Integrated Design of Responsive Chemical Manufacturing Facilities

by

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BDL.
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UNIV.

To my family

mayi sarvāṇi karmāṇi

sannyasyādhyātma – cetasā

nirāśīr nirmamo bhūtvā

yudhyasva vigata-jvaraḥ

Therefore, O Arjuna, surrendering all your works unto Me, with full knowledge of Me, without desires for profit, with no claims to proprietorship, and free from lethargy, fight.

Lord Krishna instructs Arjuna on the battlefield of Kurukshetra,
Text 30, Chapter 3, Bhagavad Gita.

Abstract

This thesis considers an integrated approach to designing responsive chemical manufacturing facilities in the presence of uncertainty.

The integrated design problem is discussed in the context of a responsive plant and literature in this area is reviewed. It is concluded that any proposed procedure should explicitly consider the dynamic operation of the process at the design stage. Potential methods for plant design and operation are reviewed and a combination of heuristic process/control structure selection and dynamic optimisation is deemed the most appropriate. A method for ensuring feasible operation over a dynamic operating envelope in the presence of uncertainty is suggested. A procedure and framework for the integrated design of responsive plants is then presented. The rationale for the procedure is to develop plant designs which operate feasibly in the face of frequent and potentially large changes in product quality and demand. The proposed design framework is demonstrated by application to two representative types of problem involving continuous multiproduct processes and a responsive reactor network capable of dealing with intermittent product demand. Finally, scope for future work is identified, both in terms of new techniques and potential case study examples.

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Chapter 1

Introduction

1.1. Background

Traditionally, in the design of chemical processes, control systems were added after the process design was largely complete, or later by retrofit to a plant once built and operated for a while. Overall system performance was then assessed during commissioning or under production conditions. Although adequately operable and economic plants usually resulted, in some instances the dynamic characteristics led to severe operating problems and significant economic penalties. A well documented case can be found in Anderson (1966), where large oscillations rendered the system inoperable when the plant was turned up to its full production rate. The root of the problem lay in the hot reactor effluent being used in a heat exchange system to vaporise and superheat the reactor feed.

Current trends in design methods, both in academia and in industry, have thus leant towards a more integrated approach, attempting to understand and exploit the appropriate synergy between the process equipment and control systems of chemical plants. Futhermore, this work intends to explore the possibility that an integration of process design and control may be employed to solve a new class of problem, making significant differences to current industrial practice.

1.2. Definition

"Responsiveness" is one of many analagous terms that have appeared in the vast fields of design and control and as such its meaning may often be open to interpretation. However for the purposes of this work it is defined thus: a responsive plant must be capable of responding **instantly** to frequent and major changes in customer demand for product quantity and/or quality.

1.3. Motivation

Conventional chemical processes tend to be oversized continuous operations that usually require a large inventory of feedstock(s) and generate a stockpile of product(s). If multiple product grades are necessary, transition times needed to switch between the plant operating points are typically in the region of hours to days, accompanied by the generation of significant quantities of off-spec material. In recent times the chemical industry has had to sell its products in a saturated and extremely competitive global market. This situation has demanded a strong need for the production of a broad range of product types and grades at time-varying capacities. As a result of these changes, industries are beginning to look at competitive and sustainable production **on demand**, and tight operating and product quality constraints. A responsive plant could, for example, replace the feedstock inventory by manufacturing reactant(s) for a conventional process *in situ*, thus reducing its environmental impact. The responsive chemical manufacturing facility is therefore defined in this work as being capable of delivering on-spec product at the required rate, on demand; i.e. the customer should be in full control of both the quantity and the quality of product being received.

For chemical plants to respond to rapid production changes, they must be extremely controllable and able to swing throughout a large operating envelope efficiently. Controllability is defined by Morari and Perkins (1995) as:

"The best dynamic performance (setpoint following and disturbance rejection) achievable for a system under closed loop control."

This implies that the control scheme for such plants plays an important role in its responsive nature. However, Ziegler and Nichols (1943) reported that process controllability is not only dependent upon the control scheme but also upon the process itself. Indeed Morari (1983a) identifies characteristics of the *process* which limit the achievable process control. The key to designing responsive chemical processes must therefore lie in tackling control issues as early as possible in any synthesis procedure, i.e. in the integration of design and control. If one considers synthesis and control at the same stage, the dynamics of the system may often be exploited in producing better designs. Soon current operation (centralised production, distribution and localised product storage) may not be able to respond quickly enough to the increasingly unpredictable customer demand profiles of the future. The contribution this work hopes to make is to develop a set of procedures for designing a restricted class of plant that must respond instantly to uncertain/intermittent demand.

1.4. Aims

The aims of this work as as follows:

1. To develop integrated **design procedures** for responsive chemical manufacturing facilities based upon an iterative, decomposed, two stage strategy, originally proposed by Walsh and Perkins (1996). The first stage determines a candidate structure capable of fulfilling the instant response criterion through heuristics, bounding properties and screening tools. The second stage determines the optimal design and operation for this candidate

using dynamic optimisation. Hence the following issues must be addressed as part of this project:

- candidate process *and* control structure selection using process engineering knowledge, from a set of design alternatives, all of which are able to respond instantly to customer demand. The limited class of problem being tackled by the proposed design procedures will require a single product (possibly with multiple grades according to customer requirements, e.g. multigrade polymer production), deliverable on-spec and on demand to the customer. The demand load(s) will be fixed **a priori** and both continuous and discrete control actions will be utilised. Issues such as reliability, safety and the environment must have their role in a "complete" set of design procedures, but are deemed outside the scope of this work.
- investigation of cyclic plant operation in the optimisation formulation, validating over multiple cycles, and ensuring feasibility under uncertainty by making use of Operational Envelopes.
- identification and development of mathematical tools within the dynamic simulation and optimisation environments used in the design and validation step.

2. To undertake representative **case studies** with the following objectives:

- demonstrate applicability and effectiveness of the proposed design methods
- provide tests for the tools used in these procedures for verification and validation purposes by comparison to published examples
- illustrate realistic scenarios where "responsiveness" may make significant improvements to plant performance
- give insights into how the design procedures may be improved.

1.5. Contributions of the work

Integrated design techniques were devised in order to solve the class of problem that combines conceptual process design, control structure selection and design under uncertainty. A major contribution this thesis makes is to provide a clear and unambiguous problem formulation for responsive plant design and operation. Other contributions improve existing integrated design methods by:

- adding heuristic control structure selection to the process design stage (section 5.5)
- refining the dynamic optimization techniques used to determine plant operation. The key contribution was in dealing with cyclic constraints by specifying ranges rather than points (section 4.2.4)
- incorporating Operational Envelopes to ensure feasibility in the presence of uncertainty (section 3.3.2).

Finally, the topic of responsiveness is introduced, defined and discussed with respect to the results obtained for the case studies and directions for future development outlined in the final chapter.

1.6. Outline of the thesis

- Chapter 2: literature review which gives an overview of various procedures used in producing conceptual flowsheets for chemical plants, and assessing their operability.
- Chapter 3: potential design procedures and associated tools are discussed, namely dynamic