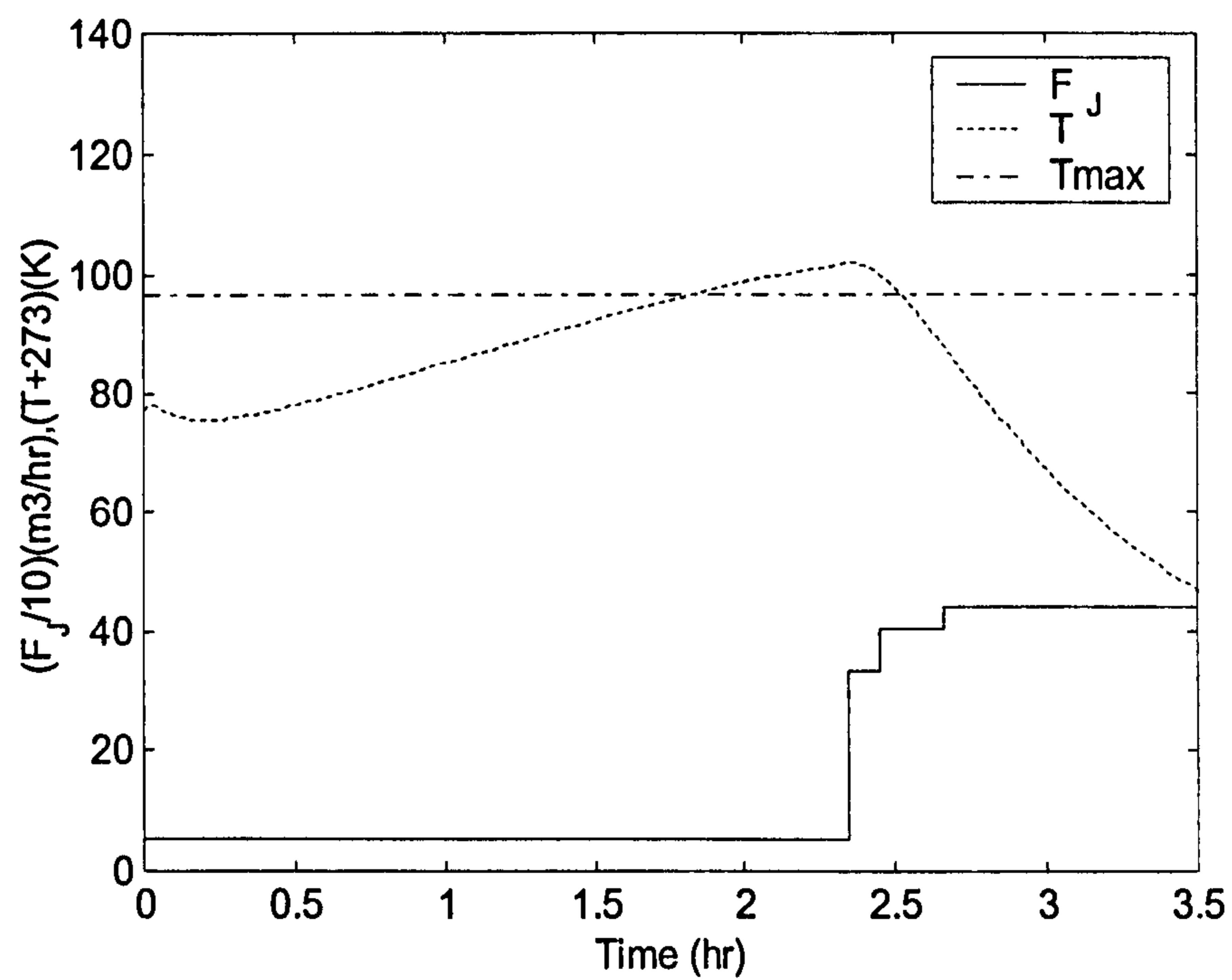


Figure 4.10: Reactor temperature and optimal coolant flow profiles for Run 2 (P1)

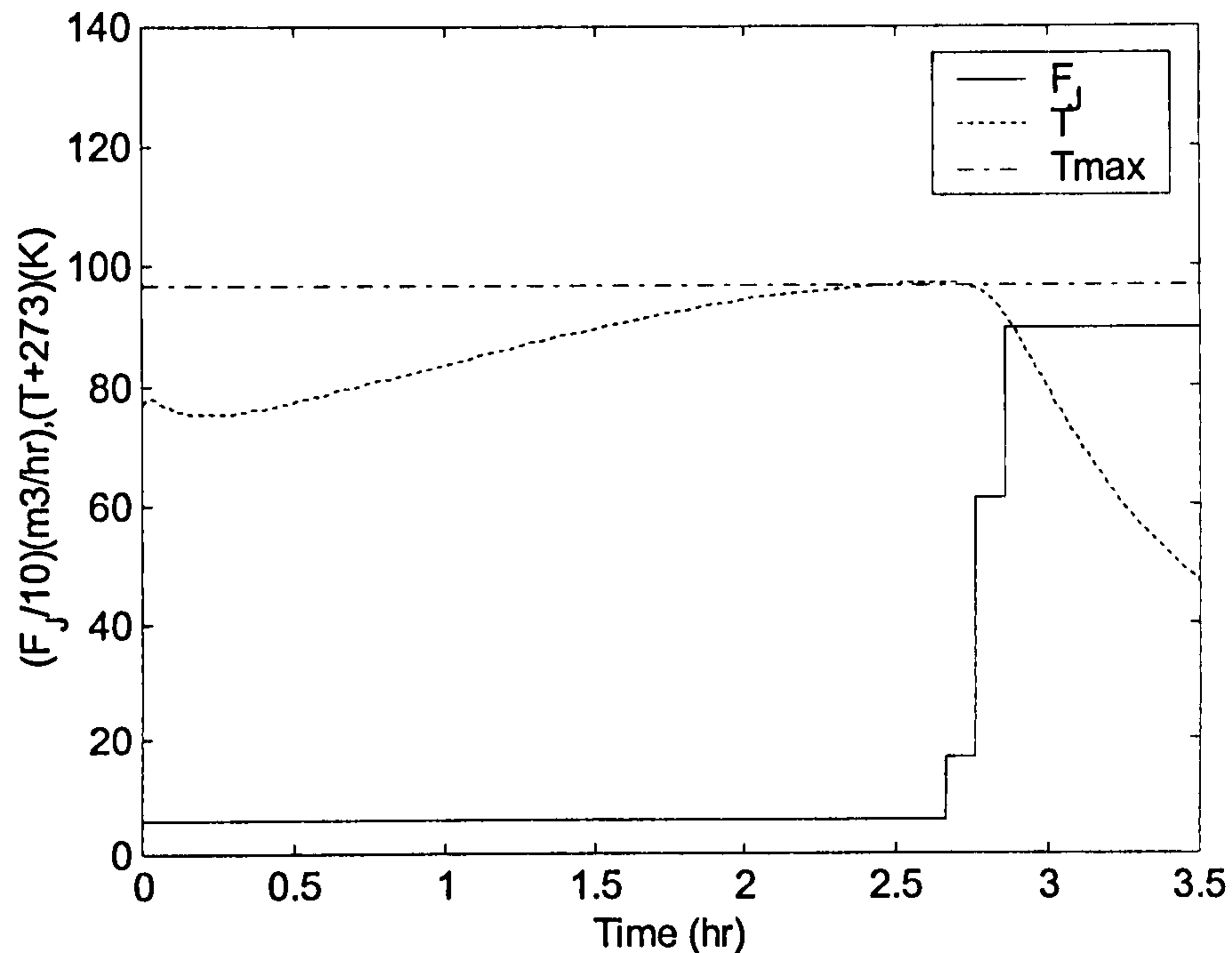


Figure 4.11: Reactor temperature and optimal coolant flow profiles for Run 3 (P1)

For Run 3 the reactor is run with a higher coolant flow (compared to Runs 1-2) for a long time (longer than Runs 1-2) to ensure that the path constraint is satisfied (Figure 4.11). Near the end of the batch the reactor is run with even higher coolant flow to ensure that the end point constraints on temperature as well as on the waste are satisfied.

Run 4 uses *one-path-interval* of 3.5 hr which is the same as the total batch time. While Run 5 uses *two-path-interval*, the length of each interval being 1.75 hr. Run 4 uses 4 control intervals (Figure 4.12) within the *path-interval* while Run 5 uses 2 control intervals within each *path-interval* (Figure 4.13). For Run 5, the optimisation problem is solved sequentially. In the first part (in *path-interval 1*) the problem is solved with path constraint but without the end point constraint on T and in the second part (in *path-interval 2*) the problem is solved using both the path constraint and end point

constraints. This strategy results to higher conversion to B in Run 5 (2.4% higher) compared to that in Run 4. This is because the reactor in Run 5 could be operated at higher temperature (low coolant flow) compared to that in Run 4 for a longer period, which results to higher conversion of B at any time during the operation (Figure 4.14).

It is to be noted that in Table 4.5 the conversion obtained for Run 3 and 4 is similar. The reason behind is, in Run 4 the value of the waste being produced is in the range of the waste constraint imposed in Run 3. The production of B and C cannot be increased in order to fulfil the path constraint on the reactor temperature.

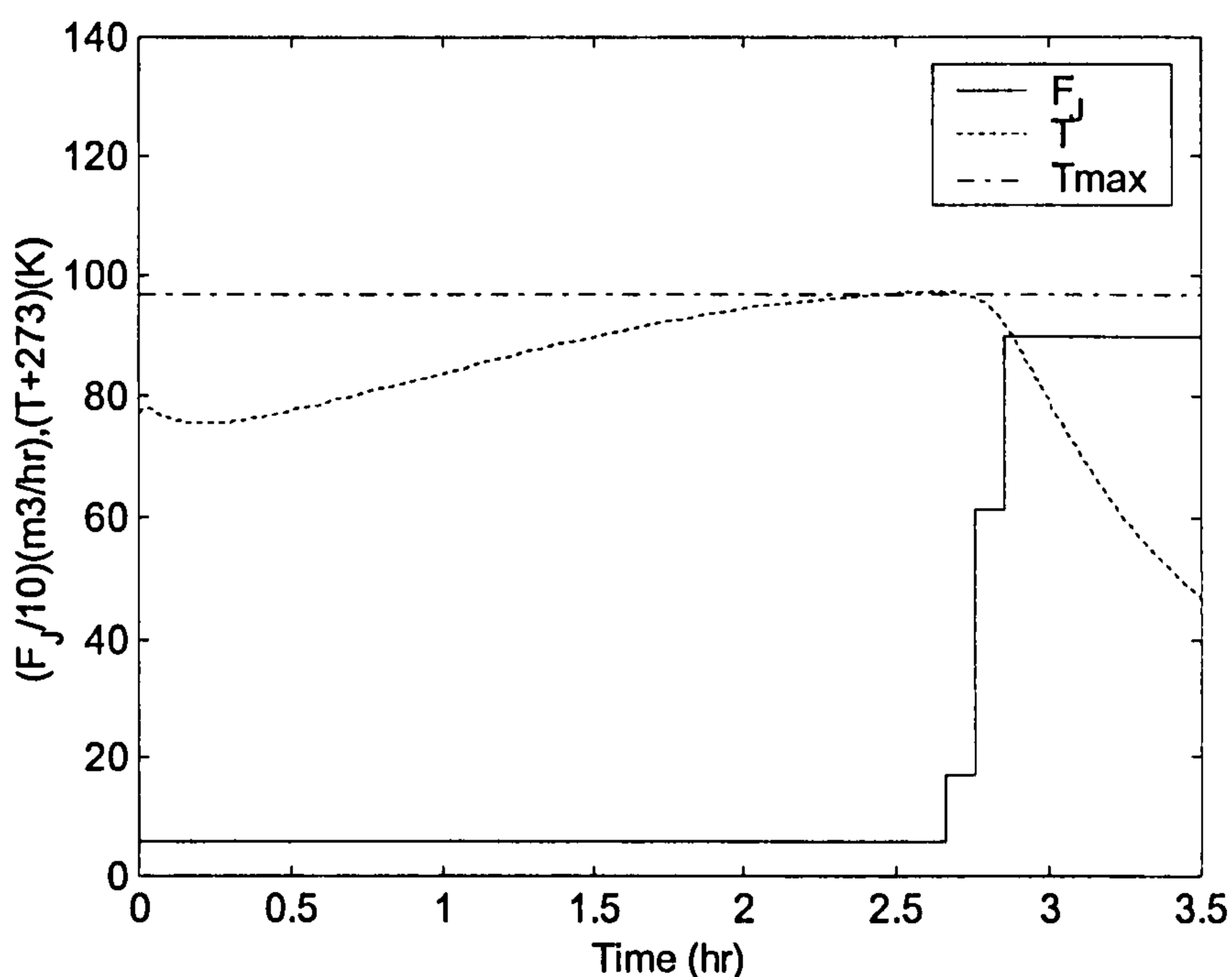


Figure 4.12: Reactor temperature and optimal coolant flow profiles for Run 4 (P1)

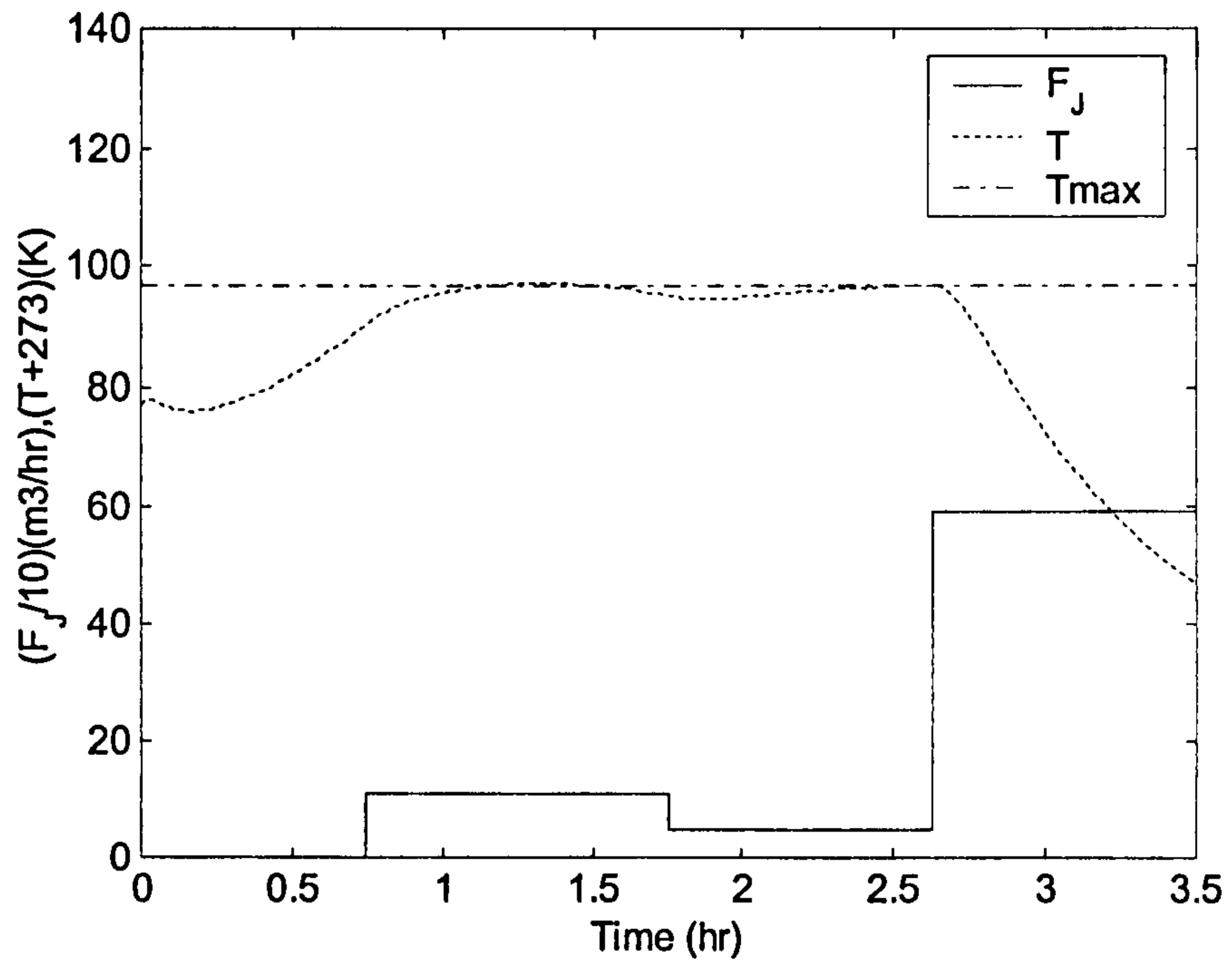


Figure 4.13: Reactor temperature and optimal coolant flow profiles for Run 5 (P1)

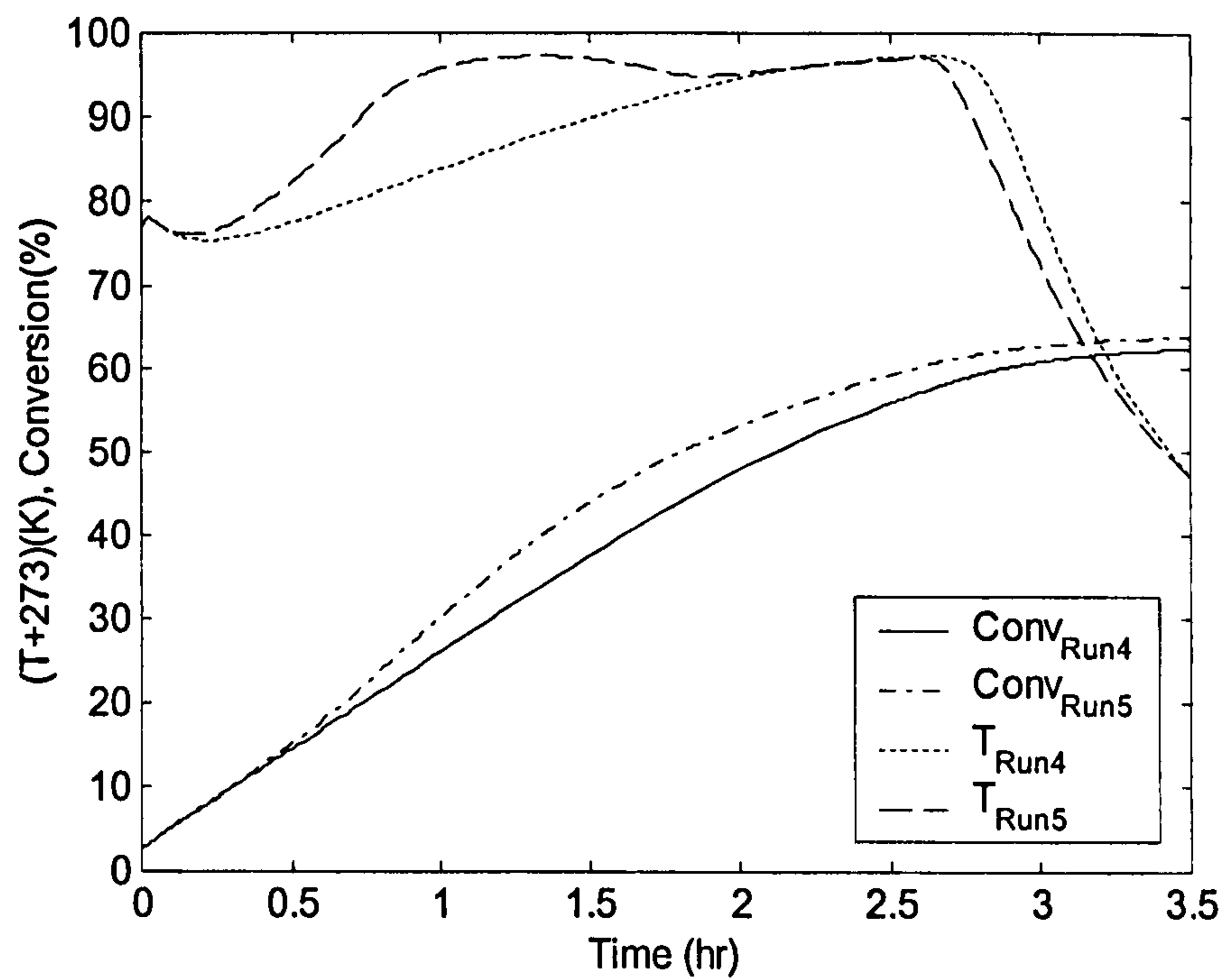


Figure 4.14: Conversion and temperature profiles for Runs 4 and 5 (P1)

Minimum Time Problem (Problem P2 or P2(j))

Here, we rerun Runs 1, 4 and 5 of Table 4.5 but with a fixed conversion of B at 0.600. The bounds on the coolant flow rate are same as before. For Run 5, in *path-interval* 1, the desired conversion of B to achieve is 0.350 and the problem is solved using only the path constraint. For *path-interval* 2, the problem is solved using both the path and end point constraints. As before, 4 control intervals are used in Runs 1 and 4 and 2 control intervals are used in each *path-interval* in Run 5. Each Run has therefore 4 control intervals which is optimised while minimising the final time. Results are summarised in Table 4.6 and Figures (4.13 – 4.16).

Table 4.6: Summary of the results – Minimum time problem

Run	Type of problem	Conv to B	Conv to C (waste)	Final time (hr)
1	With end point constraint on T	0.600	0.1071	2.46
4	With path and end point constraint on T (<i>one-path-interval</i>)	0.600	0.0809	3.21
5	With path and end point constraint on T (<i>two-path-interval</i>)	0.600	0.0845	2.91

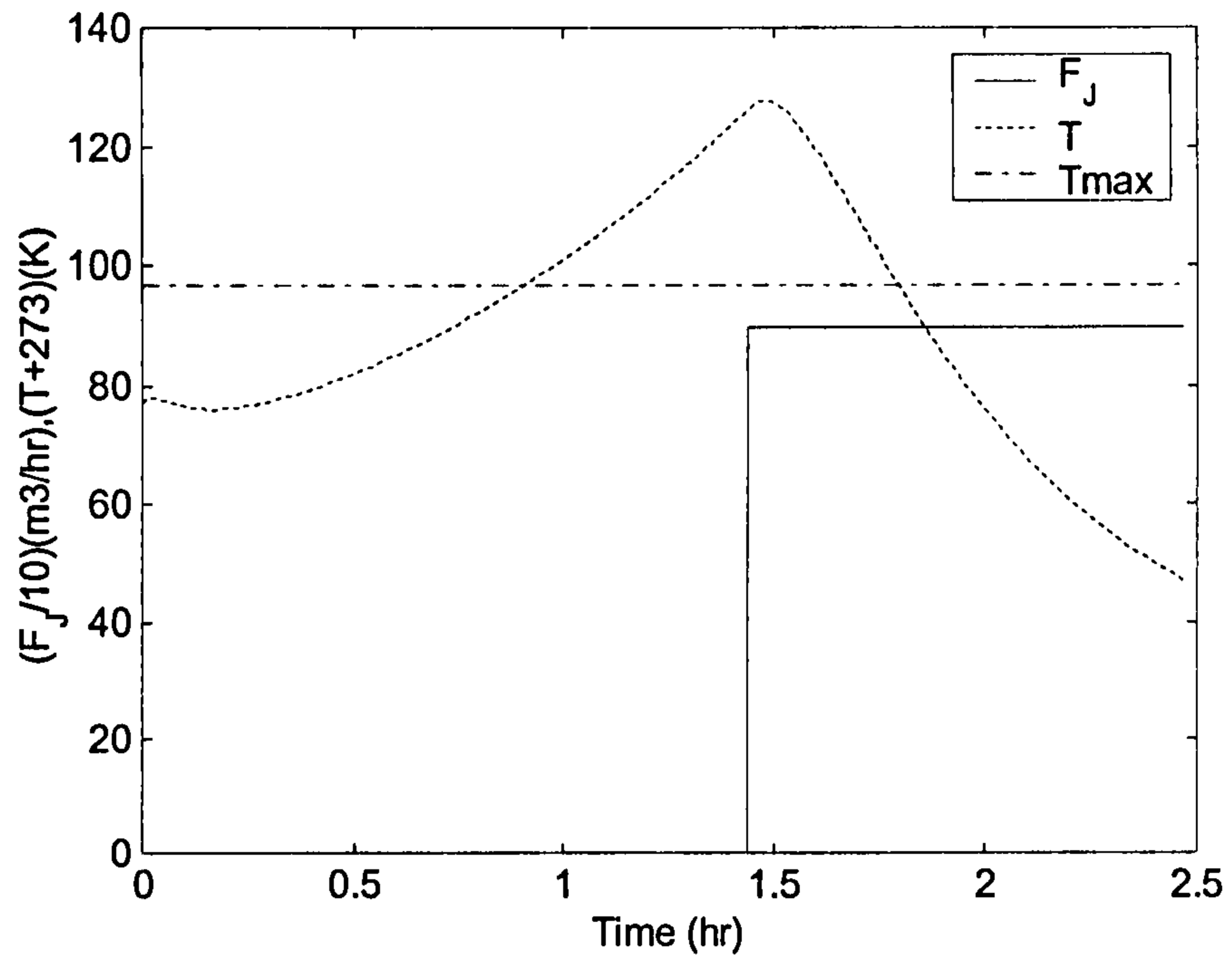


Figure 4.15: Reactor temperature and optimal coolant flow profiles for Run 1 (P2)

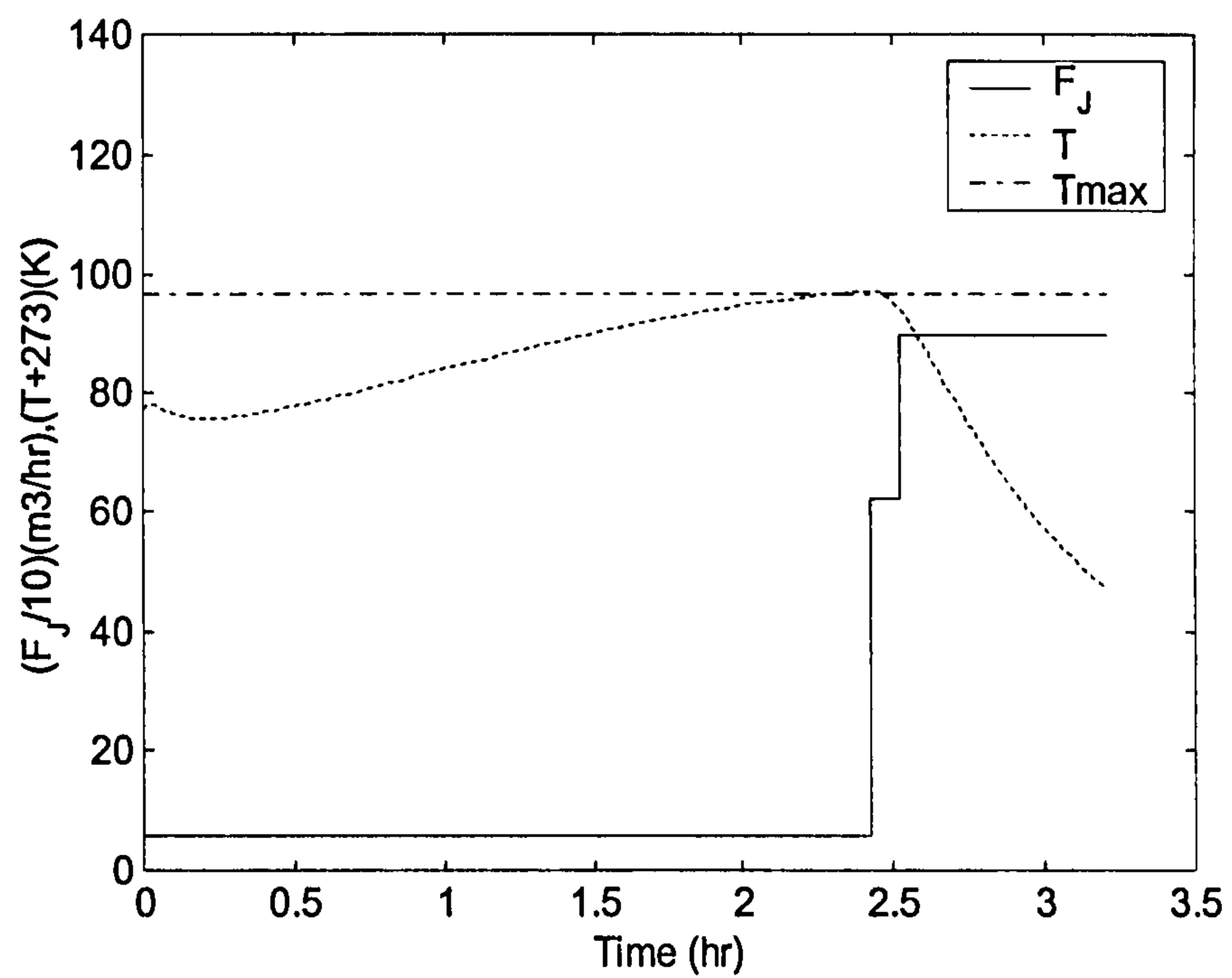


Figure 4.16: Reactor temperature and optimal coolant flow profiles for Run 4 (P2)

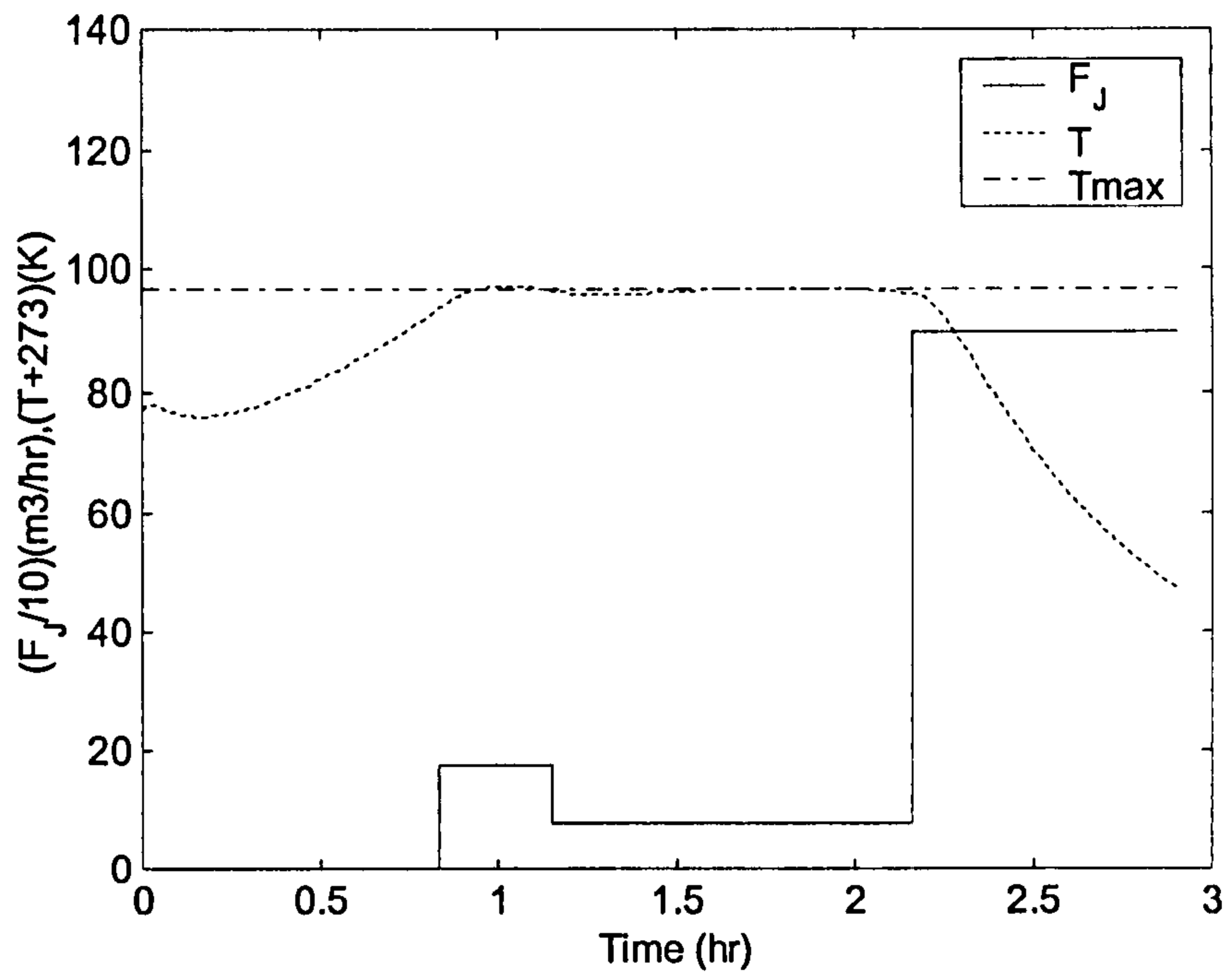


Figure 4.17: Reactor temperature and optimal coolant flow profiles for Run 5 (P2)

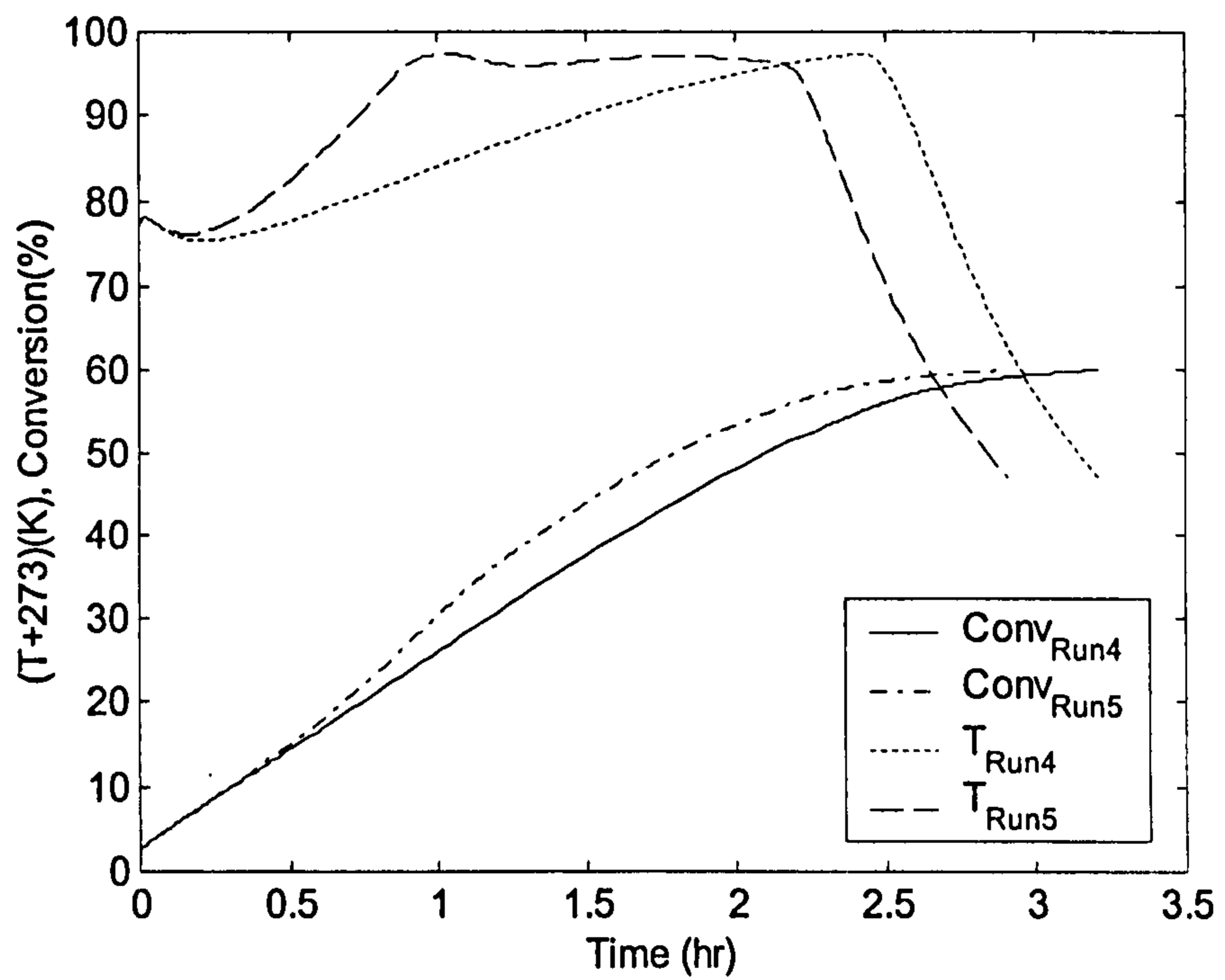


Figure 4.18: Conversion and temperature profiles for Runs 4 and 5 (P2)

From Table 4.6, it is found that the time needed to get the same conversion (0.600) while under a path constraint (Runs 4 and 5) is longer compared to the run without the path constraint (Run 1). This is because in Run 1 the reactor temperature goes above the safe limit T_{max} and achieve the desired conversion quicker (Figure 4.15). However, the batch time needed with *two-path-interval* (Run 5, Table 4.6) is lower compared to that of Run 4 (Table 4.6) which was expected. For Run 5 in the first *path-interval* (0 to 1.15 hr), there was no end point constraint and the reactor temperature could go higher than that of Run 4 at any time during the batch producing more B quicker (Figure 4.18). For the second *path-interval* (1.15 to 2.91 hr), the reactor temperature for Run 5 was higher than that of Run 4 for a considerable period thus producing more B quicker. In all Runs, the coolant flow hits its upper bound at the end of the processes to satisfy the end point constraint.

4.5.4 Case Study 4

(Part of this work has been presented at ESCAPE-11, Kolding, Denmark, 27-30 May 2001, Aziz and Mujtaba, 2001a)

In this case study, optimisation of semi-batch reactor is presented. Semi-batch reactor is also an essential unit operation in almost all batch-processing industries. As far as safety and quality are concerned, semi-batch reactors have advantages over the batch reactors. The former has a good temperature control and has the capability of minimising unwanted side reactions by maintaining low concentration of one of the reactants.

Depending on the downstream process requirement (plant schedule, quality, quantity, etc.), semi-batch reactors can also be operated in any of the three modes i.e.

fixed batch time operation, fixed conversion operation, maximum profit operation mentioned earlier and will result to dynamic optimisation problems such as *maximum conversion problem (P1), minimum time problem (P2) and maximum profit problem (P3)*.

As has been mentioned in Chapter 2, in the past, work on optimisation of semi-batch reactors has been given more attention compared to the conventional batch reactors. Some work in the optimisation of semi-batch reactors have been carried out recently by Cuthrell and Biegler (1989), Luus (1991), Levien (1992), Palanki et al. (1993), Vassiliadis (1994b), Garcia et al. (1995), Carrasco and Banga (1997), Banga et al. (1998), Luus and Hennessy (1999) and Abel et al. (2000). All authors above considered *maximum conversion problem* except Abel et al. (2000) considered *minimum time problem* in their studies.

In this work, all three types of optimisation problems are considered for an exothermic semi-batch reaction system with the reactor temperature and the flow rate of feed reactant as the control variables. A simple reactor model based on the material balances and reaction kinetics is used. It is assumed that the optimum temperature profile is obtainable by controlling external heating/cooling (not shown in the reactor model). Unlike conventional batch reactors, the reactor volume in semi-batch reactors gives an additional constraint (path constraint) in the optimisation problem (discussed in the next section). The optimal control policy in terms of feed rate and reactor temperature obtained in this work can be used to design the heating/cooling system of the reactor. The semi-batch reactor of our interest is shown in Figure 4.19.

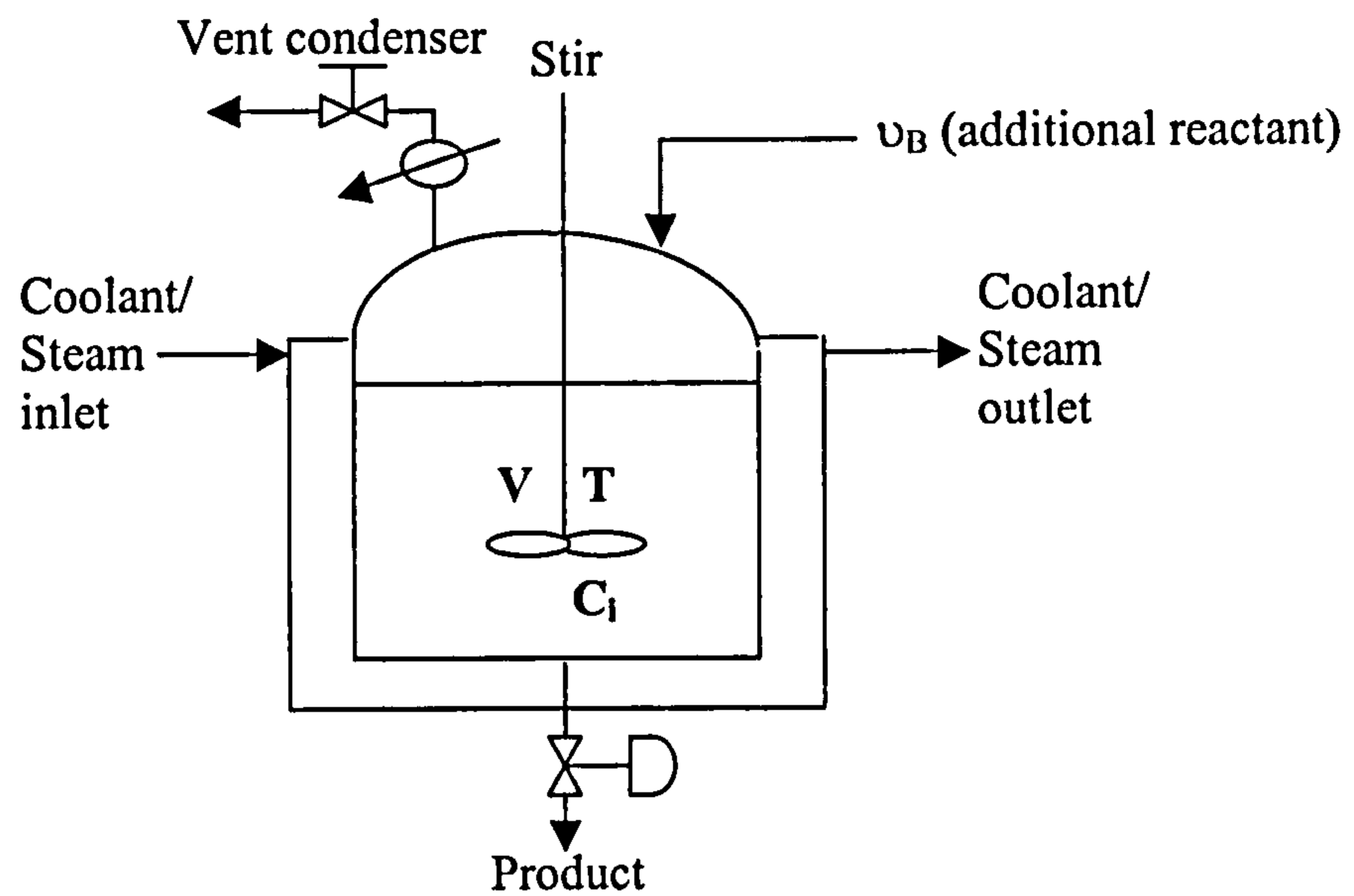


Figure 4.19: Schematic diagram of a jacketed semi-batch reactor

4.5.4.1 Optimisation Formulation of Semi-Batch Reactors

PI - Maximum Conversion Problem

$$\begin{aligned}
 &\text{Max} && DP \\
 &T(t), v_B(t) \\
 &\text{s.t.} && f(t, \underline{x}'(t), \underline{x}(t), \underline{u}(t), \underline{v}) = 0 \text{ (process model)} \\
 &&& t_f = t_f^* \\
 &&& T_L \leq T \leq T_U \text{ and } v_{BL} \leq v_B \leq v_{BU} \\
 &&& V(t) \leq V_r
 \end{aligned}$$

where DP is the desired product; T is the reactor temperature; v_B is the flow rate of feed reactant; t_f is the batch time, T_L and T_U are the lower and upper bounds of the reactor temperature, v_{BL} and v_{BU} are the lower and upper bounds of the flow rate of feed reactant. t_f^* is the fixed batch time. $V(t)$ is the volume of reactor content and V_r is the allowable reactor volume.

P2 - Minimum Time Problem

$$\begin{array}{ll}
 \text{Min} & t_f \\
 & T(t), \nu_B(t) \\
 \text{s.t.} & f(t, \underline{x}'(t), \underline{x}(t), \underline{u}(t), \nu) = 0 \text{ (process model)} \\
 & DP = DP^* \\
 & T_L \leq T \leq T_U \text{ and } \nu_{BL} \leq \nu_B \leq \nu_{BU} \\
 & V(t) \leq V_r
 \end{array}$$

where DP^* is the fixed amount of the desired product, t_f is the batch time.

P3 - Maximum Profit or Productivity Problem

$$\begin{array}{ll}
 \text{Max} & P \\
 & T(t), \nu_B(t) \\
 \text{s.t.} & f(t, \underline{x}'(t), \underline{x}(t), \underline{u}(t), \nu) = 0 \text{ (process model)} \\
 & T_L \leq T \leq T_U \text{ and } \nu_{BL} \leq \nu_B \leq \nu_{BU} \\
 & V(t) \leq V_r
 \end{array}$$

where P is the profit or productivity function.

In this work, it is assumed that the volume of the reactor content, $V(t)$ does not change because of chemical reaction but changes with the addition of external feed.

Therefore at any time $V(t)$ will be linear function of ν_B and will be given by:

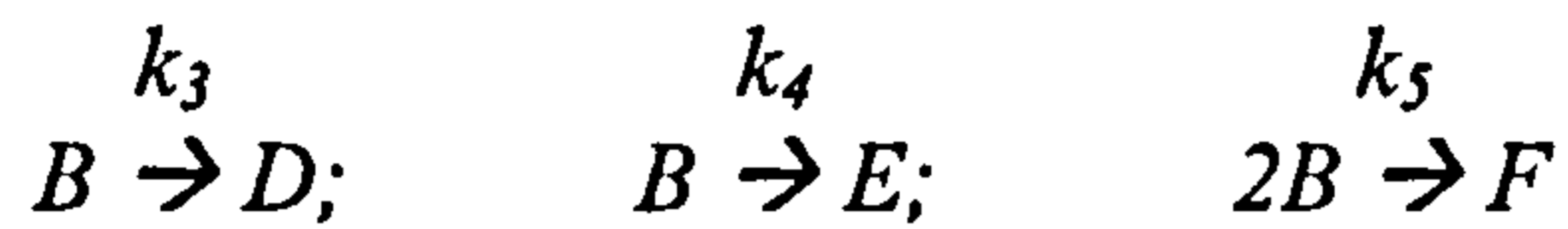
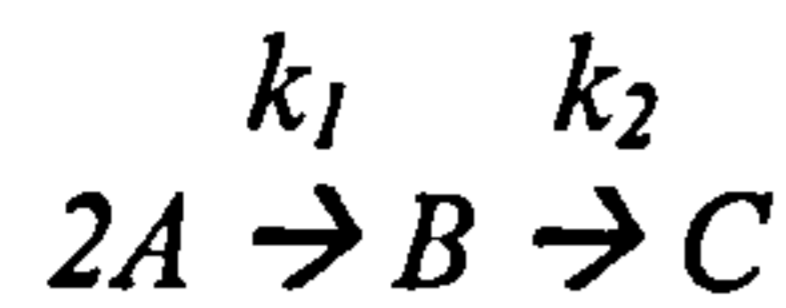
$$V(t) = V(0) + \nu_B t \quad (4.5.4.1)$$

where $V(0)$ is the initial amount of the reactor contents.

To avoid overflow of the reactor contents, following endpoint constraint $V(t_f) \leq V_r$ will ensure no overflow at any time t .

4.5.4.2 Models Equations

The exothermic semi-batch reaction scheme studied in this work is:



where A and B are the raw materials, D is the desired product, E , F and C are the by-products. The mass balances and reaction kinetics equations can be given:

$$\frac{d(C_A V)}{dt} = -2k_1 C_A^2 V \quad (4.5.4.2)$$

$$\frac{d(C_B V)}{dt} = k_1 C_A^2 V - k_2 C_B V - k_3 C_B V - k_4 C_B V - 2k_5 C_B^2 V + \nu_B C_{BF} \quad (4.5.4.3)$$

$$\frac{d(C_C V)}{dt} = k_2 C_B V \quad (4.5.4.4)$$

$$\frac{d(C_D V)}{dt} = k_3 C_B V \quad (4.5.4.5)$$

$$\frac{d(C_E V)}{dt} = k_4 C_B V \quad (4.5.4.6)$$

$$\frac{d(C_F V)}{dt} = k_5 C_B^2 V \quad (4.5.4.7)$$

$$\frac{dV}{dt} = \nu_B \quad (4.5.4.8)$$

$$k_1 = k_{10} \exp\left(-\frac{(E_1/R)}{T}\right) \quad (4.5.4.9)$$

$$k_2 = k_{20} \exp\left(-\frac{(E_2/R)}{T}\right) \quad (4.5.4.10)$$

$$k_3 = k_{30} \exp\left(-\frac{(E_3/R)}{T}\right) \quad (4.5.4.11)$$

$$k_4 = k_{40} \exp\left(-\frac{(E_4/R)}{T}\right) \quad (4.5.4.12)$$

$$k_5 = k_{50} \exp\left(-\frac{(E_5 / R)}{T}\right) \quad (4.5.4.13)$$

where C_A , C_B , C_C , C_D , C_E and C_F are concentration of component A , B , C , D , E and F respectively; k_1 , k_2 , k_3 , k_4 and k_5 are the rate constant for reaction 1 to 5 respectively. V is the total volume of the reactant and ν_B is the feed rate of reactant B .

4.5.4.3 Degrees of Freedom Analysis

Total number of variables in Equations (4.5.4.2 – 4.5.4.13) = 34:

$$\frac{d(C_A V)}{dt}, \frac{d(C_B V)}{dt}, \frac{d(C_C V)}{dt}, \frac{d(C_D V)}{dt}, \frac{d(C_E V)}{dt}, \frac{d(C_F V)}{dt}, \frac{dV}{dt}, k_1, k_2, k_3, k_4, k_5, C_A, C_B, C_C, C_D, C_E, C_F, V, \nu_B, C_{BF}, k_{10}, k_{20}, k_{30}, k_{40}, k_{50}, E_1, E_2, E_3, E_4, E_5, R, T, t$$

Degrees of Freedom,

$$\begin{aligned} \text{D.F.} &= \text{Total number of variables} - \text{Total number of equations} \\ &= 34 - 12 \\ &= 22 \end{aligned}$$

Specifications:

Parameters = 12:

$$C_{BF}, k_{10}, k_{20}, k_{30}, k_{40}, k_{50}, E_1, E_2, E_3, E_4, E_5, R$$

Independent variable = 1 = t

Initial value at time $t = 0$ for all the differential variables ($(C_A V)$, $(C_B V)$, $(C_C V)$, $(C_D V)$, $(C_E V)$, $(C_F V)$, V) to be specified = 7.

The above specifications count to 20 variables. In this work feed reactant flow rate, ν_B and reactor temperature, T are relaxed from the specifications and are optimised.

All the parameter and constant values used in the model are:

$$k_{10}=44.4 \text{ l.mol}^{-1}.\text{min}^{-1}; \quad k_{20}=4500.0 \text{ min}^{-1}$$

$$k_{30}=2500.0 \text{ min}^{-1}; \quad k_{40}=200.0 \text{ min}^{-1}$$

$$k_{50}=400 \text{ l.mol}^{-1}.\text{min}^{-1}; \quad E_1/R=2500 \text{ K}$$

$$E_2/R=5000 \text{ K}; \quad E_3/R=4000 \text{ K}$$

$$E_4/R=3800 \text{ K}; \quad E_5/R=4200 \text{ K}$$

$$C_{AF}=2.0 \text{ mol/l}; \quad C_{BF}=0.6 \text{ mol/l}$$

4.5.4.4 Results

The reactor temperature is optimised within the bounds: $20 \leq T \leq 95^\circ\text{C}$ and the feed rate of reactant B is optimised within the bounds: $0 \leq \nu_B \leq 100 \text{ l/min}$. The reactor volume (V_r) is 1.5 m^3 (1500 l) and the initial values of $[C_C V, C_D V, C_E V, C_F V]$ are $[0.0, 0.0, 0.0, 0.0]$ respectively. The initial values of $(C_A V)$ and $(C_B V)$ vary from case to case. In all runs, 4 time intervals are used within the total batch operation time. In each interval, the temperature, feed rate of reactant B and the length of the interval are optimised.

Maximum Conversion Problem (P1)

The objective of this work is to maximise the desired product D while optimising the reactor temperature and feed rate of reactant B profiles within bounds on the temperature and the feed rate respectively. The fixed batch time t_f is 60 minutes. A number of cases were run with varying amount of initial charge of B. Table 4.7 shows

the optimal temperature and feed rate of reactant B profiles and the maximum amount of product D achieved.

Run 1 is the base case. Runs 2 and 3 show the effect of initial charge of reactant B to the system. Product D increases when some amount of B is charged initially but decreases with excessive amount of initial B. The production of D depends on both the value of k_3 and the amount of B. In Run 1 where there is no B available at the beginning stage, the system operates at high temperature at the beginning of the operation to produce B. The product D will only be produced with the existence of B. Due to this reason; the amount of D achieved in Run 1 is lower compared to the amount of D in Runs 2 and 3. However with excessive amount of B at the beginning of the process (Run 3), the system operates at lower temperature for a longer period to favour reaction $B \rightarrow D$ but this reduces the conversion of B from A. The overall effect therefore is a low production of D.

Table 4.7: Summary of the results for maximum conversion problem

Run	Initial charge(mol)		V_r (m^3)	Optimum Temperature Profile					Prod D (mol)
	A	B		ν_B (l/min)	Temp.(°C)	Time, min	ν_B (l/min)	Temp.(°C)	
1	1500	0	1.5	79.14 90.77	5.99 39.87	3.90 39.80	4.13 75.13	0.0 6.39 11.39 20.35 60.0	581.08
2	1500	240	1.5	8.21 25.45	10.62 82.38	4.62E-3 74.32	4.54 76.33	0.0 5.09 17.30 22.30 60.0	616.37
3	1500	360	1.5	8.78 25.73	8.13 81.71	2.87E-2 40.36	7.19E-3 74.66	0.0 6.42 14.25 19.25 60.0	589.83

Minimum Time Problem

The objective here is to produce the same amount of D as in Table 4.7 in minimum time (Runs 1 and 2 only). The optimal temperature and feed rate of B profiles and the minimum batch time required to achieve the product are presented in Table 4.8.

It is found that minimum time needed to achieve the specified conversion in Runs 1 and 2 is less than the fixed batch time of Table 4.5. For Run 1, the time needed to achieve the same product is 15% lower. This was achieved by operating the reactor at higher temperature thus producing more B and requiring lower feed rate of B (compared to Run 1 of Table 4.7). The minimum time for Run 2 is very similar to the fixed batch time of Run 2 of Table 4.7 but initial high temperature operation reduces the total amount of external feed of reactant B (by 95%). The study of these two types of problems shows that if there is opportunity, productivity (amount of product/time) can be improved by choosing an appropriate type of operation.

Table 4.8: Summary of the results for minimum time problem

Run	Initial charge(mol)		V_r (m ³)	Optimum Temperature Profile					Final time (min)
	A	B		v_B (l/min)	Temp.(°C)	Time, min	v_B (l/min)	Temp.(°C)	
1	1500	0	1.5	v_B (l/min)	33.53	8.98E-4	1.33E-3	1.34	50.83
				Temp.(°C)	95.00	56.27	20.00	68.39	
				Time, min	0.0	21.46	26.46	32.39	
2	1500	240	1.5	v_B (l/min)	0.415	0.0	0.0	0.0	58.69
				Temp.(°C)	95.00	95.00	20.00	20.00	
				Time, min	0.0	39.13	48.69	53.69	

Maximum Profit Problem

The objective here is to determine the optimum amount of product, batch time, the reactor temperature and feed rate of reactant B profiles which will maximise a given profit function. The profit function used in this study is given below:

$$P = \frac{D \times P_D - (A_0 - A) \times P_A - (v_B C_{BF} + P_0) \times C_B + C \times P_C + E \times P_E + F \times P_F}{t} - \frac{0.95}{600} V_r^{0.6} \quad (4.5.4.14)$$

where t is the batch time in minutes. A_0 and B_0 are the amount of A and B at the beginning of the process. P_A , P_B , P_C , P_D , P_E and P_F are the prices of A , B , C , D , E and F with numerical values [0.425, 0.675, 0.5, 2.5, 0.5, 0.625] respectively. All values are in \$/mol. The last part of the Equation 4.5.4.14 denotes the capital cost of the reactor (\$/min) based on the reactor volume in litre. Two cases are run with the same initial conditions as in Runs 1 and 2 of Tables 1 and 2 and the results are compared. The optimal temperature and feed rate of B profiles, and the final product D are presented in Table 4.9.

Table 4.9: Summary of the results for maximum profit problem

Run	Optimum Temperature Profile					Prod D (mol)	Profit \$/min		
							Max Profit	Max Conv	Min Time
1	v_B (l/min)	51.59	8.31	0.0	0.0	563.2	13.54	10.74	12.80
	Temp.(°C)	95.00	78.77	36.01	78.77				
	Time,min	0.0	11.39	19.39	27.39				
2	v_B (l/min)	14.48	9.82	7.79	0.0	607.1	12.14	9.46	11.44
	Temp.(°C)	92.75	78.21	78.18	77.44				
	Time,min	0.0	11.48	21.48	31.48				

From Table 4.9, it is found that the *maximum profit problem* gives the highest profit compared to the profits based on the results of the *maximum conversion* (Table 4.7) and *minimum time problems* (Table 4.8) although the amount of product D, temperature and feed rate profile of B are all different. It shows that optimisation of both time and conversion is important to get the maximum profit of any operation, rather than specify one of them.

4.5.4.5 Concluding Remark

In all cases presented, the *maximum profit* operation gives the highest overall profit compared to the profits obtained using the results of *maximum conversion* (Table 4.7) and *minimum time* (Table 4.8) operations. The *minimum time* operation was found to have advantages over the *maximum conversion* operation, in one case it was in terms of productivity while in another case it was in terms of the total amount of reactant (B) fed in semi-continuous mode. However, the choice of a particular type of optimisation problem depends on the overall schedule of the batch reactor within the whole batch and semi-batch plant and on the requirements of the downstream process.

4.5.5 Case Study 5

The aim of this case study is to study the sensitivity of the profit function with respect to reactants and products prices. In addition, the effect of the volume on the overall profit is also taken into consideration by including it as an additional optimisation variable. Here, the same reaction scheme as in Case Study 3 is considered. However, only the maximum conversion problem is considered for the purpose of comparison with the maximum profit problem. The mathematical model of the system

and the parameters remains the same. Here the mole fraction of A, B and C (x_A , x_B , x_C) are equivalent to the concentration of A, B and C (C_A , C_B , C_C) in the mathematical model in Case Study 3. It is because at any time (throughout the batch time), the total amount of (C_A , C_B , C_C) is equal to one, which can also represent the conversion or mole fraction of the component.

4.5.5.1 Profit Function

Two types of profit functions are considered:

Profit Function 1, (PF1):

$$PF1 = \left(\frac{x_B \times P_B + x_C \times P_C + (x_{A0} - x_A) \times P_A}{t_f} \right) \times Total\ Mole - O_p C - CC \quad (4.5.5.1)$$

Profit Function 2, (PF2):

$$PF2 = \left(\frac{x_B \times P_B + (x_A + x_C) \times P_C + x_{A0} \times P_A}{t_f} \right) \times Total\ Mole - O_p C - CC \quad (4.5.5.2)$$

where

$$CC = 4.67V_r^{0.6} = \text{Capital cost} \quad (4.5.5.3)$$

$$O_p C = F_{Javg} \times P_{FJ} = \text{Operating cost} \quad (4.5.5.4)$$

P_A = price of reactant A

P_B = price of product B

P_C = price of by-product C

x_A = mole fraction of reactant A remaining in the reactor after the operation

x_B = mole fraction of product B

x_C = mole fraction of by-product C

x_{A0} = mole fraction of reactant A at initial stage

V_r = Reactor volume

F_{Javg} = average rate of coolant flow

P_{FJ} = cost of coolant

4.5.5.2 Descriptions of Different Problems

Sixteen (16) different cases were carried out:

Case 1 (base case) – in this case, the maximum conversion problem (P1) is solved.

Case 2 – same as Case 1 but the maximum profit problem (P3) is solved.

Cases 3 and 4 – to see the effect of PF1 when the product B price change.

Cases 5 and 6 – to see the effect of PF1 when the reactant A price change.

Cases 7 and 8 – to see the effect of PF1 when the by-product C price change.

Cases 9 and 10 – to see the effect of PF1 when the cost of coolant changes.

Case 11 – here the by-product is treated as waste, which has to be treated before discharging (involving some treatment cost).

Cases 12 and 13 – to see the effect of reactor volume to the PF1.

Case 14 – the reactor volume is treated as design variable to be optimised.

Cases 15 and 16 – same as Case 2 and 11 respectively but with profit function PF2.

In all cases, 4 time intervals are used and optimised together with final batch time and conversion to achieve maximum profit. The results of all cases are summarised in Table 4.10.

In all cases except Case 11 and 16, the by-product C is assumed saleable but at low price. The PF1 is used for the Cases 2 – 14. It is assumed that in these cases the

unused reactant A can be recycled. In Cases 12 – 14 the effect of reactor volume through the capital cost term is considered and in Case 14 the reactor volume is treated as design variable to be optimised. PF2 is used in Cases 15 and 16, which assume that the unused reactant A is just the waste and has been treated alike the by-product C.

4.5.5.3 Results

In Table 4.10, $P3_f$ shows the profit obtained by solving the maximum profit problem and $P1_f$ shows the profit derived using the results of the maximum conversion problem. For example, all $P1_f$ in Table 4.10 are calculated using the profit function PF1 or PF2 (as described in section 4.5.5.2) but using the same values of (x_A, x_B, x_C) as in Case 1. In different cases, $P1_f$ varies due to different values of $(P_A, P_B, P_C, P_{FJ}, V_r, t_f)$. The superiority of solving maximum profit problem over the maximum conversion problem can be clearly seen in Table 4.10. In all cases the profit gained from maximum profit problem are higher than the profit gained from the maximum conversion problem (in most cases $P3_f$ is over 30% higher than $P1_f$ (Figure 4.20)). In the maximum conversion problem, the batch time is fixed at 3.5 hr, so the way by which it can maximise the profit, is by maximising the product within the time period. On the other hand, in maximum profit problem the batch time is also optimised. Besides Cases 1 and 14, in all other cases the optimal time achieved is less than 3 hr. Although within this optimal period, the conversion of B is slightly less (except case 14) compared to that achieved in maximum conversion problem, it still leads to higher profit.

Table 4.10: Summary of results for Case Study 5

Case	P_A	P_B	P_C	P_{FJ}	V_r	x_A	x_B	x_C	Total Mole	F_{Javg}	t_f	OC	CC	P_{3f}	P_{1f}
1	24.00	135.00	13.50	3.00	1.23	0.1878	0.6463	0.1659	10	1.798	3.50	5.39	5.29	-	191.01
2	24.00	135.00	13.50	3.00	1.23	0.3400	0.5764	0.0836	10	3.731	2.36	11.19	5.29	253.07	191.01
3	24.00	202.50	13.50	3.00	1.23	0.3353	0.5790	0.0857	10	3.750	2.37	11.25	5.29	417.74	315.65
4	24.00	67.50	13.50	3.00	1.23	0.3586	0.5656	0.0758	10	3.675	2.33	11.03	5.29	88.60	66.38
5	36.00	135.00	13.50	3.00	1.23	0.3630	0.5629	0.0741	10	3.768	2.31	11.30	5.29	221.06	164.02
6	12.00	135.00	13.50	3.00	1.23	0.3202	0.5870	0.0928	10	3.699	2.41	11.10	5.29	285.53	218.00
7	24.00	135.00	20.25	3.00	1.23	0.3294	0.5822	0.0884	10	3.710	2.39	11.13	5.29	255.52	194.21
8	24.00	135.00	6.75	3.00	1.23	0.3503	0.5705	0.0792	10	3.751	2.34	11.25	5.29	250.74	187.81
9	24.00	135.00	13.50	4.50	1.23	0.3370	0.5780	0.0850	10	3.693	2.37	16.62	5.29	247.50	188.31
10	24.00	135.00	13.50	1.50	1.23	0.3430	0.5747	0.0823	10	3.776	2.36	5.66	5.29	258.70	193.71
11	24.00	135.00	-13.50	3.00	1.23	0.3809	0.5518	0.0673	10	3.809	2.27	11.43	5.29	244.30	178.21
12	24.00	135.00	13.50	3.00	1.845	0.3420	0.5752	0.0827	15	3.759	2.36	11.43	8.01	385.14	*
13	24.00	135.00	13.50	3.00	0.615	0.3341	0.5796	0.0862	5	3.658	2.38	11.28	3.49	120.15	*
14	24.00	135.00	13.50	3.00	3.653	0.1795	0.6677	0.1528	29.7	8.582	4.53	25.75	10.16	443.90	*
15	24.00	135.00	13.50	3.00	1.23	0.3281	0.5829	0.0891	10	3.817	2.39	11.45	5.29	238.36	185.38
16	24.00	135.00	-13.50	3.00	1.23	0.3342	0.5796	0.0862	10	3.697	2.38	11.09	5.29	190.51	158.09

P_{1f} = Profit calculated using the results of maximum conversion problem (\$/hr)

P_{3f} = Profit obtained by solving maximum profit problem (\$/hr)

* = The results are not comparable as V_r is changing for Cases 12-14 and are not the same as those used for P_{1f}
 x_A, x_B, x_C are mole fraction of A, B & C; P_A, P_B, P_C are in \$/kmol; P_F is in \$/m³; F_{Javg} is in m³/hr; OC, CC are in \$/hr; t_f is in hr; V_r in m³

Figure 4.21 shows the percentage improvement in profit achieved from various cases over the profit achieved in Case 2. From this observation, we can say that the main factors that affect the profit are the price of product B, P_B (Cases 3 and 4) and the reactor volume (Cases 12 – 14). However, the effect of P_A to the profit cannot be ignored (Cases 5 and 6). The effect of the other factors (i.e. P_C and P_{FJ}) is negligible (Cases 7-10). This study shows the sensitivity of different parameters that can affect the profitability of the operation

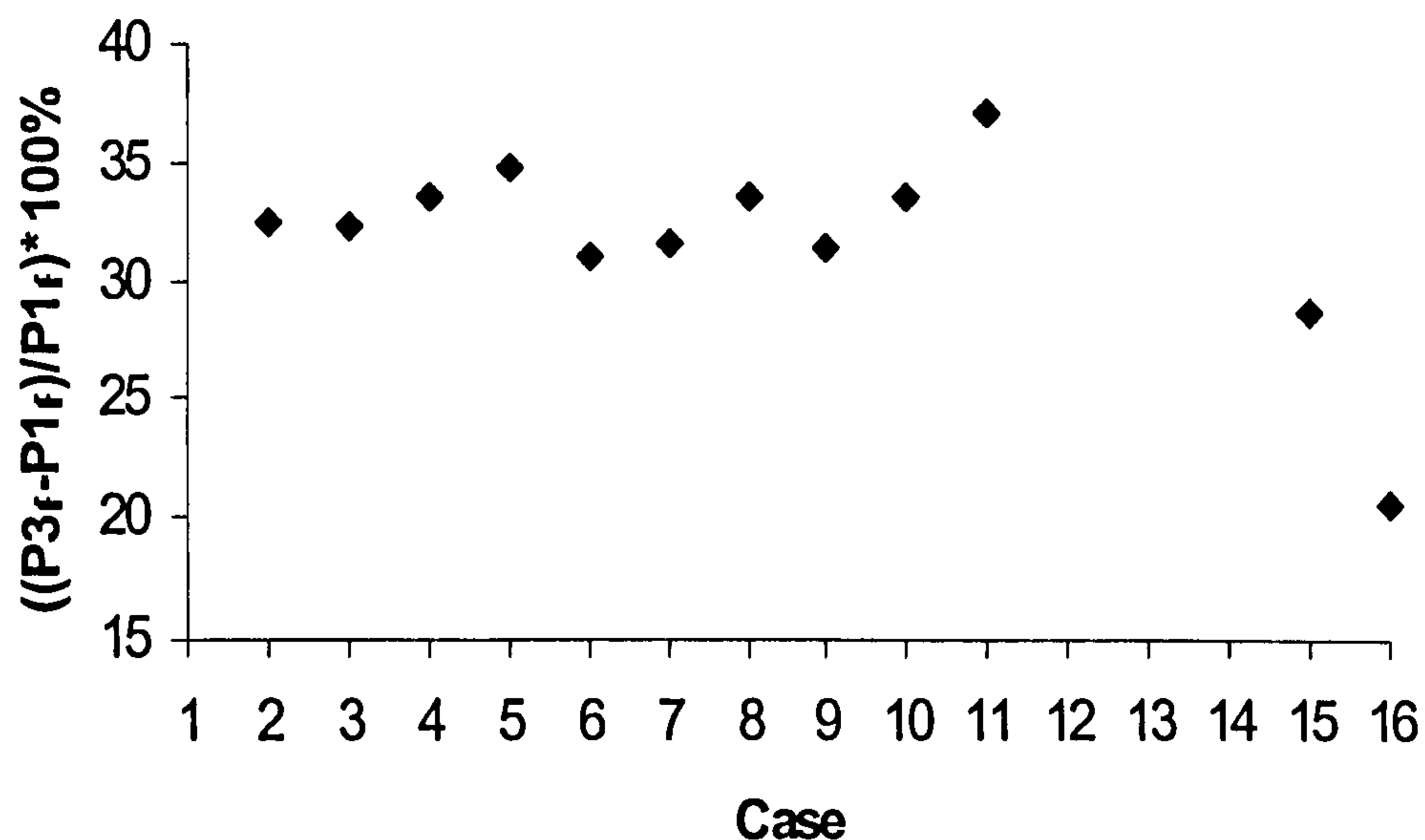


Figure 4.20: Percentage improvement in profit for P_{3f} compared to P_{1f} .

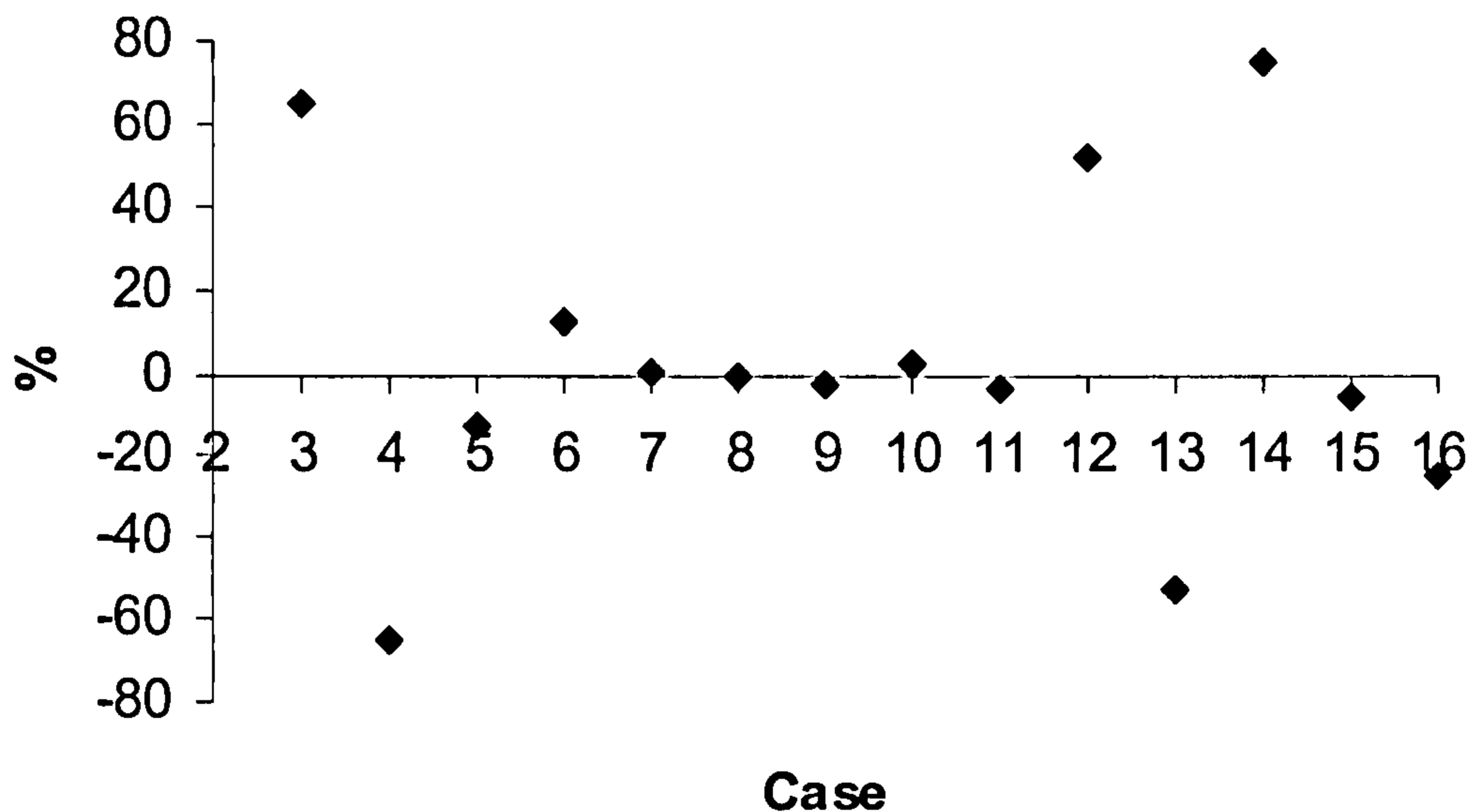


Figure 4.21: Percentage improvement in profit for various cases over the profit obtained in Case 2.

Cases 12 and 13 show the effect of the reactor volume on the profit. It is found that there is a potential to increase the profit by increasing the reactor volume. For this reason, Case 14 is carried out to find the optimal volume and batch time that will maximise the profit. Table 4.10 shows the optimal volume and time to achieve a profit of \$443.90/hr are 3.653 m^3 and 4.53 hr respectively. When the system is given a longer operation period and more room, it is able to utilise them better in order to maximise the profit. Although the amount of coolant flow needed is doubled and the operating cost is increased, increase in revenue due to high conversion of product B outweighs significantly the operating cost and leads to higher overall profit.

Cases 15 and 16 are similar to Cases 2 and 11 respectively. However, the profit function used is different. Cases 15 and 16 used the profit function PF2 and Cases 2 and 11 used the profit function PF1 respectively. PF1 assumes that the reactant A left after

the operation can be recycled and used in the next batch. In contrast the PF2 assumes that the un-reacted reactant A, will become waste and is treated alike as by-product C. This is usually the case when the separation task for A is very costly.

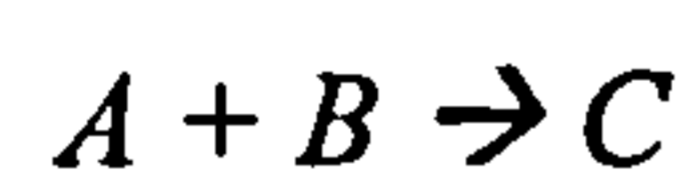
In Case 11, C is penalised so less amount of C (x_C) is produced. However, it still maintains similar level of profit (as in Case 2) by minimising t_f little bit and producing slightly less amount of B. Similar condition is observed in Cases 15 and 16 but in Case 16, profit obtained is decreased considerably due to penalty on both A and C. Since in Case 16 both (A+C) are penalised compared to Case 11 (only C is penalised), amount of A and C ($x_A + x_C$) decreased by 6.2% but the amount of B is increased by 5.04%.

4.7.6 Case Study 6

(Part of this work was presented in ESCAPE-10, Florence, Italy, 7-10 May 2000, Aziz et al., 2000a and PSE2000, Keystone, Colorado, USA, 16-21 July 2000, Aziz et al., 2000b)

Here the reaction scheme is same as that used by Cott and Macchietto (1989).

The reaction scheme is:



where A, B are the raw materials, C is the desired product and D is the waste product.

The aim of this study is to obtain an optimum temperature profile, which will maximise the desired product C. The optimum temperature profile will then be used as set points for designing controllers for on-line tracking in Chapter 5.

4.5.6.1 Model Equations

The model equations are same as those used in the original reference but can be derived using the Option 1 in GBRM presented in Chapter 3. The number of components, $m = 6$ and the number of reaction, $n = 2$. The values of ν_{11} , ν_{12} , ν_{21} , and ν_{23} are equal to -1 ; the values of ν_{13} and ν_{24} are equal to 1 and the rest of ν_{ji} are equal to zero. The values of β_{11} , β_{21} , β_{12} and β_{32} are equal to 1 . The rest of β_{ij} are equal to zero.

Using M_i for C_i in equations 3.27 and 3.28 in Chapter 3 one obtains:

$$\frac{dM_A}{dt} = -r_1 - r_2 \quad (4.5.6.1)$$

$$\frac{dM_B}{dt} = -r_1 \quad (4.5.6.2)$$

$$\frac{dM_C}{dt} = r_1 - r_2 \quad (4.5.6.3)$$

$$\frac{dM_D}{dt} = r_2 \quad (4.5.6.4)$$

$$r_1 = k_1 M_A M_B \quad (4.5.6.5)$$

$$r_2 = k_2 M_A M_C \quad (4.5.6.6)$$

Equation 3.29 in Chapter 3 for $j = 1$ results in:

$$k_1 = k_{10} \exp\left(-\frac{E_1}{RT}\right) \quad \text{which is equivalent to:}$$

$$k_1 = \exp\left(k_1^1 - \frac{k_1^2}{(T_r + 273.15)}\right) \quad (4.5.6.7)$$

$$\text{where } k_{10} = \exp(k_1^1) \text{ and } \exp\left(-\frac{E_1}{RT}\right) = \exp\left(-\frac{k_1^2}{(T_r + 273.15)}\right)$$

Similarly for $j = 2$:

$$k_2 = \exp\left(k_2^1 - \frac{k_2^2}{(T_r + 273.15)}\right) \quad (4.5.6.8)$$

4.5.6.2 Degrees of Freedom Analysis

Total number of variables in Equations (6.7.6.1 – 6.7.6.8) = 18:

$$\frac{dM_A}{dt}, \frac{dM_B}{dt}, \frac{dM_C}{dt}, \frac{dM_D}{dt}, r_1, r_2, k_1, k_2, M_A, M_B, M_C, M_D, k_1^1, k_1^2, k_2^1, k_2^2, T_r, t$$

Degrees of Freedom,

$$\begin{aligned} \text{DF} &= \text{Total number of variables} - \text{Total number of equations} \\ &= 18 - 8 \\ &= 10 \end{aligned}$$

Specifications:

Parameters = 4:

$$k_1^1, k_1^2, k_2^1, k_2^2$$

Independent variable = 1 = t

Initial value at time $t = 0$ for all the differential variables M_A, M_B, M_C, M_D to be specified = 4.

The above specifications count to 9 variables. In this work reactor temperature, T_r is relaxed from the specifications and is optimised.

All the constant values used in the model are:

$$\begin{aligned} k_1^1 &= 20.9057; & k_1^2 &= 10000 \\ k_2^1 &= 38.9057; & k_2^2 &= 17000 \end{aligned}$$

4.5.6.3 Results

Here an off-line dynamic optimisation problem (P1) is solved to find the optimum temperature profile that will maximise the product “C” and minimise the by-product “D”. Two runs were carried out; RUN1 uses one control interval (time) and RUN2 uses three fixed control intervals. The batch time is 120 minutes and the initial values of $[M_A, M_B, M_C, M_D, T_r, T_j]$ are $[12.0, 12.0, 0.0, 0.0, 20.0, 20.0]$ respectively. The reactor temperature is used as the controlled variable and is bounded between 20 and 100°C.

The results (optimal temperature profiles) for both runs will be used as the set point to be tracked by various types of controllers in the next chapter. The results are summarised in Table 4.11.

Table 4.11: Summary of the Results for Case Study 6

Run	Optimum Temperature Profile		M_C
1	Temperature, °C	92.46 -----	6.5126
	Switching time, min	t = 0 120.0	
2	Temperature, °C	92.83 91.17 93.41 ----- ----- -----	6.5171
	Switching time, min	t = 0 40.0 80.0 120.0	

Table 4.11 shows that by using three control intervals, the amount of product achieved is slightly higher than that obtained using one control interval meaning that for the type of problem and batch time chosen, the maximum conversions achieved are insensitive to the number of control intervals used. It is due to the fact that the temperature profiles obtained in RUN2 is very similar to that of RUN1. In fact, the average temperature in RUN2 is nearly equal to that in RUN1.

4.6 Summary and Conclusions

In the past, most of the previous researchers were only interested to demonstrate the ability and the effectiveness of the numerical methods they proposed to solve optimisation problem. For this reason, most of them have studied only maximum conversion and/or minimum time problem with very simple model. In contrast, in this chapter, dynamic optimisation (or also known as optimal control) of various reaction schemes in batch reactor has been studied by using one of the promising techniques i.e. CVP. We studied all types of optimisation problems i.e. maximum conversion, minimum time and maximum profit problems. We also considered practical operating constraints on safety, waste production and the quality of product. A new approach of solving optimisation problem with safety constraint was introduced. To summarise, the materials presented in this chapter are:

1. Two case studies were carried out to study the efficiency of CVP technique chosen. The results achieved were compared with one of the other techniques available i.e. IDP. It was found CVP method is as accurate as IDP but results to practical and easily implementable control.
2. Two types of models (simple and detailed) based on first principles are used. Their applications were further discussed and stressed. The simple model is mainly used to obtain optimum reactor temperature, which can be used as a basis for a reactor design. On the other hand, the detailed model should be used to optimise operating condition of the existing reactor and can be used for designing new reactors.
3. A new solution approach to solve a path (safety) constrained problem is introduced. Traditionally, path constrained problems have been solved as a one

step method over the time horizon concerned. Here we proposed a multiple steps method (sequential solution approach). The results achieved showed the merit of the latter over the traditional approach.

4. All types of optimisation problems have been solved. The importance of each type of problem and when they should be used are highlighted.
5. The design variable was included in the optimisation problem for first time. Here the reactor volume was considered as design variable that has to be optimised in order to maximise the profitability of the investment (capital cost) and operation.
6. A case study was carried out to study the sensitivity of the profitability with respect to product price, reactant cost, by-product and operating cost.

All optimisation studies presented in this chapter were carried off-line (open loop system). To implement these results on-line, Case Study 6 has been chosen which is detailed in the next chapter.

Chapter Five

5.0 Control

(On-line Tracking of Optimal Profiles)

5.1 Introduction

In the past, many researchers have studied the dynamic optimisation (optimal control) of batch reactors (Chapter 4). They determined the optimum reactor temperature for different reaction schemes, which maximises the yield, productivity, profit, etc. However, all these researchers considered only the off-line optimisation problems. None of them have implemented these results on-line. Designing controllers to implement the optimal control profiles or tracking the dynamic set points on-line is an important area of research for inherently dynamic batch processes. The non-linear nature of most dynamic systems in the real world has demanded the incorporation of non-linear control techniques to improve the controller performance. Although many different techniques, involving non-linear models have been proposed and applied for controls of non-linear systems (Bequette, 1991), in reality non-linear system modelling and identification is much more complex and difficult to obtain when compared to linear systems. Here we implement the optimal profiles achieved in the previous chapter to design and evaluate different types of non-linear controllers.

5.2 Generic Model Control (GMC) Strategy

(Part of this work was presented at:

a) ESCAPE-10, Florence, Italy, 7-10 May 2000, Aziz et al., 2000a

b) PSE2000, Keystone, Colorado, USA, 16-21 July 2000, Aziz et al., 2000b

c) In "Application of neural networks and other learning technologies in process engineering, Eds. I.M.Mujtaba and M.A.Hussain", Aziz et al., 2001a)

In this work, we implement the GMC controller coupled with on-line neural network heat-release estimator. The optimum temperature profile thus obtained (off-line) is used as the set point to be tracked (on-line) by the GMC controller.

5.2.1 Theoretical Formulation

Generic Model Control (GMC), a model-based control strategy developed by Lee and Sullivan (1988) is one of several advanced process control algorithms developed recently. The GMC uses non-linear models of a process to determine the control action. The desired response can be obtained by incorporating two tuning parameters. There are several advantages that make GMC a good framework for developing reactor controllers (Cott and Macchietto, 1989):

1. The process model appears directly in the control algorithm.
2. The process model does not need to be linearised before use, allowing for the inherent non-linearity of exothermic batch reactor operation to be taken account.
3. By design, GMC provides feedback control of the rate of change of the control variable. This suggests that the rate of temperature change, which is very important in heat-up operations, can be used directly as a control variable.

4. The relationship between feed forward and feedback control is explicitly stated in GMC algorithm.
5. The GMC algorithm is relatively easy to implement.
6. Importantly, the GMC framework permits us to develop a control algorithm that can be used for both heat-up and temperature maintenance and therefore eliminates the need for switching criterion between algorithms; this should result in much more robust control strategy.

The GMC control algorithm can be written as:

$$\frac{dx}{dt} = K_1(x_{sp} - x) + K_2 \int_0^t (x_{sp} - x) dt \quad (5.2.1)$$

where x is the current value and x_{sp} is the desired value of the control variable.

The first expression in the algorithm $K_1(x_{sp} - x)$ is to bring the process back to steady state due to change in $\frac{dx}{dt}$. In order to make the process have a zero offset, the

second expression $K_2 \int_0^t (x_{sp} - x) dt$ is introduced. Details of this GMC method can be

found in Lee and Sullivan (1988).

For temperature control of the batch reactor, a process model relating the reactor temperature, T to the manipulated variable i.e. the jacket temperature, T_j is required. Assuming that the amount of heat retained in the walls of the reactor is small in comparison to the heat transferred in the rest of the system, an energy balance around the reactor contents gives the following model:

$$\frac{dT}{dt} = \frac{Q_r + UA(T_j - T)}{\rho_r c_p V_r} \quad (5.2.2)$$

Replacing T for x and T_{sp} for x_{sp} in Equation 5.2.1, combining Equations 5.2.1 and 5.2.2 and finally solving for the manipulated variable, T_J the control formulation under the GMC is given by:

$$T_J = T + \frac{V_r c_p \rho_r}{UA} \left[K_1 (T_{sp} - T) + K_2 \int_0^t (T_{sp} - T) dt \right] - \frac{Q_r}{UA} \quad (5.2.3)$$

where T_J gives the jacket temperature trajectory required so that the reactor temperature, T follows the desired trajectory incorporating the values of GMC tuning parameters, K_1 and K_2 .

The discrete form of Equation 5.2.3 for the k^{th} time interval is implemented for the on-line control and is given by:

$$T_J(k) = T(k) + \frac{V_r c_p \rho_r}{UA} \left[K_1 (T_{sp} - T(k)) + K_2 \sum_0^k (T_{sp} - T(k)) \Delta t \right] - \frac{Q_r}{UA} \quad (5.2.4)$$

where Δt is the sampling time.

However, Equation 5.2.4 gives the actual jacket temperature, $T_J(k)$ which is not the jacket temperature set point, $T_{Jsp}(k)$, needed to control the reactor temperature at its set point T_{sp} . It is reasonable to assume that the dynamics of the jacket temperature control are approximately first order (Liptak, 1986) with time constant τ_J and hence, the T_{Jsp} can be further calculated using the following equation:

$$T_{Jsp}(k) = T_J(k-1) + \frac{\tau_J}{\Delta t} [T_J(k) - T_J(k-1)] \quad (5.2.5)$$

5.2.2 On-Line Estimation of the Heat-Release using Neural Network Techniques (GMC Strategy)

The success of the GMC controller as formulated in Equation 5.2.4 is largely dependent on the ability to measure, estimate, or predict the heat-release, Q_r at any given time. As neural networks have been proven to be an accurate and fast on-line dynamic estimator, they have been used to carry out the task in this work (Hussain, 1999). A multi-layered feed forward network is used which is trained using the back-propagation method. The back-propagation method (see Appendix B) has been chosen since it is the most well known and widely used algorithm associated with the training of a feed forward neural network. The neural network systems identification steps can be seen in Figure 5.1.

The multi-layered feed forward neural network (Figure 5.2) consists of a set of nodes, which are arranged in layers. The nodes in each layer are connected to all the nodes in the layer above/next to it and all the signals propagate in a forward direction through the network layers. There are no self-connections, lateral connections or back connections. In each node (in hidden and output layer), a constant bias is added. The outputs of nodes in one layer are transmitted to nodes in another layer through connections, which incorporate weighting factors that amplify or attenuate such outputs. The net input to each node (except for input layer nodes) is the sum of the weighted outputs of the nodes in the prior layer. Each node is activated in accordance with the input to the node, the activation function and the bias/threshold of the node. There are various types activation functions available but in this work, the log-sigmoid function has been used in both the hidden and output layers. The architecture of the multi-layered feed forward neural network can be seen in Figure 5.2.

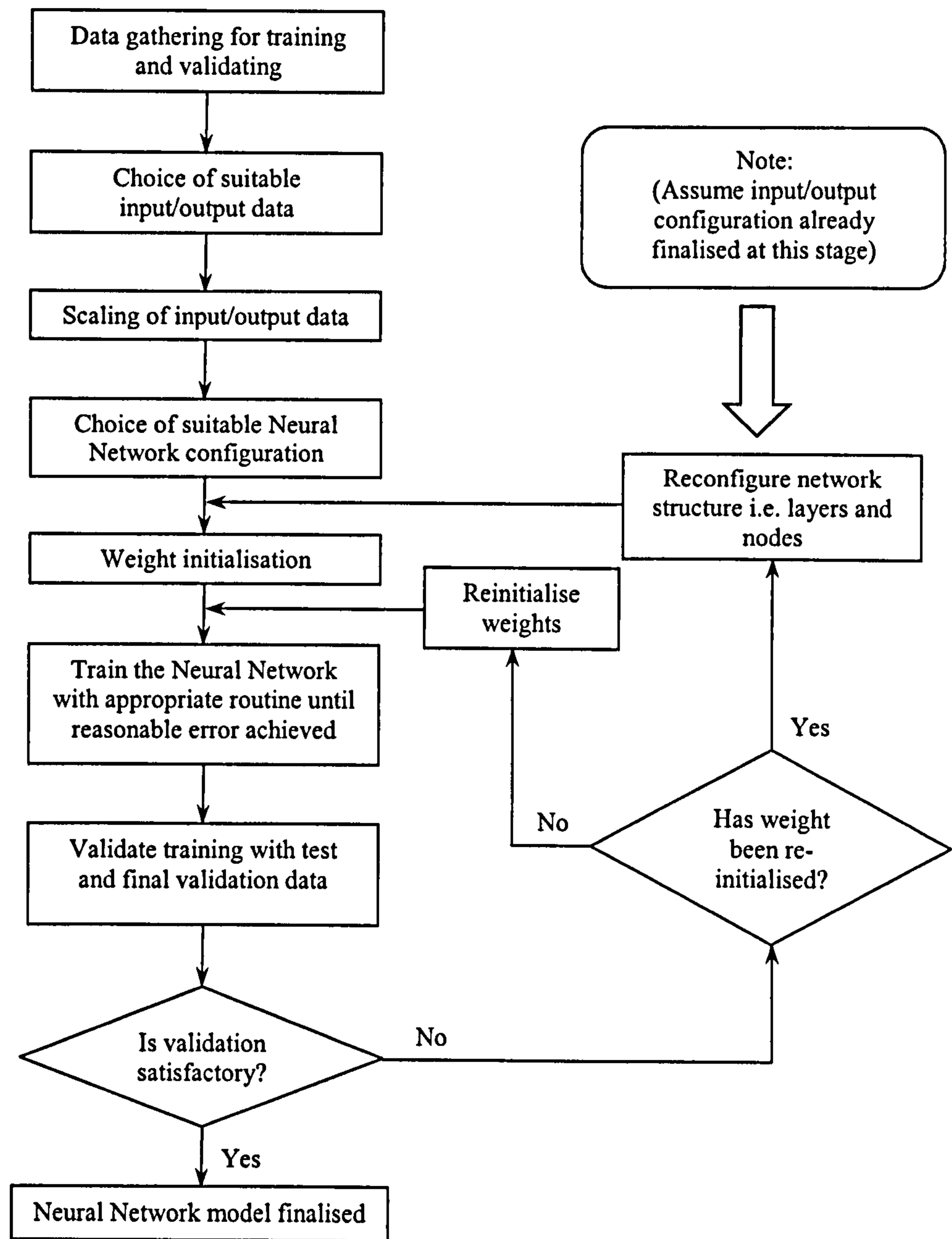


Figure 5.1: Neural network systems identification – Basic steps

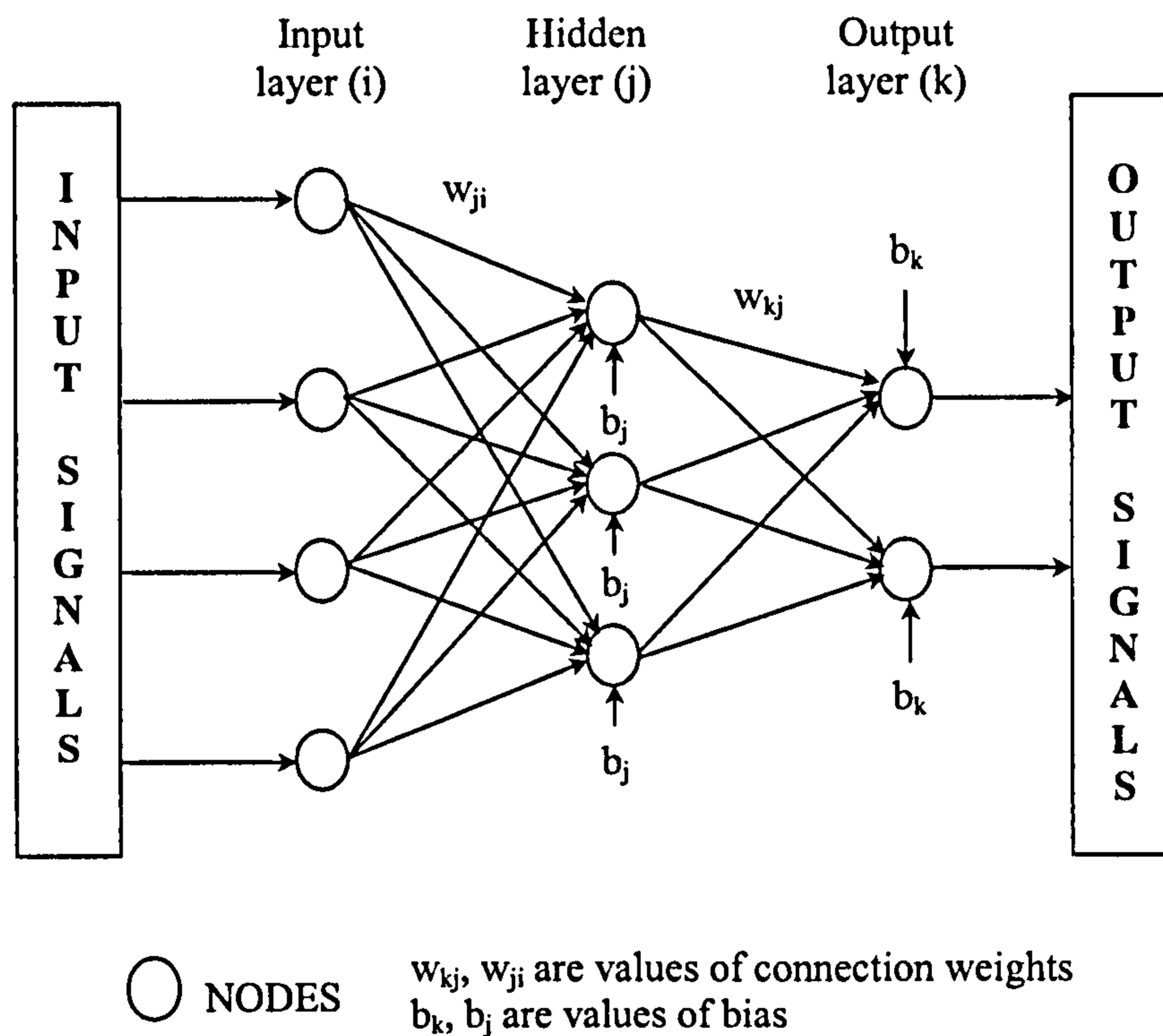


Figure 5.2: Multi-layered feed forward neural network topology

The numbers of hidden layers and nodes may vary in different applications and depend on the user specifications. No specific technique is available to decide the optimum number and it is usually carried out through trial-and-error procedure. Since the process being studied is a dynamic system, it is necessary to feed the neural network with past historical data. Here the input layers consists of the present and past values of T ($T(k-2)$, $T(k-1)$, $T(k)$), T_J ($T_J(k-1)$, $T_J(k)$) and the past values of Q_r ($Q_r(k-1)$) and the output layer estimates the value of the heat-release, Q_r at time interval k .

With these 6 inputs, the neural network is trained through forward modelling methodology to obtain the value of the output i.e. present value of $Q_r(k)$. Forward modelling is the most popular and straightforward approach to augment the network inputs with corresponding discrete past inputs and past outputs data signals from the

model or system being identified. Other state variables that are considered as part of the inputs can also be fed into the network. All the data are moved forward at one discrete-time interval until all of them are fed into the network in a moving window scheme. All data are fed into the neural network repeatedly until the training error criterion is achieved. After this step, the designed neural network with its weights, biases and chosen functions is validated/tested with a new set of data before being used in the GMC strategy. The input-output map for the neural network training can be seen in Figure 5.3. Here the neural network is placed in parallel with the estimator for Q_r and the error between them and neural network output (i.e. prediction error) is used as the training signal for the neural network (see Figure 5.4). In Figure 5.4, $u(k)$ refers to the 6 inputs mentioned above i.e. T , T_j and Q_r while $y(k+1)$ and $\hat{y}(k+1)$ refer to the actual (reference) value of $Q_r(k+1)$ from model and predicted output of $Q_r(k+1)$ from neural network respectively. z^{-1} and z^{-k} refer to the past values at the previous time step and previous k^{th} time step respectively for inputs and outputs. The estimated Q_r is then used in Equation 5.2.4 to estimate the value of T_j and is then applied in the GMC strategy as illustrated in Figure 5.5.

	$T(k-2)$	$T(k-1)$	$T(k)$
□ Input		$T_J(k-1)$	$T_J(k)$
■ Output		$Q_r(k-1)$	$Q_r(k)$

Figure 5.3: Input/output map of neural network estimator

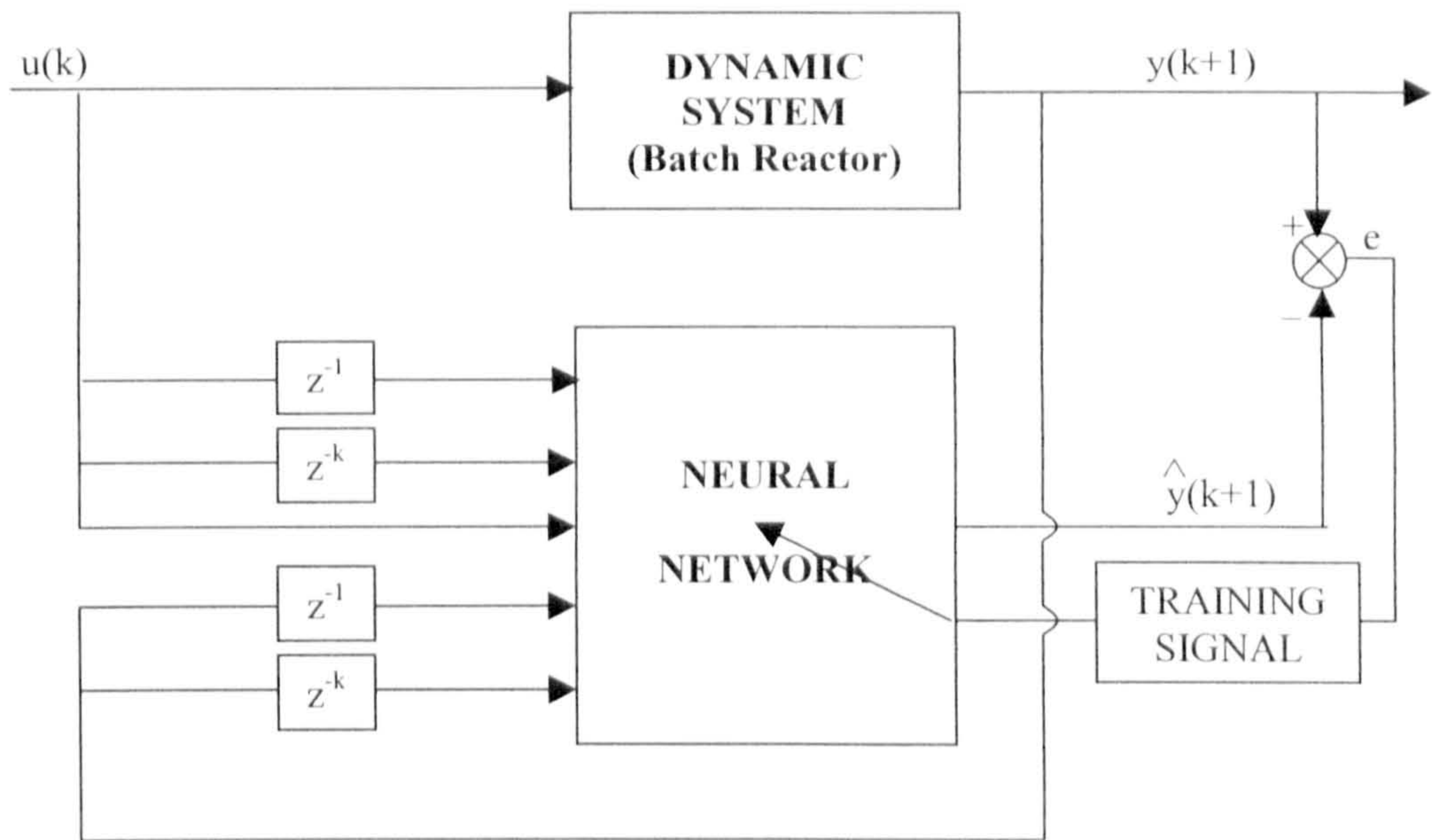


Figure 5.4: Forward modelling of heat-release by neural network

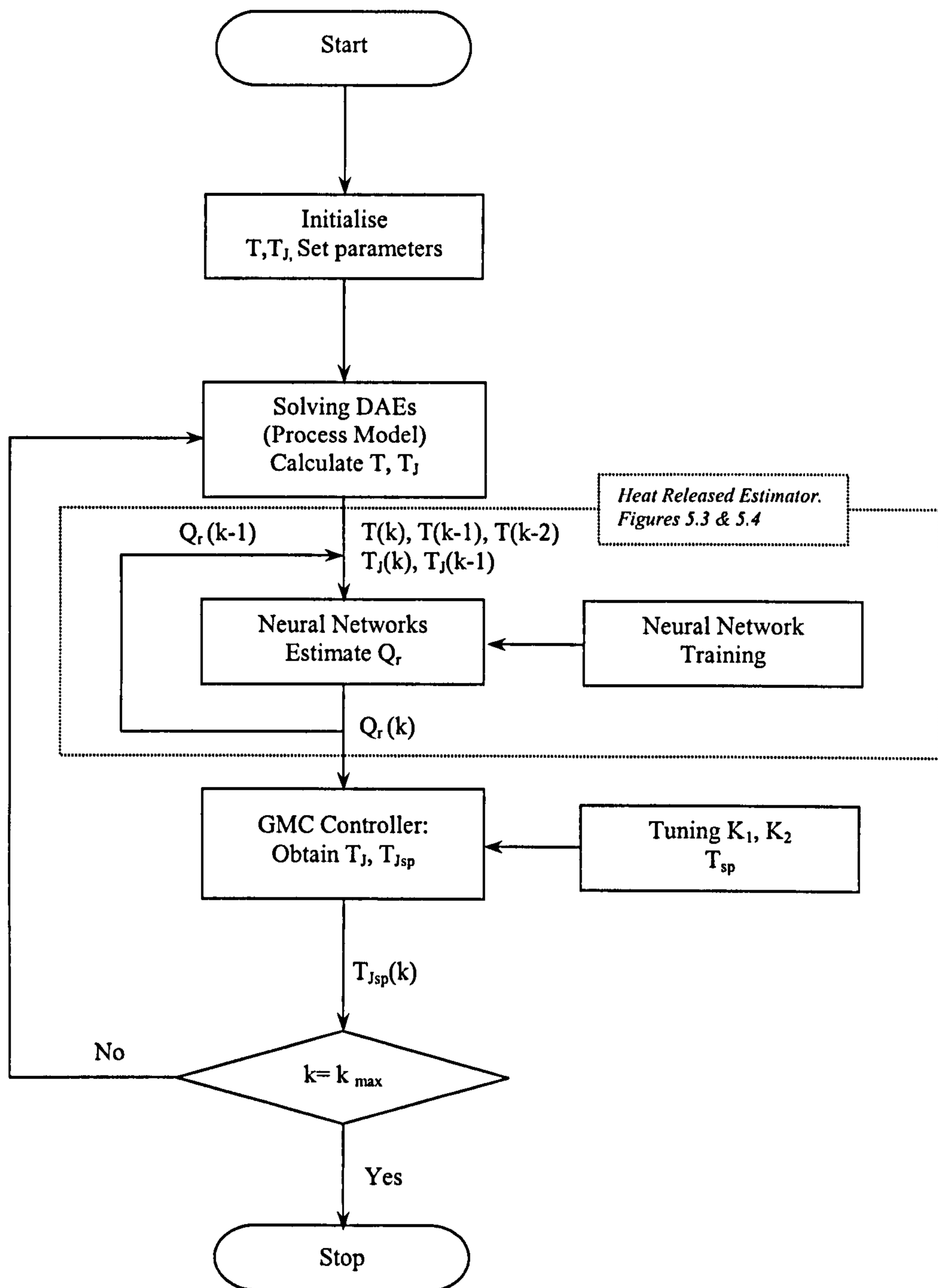


Figure 5.5: GMC-NN strategy in controlling batch reactor

In Case Study 1, four sets of data have been used for training and one set of data is used for validation. The specification for this neural network estimator is:

Neural Network Specifications:

Number of input layer : 6
 Number of hidden layer : 20
 Number of output layer : 1
 Activation function : Sigmoid function applied in both layers
 Error criteria :

	No. of samples	Mean Squared Error (MSE)	Sum Squared Error (SSE)
Training Data 1	350	9.8778×10^{-9}	3.4869×10^{-6}
Training Data 2	350	8.7857×10^{-5}	0.0310
Training Data 3	350	2.5873×10^{-5}	0.0090
Training Data 4	350	3.2419×10^{-5}	0.0114
Validation Data	350	4.0117×10^{-5}	0.0142

In Case Study 2, four sets of data have been used for training and two sets of data are used for validation. The specification for this neural network estimator is:

Neural Network Specifications:

Number of input layer : 6
 Number of hidden layer : 18
 Number of output layer : 1
 Activation function : Sigmoid function applied in both layers

Error criteria :

	No. of samples	Mean Squared Error (MSE)	Sum Squared Error (SSE)
Training Data 1	600	6.9585×10^{-9}	4.1960×10^{-6}
Training Data 2	600	4.4512×10^{-8}	2.6841×10^{-5}
Training Data 3	600	2.1747×10^{-6}	0.0013
Training Data 4	600	3.9412×10^{-6}	0.0024
Validation Data 1	600	3.0259×10^{-8}	1.8216×10^{-5}
Validation Data 2	600	3.8796×10^{-6}	0.0023

5.3 Neural Controller

Since neural network is able to model non-linear systems with some desired accuracy, it has high potential to fulfil the control task (as non-linear controller). Moreover, it is very versatile in nature that made it easily be incorporated in various conventional and advanced control strategies for non-linear systems (Hussain and Kershenbaum, 2000).

In this work, a Direct Inverse control strategy and an inverse-model-based control strategy (IMBC) are used to control the optimal temperature profiles of a complex exothermic reaction. The performance of these control strategies will be compared with the performance of the GMC coupled with neural network (GMC-NN).

5.3.1 Forward Model

As for the neural network estimator in GMC-NN strategy, forward modelling approach is also used here to predict the future value of the reactor temperature, T. This neural network will be used as the model in the IMBC strategy. The input/output pattern

for this forward model (for the batch reactor under consideration) is shown in Figure 5.6. There are two sets of data have been used for training and one set is used for validation. The neural network specification for this forward model is:

Neural Network Specifications:

Number of input layer	: 7
Number of hidden layer	: 15
Number of output layer	: 1
Activation function	: Sigmoid function applied in both layers
Error criteria	:

	No. of samples	Mean Squared Error (MSE)	Sum Squared Error (SSE)
Training Data 1	600	6.6165×10^{-9}	3.9897×10^{-6}
Training Data 2	600	9.6036×10^{-9}	5.7910×10^{-6}
Validation Data	600	6.3014×10^{-8}	3.7997×10^{-5}

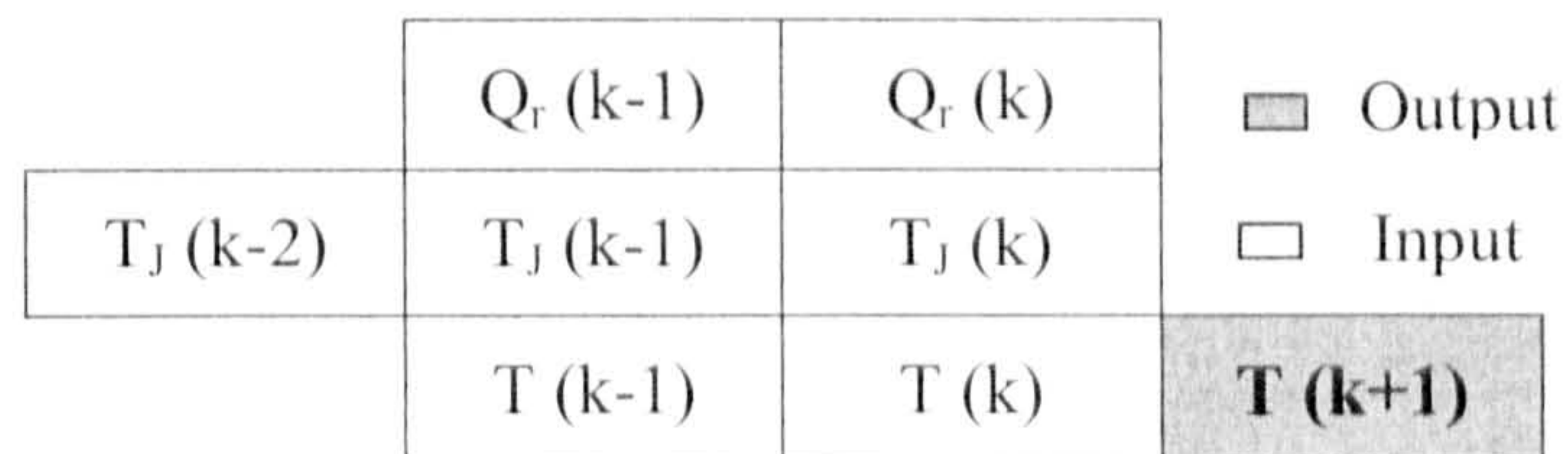


Figure 5.6: Input/output patterns for forward model in IMBC strategy

5.3.2 Inverse Model

Inverse model is basically the neural network structure representing the inverse of the system dynamics at the region of the training or identification. During training the network is fed with the required future or reference output together with the past inputs and past outputs to predict the current input or control action, $u(k)$ (Figure 5.7). Here, control action, $u(k)$ is jacket temperature, $T_j(k)$. This trained network represents the inverse model of the system. The assignment of the input nodes is same as that of forward model but with the prediction of $y(k+1)$ replaced by the control input, $u(k)$ as the network input. The input/output pattern for this inverse model (for the batch reactor under consideration) is shown in Figure 5.8. It is to be noted that at any k , $T_{sp}(k+1)$ and $T_{sp}(k+2)$ are also known in advance.

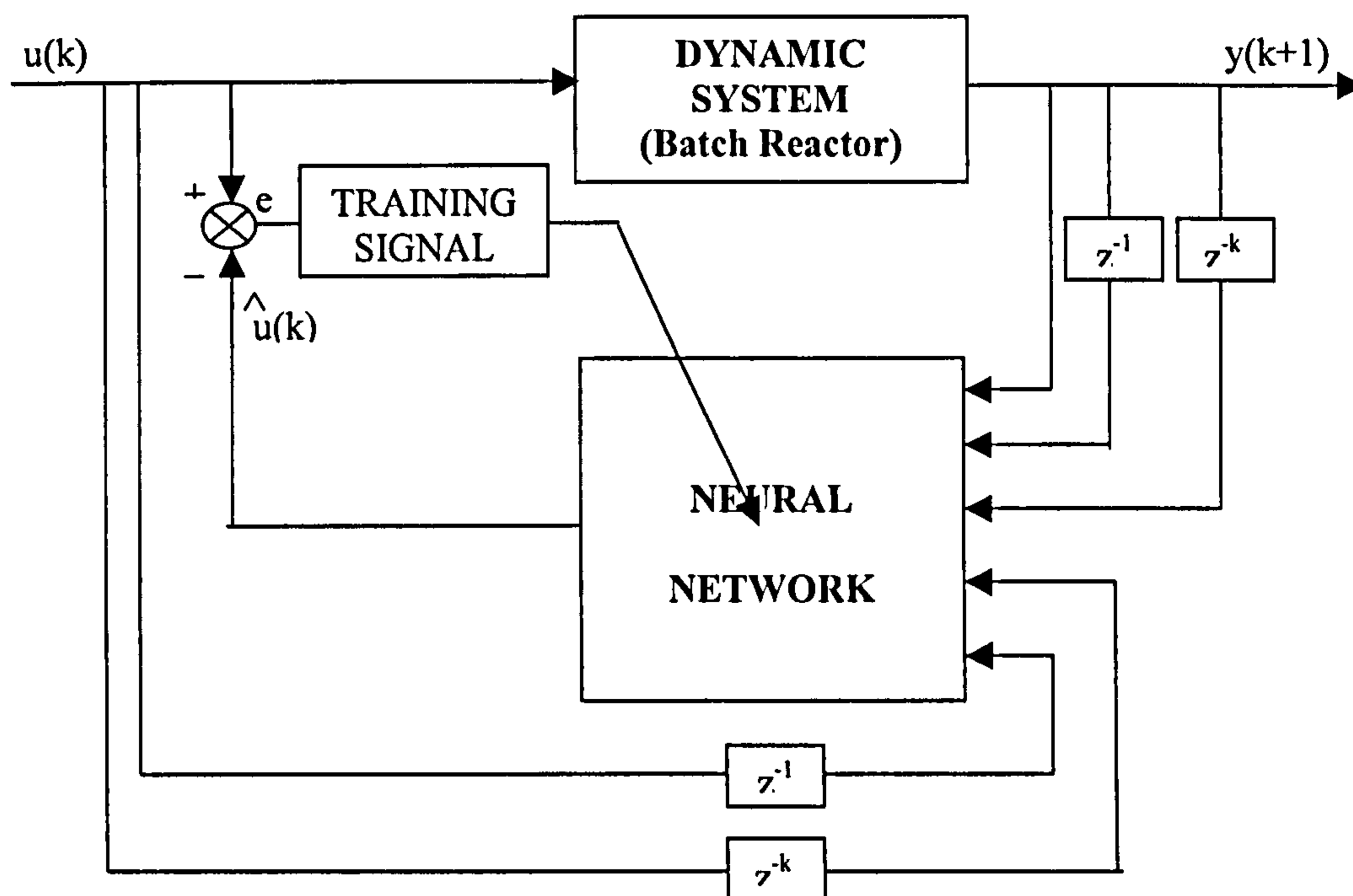


Figure 5.7: Method for the training of the inverse model for control

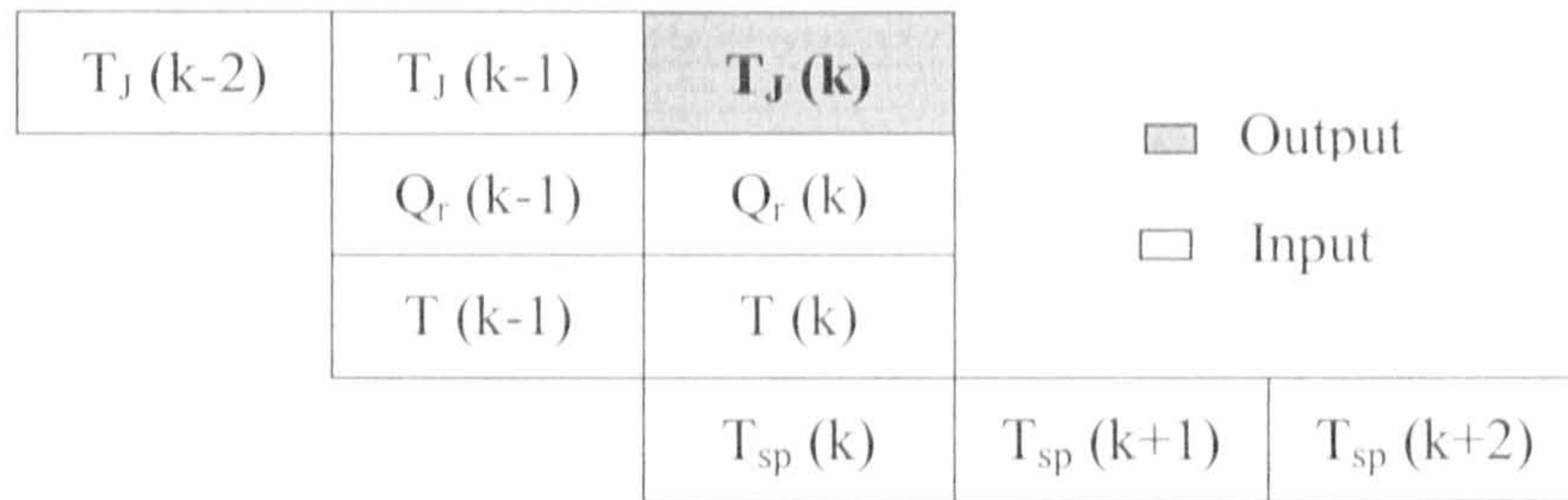


Figure 5.8: Input/output patterns for inverse model

There are two sets of data have been used for training and one set is used for validation. The neural network specification for this inverse model is:

Neural Network Specifications:

Number of input layer	: 9
Number of hidden layer	: 20
Number of output layer	: 1
Activation function	: Sigmoid function applied in both layers
Error criteria	:

	No. of samples	Mean Squared Error (MSE)	Sum Squared Error (SSE)
Training Data 1	600	5.2681×10^{-6}	0.0032
Training Data 2	600	1.1322×10^{-4}	0.0684
Validation Data	600	8.9409×10^{-5}	0.0540

5.3.3 Direct Inverse Control Technique

Here, the inverse model obtained in the previous section will act as the controller in cascade with the system under control, without any feedback (Figure 5.9). The neural network, acting as the controller, has to learn to supply at its output the appropriate

control parameters to achieve the desired targets (set points). In these control scheme the desired set points (y_{sp}) are fed to the network together with the past plant inputs and outputs to predict the desired control parameters, $u(k)$ which will produce expected output ($y(k+1)$) from the model/system (which should be same as y_{sp}). The neural network estimator for Q_r (designed for GMC-NN strategy) is also implemented in this strategy to supply the present and past Q_r values, which will provide the input data to the inverse model. It will also be used in the IMBC strategy. In Figure 5.9 the plant is represented by a first principle based batch reactor model.

5.3.4 Inverse-Model-Based Control Strategy (IMBC)

Internal model control (IMC) strategy is one of the many control strategies that can be applied to various chemical process plants. It promises to offer better control and reliability compared to other control strategies (Hussain, 1999). In this scheme, both the forward and inverse models are used directly as elements within the feedback loop. Here we named it as IMBC. The neural network inverse model is utilised in the control strategy by simply cascading it with the controlled system or plant. In this case the neural network acting as the controller, has to learn to supply at its output, the appropriate control parameters, $u(k)$ to achieve the desired targets, y_{sp} . In addition, the forward model is placed in parallel with the plant, to cater for plant model mismatches and the error between the plant output and neural network forward model is subtracted from the set point before being feedback into the inverse model (Figure 5.10). The inverse and forward models obtained a prior will be incorporated in this IMBC strategy. In Figure 5.10 the plant is again represented by a first principle based batch reactor model.

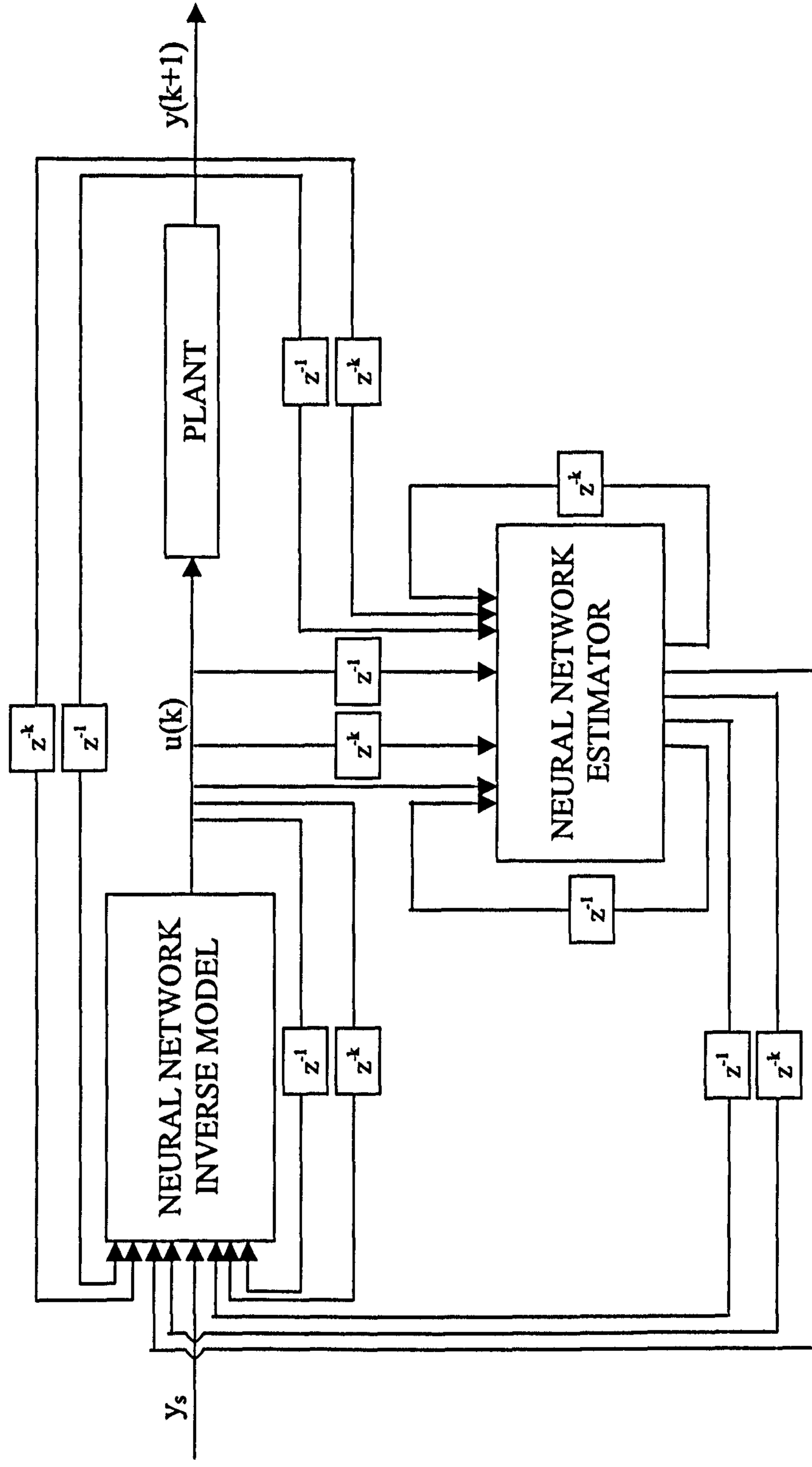


Figure 5.9: Direct Inverse control strategy

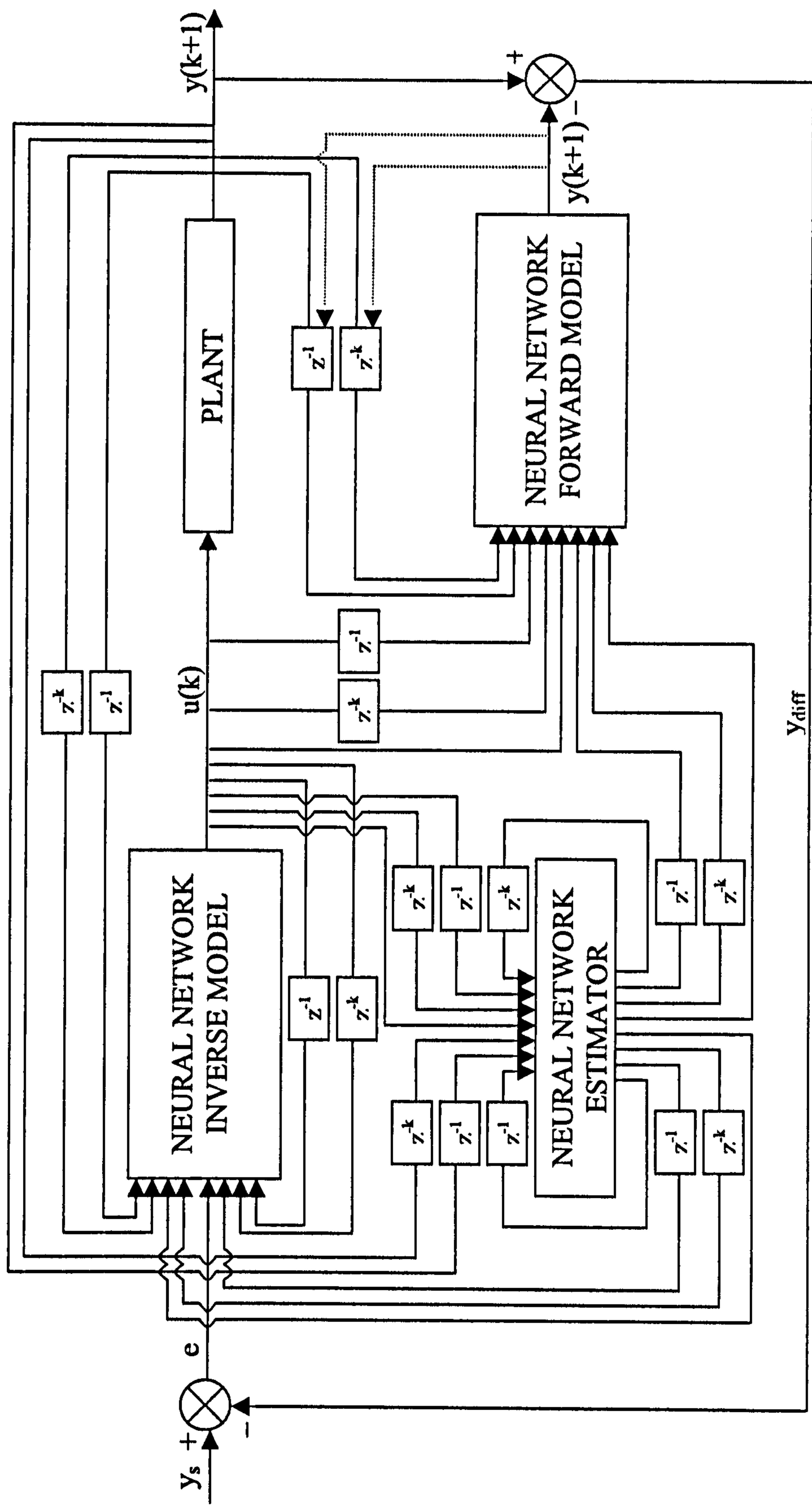


Figure 5.10: Inverse-model-based control (IMBC) strategy

5.4 Dual-Mode (DM) Control Strategy

Dual-mode (DM) control is the most commonly used strategy in batch reactors that have initial heat-up (i.e. for exothermic reaction). The object of the DM control strategy implementation here is to provide a standard comparison for its performance with advanced control strategies mentioned earlier (GMC-NN, IMBC and Direct Inverse). DM control strategy is combination of on-off and conventional control type strategy. First, maximum heating (on) is applied until the reactor temperature is within a specified degree of the set point and then maximum cooling (off) is applied when the temperature has reached its final desired set point. At this point, standard feedback controllers are switched on and used to maintain the temperature (constant or dynamic set points). In the standard DM strategy, the PID controller is normally used. However in this study both PID and PI controllers are used. Details of these controllers can be found in Seborg et al. (1989). In both controllers, jacket temperature, T_j is the manipulated variable at time k and $T_{j\text{sp}}(k)$ is calculated using Equation 5.2.5 which is same as in the GMC-NN.

The DM control strategy consists of a sequence of control actions, each one carried out after the reactor has reached a certain condition. The sequence of operations is as follows:

1. Full heating is applied until the reactor temperature is within a certain percent (E_m) of its set point temperature.
2. Full cooling is then applied for a certain period of time (TD-1).
3. The jacket set point temperature ($T_{j\text{sp}}$) of controller is then set to the pre-load temperature (PL) for a certain period of time (TD-2).

4. A temperature controller (PID or PI) is cascaded to the jacket temperature controller and its set point is set to T_{sp} .

There are two steps applied in order to tune the DM control strategy. First, PID or PI tuning parameters were tuned by performing an open-loop step response test. The Cohen and Coon method was then applied to estimate the value of the tuning parameters (K_c , τ_I and τ_D). However the tuning parameters have been fine-tuned to make the control less drastic (through a few simulation runs with small changes on the estimated tuning parameters value). Second, the remaining four constants (E_m , TD-1, TD-2 and PL) were determined by running a series of simulation runs. The details of DM control strategy and its tuning can be found in Liptak (1986).

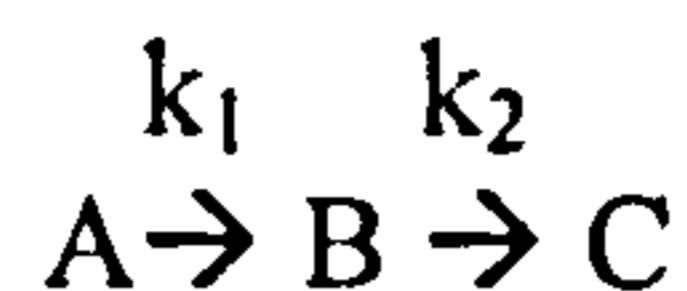
5.5 Applications (On-line tracking of optimal profiles)

Here, we consider two case studies. The first case study deals with consecutive exothermic batch reactions and the second case study uses the reaction scheme considered by Cott and Macchietto (1989). In both cases an off-line dynamic optimisation problem is solved with fixed batch time to find the optimum temperature profile that will maximise the conversion of the desired product (Chapter 4). In Case Study 1, the optimisation problem is solved with only one time interval but in Case Study 2 both one time interval and three time intervals are used. In Case Study 1, only GMC-NN and PID controllers were implemented. On the other hand, in Case Study 2 all types of controllers, which have been discussed in the earlier sections, are implemented. In both cases, various tests were carried out to evaluate the robustness of the controller applied.

5.5.1 Case Study 1

In this example, a consecutive exothermic batch reaction scheme is considered.

The reaction type is:



where A is a raw material, B is the desired product and C is a waste or by-product.

The reaction scheme considered is same as in Case Study 3 of Chapter 4.

However the heat retention in the wall is not considered (Option 3 of GBRM).

5.5.1.1 Models Equations

Option 3 model of Chapter 3 with $m = 3$ and $n = 2$ is considered.

The values of v_{11} and v_{22} are equal to -1 ; the values of v_{12} and v_{23} are equal to 1 and the rest of v_{ji} are equal to zero. The values of β_{11} and β_{22} are equal to 1 . The rest of β_{ij} are equal to zero. However, here the expression of $\frac{F_J}{V_J}$ and T_{J0} in Equation 3.42 in Chapter

3 is replaced by $\frac{1}{\tau_J}$ and T_{Jsp} respectively. The equations 3.36 and 3.42 in Chapter 3 will

then result to:

$$\frac{dC_A}{dt} = -k_1 C_A \quad (5.5.1)$$

$$\frac{dC_B}{dt} = k_1 C_A - k_2 C_B \quad (5.5.2)$$

$$\frac{dC_C}{dt} = k_2 C_B \quad (5.5.3)$$

$$\frac{dT}{dt} = \frac{(Q_r - Q_J)}{\rho c_p V} \quad (5.5.4)$$

$$\frac{dT_J}{dt} = \frac{(T_{Jsp} - T_J)}{\tau_J} + \frac{Q_J}{V_J \rho_J c_{pJ}} \quad (5.5.5)$$

$$k_1 = k_{10} \exp\left(-\frac{E_1}{RT}\right) \quad (5.5.6)$$

$$k_2 = k_{20} \exp\left(-\frac{E_2}{RT}\right) \quad (5.5.7)$$

$$Q_r = -\Delta H_1 (k_1 C_A) V - \Delta H_2 (k_2 C_B) V \quad (5.5.8)$$

$$Q_J = UA(T - T_J) \quad (5.5.9)$$

All constant parameter values are as given in Table 5.1.

Table 5.1: The constant parameter values of the model and control equation

$\Delta H_1 = -6.50E8 \text{ J/kmol}$	$k_{10} = 4.38E4 \text{ h}^{-1}$	$c_p = 4200.0 \text{ J/kgK}$
$\Delta H_2 = -1.20E8 \text{ J/kmol}$	$k_{20} = 3.94E5 \text{ h}^{-1}$	$c_{pJ} = 4200.0 \text{ J/kgK}$
$E_1 = 3.49E7 \text{ J/kmol}$	$\rho = 800.0 \text{ kg/m}^3$	$\tau_J = 0.075 \text{ h}$
$E_2 = 4.65E7 \text{ J/kmol}$	$R = 8314.0 \text{ J/kmol.K}$	$\rho_J = 1000.0 \text{ kg/m}^3$
$A = 5.25 \text{ m}^2$	$U = 8.18E6 \text{ J/h.K.m}^2$	$\Delta t = 0.01 \text{ h}$
$V = 1.23 \text{ m}^3$	$V_J = 0.53 \text{ m}^3$	

The objectives of this study are:

1. To obtain optimum reactor temperature profile to maximise the conversion to the desired product B. This does not require the full model (only requires Equations 5.5.1 – 5.5.3, 5.5.6 – 5.5.7) to be used.
2. To track the optimum temperature profile obtained in (1) using GMC-NN and PID controllers. This requires the full model to be used (5.5.1 – 5.5.9).
3. To compare the performance of the GMC-NN controller with PID controller.
4. To test the robustness of both controllers.

It is assumed that the reactants are being pre-heated before they are charged into the reactor. The initial values of $[C_A, C_B, C_C, T, T_J]$ are $[0.975, 0.025, 0.0, 350K, 300K]$ respectively. The total batch time is 3.5 hours. The reactor temperature, T (controlled variable) and jacket temperature, T_J (manipulated variable) are bounded between 300 and 400K. The results are summarised in Table 5.2. It is found that the maximum conversion achieved is 0.6613 (off-line conversion achieved by solving the dynamic optimisation problem P1). The optimum temperature is 369.40, which has been used as the T_{sp} to be tracked by GMC-NN (Equation 5.2.4) and PID controllers in the on-line control implementation.

Table 5.2: Summary of the result for Case Study 1

Off-line Optimum Temperature Profile		C_B^*
Temperature, K	369.40	0.6613
Switching time, h	t = 0 3.5	
Controller	C_B	CP
PID	0.6602	99.83
GMC-NN	0.6602	99.83
Tuning Parameters		
GMC-NN	DM with PID	
$K_1 = 22.22 \text{ h}^{-1}$	$E_m = 0.5\%$	$K_c = 19.68 \text{ h}$
$K_2 = 1.235 \text{ h}^{-2}$	PL = 345 K	$\tau_I = 0.07 \text{ h}$
	TD-1 = 0.02 h	$\tau_D = 0.0007 \text{ h}$
	TD-2 = 0.01 h	
C_B On-line conversion to B		
C_B^* Off-line conversion to B		
CP Controller Performance (%) $\{(C_B/C_B^*) \times 100\}$		

Tuning procedure for GMC tuning parameters can be found in Lee and Sullivan (1988) and Cott and Macchietto (1989).

5.5.1.2 Results

It can be clearly seen (Table 5.2) that the conversion of 0.6602 achieved by the GMC-NN is very close to that achieved by off-line dynamic optimisation (0.6613). The GMC-NN was also able to track the given set point very well. The response of the GMC-NN controller is shown in Figure 5.11.

The performance of the GMC-NN controller is strongly dependent on the estimation of the heat released by the reaction, Q_r . Figure 5.12 shows that the neural network was able to give a very good estimation of the heat released by the reaction and hence guarantee the good performance of the GMC-NN controller.

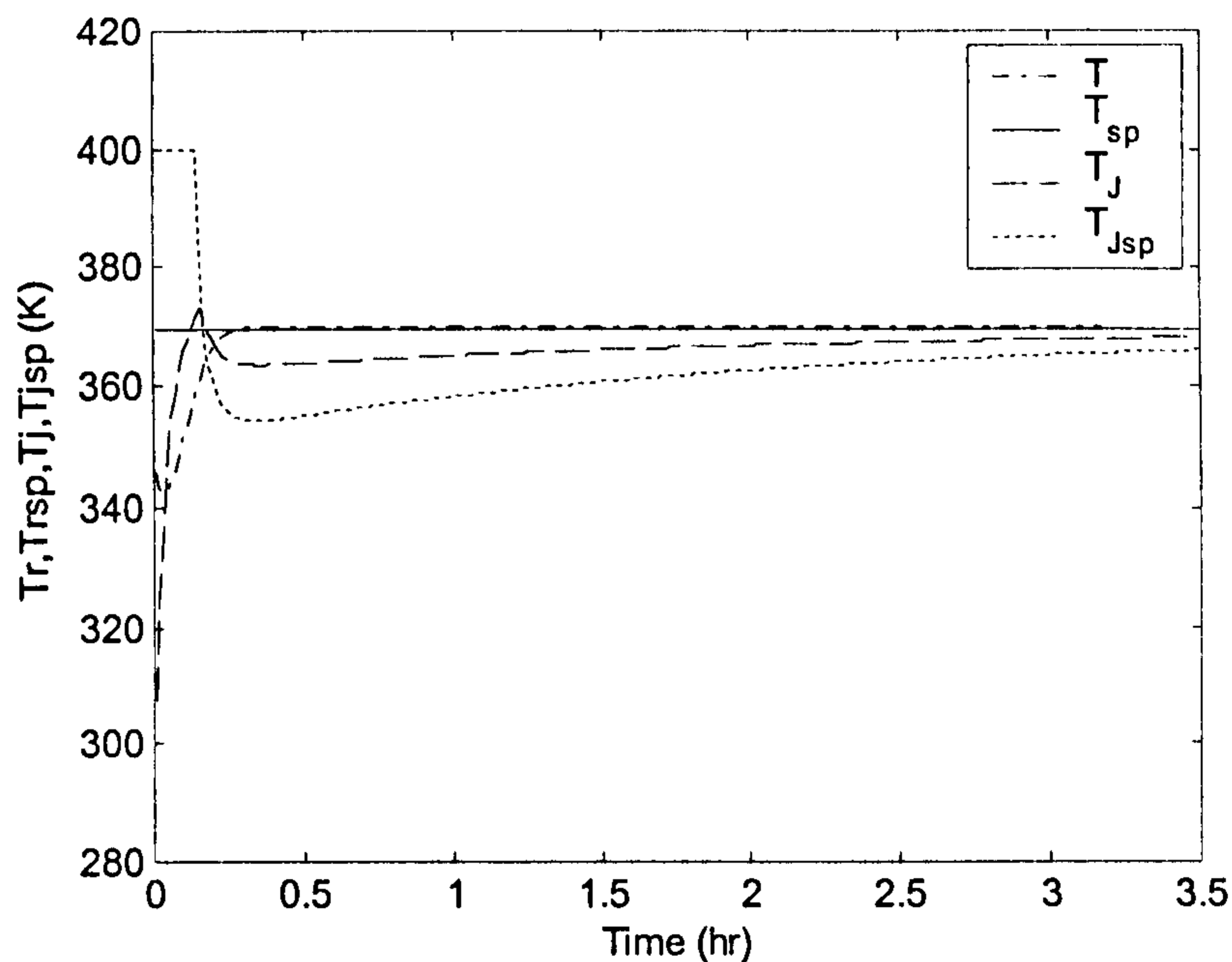


Figure 5.11: GMC-NN response (Case Study 1)

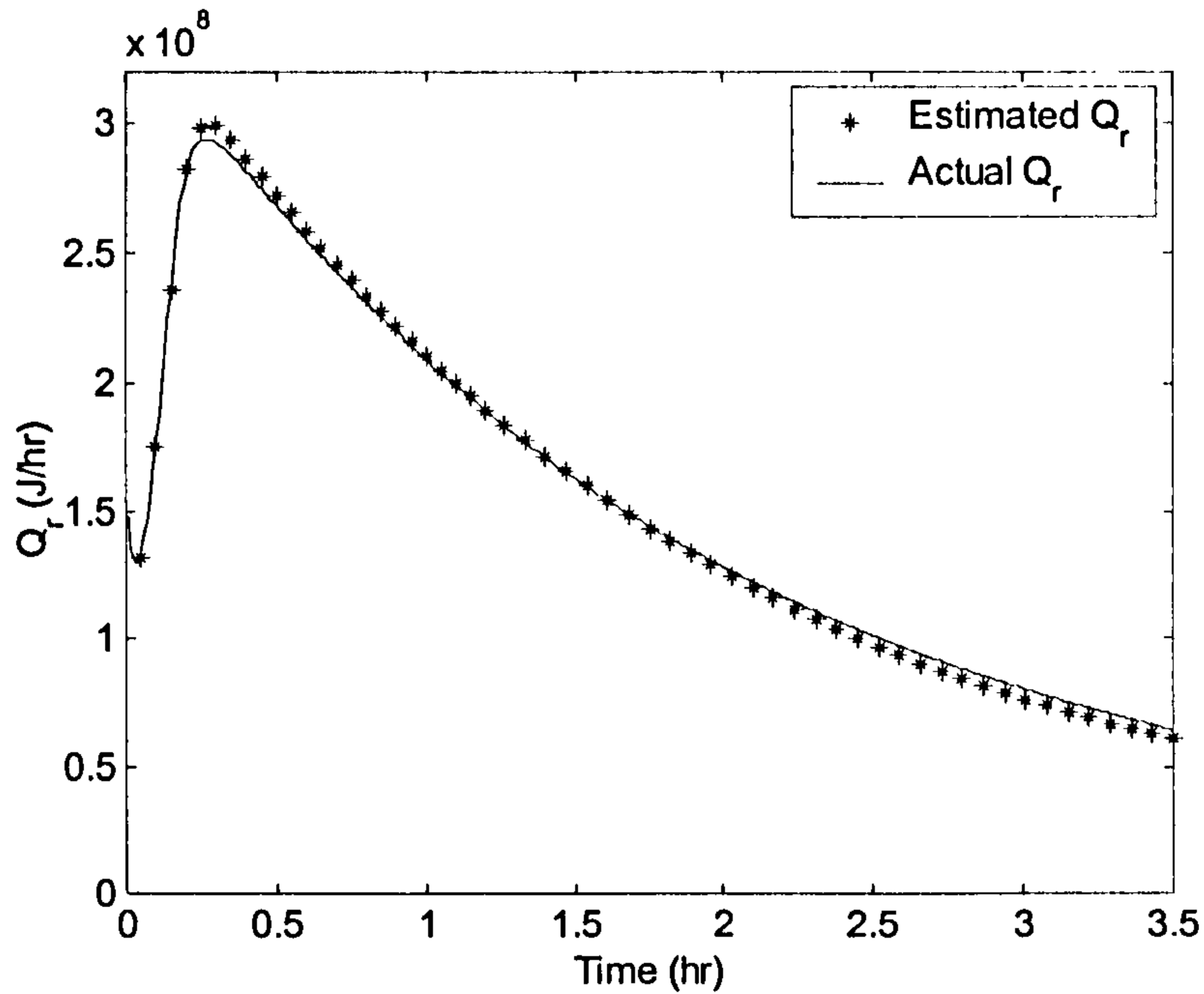


Figure 5.12: Performance of heat-release estimator (Case Study 1)

It was found that the PID controller was also able to track the given set point very well but the response was more sluggish and with oscillations compared to the GMC-NN strategy (see Figure 5.13). The controller performance (CP) of the PID controller was the same as that of the GMC-NN controller (99.83%).

In order to study further the robustness of the controller, four further tests have been carried out by changing some of the process parameters. In this case, the GMC-NN and PID controllers (tuned as before) were used to control an operation where some of the conditions have been changed from their nominal values.

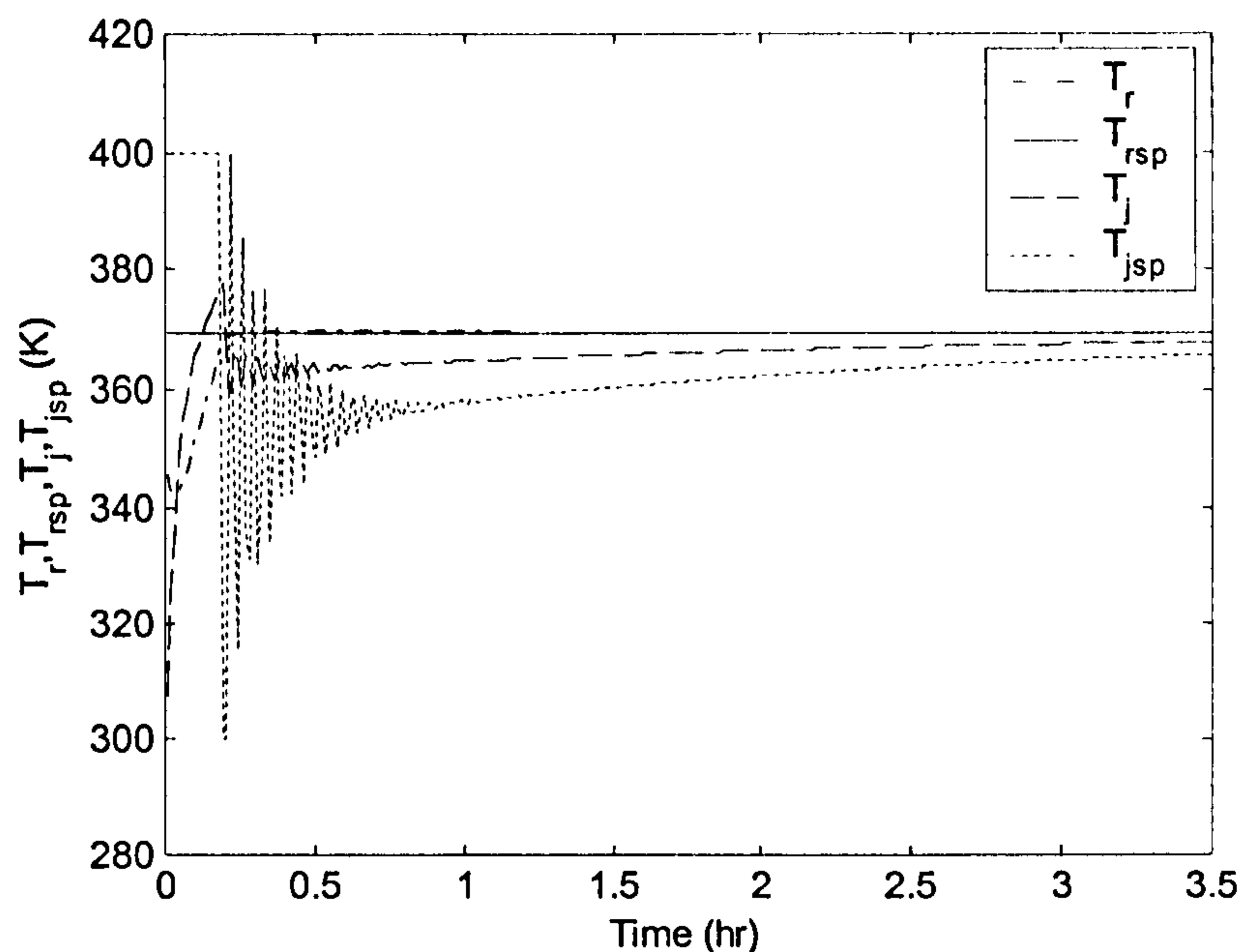


Figure 5.13: PID response (Case Study1)

In the first test (TEST1), the heat of reactions was increased by 25%. This is equivalent to the presence of un-modelled reactions. The second test (TEST2) involved the increment of the rate constant by 25% from the true value. The third test (TEST3) involved 30% reduction in the weight of the initial quantities of reactants. It represents a change in operating conditions that could be caused by a deliberate change in product demand or an accidental failure of the charging system. Finally, the fourth test (TEST4) involved the reduction of heat transfer coefficient by 40% from its original value, which might happen due to fouling of the heat-transfer surfaces.

The results for all the tests are shown in Figures (5.14-5.21). They show that both the GMC-NN and PID controllers were able to accommodate these changes. The neural network also gave a very good estimation of heat released on-line in every test.

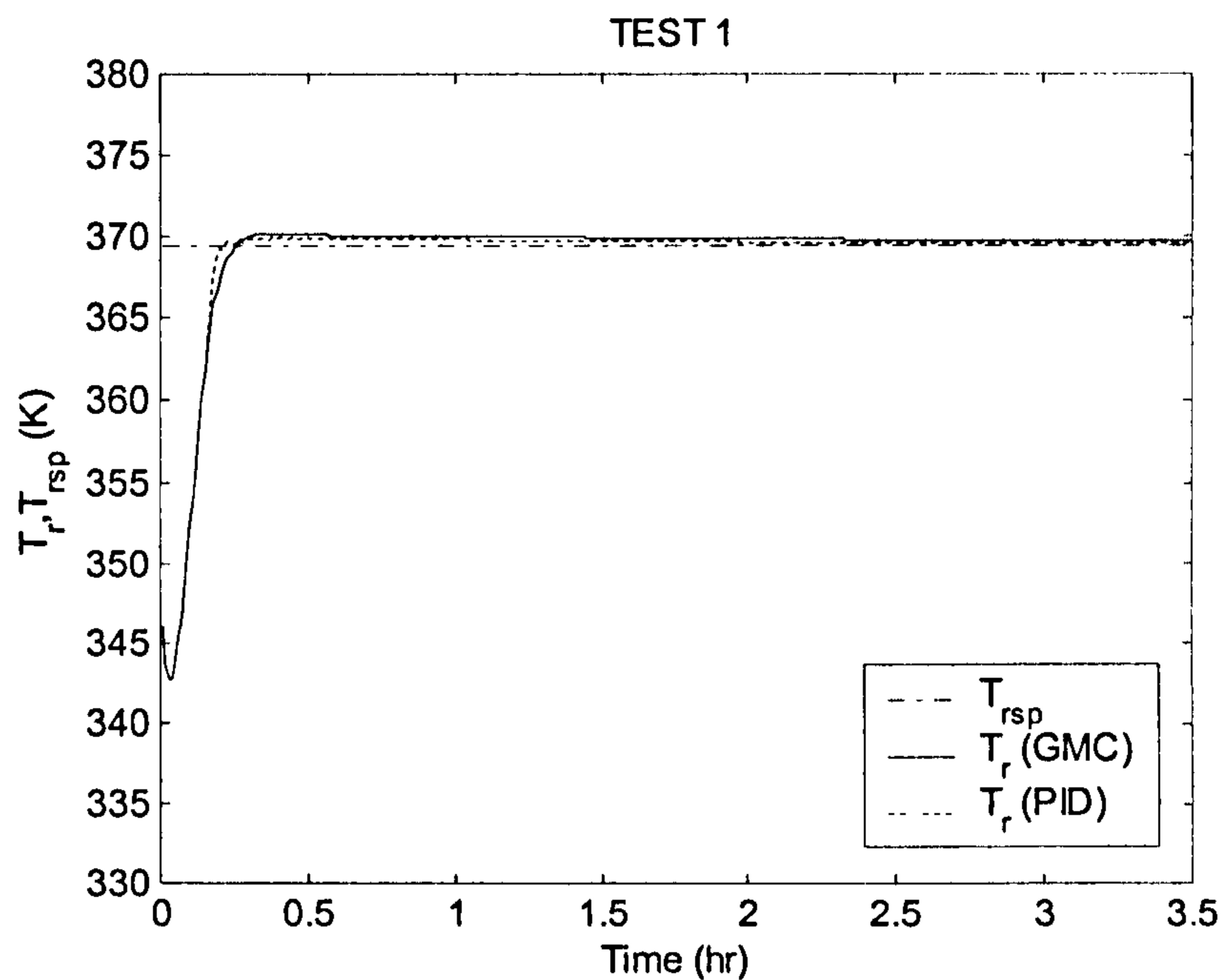


Figure 5.14: Controllers responses for heat reaction change (Case Study 1)

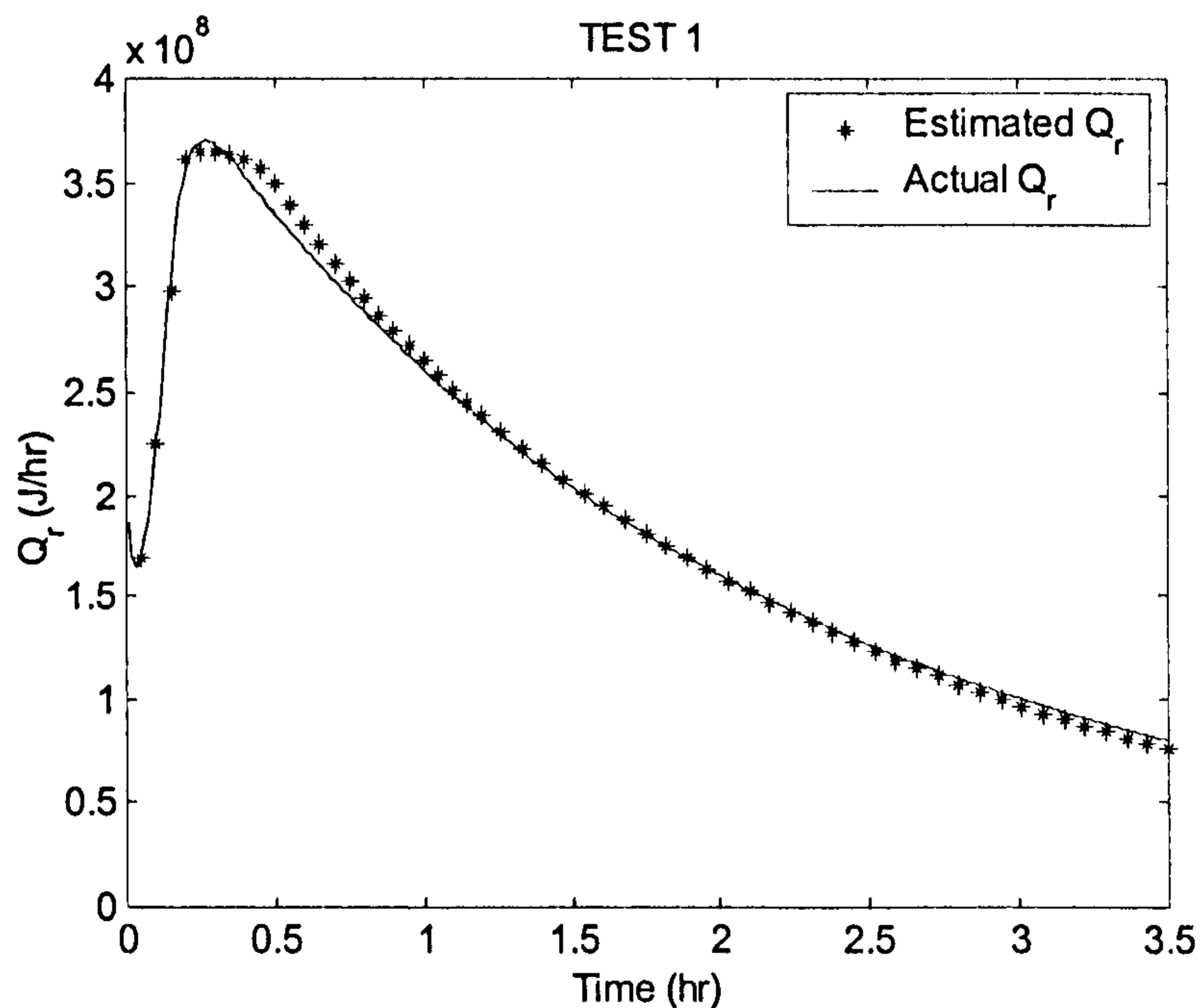


Figure 5.15: Heat release estimator performance for heat reaction change (Case Study 1)

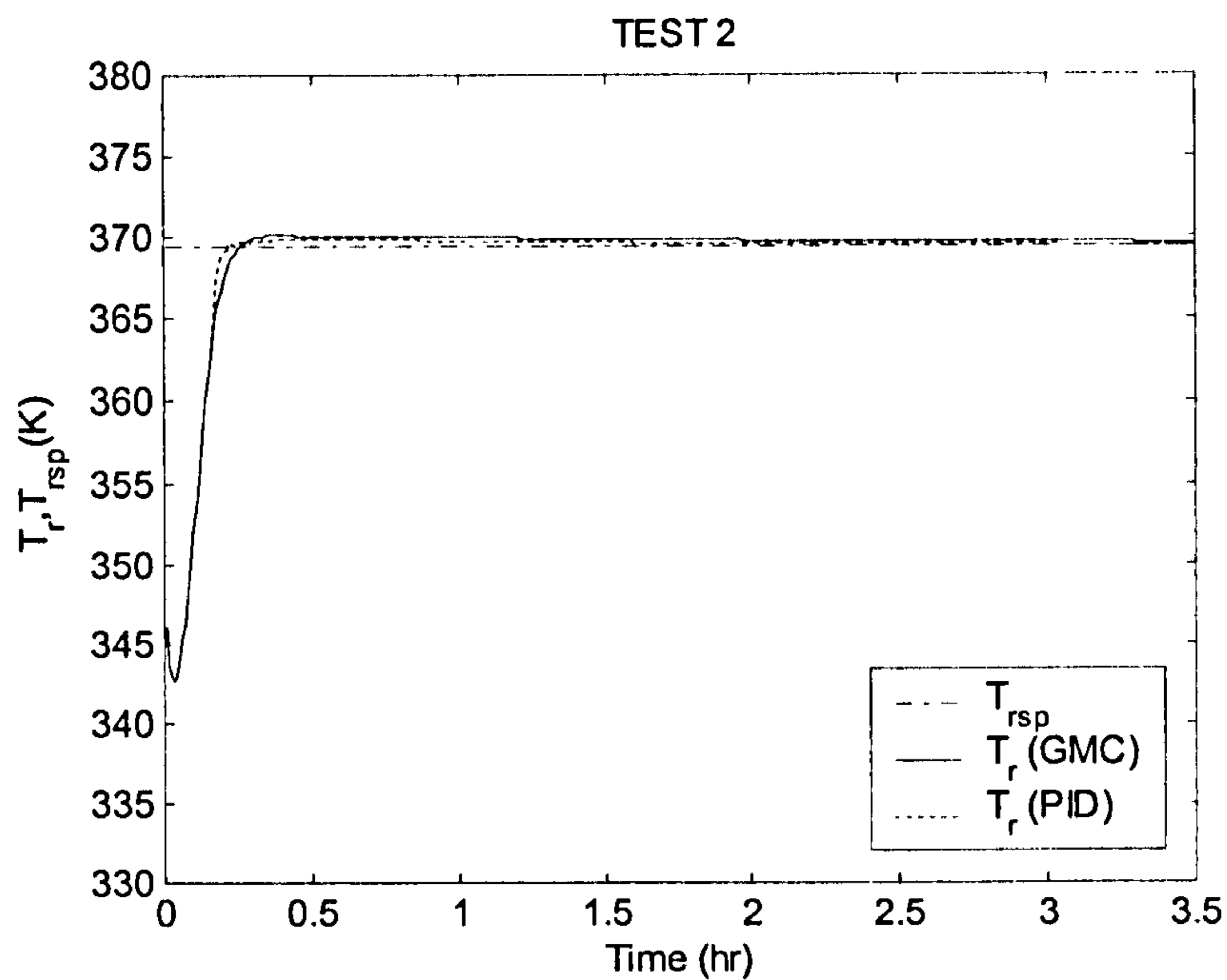


Figure 5.16: Controllers responses for rate constant change (Case Study 1)

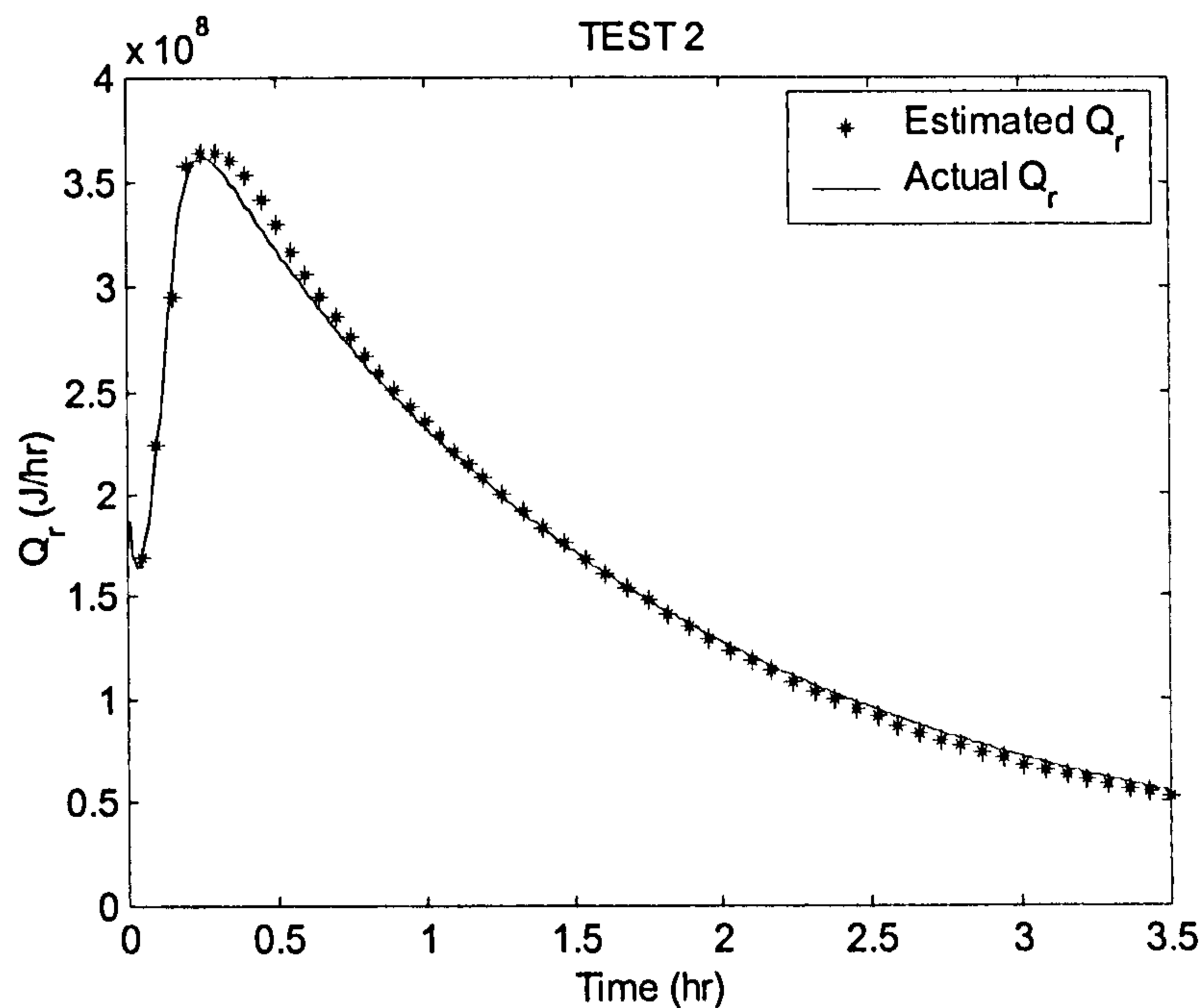


Figure 5.17: Heat release estimator performance for rate constant change (Case Study 1)

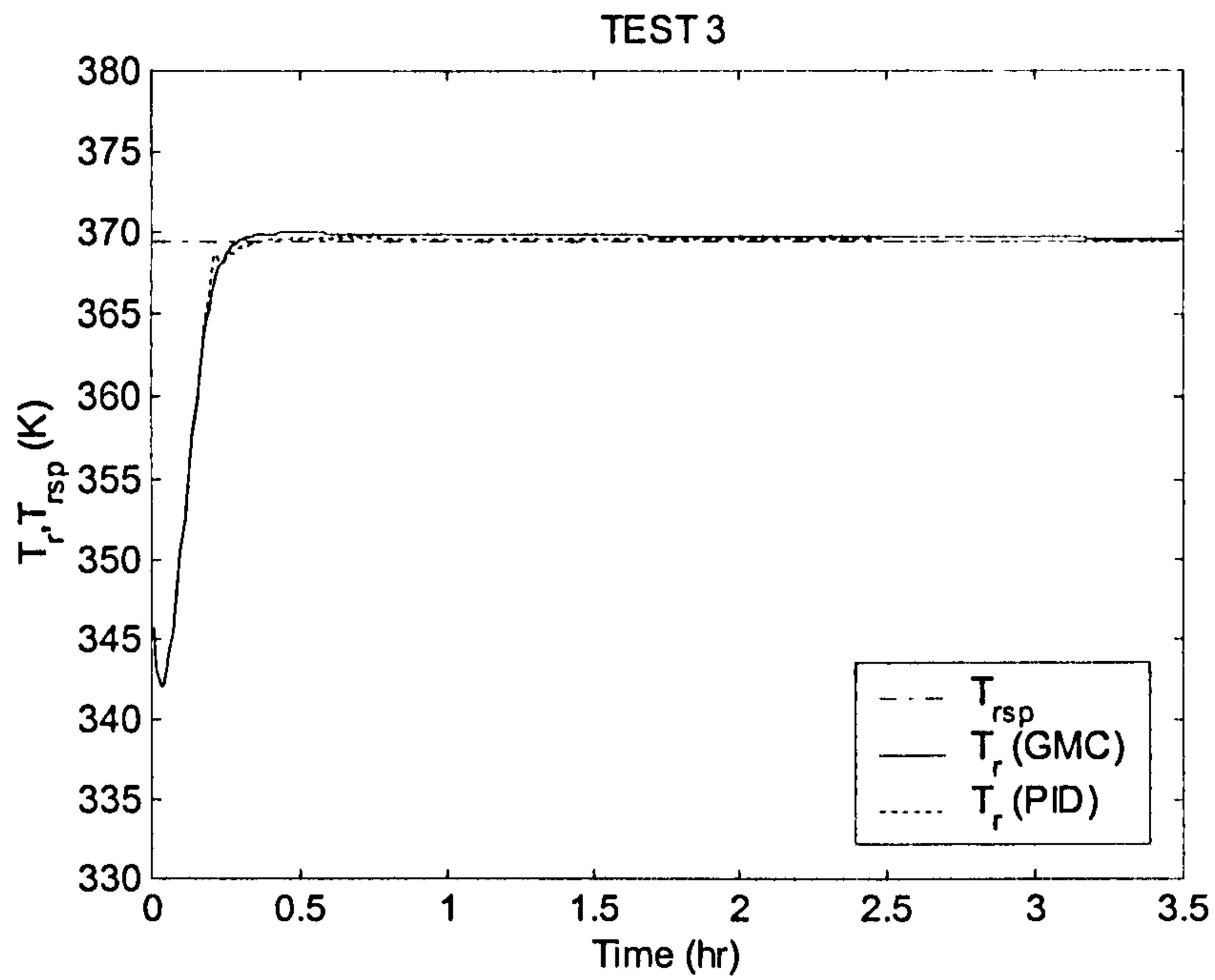


Figure 5.18: Controllers responses for weight change (Case Study 1)

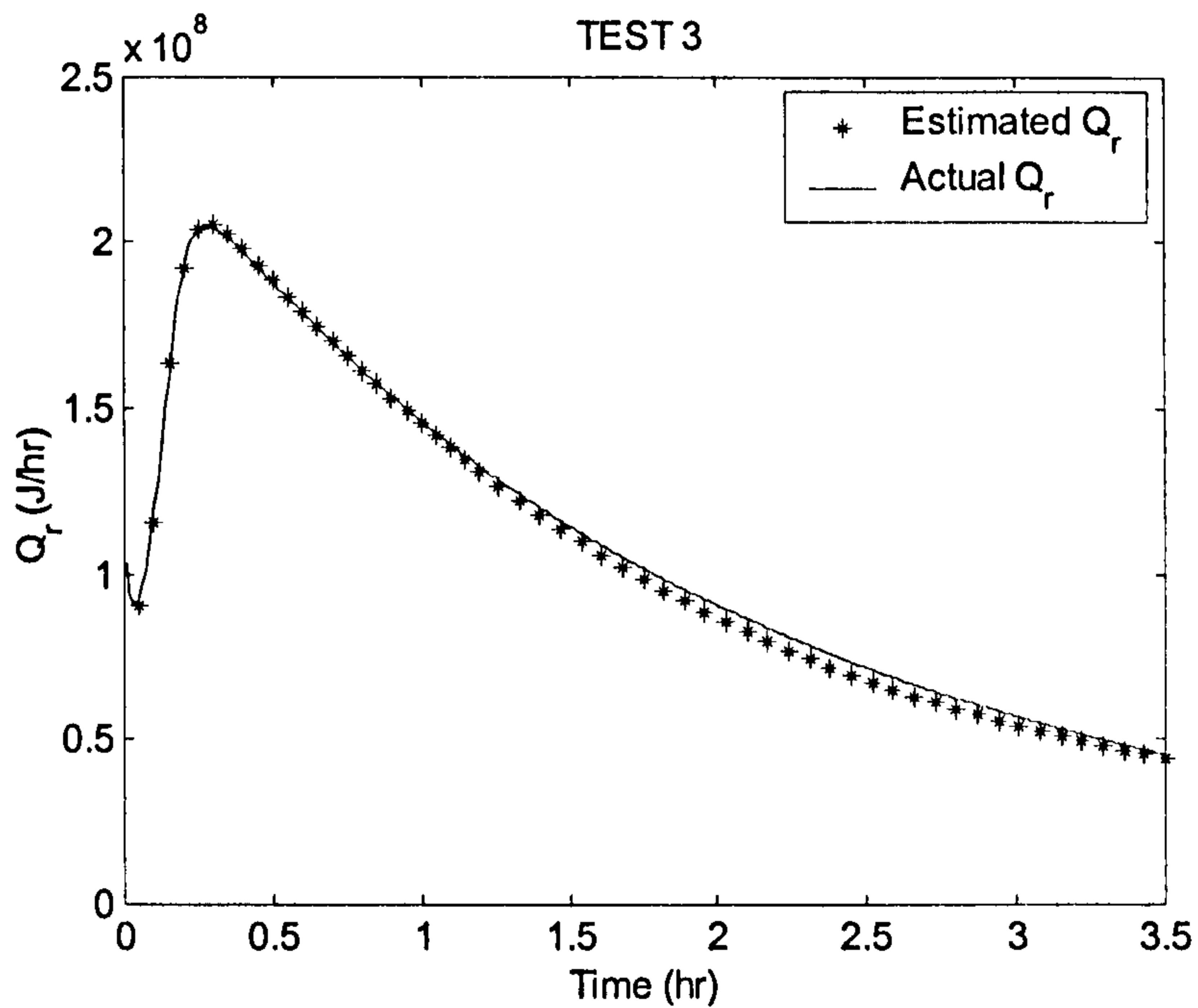


Figure 5.19: Heat release estimator performance for weight change (Case Study 1)

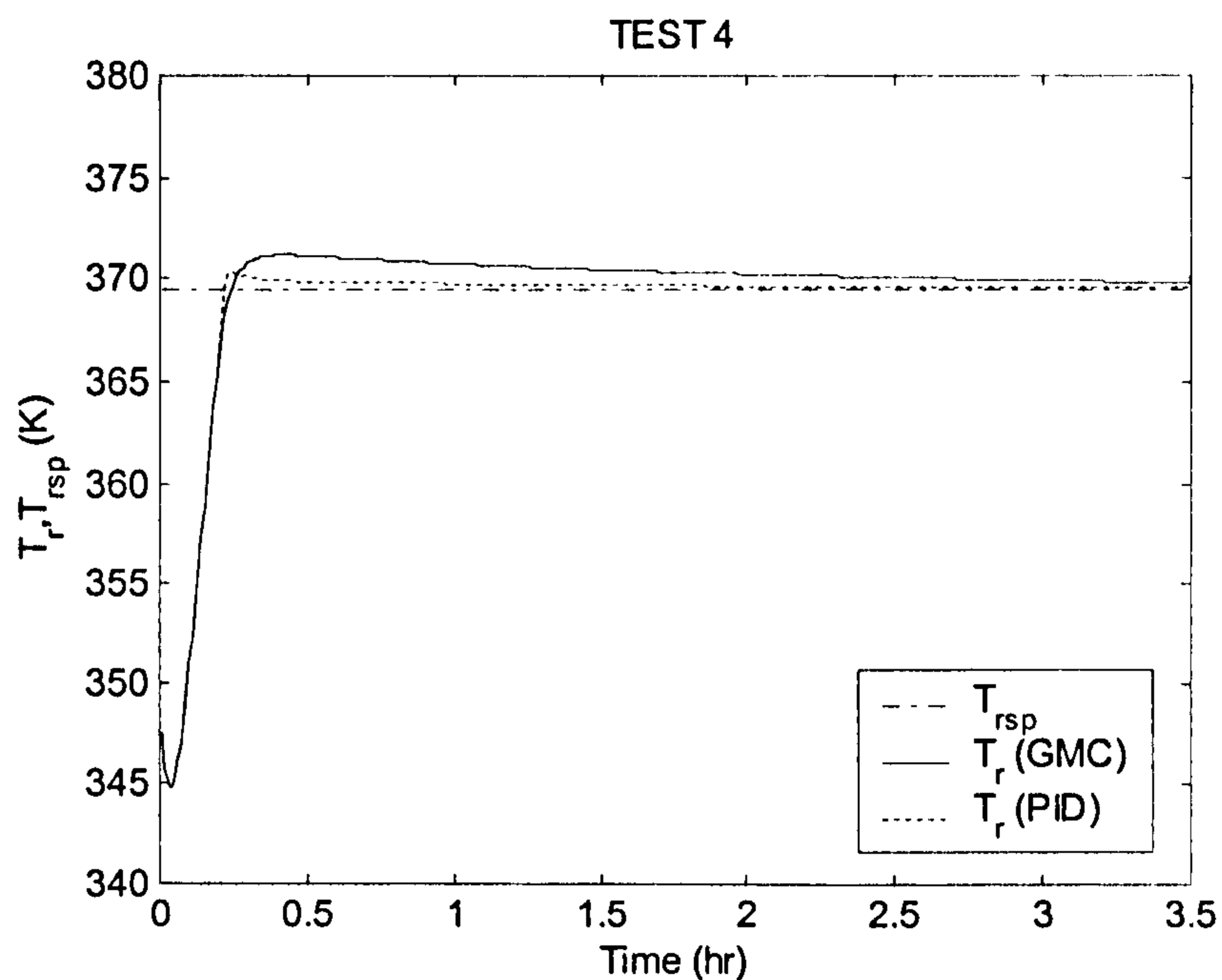


Figure 5.20: Controllers responses for heat transfer coefficient change (Case Study 1)

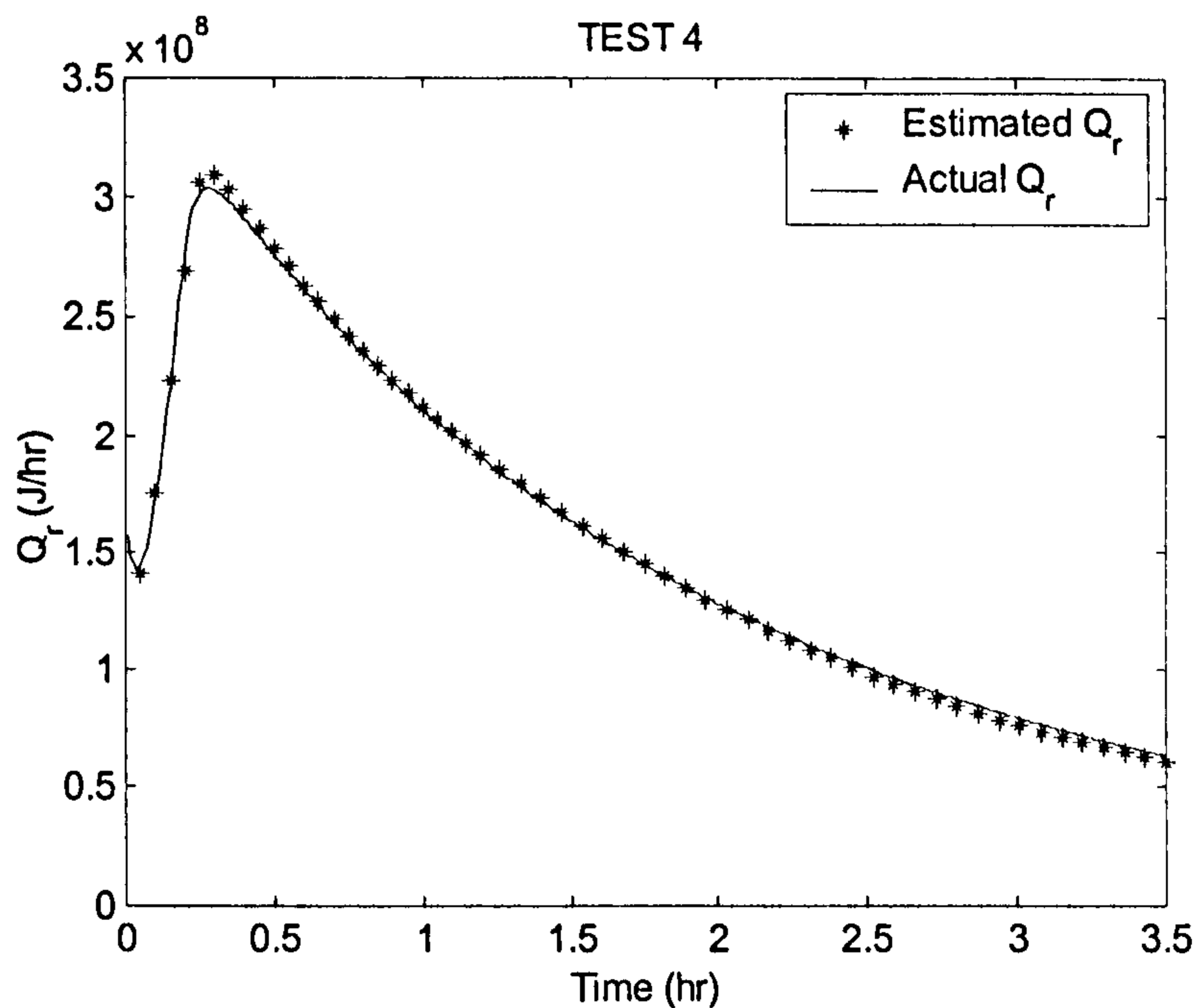


Figure 5.21: Heat release estimator performance for heat transfer coefficient change (Case Study 1)

5.5.2 Case Study 2

Here the reaction scheme is same as that used by Cott and Macchietto (1989).

The reaction scheme is:



where A, B are the raw materials, C is the desired product and D is the waste product.

5.5.2.1 Model Equations

The model equations for the batch reactor can be written as:

$$\frac{dM_A}{dt} = -R_1 - R_2 \quad (5.5.10)$$

$$\frac{dM_B}{dt} = -R_1 \quad (5.5.11)$$

$$\frac{dM_C}{dt} = +R_1 - R_2 \quad (5.5.12)$$

$$\frac{dM_D}{dt} = +R_2 \quad (5.5.13)$$

$$\frac{dT}{dt} = \frac{(Q_r + Q_J)}{M_r c_{pr}} \quad (5.5.14)$$

$$\frac{dT_J}{dt} = \frac{(T_{Jsp} - T_J)}{\tau_J} - \frac{Q_J}{V_J \rho_J c_{pJ}} \quad (5.5.15)$$

$$R_1 = k_1 M_A M_B \quad (5.5.16)$$

$$R_2 = k_2 M_A M_C \quad (5.5.17)$$

$$k_1 = \exp\left(k_1^1 - \frac{k_1^2}{(T + 273.15)}\right) \quad (5.5.18)$$

$$k_2 = \exp\left(k_2^1 - \frac{k_2^2}{(T + 273.15)}\right) \quad (5.5.19)$$

$$Q_r = -\Delta H_1 R_1 - \Delta H_2 R_2 \quad (5.5.20)$$

$$M_r = M_A + M_B + M_C + M_D \quad (5.5.21)$$

$$c_{pr} = \frac{c_{pA} M_A + c_{pB} M_B + c_{pC} M_C + c_{pD} M_D}{M_r} \quad (5.5.22)$$

$$Q_J = UA(T_J - T) \quad (5.5.23)$$

All the parameter and constant values used in the model and control equation are given in Table 5.3.

Table 5.3: The constant parameter values of the model and control equation

$c_{pB} = 167.3 \text{ kJ/kmol}^\circ\text{C}$	$k_1^1 = 20.9057$
$c_{pC} = 217.6 \text{ kJ/kmol}^\circ\text{C}$	$k_1^2 = 10000$
$c_{pD} = 334.7 \text{ kJ/kmol}^\circ\text{C}$	$k_2^1 = 38.9057$
$\Delta H_1 = -41840.0 \text{ kJ/kmol}$	$k_2^2 = 17000$
$\Delta H_2 = -25104.0 \text{ kJ/kmol}$	$V_J = 0.6921 \text{ m}^3$
$c_p = 1.8828 \text{ kJ/kg}^\circ\text{C}$	$A = 6.24 \text{ m}^2$
$c_{pJ} = 1.8828 \text{ kJ/kg}^\circ\text{C}$	$\Delta t = 0.2 \text{ min}$
$U = 40.84 \text{ kJ/min.m}^2.\circ\text{C}$	$\tau_J = 3.0 \text{ min}$
$\rho_J = 1000.0 \text{ kg/m}^3$	$W_r = 1560.0 \text{ kg}$

Here again an off-line dynamic optimisation problem (P1) is solved to find the optimum temperature profile that will maximise the product “C” and minimise the by-product “D” (Case Study 6 – Chapter 4). Two runs were carried out; RUN1 uses one control interval (time) and RUN2 uses three fixed control intervals. The batch time is 120 minutes and the initial values of $[M_A, M_B, M_C, M_D, T, T_J]$ are $[12.0, 12.0, 0.0, 0.0, 20.0, 20.0]$ respectively. The reactor temperature is used as the controlled variable and is bounded between 20 and 100°C. The manipulated variable, T_J is bounded between 20

and 120°C. The model in the dynamic optimisation problem does not require Equation (5.5.14 – 5.5.15) and (5.5.20 – 5.5.23) to be used (Case Study 6 in Chapter 4).

The results (optimal temperature profiles) for both runs are then used as the set point to be tracked by all types of controllers that have been discussed earlier (GMC-NN controller (Equation 5.5.24), neural controller in Direct Inverse and IMBC strategies and DM with PID and PI controllers).

$$T_j(k) = T(k) + \frac{W_r c_p}{UA} \left[K_1 (T_{sp} - T(k)) + K_2 \sum_0^k (T_{sp} - T(k)) \Delta t \right] - \frac{Q_r}{UA} \quad (5.5.24)$$

The results are summarised in Table 5.4. The first part (off-line results) of the Table 5.4 is same as those in Table 4.11 (Chapter 4)

5.5.2.2 Results

GMC coupled with NN estimator response

Figures (5.22 and 5.23) show the response of the GMC-NN controller in tracking the set points (T_{sp}) and the performance of the neural network in estimating heat-released for RUN1 and Figures (5.24 and 5.25) show those for RUN2. It can be seen that the GMC-NN was able to accommodate very well both constant and dynamic set points although a little sluggishly for the latter. Again Figures 5.23 and 5.25 show that the neural network gave a very good estimation for the heat released by the reaction.

Table 5.4 shows that for both runs the amount of desired product obtained on-line (after implementing the GMC-NN controller) was within 4% of that obtained by off-line dynamic optimisation. This clearly shows the effectiveness of implementing the GMC-NN controller combined with the neural network estimator.

Table 5.4: Summary of the results for Case Study 2

Run	Off-line Optimum Temperature Profile		M_C^*
1	Temperature, °C	-----92.46-----	6.5126
	Switching time, min $t = 0$	120.0	
2	Temperature, °C	-----92.83----- -----91.17----- -----93.41-----	6.5171
	Switching time, min $t = 0$	40.0 80.0 120.0	
Run	Controller	M_C	CP
1	PI	6.3064	96.83
	PID	6.3392	97.34
	GMC-NN	6.3270	97.15
	Direct Inverse	6.3389	97.33
	IMC	6.3389	97.33
2	PI	6.3053	96.75
	PID	6.3409	97.30
	GMC-NN	6.3309	97.14
	Direct Inverse	6.3397	97.28
	IMC	6.3398	97.28
Tuning Parameters			
GMC-NN		DM with PID and PI	
$K_1 = 0.20 \text{ min}^{-1}$		$E_m = 5.0\%$	
$K_2 = 1.00E-4 \text{ min}^{-2}$		PL = 46 °C	
		TD-1 = 2.8 min	PID:
		TD-2 = 2.4 min	$K_c = 26.5381 \text{ min}$
			$\tau_I = 2.8658 \text{ min}$
			$\tau_D = 0.4284 \text{ min}$
			PI:
			$K_c = 10.7574 \text{ min}$
			$\tau_I = 53.4882 \text{ min}$
M_C On-line Product			
M_C^* Off-line Product			
CP Controller Performance (%) $\{(M_C/M_C^*) \times 100\}$			

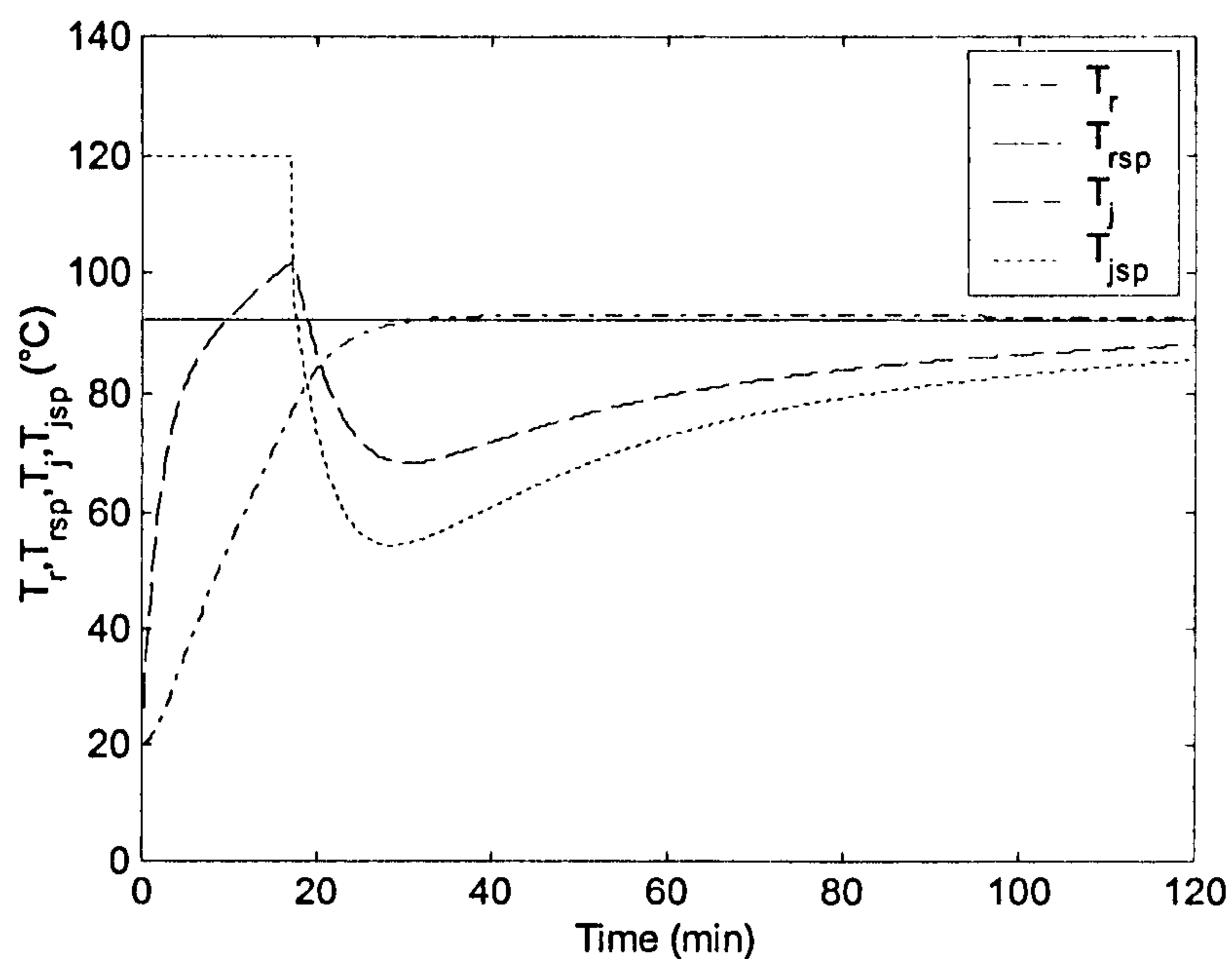
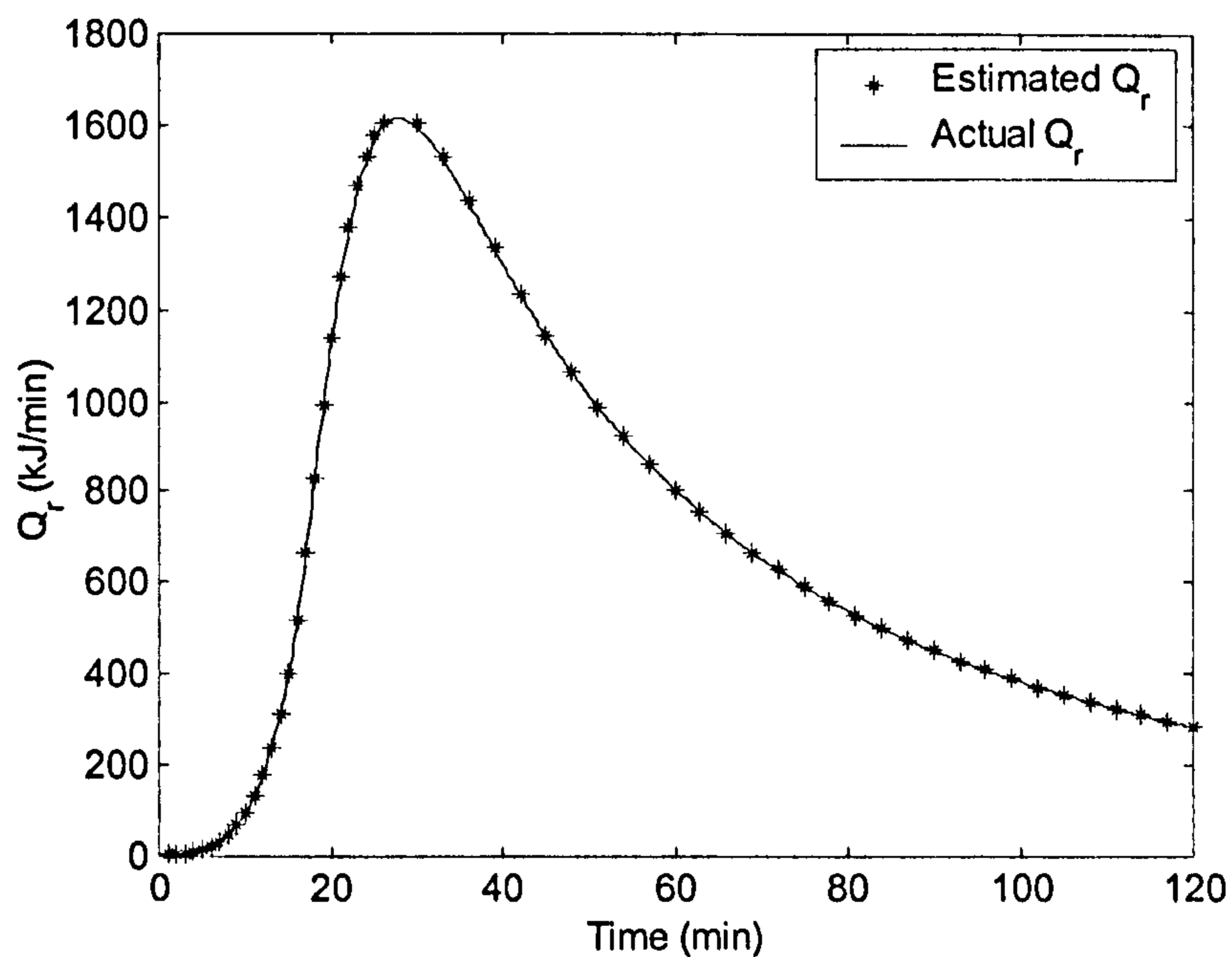
Figure 5.22: GMC-NN response for 1 T_{sp} (Case Study 2 - RUN1)

Figure 5.23: Performance of heat-release estimator (Case Study 2 - RUN1)

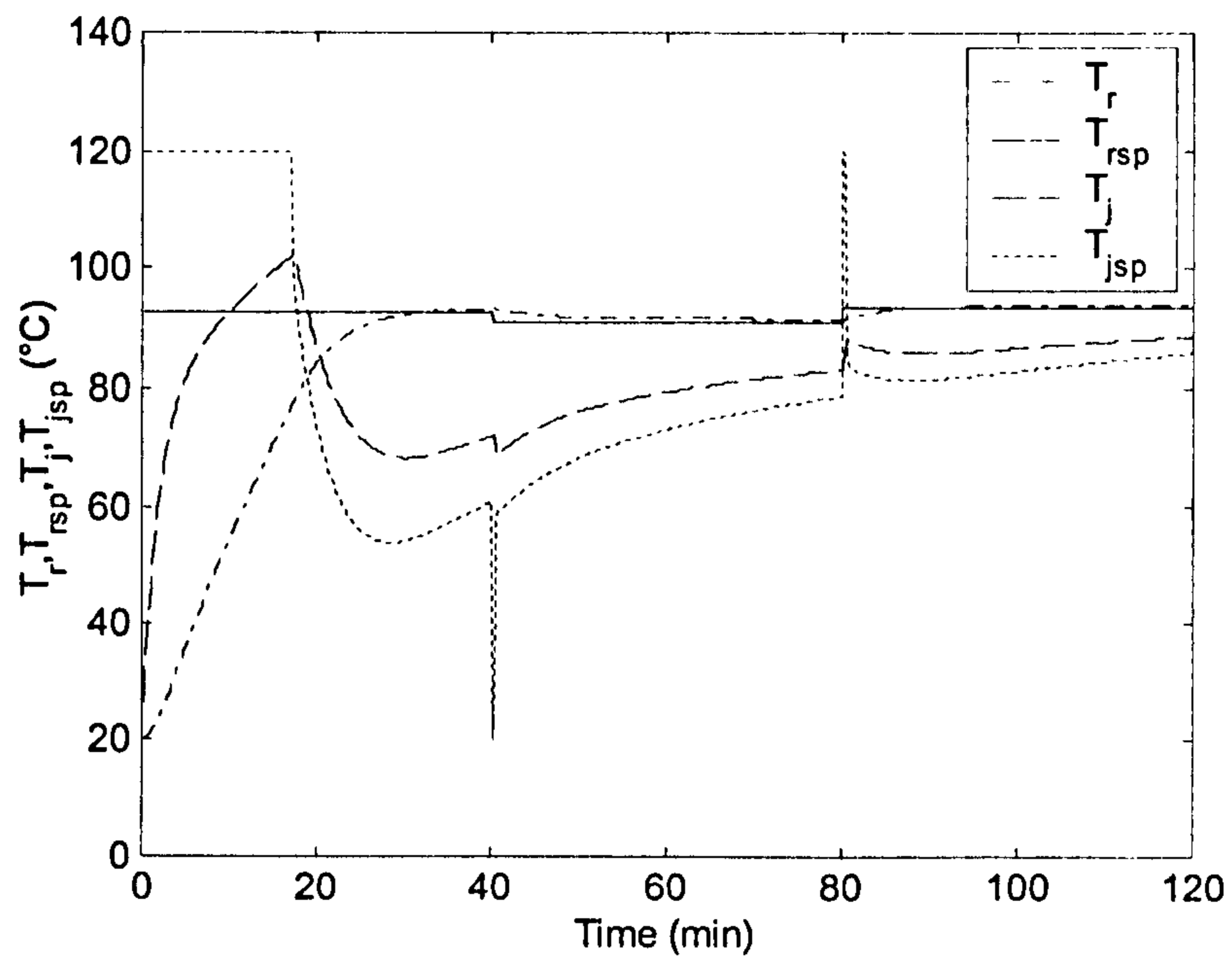


Figure 5.24: GMC-NN response for dynamic set points (Case Study 2 - RUN2)

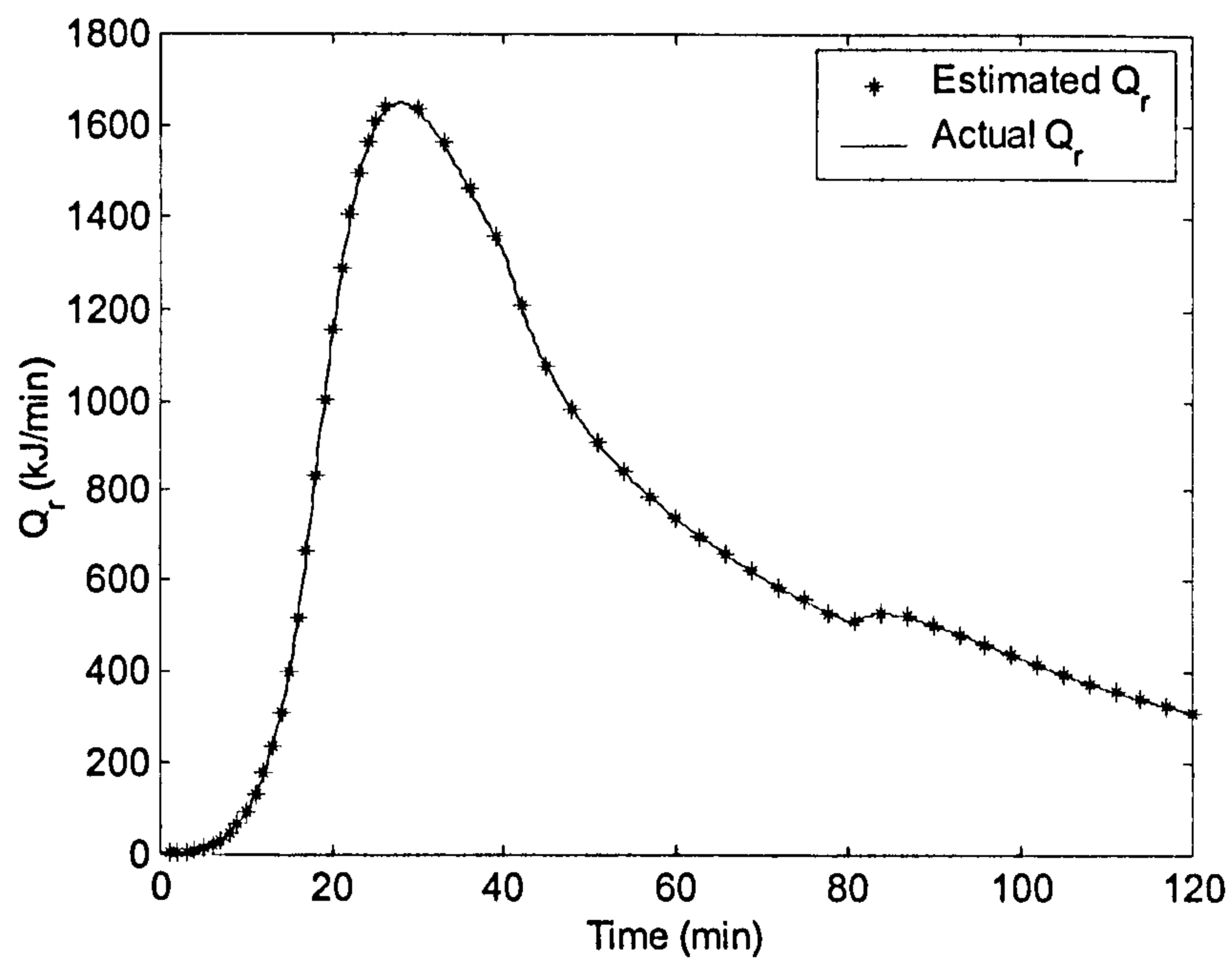


Figure 5.25: Performance of heat-release estimator (Case Study 2 - RUN2)

Comparison between GMC-NN and DM with PID and PI controllers

(Part of this work has been presented in PSE2000, Keystone, Colorado, USA, 16-21

July 2000, Aziz et al., 2000b)

Figures 5.26 and 5.27 show the response of the PI controllers in tracking the dynamic set points (T_{sp}) for RUN1 and RUN2. The responses of the PID controller for RUN1 and RUN2 are shown in Figures 5.28 and 5.29.

Large offset in reactor temperature was observed using PI controller (Figure 5.26) while the PID and GMC-NN controllers were able to track the reactor temperature with little offsets. However, based on the amount of desired products achieved (Table 5.4), the controller performance (CP) using the PID is found to be slightly better than that obtained by using the GMC-NN. It is due to the larger rise time for the GMC-NN control strategy compared to the PID controller. The heat-up process used by the DM controller was quicker compared to the GMC-NN controller. However the GMC-NN controller provided less drastic change in the jacket temperature set point compared to PID controller. Also it was found that the performance of GMC-NN controller is more stable compared to other two controllers (in order to track both types of set point profiles). The responses of PI and PID controllers were more sluggish than GMC-NN in tracking the dynamic set points. Another advantage of the GMC-NN is that only two parameters have to be tuned compared to six and seven parameters for the DM with PI and PID controllers respectively. The performance of the GMC-NN controller is strongly dependent on the estimation of the heat released (Q_r) by the reaction. The neural networks used in this work gave a very good estimation of the heat released by the reaction (Figure 5.23 and 5.25) and hence guaranteed the good performance of the GMC-NN controller.

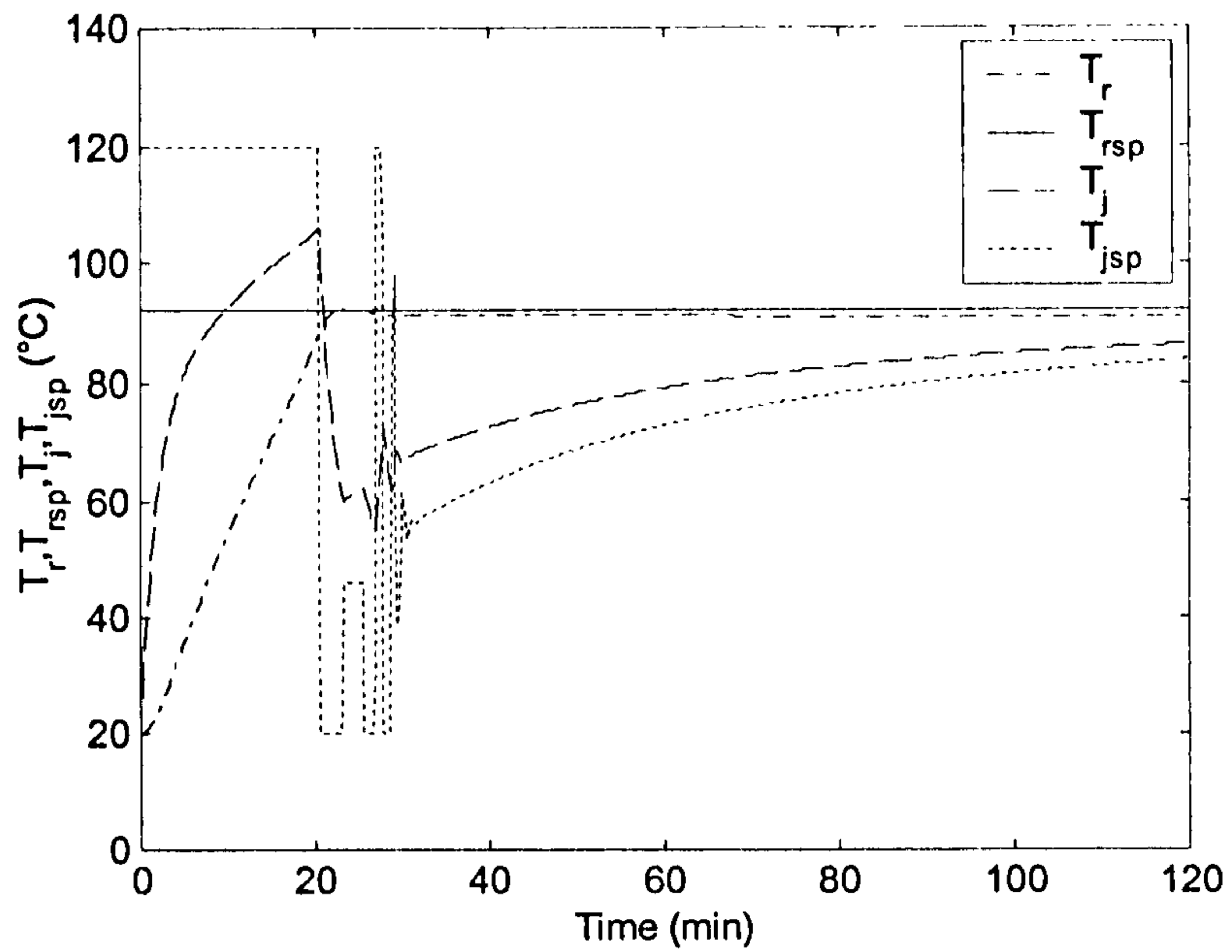


Figure 5.26: PI response for 1 T_{sp} (Case Study 2 - RUN1)

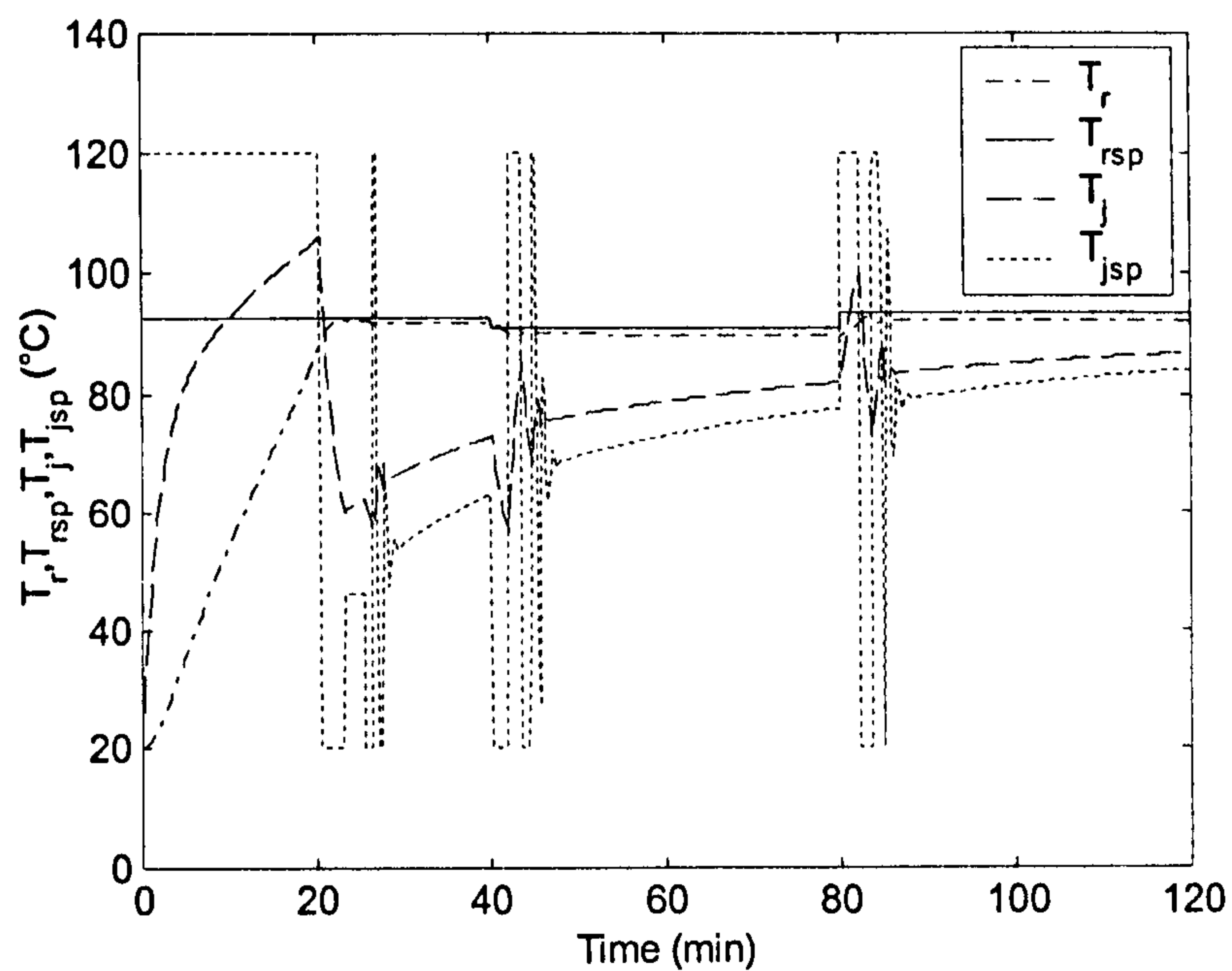


Figure 5.27: PI response for dynamic set points (Case Study 2 - RUN2)

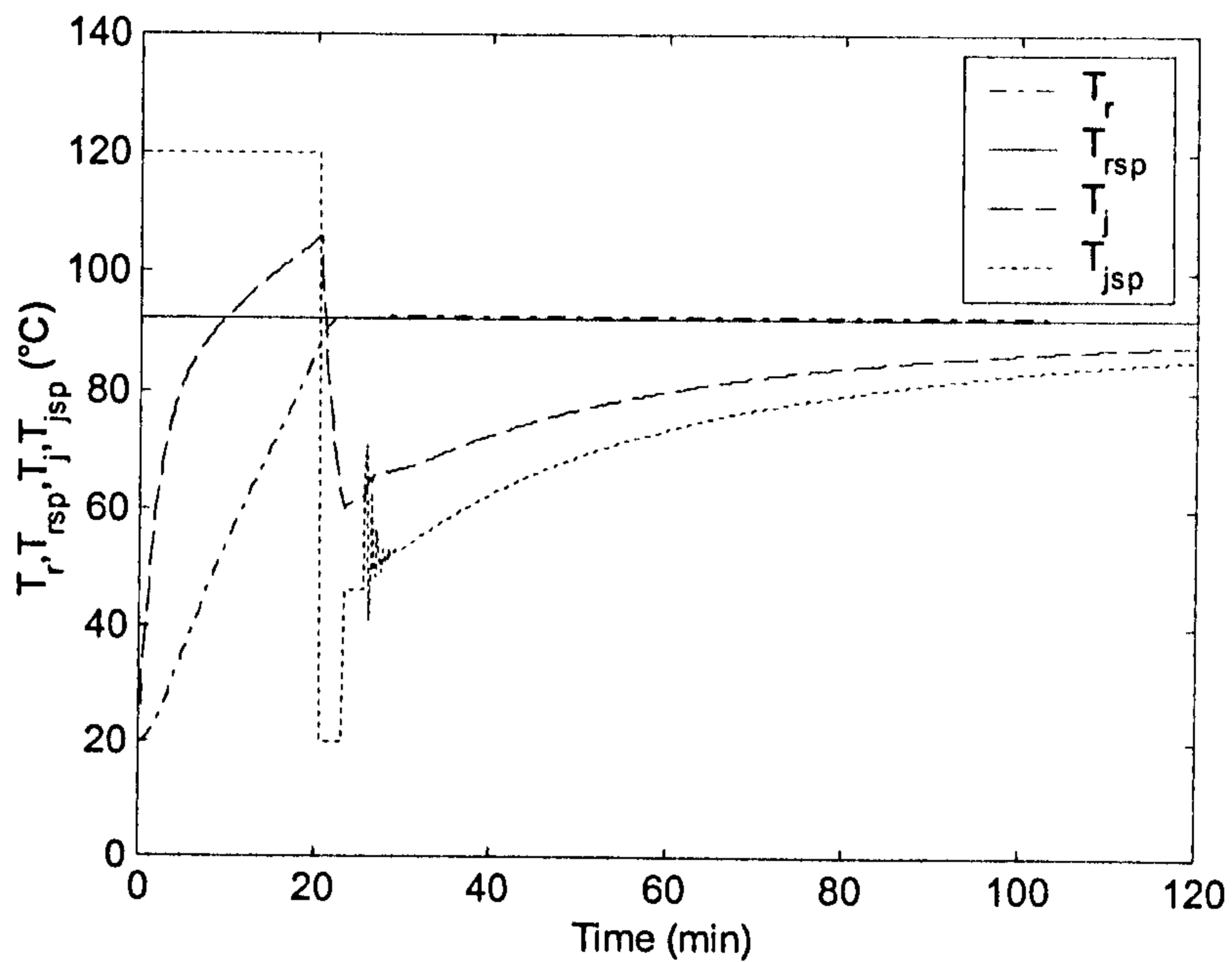
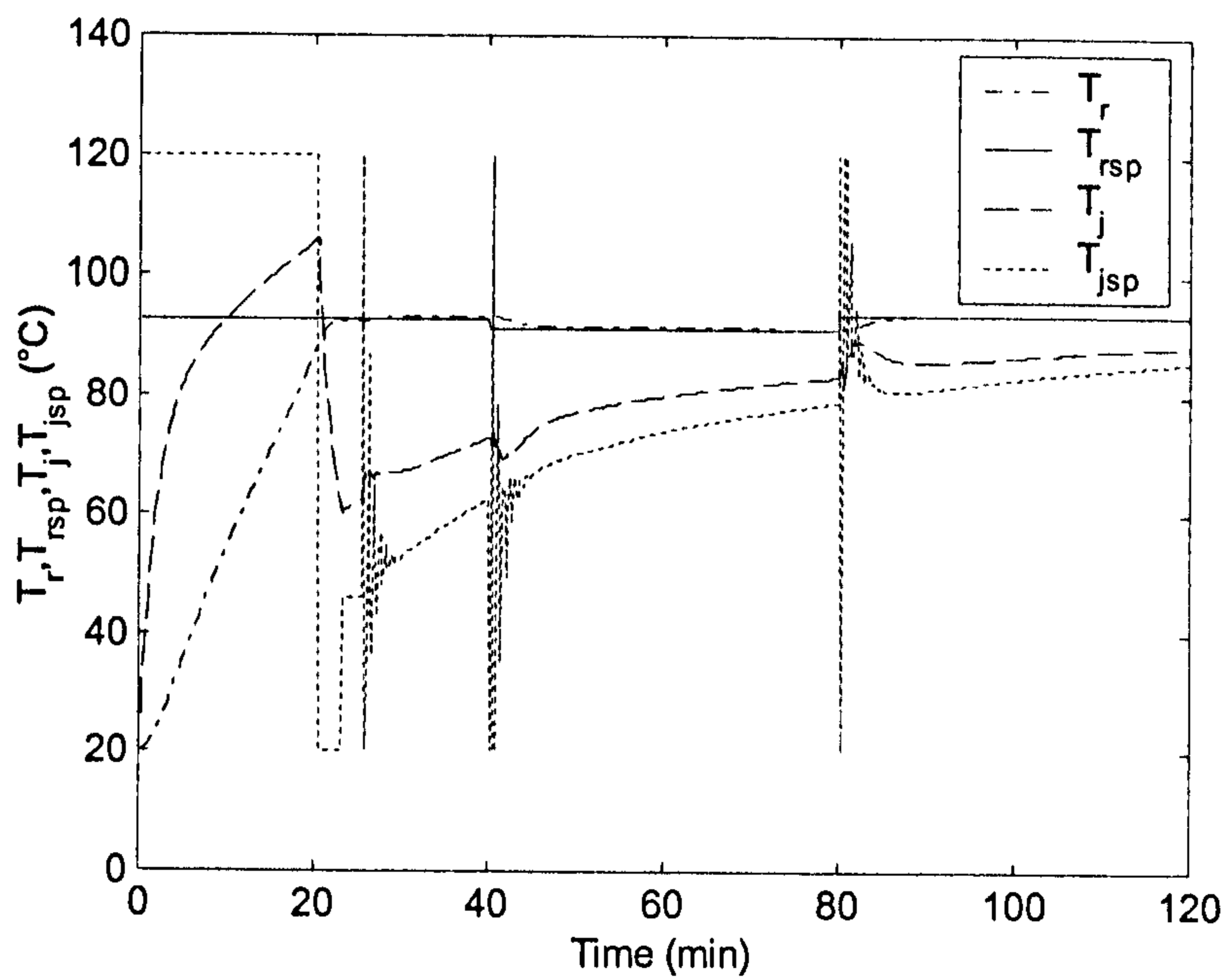
Figure 5.28: PID response for 1 T_{sp} (Case Study 2 - RUN1)

Figure 5.29: PID response for dynamic set points (Case Study 2 - RUN2)

Direct Inverse and IMBC Controllers Responses

Figures 5.30 and 5.31 show the response of the Direct Inverse controller in tracking the set points (T_{sp}) for RUN1 and RUN2. The responses of the IMBC controller for RUN1 and RUN2 are shown in Figures 5.32 and 5.33. It can be seen that the both controllers, Direct Inverse and IMBC controllers were able to track the constant and dynamic set points very well. Even though the responses of both Direct Inverse and IMBC controllers look very similar to each other, in fact there is some improvement made by the latter as it reduced the value of offset to some extent. The amounts of the product obtained using both types of controllers (shown in Table 5.4) strengthen this observation i.e. the amount of product from the IMBC (for the dynamic set points) is slightly higher compared to that obtained using Direct Inverse. It is because in the IMBC strategy, its error information (using plant-model mismatch) is considered and supplied to the controller through the feedback loop.

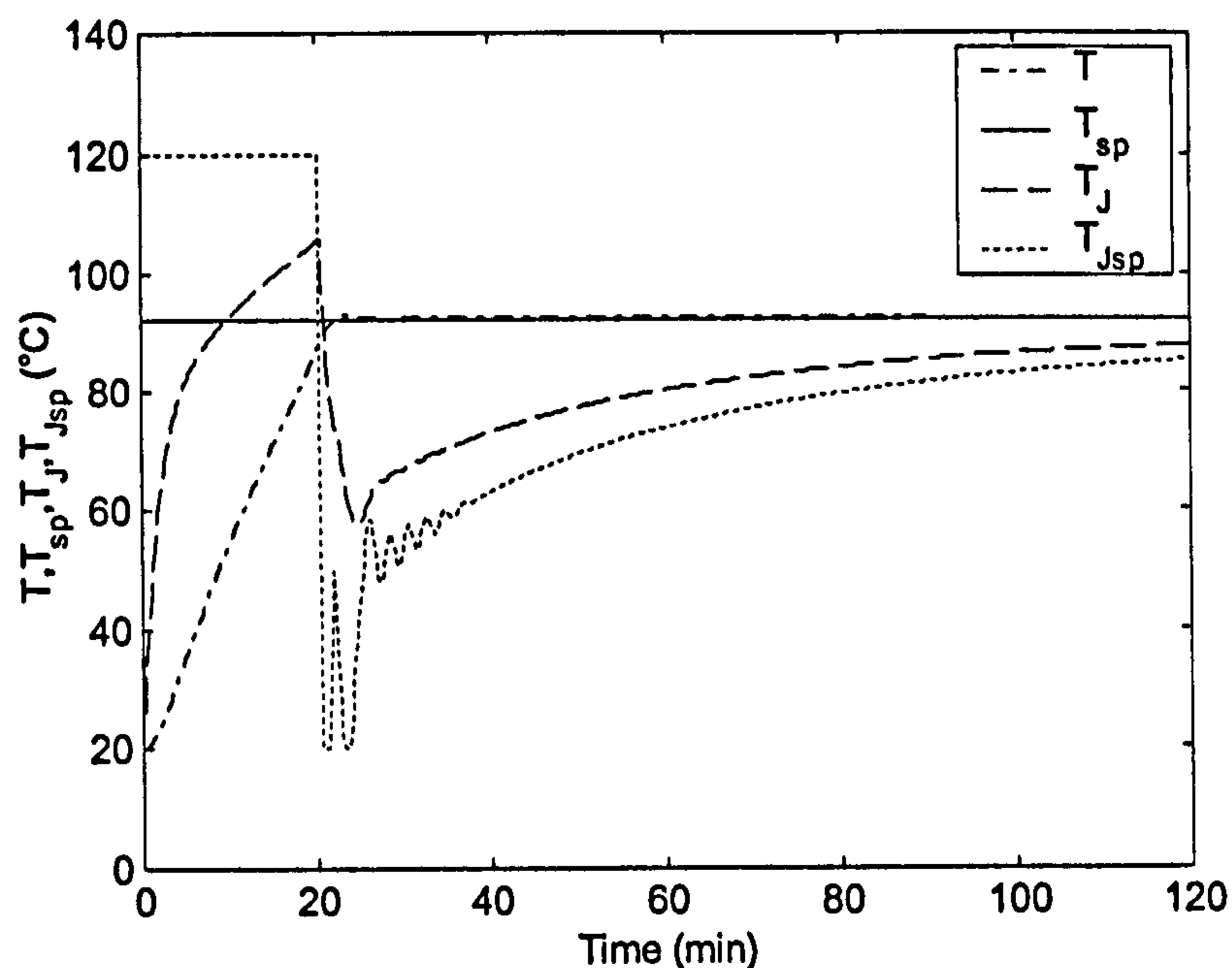


Figure 5.30: Direct Inverse response for 1 T_{sp} (Case Study 2 - RUN1)

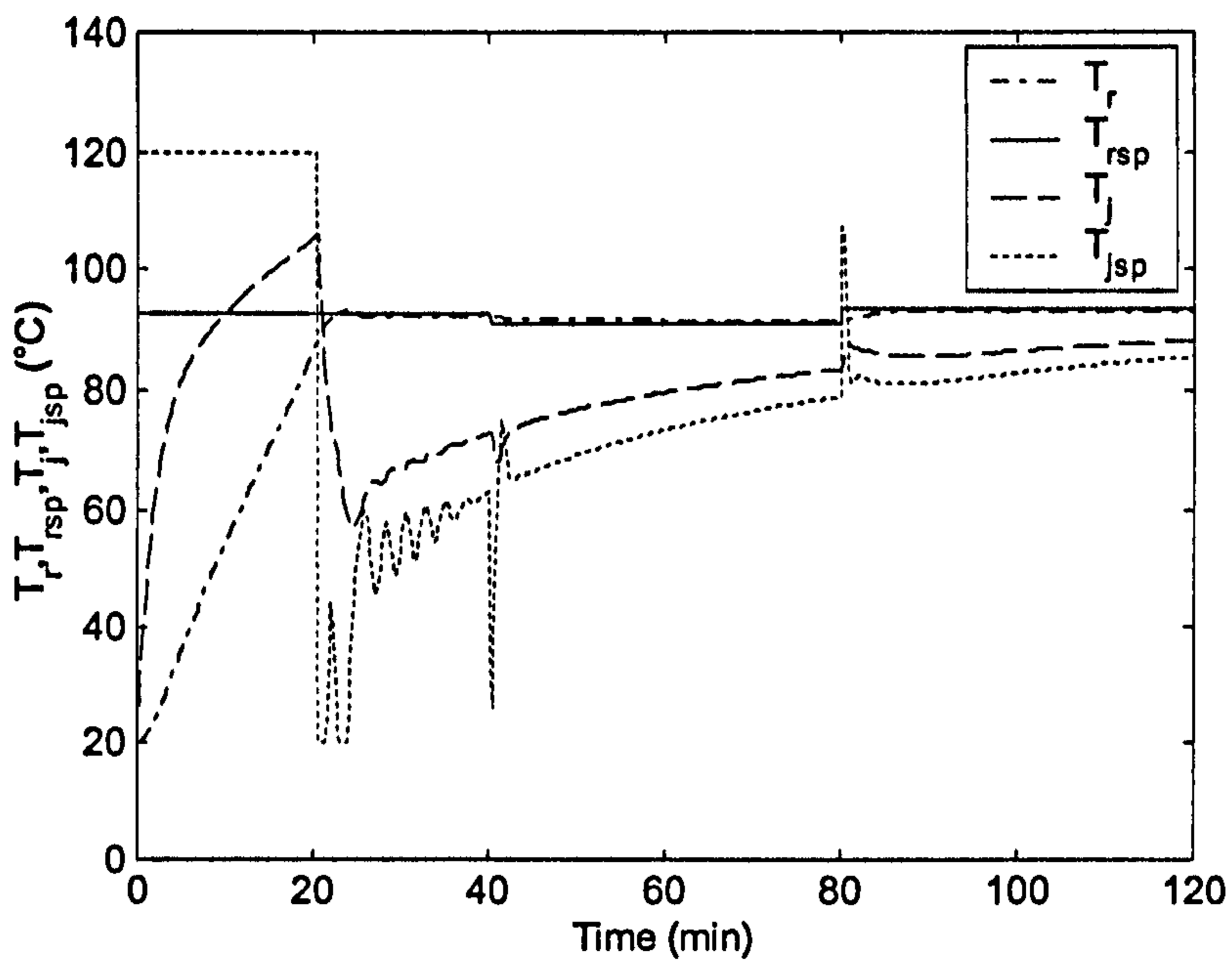


Figure 5.31: Direct Inverse response for dynamic set points (Case Study 2 - RUN2)

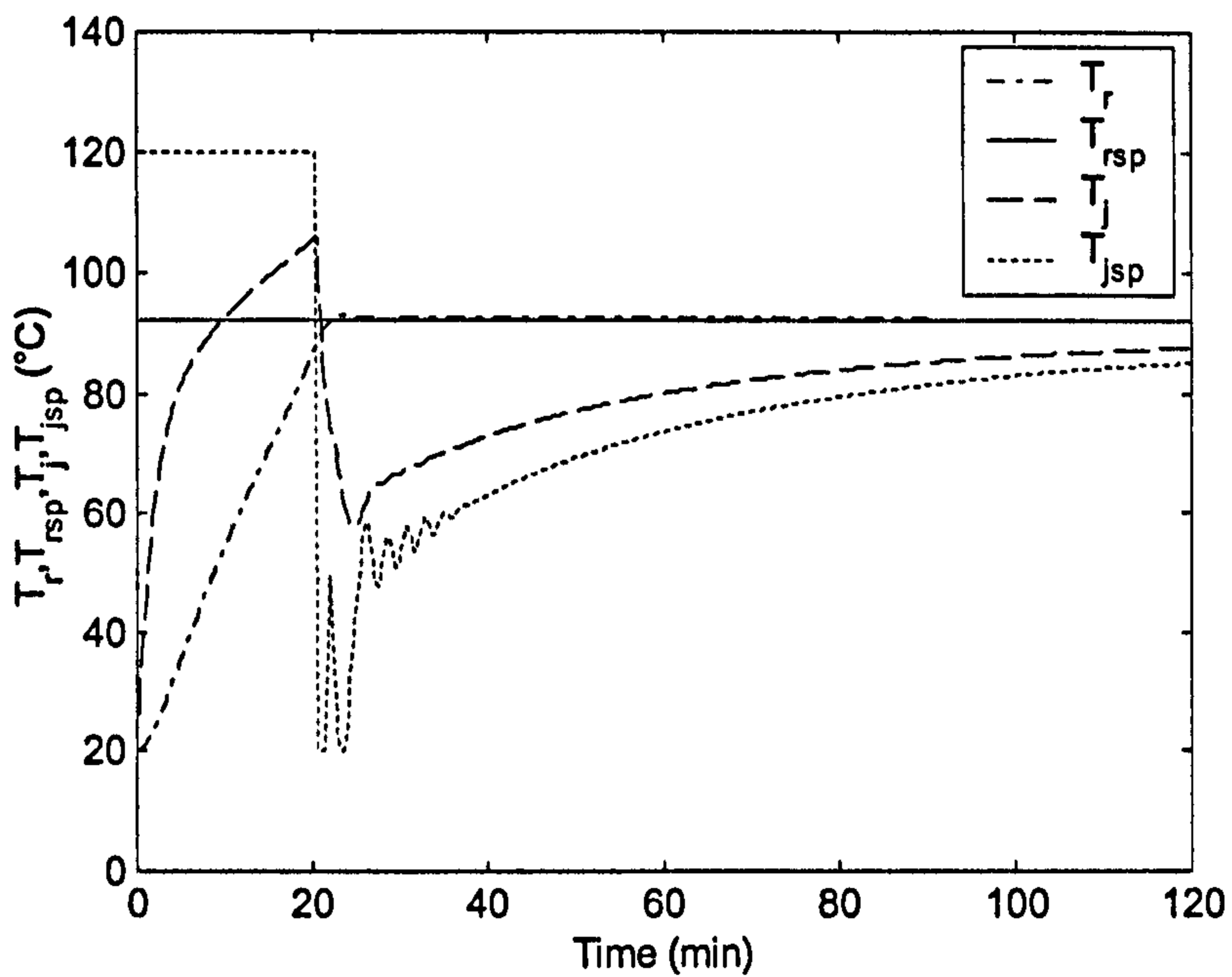


Figure 5.32: IMBC response for 1 T_{sp} (Case Study 2 - RUN1)

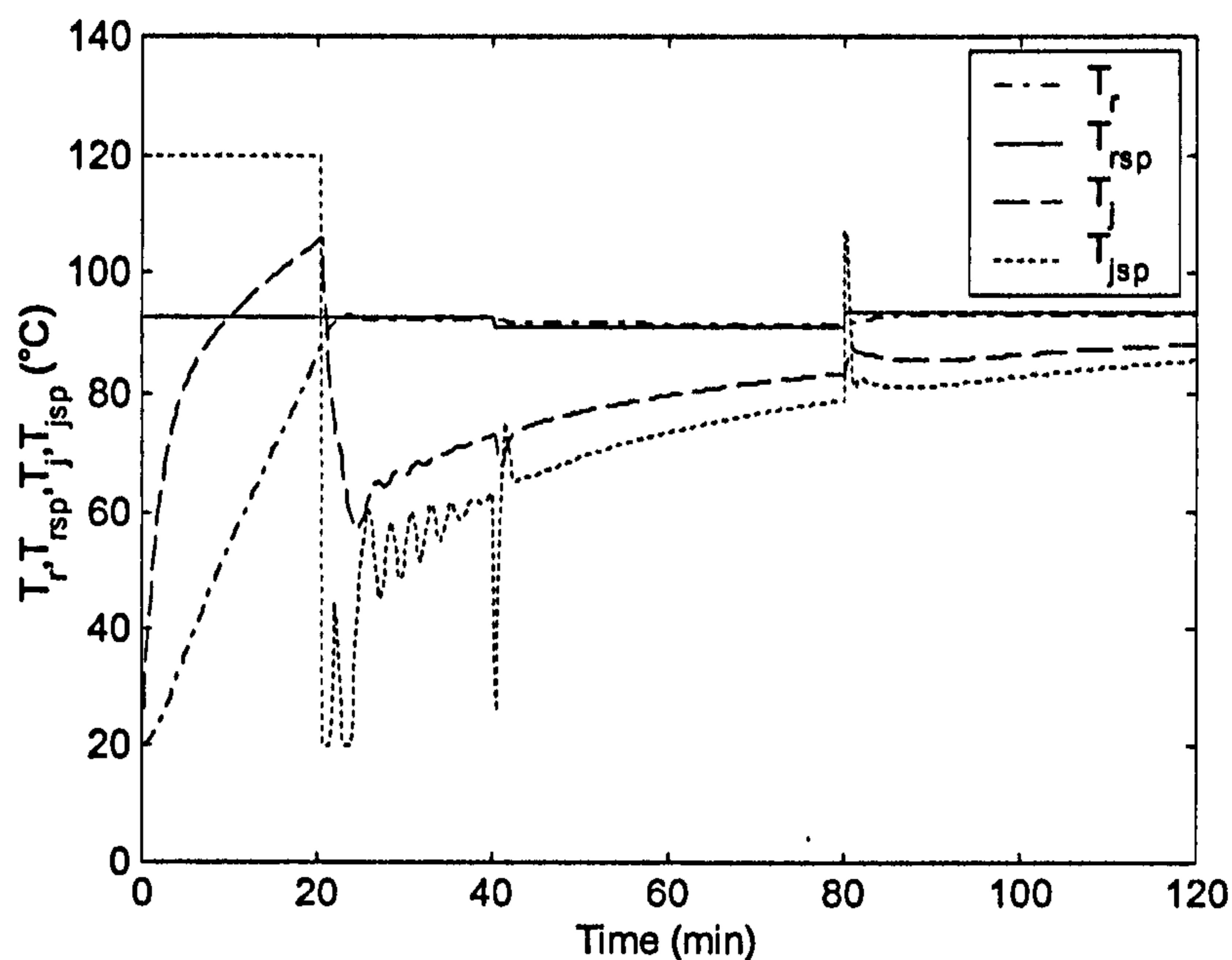


Figure 5.33: IMBC response for dynamic set points (Case Study 2 - RUN2)

Comparison between GMC-NN and Neural Controllers (Direct Inverse and IMBC controllers)

(Part of this work was presented at the 6th World Congress of Chemical Engineering, Melbourne, Australia, 23-27 September 2001, Aziz et al., 2001b)

In this case study, neural controllers and GMC-NN controller performed well in tracking the constant and dynamic set points. However, the GMC-NN controller ends up with larger offset and greater rise time. Due to these reasons, the value of product B using GMC-NN (shown in Table 5.4) is slightly lower compared to those obtained using the neural controllers. The controller performance, CP for the GMC-NN controller is 97.15% and 97.14% for the constant and dynamic set points respectively. On the other hand the CP for neural controllers is 97.33% and 97.28% for the constant and dynamic set points respectively. Even though the neural controllers outperformed

the GMC-NN controller in terms of tracking the set points, but it shows more drastic changes in the controller actions when compared with GMC-NN. This is due to the use of piecewise constant (constant over a time interval) value of T_j in the training of inverse model network. T_j values were obtained by solving the following optimisation problem:

$$\text{Min} \quad \sum_{j=1}^N (T - T_{sp})_j$$

T_j

Subject to: constraints (model equations etc.)

N is the number of intervals within the batch time of interest.

The drastic changes in the control actions can be overcome if smaller time intervals for calculating T_j are used. It is also observed that there is a little overshoot in the neural controllers' response. After all, the neural controllers do not need any parameter tuning as in the case in GMC-NN.

Comparison between Neural Controllers (Direct Inverse and IMBC controllers) and PID Controller

This study shows that both the neural controllers and PID can track the set points well. In tracking the constant set points, the former gives less offset but was more sluggish in the control action. However in tracking the dynamic set points, the neural controllers end up with greater offset and were less sluggish in control action as compared to the PID controller. In terms of the controller performance, both types of controllers result in a very similar performance (Table 5.4). Again, neural controllers have the advantage over PID as they do not need any parameters to be tuned.

Robustness Test

Here, again the robustness of the controllers applied in this case study has been tested. Three tests were carried out by changing the process parameters. In all tests the controllers (tuned as before) were used to control an operation where some of the conditions have been changed from their nominal values. In the first test (TEST1), the heat of reactions was increased by 25%. In the second test (TEST2) the heat transfer coefficient is reduced by 40% of its original value. The third test (TEST3) involves 30% reduction in the molar (or mass) of reactants. In all tests, a constant reactor temperature set point (RUN1, Table 5.2) is to be tracked by all controllers.

The results for all these tests are shown in Figures (5.34-5.39). For all tests, it can be seen that the GMC-NN controller was able to accommodate all the changes very well compared to the other types of controllers. In TEST1 and TEST2, it is found that the response of neural controllers is worst (greater overshoot) than the conventional PI and PID controllers (Figures 5.34-5.37). It is due to the inherent property of neural networks i.e. it is good in interpolation but not in extrapolation. The capability of the neural controllers are solely dependent on the range of the parameter values used for training. In this work, the neural controllers were trained between the reactor temperature range of 90 and 95°C only. However in TEST3 (Figures 5.38-5.39) the changes were within the limit of the training therefore the neural controllers perform well (as GMC-NN). In fact the former reaches to the set point quicker than the latter. On the other hand, in this test the response by PI and PID controllers were initially sluggish.

5.5.3 Remarks from the Applications (Case Study 1 and 2)

Both cases studies clearly show the robustness and the stability of the GMC-NN controller in controlling various kinds of reaction schemes. PID controller did well in Case Study 1 but unable to cope with the robustness test in Case Study 2. From this observation, we can conclude that PID can be a good controller for some type of reaction and condition only. PI controller and neural controllers in Direct Inverse and IMBC strategies have been applied and tested only in Case Study 2. From the study, it is obvious that the PI controller was unable to perform well compared to other types of controllers. In other words, it could not cope with the non-linear characteristics of the batch reactor. Neural controllers have very high potential to be applied as a robust controller but must be trained very well and the training range should cover all possible conditions that can happen to the system or process.

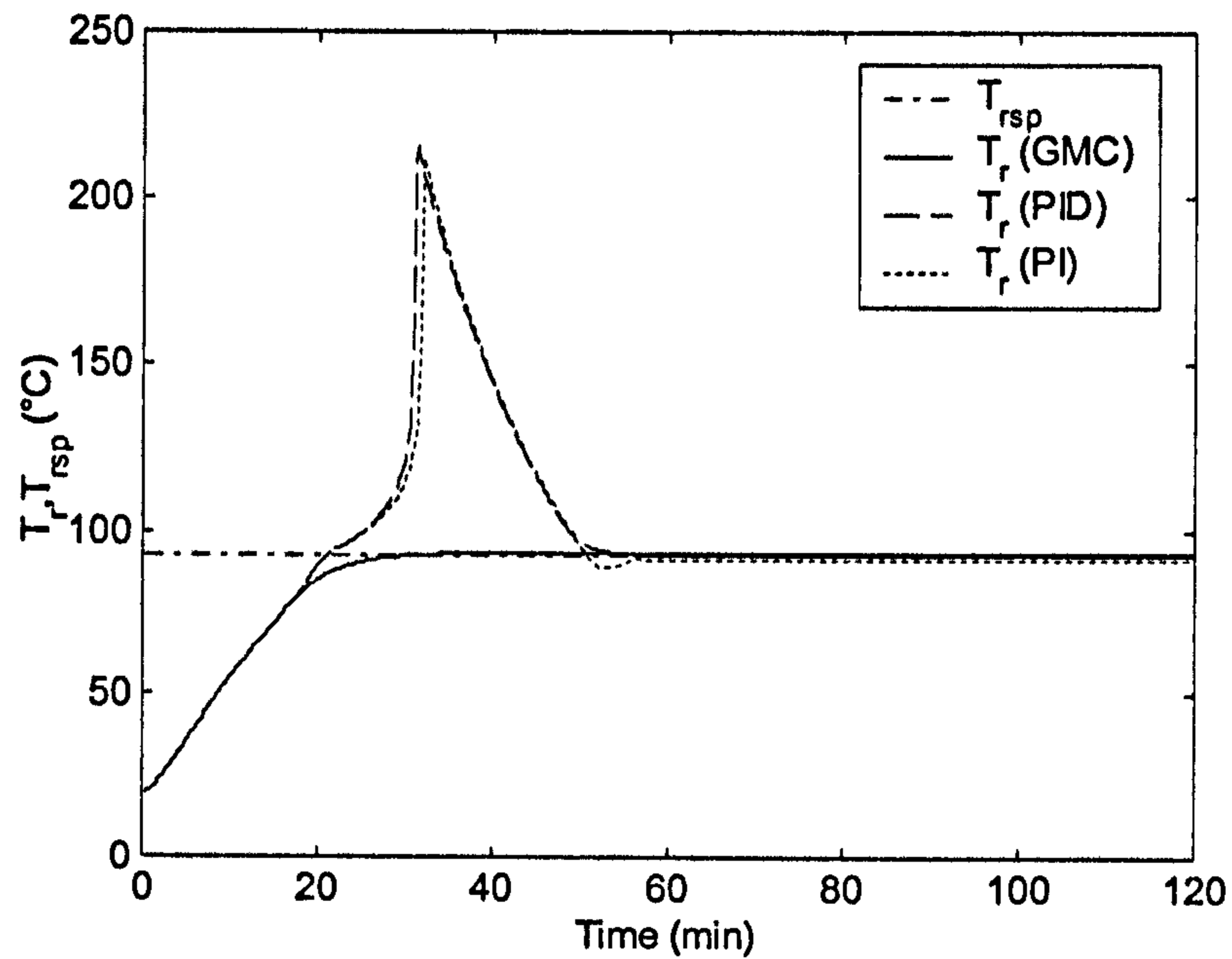


Figure 5.34: Controllers responses for heat reaction change (Case Study 2) – I

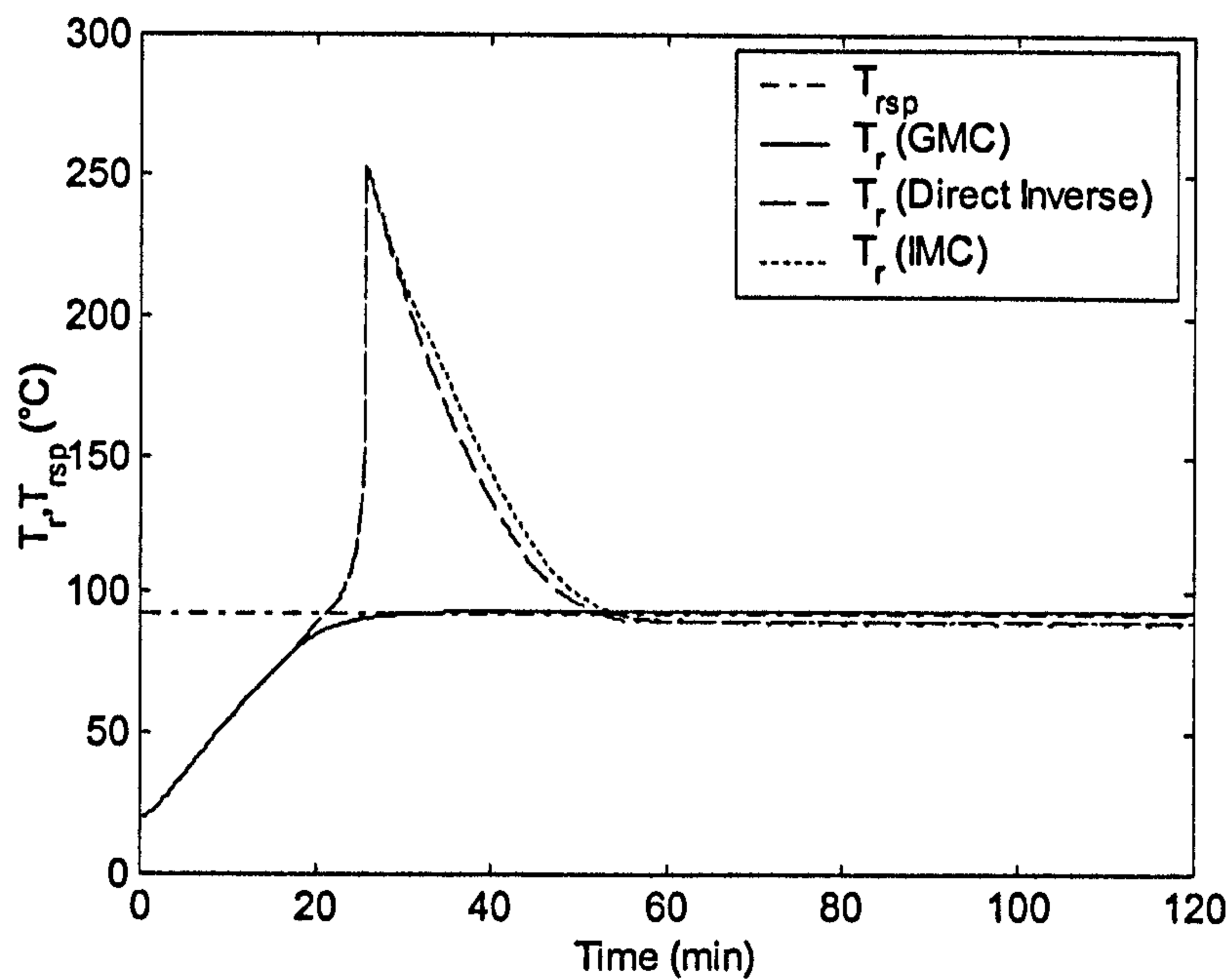


Figure 5.35: Controllers responses for heat reaction change (Case Study 2) – II

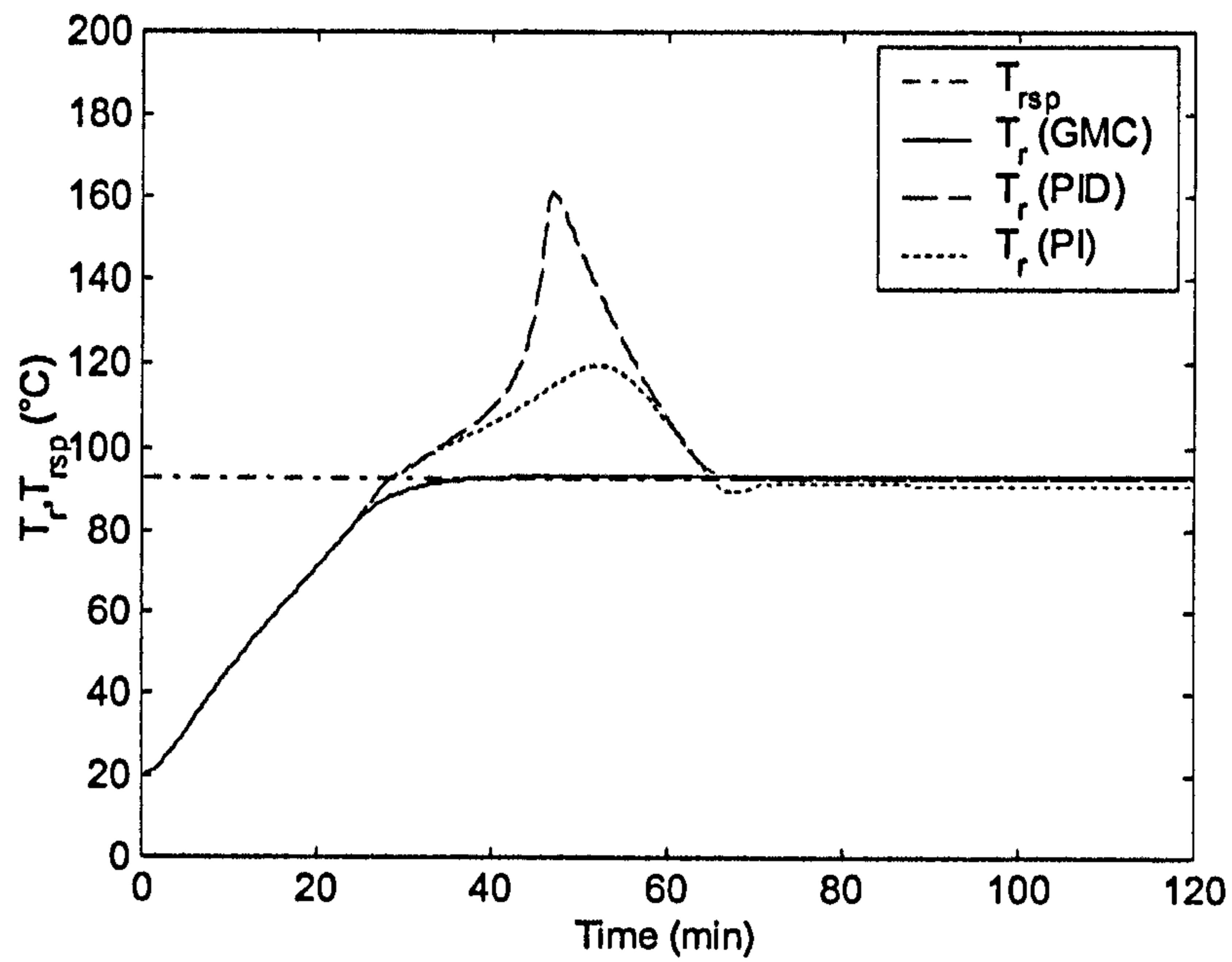


Figure 5.36: Controllers responses for heat transfer coefficient change (Case Study 2) - I

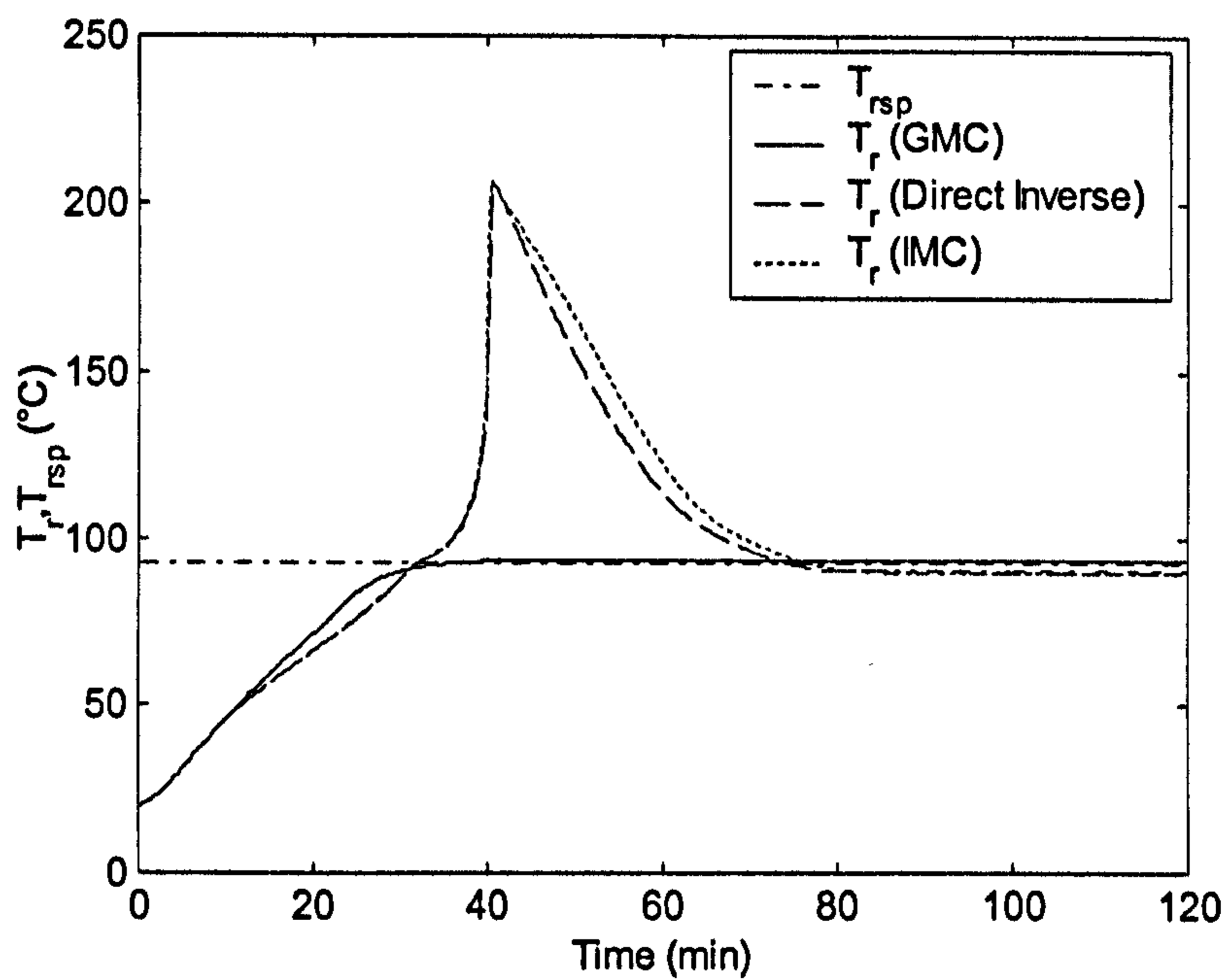


Figure 5.37: Controllers responses for heat transfer coefficient change (Case Study 2)-II

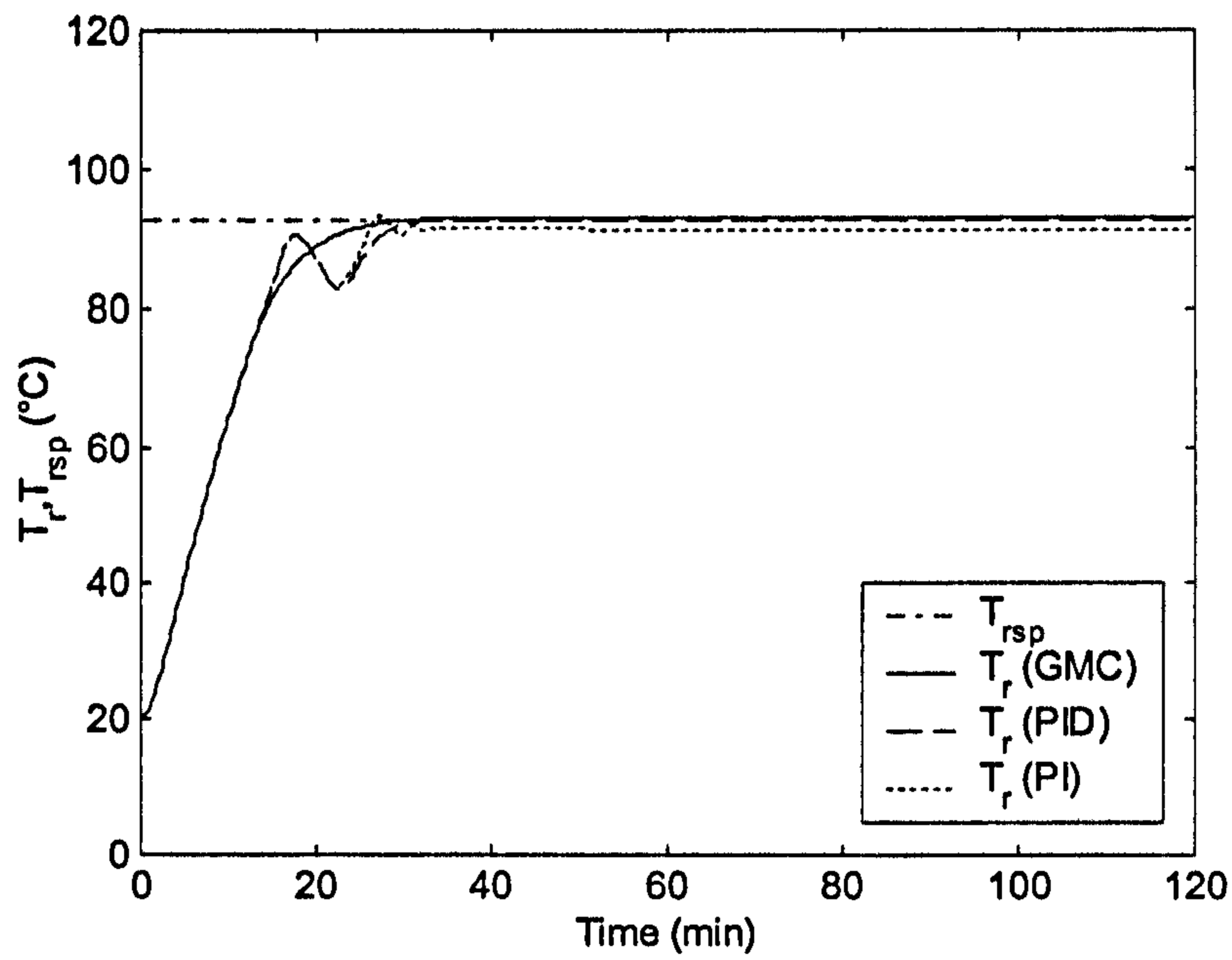


Figure 5.38: Controllers responses for molar/weight change (Case Study 2) – I

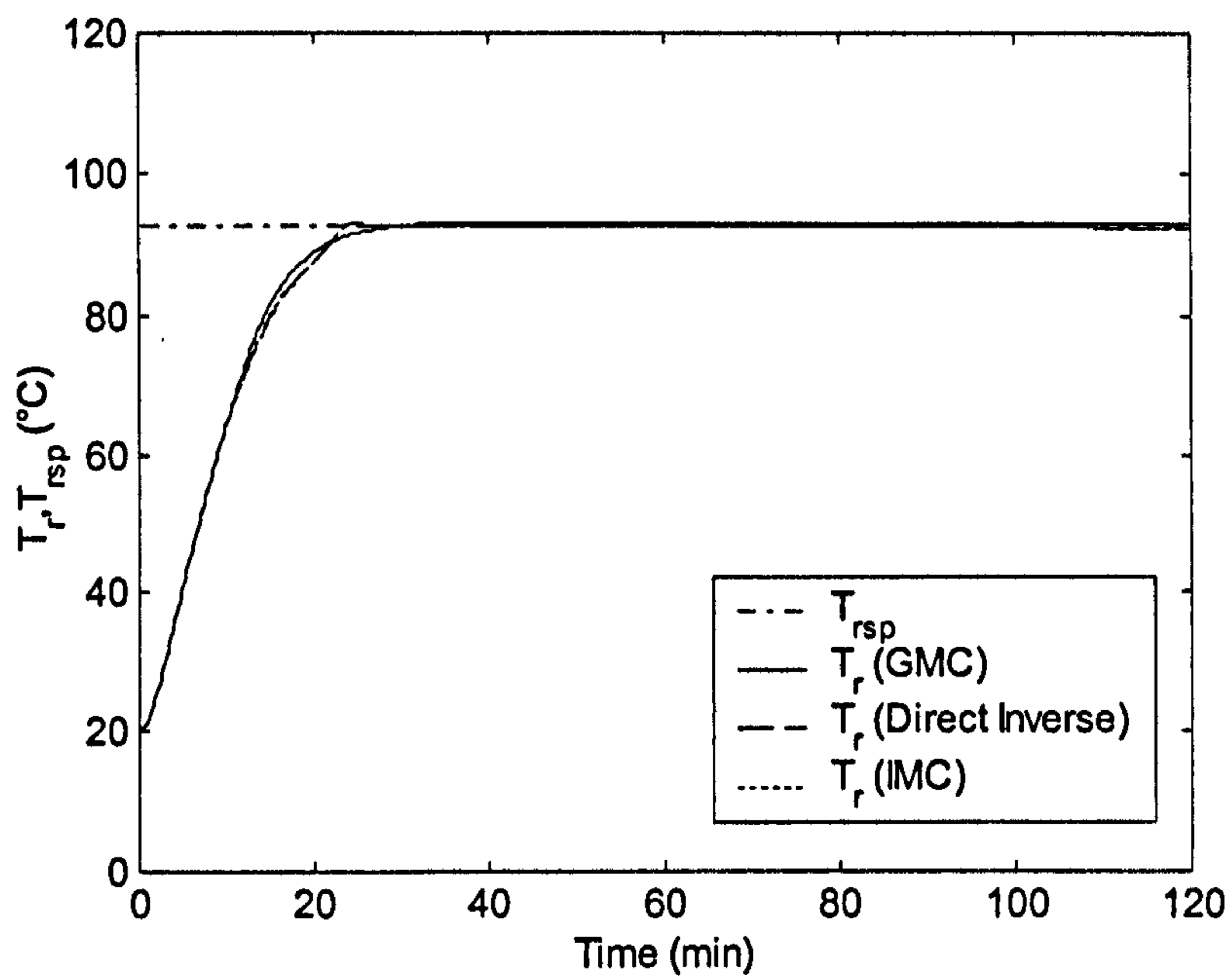


Figure 5.39: Controllers responses for molar/weight change (Case Study 2) – II

5.6 Conclusions

This chapter deals with the batch reactor control issue. It links up between the off-line optimisation of Chapter 4 with the on-line tracking of the optimum profiles. Two conventional types of controllers i.e. PI and PID controllers and three types of advanced non-linear control strategies i.e. GMC-NN and neural controllers in Direct Inverse and IMBC strategies have been applied and tested in this section. Rather than considering on-line tracking of constant set points profile only (as have been considered by most of the previous researchers), on-line tracking the dynamic set points was considered as well. However, in this study, it was limited to three step changes only. Two case studies have been presented and the robustness tests have also been made to test the robustness of each controller under consideration in terms of process parameter changes. It was found that the GMC-NN controller outperformed other types of controllers. However, the neural controllers is also promising as a robust controller in future if they have been trained a priori to cover all possible conditions that can happen to the process. To summarise, the items presented in this chapter are:

1. We considered both off-line dynamic optimisation and the on-line set point tracking of batch reactors. To bridge the gap (in the literature), we implemented the off-line optimal profiles to be tracked by various types of advanced non-linear controllers.
2. Both a constant and dynamic set points have been considered as set points to be tracked by different types of controllers. This provided more room to evaluate the stability and the robustness of the controllers applied.
3. Various advanced control strategies i.e. GMC-NN and neural controllers (inverse model) in Direct Inverse and IMBC strategies have been implemented.

Their performances (based on their responses and the amount of the desired product) were evaluated and compared with conventional DM control strategy with PID and PI controllers.

4. Robustness tests were conducted for each of the controller applied with changes in process parameters i.e. value of heat of reactions, heat transfer coefficient, reaction rate constant and weight or molar change.
5. Versatility of neural networks as a tool to be used as estimator, predictor and controller are shown using two case studies.
6. For the first time, evaluation and comparison between three advanced non-linear control strategies i.e. GMC-NN controller and neural controllers (in Direct Inverse and IMBC strategies) have been made in batch reactor systems using two different case studies with different types of reaction schemes. The first case study represented the consecutive reaction scheme with pre-heated reactant and second case study dealt with a complex reaction scheme with heating and cooling element.

Designing a controller for batch reactor is a very challenging work because of its dynamic nature. Generally, the conventional and linear controllers are unable to cope with the unsteady state nature of this type of system. However the use of advanced control strategies such as the GMC-NN, neural network inverse model based controllers have been shown to be a promising alternate to the control of batch reactor system.

Chapter Six

6.0 BATCH_REACT: An Integrated Framework for Modelling, Simulation, Optimisation and Control of Batch Reactors

(Part of this work was presented at the EURECHA Session & AGM, Denmark, 27 May 2001)

6.1 Overview

The aspects of modelling, simulation, optimisation and control of batch reactors have been dealt separately in the past (see Chapter 2). For this reason, in this chapter, we propose to develop an integrated framework BATCH_REACT, which will consider all aspects mentioned above. With minimum information from the users on reaction schemes, e.g. series, parallel or complex reactions, reversible or irreversible reactions, kinetic information the BATCH_REACT will:

1. Generate model equations to describe the batch reactor operation (detailed or short-cut) – using GBRM (Chapter 3)
2. Simulate the reactor
3. Generate off-line optimal operation policies (set points)
4. Design controllers to track the optimal set points on-line

In other words, all the works done in Chapters 3-5 will be combined together in BATCH_REACT. The proposed software can be used for teaching, research and for commercial exploitation.

6.2 Four-Level BATCH_REACT Framework

There are four levels of activities in the proposed framework (Figure 6.1) i.e. modelling, simulation, optimisation and control. Here, we will explain all the levels, how they function and their limitations.

In the modelling level (Level 1), two types of models i.e. short-cut model and detailed model will be considered with all the options presented in Chapter 3. There are five options available in this level, which cover both simple and detailed models. In the short-cut model, only mass balance and kinetic equations are generated. In the detailed model all the equations in short-cut model plus the energy balance equations will be included. At this level, the user has to supply a very limited information such as reaction schemes, e.g. series, parallel or complex reactions, reversible or irreversible reactions and kinetic information (i.e. the user will choose the option). Besides the equations mentioned above, at this level, the non-zero matrix and the gradient information of the model will also be generated, so that these can be coupled to a solver.

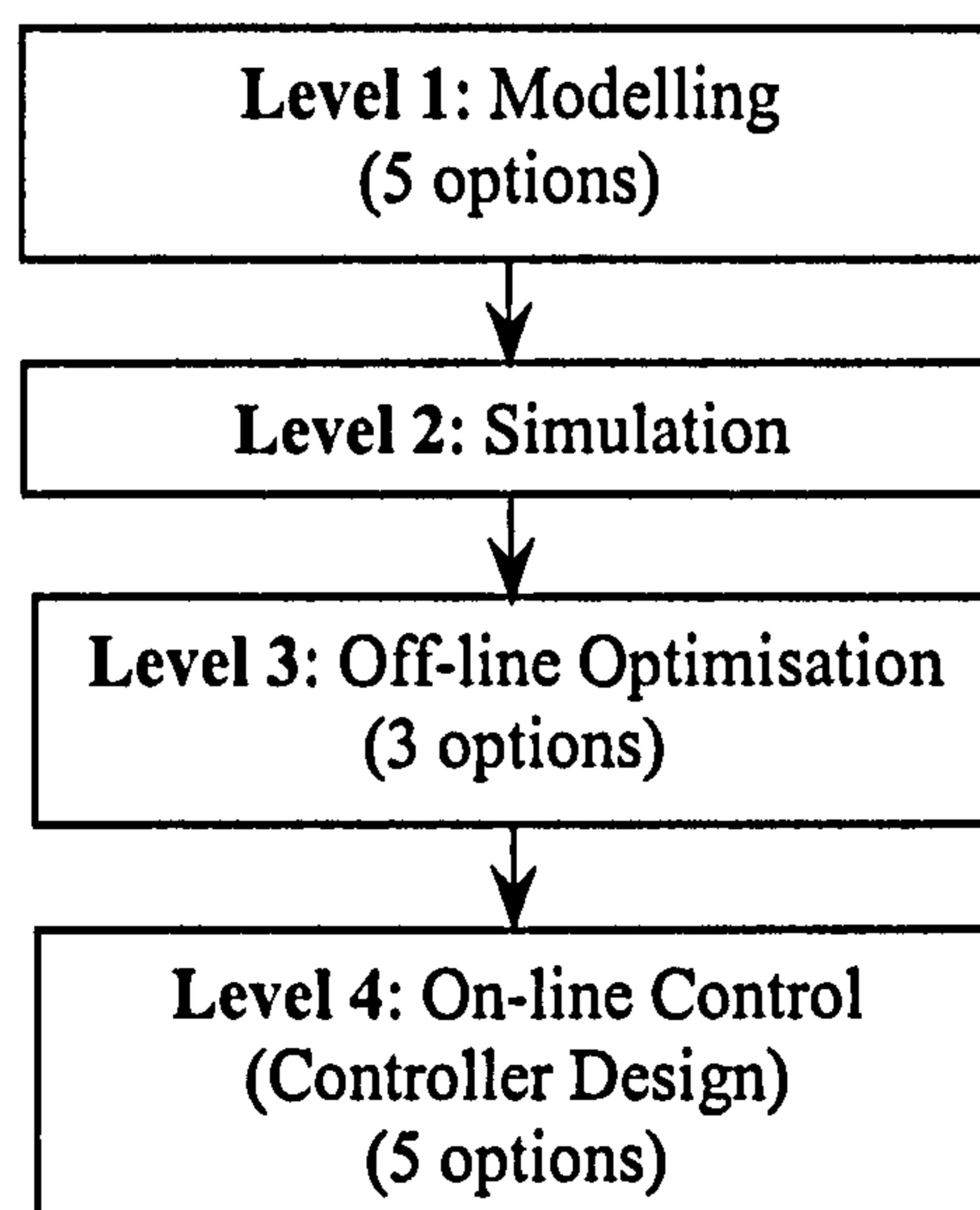


Figure 6.1: Four Levels BATCH_REACT Framework

Since a batch reactor is inherently an unsteady state process, the model equations generated in Level 1 will result to a number of differential and algebraic equations (DAEs), which can be solved using Gear's-type integrator. Based on the model chosen and the information supplied by the user, the process can be simulated (Level 2). At this level, data generation for neural network based model or controller or sensitivity analysis of the process can be made.

In Level 3, the optimisation of the process will take place. There are three optimisation options offered in this level (maximum conversion, minimum time and maximum profit problems). However, based on the model chosen by the user in Level 1, the control variable will be fixed. It will be either the reactor temperature, jacket temperature or jacket (coolant or heating) flow rate. The user can decide the number of time intervals with the option of optimising them or not, the final batch time or the desired conversion (if maximum conversion problem or minimum time problem is chosen). With all these information, the off-line optimisation will be carried out. As explained in Chapter 4, the control vector parameterisation (CVP) method with successive quadratic programming (SQP) can be used in this level. Other optimisation techniques can be implemented in future if needed.

In Level 4, the optimal profiles achieved in Level 3 will be tracked on-line by the type of controller chosen by the user and the chosen controller will be designed. However, currently five types of controllers (options) are provided i.e. dual mode (DM) with PI and PID, GMC and neural controllers (Direct Inverse and IMBC).

6.3 Steps to Follow by the User

In brief the steps that have to be taken by the users in order to use BATCH_REACT will be:

1. Provide the reaction schemes and reaction kinetic information with all the parameters and constant values.
2. Choose the available model option.
3. Choose BATCH_REACT level (2, 3 or 4)
4. Choose option for the type of the optimisation problem to be solved in Level 3, e.g. maximum conversion, minimum time or maximum profit problems. Provide the optimisation specifications such as the number of control intervals, final batch time, desired conversion, accuracy etc.
5. Choose option for the type of controller to be used in Level 4. If neural network based controller is chosen, Level 2 cannot be omitted (data for neural network training are to be generated and the training has to be done off-line). If chosen to use optimal operating profile, the Level 3 will also have to be chosen. If not user has to supply the set points. Change some parameters in the process if the robustness tests need to be done.

The flow diagram of the BATCH_REACT can be seen in Figure 6.2.

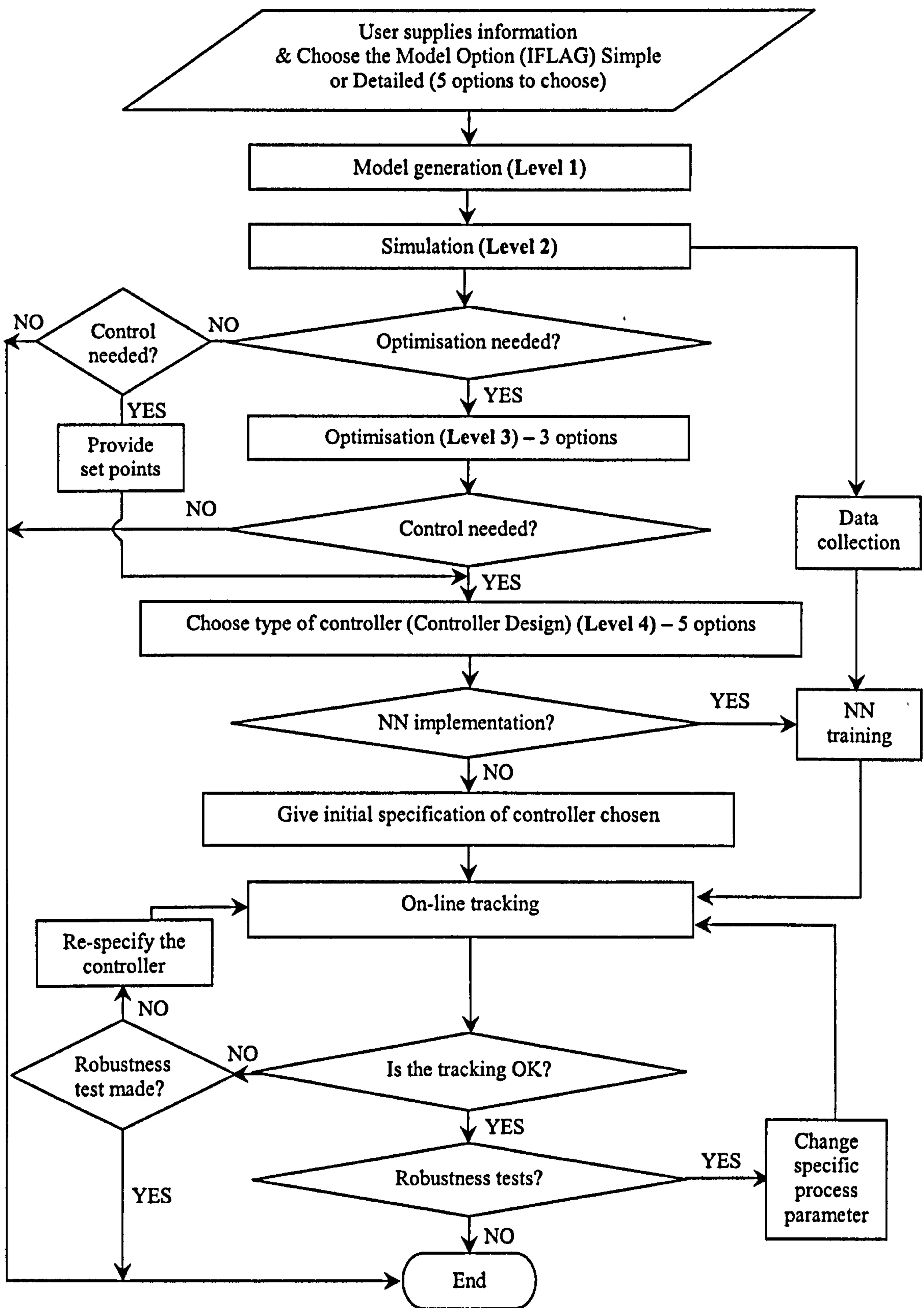


Figure 6.2: Flow diagram of the BATCH_REACT

6.4 Current Limitations of the Proposed BATCH_REACT

Some of the current limitations of the proposed software is listed here:

1. The mass balance equation generated by GBRM is in term of reactant and product concentration.
2. The control variable is fixed based on the model option chosen. It is either the reactor temperature, the jacket temperature or the jacket flow rate.
3. Data for neural network based controller are obtained by simulation – can be replaced by experiments
4. Only five types of control strategies (DM with PI and PID controllers, GMC, Direct Inverse and IMBC) are offered in the control level.

6.5 Future Development

To overcome the limitations mentioned in the previous section following improvements have to be done:

- 1 Mole or mole fraction based mass balance equations should be incorporated. Extension to semi-batch reactor system should be considered.
- 2 The user should be given more flexibility in choosing the control variable.
- 3 An interface should be developed for neural network with the proposed software framework where the data collection and the training can be done on-line – the adaptive concept can be implemented or the software can be interfaced with MATLAB program.
- 4 Other advanced controller schemes such as hybrid and adaptive schemes should be incorporated.

Chapter Seven

7.0 Conclusions and Future Work

7.1 Conclusions and Contributions

The detailed summaries and conclusions for various parts of this thesis have been outlined at the end of each chapter. Here we summarise the important points relating to the scope of each chapter respectively.

In Chapter 2, the dynamic optimisation (off-line optimal control) and on-line control of batch reactors were reviewed. Dynamic optimisation of batch reactors refers to the reaction with known kinetics and the aim is to maximise or minimise a certain objective function and control of batch reactors refers to tracking the set-point profile and disturbance rejection. The review on control application was focused mainly on three strategies i.e. GMC, Direct Inverse and IMBC strategies. Process modelling issues with respect to simple and detailed models and their applications were also highlighted. It has been observed that there were limited work on optimisation of batch reactors and was mainly focused on the numerical methods. Most of the optimisation formulations were simple, based on simple process model without considering the path and/or end point constraints and limited to the maximum conversion or minimum time problems only. It was also observed that the control vector parameterisation (CVP) technique is still one of the most popular methods to solve the dynamic optimisation problem. Its merits over other methods or approaches were also discussed in this chapter. In the past, no attempt has been made to implement the optimal profiles (obtained by solving off-line dynamic optimisation problem) on-line (as a basis for the controller design). From this review, it was found that the three control techniques used in this work (GMC,

in this work (GMC, Direct Inverse and IMBC) have a very limited implementation in batch reactor even though they were proved as robust controllers in this work and in others as well.

In Chapter 3, we focused on the dynamic modelling of batch reactors based on the mechanistic first-principle-model. A general batch reactor model (GBRM) was developed and coded incorporating several options which results in models of different complexity (simple to detailed). Also most common types of reaction schemes (consecutive, parallel, complex, reversible and irreversible) were built in GBRM. Given the options, GBRM will automatically build up the required dynamic model (the mass and energy balances, the reaction rate and reaction rate constant equations, etc.) of batch reactors. All options available in this code were discussed using various examples and the degrees of freedom analysis of the resulting models were analysed. Finally some simulation results of various types of batch reaction models using GBRM were presented and compared with those using the process-specific models.

In Chapter 4, classical formulation of dynamic optimisation problem (maximum conversion, minimum time and maximum profit problems) for batch reactors were presented and the solution of the problems using CVP technique was explained. Six case studies with different aims and objectives were presented. In Case Studies 1 and 2, maximum conversion and the minimum time problems for two different reaction schemes were presented. The aim of these studies was to verify and compare the efficiency of CVP technique as opposed to iterative dynamic programming (IDP) technique. In Case Study 3, again both the maximum conversion and minimum time problems were solved. However, here safety and environmental constraints were taken into consideration. To tackle the safety constraint (path constraint), a new solution

approach was introduced, which was found advantages over the classical method. In Case Study 4, an exothermic semi-batch reactor was considered. Here, all types of optimisation problems were solved. The comparison on the results achieved from all modes of operations was made and the suitability of each type of operation under specified circumstances was discussed. Here, both the feed flow rate of one of the reactants and reactor temperature were optimised. In Case Study 5, a sensitivity analysis of the profit function (in maximum profit problem) in batch reactors was studied. How the prices of reactant, product, by-product, operating cost and reactor volume (capital cost) influence the overall profit were analysed. Here, the reactor volume has also been introduced as a design variable to be optimised. Finally, in Case Study 6, a maximum conversion problem with fixed time interval with a complex reaction scheme was carried out. The aim of this case study was to provide set points in terms of reactor temperature for on-line tracking of these set points by various types of controllers.

In Chapter 5, we implemented the off-line optimisation results obtained in Chapter 4 as set points to be tracked by various types of controllers. Three advanced control strategies were used; they were GMC-NN, neural controllers in Direct Inverse and IMBC control strategies. However, the DM control strategy with PI and PID controllers was also implemented for comparison with the other control strategies. The theory of all types of controllers implemented in this work was discussed. The control algorithm, merits and implementation procedure for each control strategy were briefly explained and addressed. Two case studies were presented. In Case Study 1, a consecutive exothermic batch reaction scheme with pre-heated reactants and in Case Study 2 a complex reaction scheme with both heating and cooling elements were dealt with. In Case Study 1, GMC-NN and PID controllers were designed to track a constant

set point. In Case Study 2 all the proposed controllers were designed to track both constant and dynamic set points. In both cases, the robustness tests were carried out and the results achieved were analysed and compared. Also, in this section the versatility in the use of neural network as on-line estimator and as controller was demonstrated.

In Chapter 6, steps to develop an integrated framework (software) called BATCH_REACT for modelling, simulation, optimisation and controller were discussed. With minimum information from the users on reaction schemes, e.g. series, parallel or complex reactions, reversible or irreversible reactions, kinetic information, the BATCH_REACT will generate model equations to describe the batch reactor operation (detailed or short-cut models), simulate the reactor, generate optimal operation policies off-line and design controllers to track the optimal set points. The work presented in Chapters 3-5 will be the major building blocks of the proposed software.

7.2 Suggestions for future work

"This is not the end. It is not even the beginning of the end. But it is the end of the beginning", Winston Churchill (Element of Chemical Reaction Engineering, pg. 805, Prentice-Hall).

Some future work arising naturally from this work include:

1. Methodologies for obtaining off-line optimal operating policies and for designing controller with general reaction schemes in conventional batch reactors are mainly considered here. Future work should apply the methods in real applications such as polymerisation, fermentation, etc. Methodologies

should also be fully explored for semi-batch reactors, especially on-line tracking of optimal control profiles.

2. Only three types of advanced of control strategies were applied and evaluated in this work. Since versatility of neural network has been proved, in the future, it will be implemented together with these new-advanced control techniques such as hybrid scheme with GMC, adaptive and also predictive techniques.
3. The proposed control strategies should be implemented and validated in real system.
4. The building blocks (as presented in Chapters 3-5) of the proposed software BATCH_REACT need to be coupled together. The software has to be tested using simulations and real life application.

References

Abel, O., Helbig, A., Marquardt, W., Zwick, H. and Daszkowski, T., 2000. Productivity optimization of an industrial semi-batch polymerization reactor under safety constraints. *Journal of Process Control*, 10, pp. 351-362.

Aris, R., 1960. On Denbigh's optimum temperature sequence. *Chemical Engineering Science*, 12, pp. 375-382.

Atkinson, K., 1993. *Elementary numerical analysis*. 2nd ed. Singapore: Wiley.

Aziz, N. and Mujtaba, I.M., 1998. Optimal control of batch reactors, *Symposium on Advances in Process Control 5*, University of Wales, Swansea, 2-3 September 1998. IChemE, pp 201-209.

Aziz, N. and Mujtaba, I.M., 2001a. Optimal control of semi-batch reactors. *In Proc of Computer-Aided Chemical Engineering*, 9 (ESCAPE-10). Eds. R. Gani and S.B. Jorgensen. Elsevier, pp. 609-614.

Aziz, N. and Mujtaba, I.M., 2001b. Optimal operation policies in batch reactors. *Chemical Engineering Journal*, accepted for publication.

Aziz, N., Hussain, M.A. and Mujtaba, I.M., 2000a. Optimal control of batch reactors using Generic Model Control (GMC) and neural network. *In Proc of Computer-Aided Chemical Engineering*, 8 (ESCAPE-9). Ed. S. Pierucci. Elsevier, pp. 175-180.

Aziz, N., Hussain, M.A. and Mujtaba, I.M., 2000b. Performance of different types of controllers in tracking optimal temperature profiles in batch reactors. *Computers Chem. Engng.* 24 (2-7), July 15, pp. 1069-1075.

Aziz, N., Hussain, M.A. and Mujtaba, I.M., 2001a. Set point tracking in batch reactors: use of PID and general model control with neural network techniques. *In Application of neural networks and other learning technologies in process engineering*. Eds. I.M.Mujtaba and M.A.Hussain. London: Imperial College Press, 2001. pp. 217-241.

Aziz, N., Hussain, M.A. and Mujtaba, I.M., 2001b. Optimal control of batch reactor: comparison of neural network based GMC and inverse model control approach. Accepted for 6th World Congress of Chemical Engineering, Melbourne, Australia, 23-27 September.

Banga, J.R., Irizarry-Rivera, R. and Sieder, W.D., 1998. Stochastic optimization for optimization for optimal and model-predictive control. *Computers Chem. Engng.*, **22** (4/5), pp. 603-612.

Barton, P.I., Allgor, R.J., Feehary, W.F. and Galan, S., 1998. Dynamic optimization in a discontinuous world. *Ind. Eng. Chem. Res.*, **37** (3), pp. 966-981.

Bequette, B.W., 1991. Nonlinear control of chemical process: A review. *Ind. Eng. Chem. Res.*, **30** (7), pp. 1391-1413.

Berber, R., 1996. Control of batch reactors: a review. *Trans IChemE.* **74** (A), January, pp. 3-20.

Bhat, J., Chidambaram, M. and Madhavan, K.P., 1990. Robust control of batch reactors. *Chem. Eng. Comm.*, **87**, pp. 195-204.

Biegler, L.T., 1984. Solution of dynamic optimization problems by successive quadratic programming and orthogonal collocation. *Computers Chem. Engng.*, **8** (3/4), pp. 243-248.

Bonvin, D., 1998. Optimal operation of batch reactors – a personal view. *Journal of Process Control*, **8** (5-6), pp. 355-368.

Bryson, A.E. and Ho, Y.C., 1975. *Applied optimal control: optimization, estimation and control*. Revised ed. Washington, DC: Wiley.

Calderon, Z., Espuna, A. and Puigjaner, L., 1998. Waste analysis and minimization in batch and semibatch reactor operation through dynamic simulation and neural networks. *Computers Chem. Engng.*, **22** (Suppl.), pp. S977-S980.

Carrasco, E.F. and Banga, J.R., 1997. Dynamic optimization of batch reactors using adaptive stochastic algorithms. *Ind. Eng. Chem. Res.*, **36** (6), pp. 2252-2261.

Chen, C.L., 1988. *A class of successive quadratic programming methods for flowsheet optimisation*. PhD thesis. University of London.

Cott, B.J. and Macchietto, S., 1989. Temperature control of exothermic batch reactors using Generic Model Control. *Ind. Eng. Chem. Res.*, **28** (8), pp. 1177-1184.

Cott, B.J., Durham, R.G., Lee, P.L. and Sullivan, G.R., 1989. Process model-based engineering. *Computers Chem. Engng.*, **13** (9), pp. 973-984.

Coughanowr, D.R., 1991. *Process systems analysis and control*. 2nd ed. New York: McGraw-Hill.

Crescitelli, S. and Nicoletti, B., 1973. Near optimal control of batch reactors. *Chemical Engineering Science*, **28**, pp. 463-471.

Cuthrell, J.E. and Biegler, L.T., 1989. Simultaneous optimization and solution methods for batch reactor control profiles. *Computers Chem. Engng.*, **13** (1/2), pp. 49-62.

Dadebo, S.A. and McAuley, K.B., 1995. Dynamic optimization of constrained chemical engineering problems using dynamic programming. *Computers Chem. Engng.*, **19** (5), pp. 513-525.

Denbigh, S.G., 1958. Optimum temperature sequences in reactors. *Chemical Engineering Science*, **8**, pp. 125-132.

Dirion, J.L., Cabassud, M., LeLann, M.V. and Casamatta, G., 1995. Design of a neural controller by inverse modelling. *Computers Chem. Engng.*, 19 (Suppl.), pp. S797-S802.

Dirion, J.L., Cabassud, M., LeLann, M.V. and Casamatta, G., 1996. Development of adaptive neural networks for flexible control of batch processes. *The Chemical Engineering Journal*, 63, pp. 65-77.

Dirion, J.L., Ettetdgui, B., Cabassud, M., LeLann, M.V., and Casamatta, G., 1996. Elaboration of neural networks system for semi-batch reactor temperature control: an experimental study. *Chemical Engineering and Processing*, 31, pp. 225-234.

Eaton, J.W. and Rawlings, J.B., 1990. Feedback control of chemical processes using on-line optimization techniques. *Computers Chem. Engng.*, 14 (4/5), pp. 469-479.

Edgar, T.F. 1998. Process Modeling and Control: A vision of the future. *Presented at Advances in Process Control 5 Symposium*. University of Wales, Swansea, 2-3 September 1998. IChemE.

Edgar, T.F. and Himmelblau, D.M., 1989. *Optimization of chemical processes*. New York: McGraw-Hill.

Fikar, M., Latifi, M.A., Fournier, F. and Creff, Y., 1998. Control vector parametrisation versus iterative dynamic programming in dynamic optimisation of a distillation column. *Computers Chem. Engng.*, 22 (Suppl.), pp. S625-S628.

Fisher, D.G., 1991, Process control: An overview and personal perspective. *Can. J. Chem. Eng.*, 69, pp. 5-26.

Fogler, H., 1992. *Elements of chemical reaction engineering*. 2nd ed. New Jersey: Prentice-Hall.

- Fournier, F., Latifi, M.A. and Valentin, G., 1999. Methodology of dynamic optimization and optimal control of batch electrochemical reactors. *Chemical Engineering Science*, **54**, pp. 2707-2714.
- Galvan, I.M. and Zaldivar, J.M., 1998. Application of recurrent neural network in batch reactors Part II: Nonlinear inverse and predictive control of the heat transfer fluid temperature. *Chemical Engineering and Processing*, **37**, pp. 149-161.
- Garcia, C.E. and Morari, M., 1982. Internal model control. *Ind. Eng. Chem. Processing Des. Dev.*, **21**, pp. 308-323.
- Garcia, V., Cabassud, M., LeLann, M.V., Pibouleau, L. and Casamatta, G., 1995. Constraint optimization for fine chemical productions in batch reactors. *The Chemical Engineering Journal*, **59**, pp. 229-241.
- Gentric, C., Pla, F., Latifi, M.A. and Corriou, J.P., 1999. Optimization and non-linear control of a batch emulsion polymerisation reactor. *Chemical Engineering Journal*, **75**, pp. 31-46.
- Goh, C.J. and Teo, K.L., 1988. Control parameterization: a unified approach to optimal control problems with general constraints. *Automatica*, **24**, pp 3-18.
- Grau, M.D., Nougues, J.M. and Puigjaner, L., 2000. Batch and semi-batch reactor performance for an exothermic reaction. *Chemical Engineering and Processing*, **39**, pp. 141-148.
- Gritsis, D., 1990. *The dynamic simulation and optimal control of systems described by index two differential-algebraic equations*. PhD thesis. University of London.
- Holland, C.D. and Anthony, R.G., 1979. *Fundamental of chemical reaction engineering*. New Jersey: Prentice-Hall.

- Hussain, M.A., 1996. *Inverse-model control strategies using neural networks: analysis, simulation and on-line implementation*. PhD thesis. University of London.
- Hussain, M.A., 1999. Review of the applications of neural networks in chemical process control - simulation and online implementation. *Artificial Intelligence in Engineering*, **13**, pp. 55-68.
- Hussain, M.A. and Kershenbaum, L.S., 2000. Implementation of an inverse-model-based control strategy using neural networks on a partially simulated exothermic reactor. *Trans IChemE*, **78** (A), pp. 299-311.
- Juba, M.R. and Hamer, J.W., 1986. Progress and challenges in batch process control. *In Proceedings of 3rd International Conference on Chemical Process Control, CPC III*, Amsterdam. Eds. by M.Morari and T.J.McAvoy. AIChE, CACHE Austin TX and Elsevier, pp. 139-183.
- Jutan, A. and Uppal, A., 1984. Combined feedforward-feedback servo control scheme for an exothermic batch reactor. *Ind. Eng. Chem. Process Des. Dev.*, **23** (3), pp. 597-602.
- Jutan, A., 1989, A nonlinear PI(D) controller. *Can. J. Chem. Eng.*, **67**, pp. 485-493.
- Kershenbaum, L.S. and Kittisupakorn, P., 1994. The use of a partially simulated exothermic (PARSEX) reactor for experimental testing of control algorithms. *Trans IChemE*, **72** (A), pp. 55-63.
- Kittisupakorn, P., Hussain, M.A. and Arpornwichanop, A., 2000. Temperature control of an exothermic batch reactor by model predictive control, *The 3rd Asian Control Conference*, Shanghai, China, 4-7 July 2000. pp 2506-2511.
- Ko, D.Y.C. and Stevens, W.F., 1971. Studies of singular solutions in dynamic optimization: II. Optimal singular design of a plug-flow tubular reactor. *AIChE Journal*, **17**, pp. 160-166.

Krothapally, M. and Palanki, S., 1997. A neural network strategy for batch process optimization. *Computers Chem. Engng.*, **21** (Suppl.), pp. S463-S468.

Laguerie, C. and Delmas, H., 1989. Chemical reactors. *In Process modeling and control in chemical engineering*. Ed. K.Najim. New York: Dekker, 1989. pp. 1-103.

Le Lann, M.V., Cabassud, M. and Casamatta, G., 1999. Modelling, optimization and control of batch chemical reactors in fine chemical production. *Annual Reviews in Control*, **23**, pp. 25-34.

Lee, P.L. and Sullivan, G.R., 1988. Generic model control. *Computers Chem. Engng.*, **12** (6), pp. 573-580.

Lee, P.L., Newell, R.B. and Sullivan, G.R., 1989, Generic model control – a case study. *Can. J. Chem. Eng.*, **67**, pp. 478-484.

Lee, T.H, Adams, G.E. and Gaines, W.M., 1968. *Computer process control: modeling and optimization*. New York: John Wiley.

Levien, K., 1992. Maximizing the product distribution in batch reactors: Reaction in parallel. *Chemical Engineering Science*, **47** (7), pp. 1751-1760.

Liptak, G., 1986. Controlling and optimizing chemical reactors. *Chemical Engineering*, May, pp. 69-81.

Loeblein, C., Perkins, J.D., Srinivasan, B. and Bonvin, D., 1997. Performance analysis of on-line batch optimization systems. *Computers Chem. Engng.*, **21** (Suppl.), pp. S867-S872.

Logsdon, J.S., 1990. *Efficient determination of optimal control profiles for differential algebraic systems*. PhD thesis. Carnegie Mellon University.

Logsdon, J.S. and Biegler, L.T., 1989. Accurate solution of differential-algebraic optimization problems. *Ind. Eng. Chem. Res.*, **28** (11), pp. 1628-1639.

- Logsdon, J.S. and Biegler, L.T., 1993. A relaxed reduced space SQP strategy for dynamic optimization problems. *Computers Chem. Engng.*, 17 (4), pp. 367-372.
- Luus, R., 1990. Application of dynamic programming to high-dimensional non-linear optimal control problems. *Int. J. Control*, 52, pp. 239-250.
- Luus, R., 1991. Effect of the choice of final time in optimal control of nonlinear systems. *Can. J. Chem. Eng.*, 69 (Feb), pp. 144-151.
- Luus, R., 1994. Optimal control of batch reactors by iterative dynamic. *Journal of Process Control*, 4 (4), pp. 218-226.
- Luus, R. and Hennessy, D., 1999. Optimization of fed-batch reactors by the Luus-Jaakola optimization procedure. *Ind. Eng. Chem. Res.*, 38 (5), pp. 1948-1955.
- Luus, R. and Okongwu, O.N., 1999. Towards practical optimal control of batch reactors. *Chemical Engineering Journal*, 75, pp. 1-9.
- Marroquin, G. and Luyben, W.L., 1973. Practical control studies of batch reactors using realistic mathematical models. *Chemical Engineering Science*, 28, pp. 993-1003.
- Martinez, E.C., 2000. Batch process modeling for optimization using reinforcement learning. *Computers Chem. Engng.*, 24 (2-7), pp. 1187-1193.
- Morison, K.R., 1984. *Optimal control of processes described by systems of differential-algebraic equations*. PhD thesis. University of London.
- Mujtaba, I.M. and Hussain, M.A., 1998. Optimal operation of dynamic processes under process-model mismatches: Application to batch distillation. *Computers Chem. Engng.*, 22 (Suppl.), pp. S621-S624.
- Mujtaba, I.M. and Macchietto, S., 1998. Holdup issues in batch distillation – binary mixtures. *Chemical Engineering Science*, 53 (14), pp. 2519-2530.

Murty, B.S.N., Gangiah, K. and Husain, A., 1980. Performance of various methods in computing optimal control policies. *Chemical Engineering Journal*, **19**, pp. 201-208.

Nikraves, M., Farrell, A.E. and Stanford, T.G., 1997. Dynamic neural network control for non-linear systems: optimal neural network structure and stability analysis. *Chemical Engineering Journal*, **68**, pp. 41-50.

Octave, L., 1999. *Chemical reaction engineering*. 3rd ed. New York: John Wiley.

Palanki, S., Kravaris, C. and Wang, H.Y., 1993. Synthesis of feedback laws for end-point optimization in batch process. *Chemical Engineering Science*, **48** (1), pp. 135-152.

Palanki, S., Kravaris, C. and Wang, H.Y., 1994. Optimal feedback control of batch reactors with a state inequality constraint and free terminal time. *Chemical Engineering Science*, **49** (1), pp. 85-97.

Perry, R.H. and Green, D., 1984. *Perry's chemical engineers' handbook*. 6th ed. New York: McGraw-Hill.

Peters, M.S. and Timmerhaus, K.D., 1991. *Plant design and economics for chemical engineers*. New York: McGraw-Hill.

Rama, N.R. and Carol, A.Z., 1994. *Fortran 77 with 90: applications for scientist and engineers*. 2nd ed. Minneapolis: West Pub.

Ray, W.H., 1981. *Advanced process control*. New York: McGraw-Hill, pp. 84-118.

Reddy, K.V. and Husain, A., 1981, Computation of optimal control policy with singular subarc. *Can. J. Chem. Eng.*, **59**, pp. 557-559.

Reklaitis, G.V., Ravindran, A. and Ragsdell, K.M., 1983. *Engineering Optimization methods and applications*. New York: John Wiley.

- Rippin, D.W.T., 1983. Simulation of single and multiproduct batch chemical plants for optimal design and operation. *Computers Chem. Engng.*, **7** (3), pp. 137-156.
- Rodriguez-Toral, M.A., Morton, W. and Mitchell, D.R., 2000. Using new packages for modeling, equation oriented simulation and optimization of a cogeneration plant. *Computers Chem. Engng.*, **24**, pp. 2667-2685.
- Ruppen, D., Benthack, C. and Bonvin, D., 1995. Optimization of batch reactor operation under parametric uncertainty – computational aspects. *Journal of Process Control*, **5** (4), pp. 235-240.
- Rusnak, A., Fikar, M., Latifi, M.A. and Meszaros, A. 2001. Receding horizon iterative dynamic programming with discrete time models. *Computers Chem. Engng.*, **25**, pp. 161-167.
- Sargent, R.W.H and Sullivan, G.R., 1977. The development of an efficient optimal control package. *In Proc. IFIP Conf. Optim. Tech.*, **8**, part 2.
- Seborg, D.E., Edgar, T.F. and Mellichamp, D.A., 1989. *Process dynamics and control*. New York: John Wiley.
- Sharif, M., 1999. *Design of integrated batch processes*. PhD thesis. University of London.
- Shukla, P.K. and Pushpavanam, S., 1994. Parametric sensitivity, runaway, and safety in batch reactors: experiments and models. *Ind. Eng. Chem. Res.*, **33** (12), pp. 3202-3208.
- Siebenthal, C.D. and Aris, R., 1964. Studies in optimization – VII The application of Pontryagin's methods to the control of batch and tubular reactors. *Chemical Engineering Science*, **19**, pp. 747-761.
- Slotine, J.J.E, 1991. *Applied nonlinear control*. London: Prentice-Hall.

Soroush, M. and Kravaris, C., 1993a. Optimal design and operation of batch reactors. 1. Theoretical Framework. *Ind. Eng. Chem. Res.*, **32** (5), pp. 866-881.

Soroush, M. and Kravaris, C., 1993b. Optimal design and operation of batch reactors. 1. A case study. *Ind. Eng. Chem. Res.*, **32** (5), pp. 882-893.

Terwiesch, P., Agarwal, M. and Rippin, D.W.T., 1994. Batch unit optimization with imperfect modelling: a survey. *Journal of Process Control*, **4** (4), pp. 239-258.

Thomas, E.M., 1995. *Process control: designing processes and control systems for dynamic performance*. New York: McGraw-Hill.

Toulouse, C., Cezerac, J., Cabassud, M., LeLann, M.V. and Casamatta, G., 1996. Optimisation and scale-up of batch chemical reactors: impact of safety constraints. *Chemical Engineering Science*, **51** (10), pp. 2243-2252.

Vasantharajan, S. and Biegler, L.T., 1988. Large-scale decomposition for successive quadratic programming. *Computers Chem. Engng.*, **12** (11), pp. 1087-1101.

Vassiliadis, V., 1993. *Computational solution of dynamic optimization problems with general differential-algebraic constraints*. PhD thesis. University of London.

Vassiliadis, V.S., Sargent, R.W. and Pantelides, C.C., 1994a. Solution of a class of multistage dynamic optimization problems. 1. Problems without path constraints. *Ind. Eng. Chem. Res.*, **33** (9), pp. 2111-2122.

Vassiliadis, V.S., Sargent, R.W. and Pantelides, C.C., 1994b. Solution of a class of multistage dynamic optimization problems. 2. Problems with path constraints. *Ind. Eng. Chem. Res.*, **33** (9), pp. 2123-2133.

Wen, C.S. and Yen, T.F. 1977. Optimization of oil shale pyrolysis. *Chemical Engineering Science*, **32**, pp. 346-349.

William, W.L., 1990. *Process modelling, simulation and control for chemical engineers*. 2nd ed. Singapore: McGraw-Hill.

Zaldivar, J.M., Hernandez, H. and Panetsos, F., 1992. Control of batch reactors using neural networks. *Chemical Engineering and Processing*, 31, pp. 173-180.

Appendices

Appendix A

Successive Quadratic Programming (SQP) method has been regarded as one of the most effective methods for nonlinearly constrained optimisation problems in terms of function evaluations (Chen, 1988). The method was motivated by the application of Newton-type methods to the first order optimality conditions of the original problem.

Basic SQP Algorithm

1. Given initial conditions.
2. Form a quadratic programming sub problem QP B.I and solve it for the search directions and the estimated Lagrangian multipliers.

$$\begin{array}{ll} \min & \nabla_x f(x_k)^T d_{xk} + \frac{1}{2} d_{xk}^T B_k d_{xk} \\ \text{QP B.I} \quad \text{s.t.} & h(x_k) + \nabla_x h(x_k)^T d_{xk} = 0 \\ & g(x_k) + \nabla_x g(x_k)^T d_{xk} \geq 0 \end{array}$$

where

$\nabla_x f(x)$ is the gradient of the objective function

$\nabla_x h(x)$ is the gradient of equality constraints

$\nabla_x g(x)$ is the gradient of inequality constraints

B_k is an approximation to the Hessian of the Lagrangian function at the k^{th} iteration

d_x is the search direction

3. Check the convergence criteria. If this criterion is satisfied, then exit. Otherwise the algorithm continues.
4. Employ a procedure to ensure convergence and obtain a new estimate of the solution.
5. Updated the Hessian of the Lagrangian function.
6. Go to step 2.

Assumptions

1. The objective function $f(x)$ and constraints $h(x)$ and $g(x)$ are continuously differentiable in the range of interest.
2. The constraints are consistent, i.e. nonempty set of feasible points exists within these constraints.
3. A local solution of the original problems exists.
4. The gradient of the active constraints are linearly independent in the range of interest. This assumption is often referred to as a constraint qualification.

A Complete SQP algorithm

Begin *<Initialisation Section>*

Given an initial estimate x_0 .

Given a tolerance ϵ .

Given maximum number of iterations allowed \max_f .

Given maximum feasibility steps allowed \max_s .

Choose a merit function.

Choose either a line search method or a trust region method.

Choose an initial Hessian.

Set $k = 1$

End

Repeat

Set $k = k + 1$

Begin *<Construct and solve QP>*

Solve QP

End

Begin *<Check convergence criteria>*

If inequality equation is satisfied **Then** Exit (Solution found)

End

While the line search method is requested **Do**

Begin *<Perform line search>*

End

While the trust region method is requested **Do**

Begin *<Perform trust region>*

End

Begin *<Update the Hessian>*

End

Until $k \geq \max_f$.

Complete Successive Reduced Quadratic Programming SRQP algorithm using in the optimiser code**Begin** *<Initialisation Section>*Given an initial estimate x_0 .Given a tolerance ϵ .Given maximum number of iterations allowed \max_f .Given maximum feasibility steps allowed \max_s .

Choose design variables manually or automatically.

Choose a reduced model

Choose a merit function.

Choose either a line search method or a trust region method.

Choose an initial Hessian.

Set $k = 1$ **End****Repeat**Set $k = k + 1$ **Begin** *<Construct and solve reduced QP>*

Solve reduced model

End**Begin** *<Check convergence criteria>***If** inequality equation is satisfied **Then** Exit (Solution found)**End**

While the line search method is requested **Do**

Begin *<Perform line search>*

End

Begin *<Update the Hessian>*

End

Until $k \geq \max_f$.

Appendix B

Backpropagation Algorithm

Basically a gradient descent, parallelly distributed optimisation technique to minimise the error between network and target output by manipulation of the connection weights (parameters) of network. Figure below shows the main steps of back propagation algorithm (for one node and one input/output pattern)

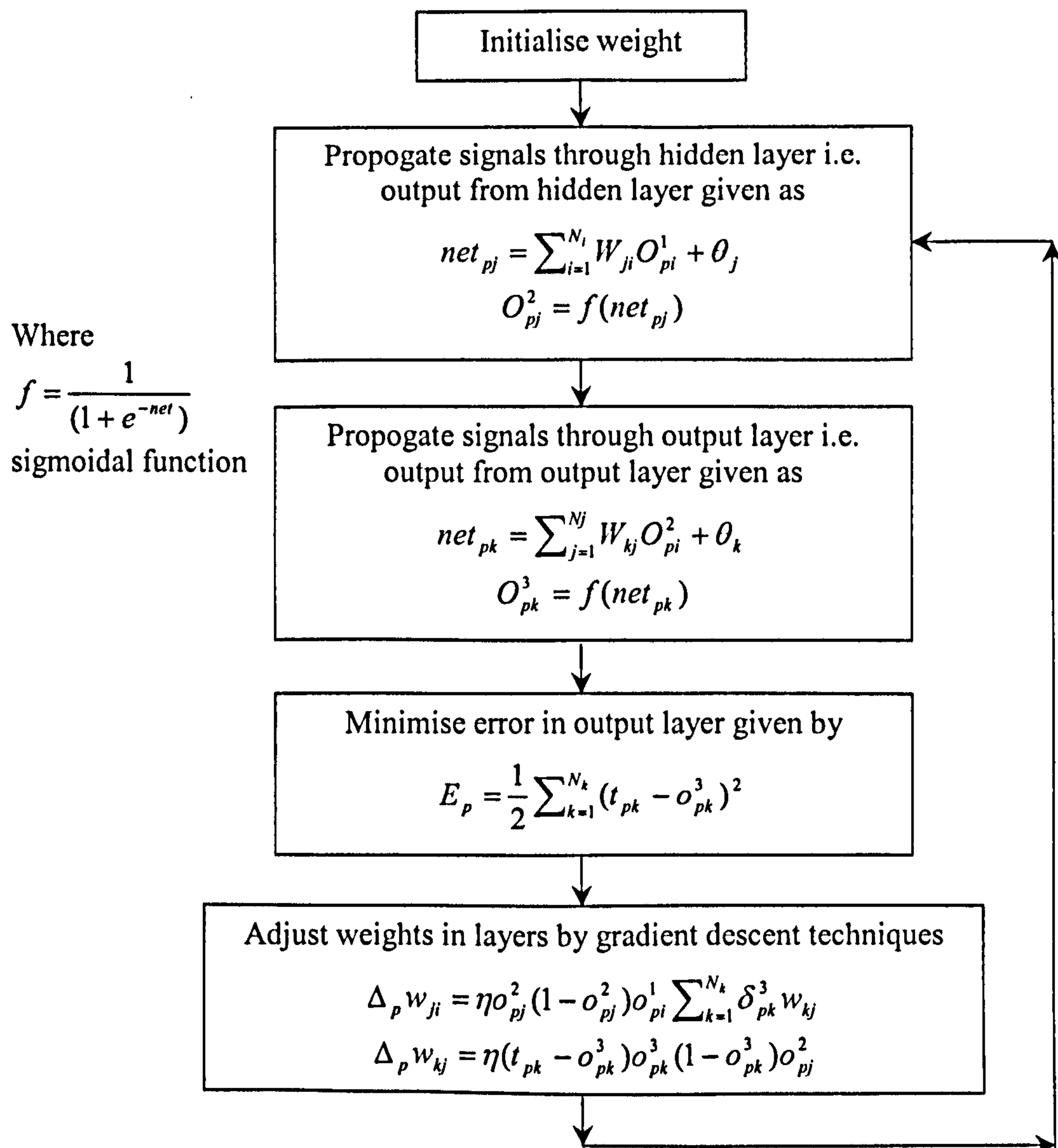


Figure B: Main steps of the main steps of back propagation algorithm (for one node and one input/output pattern)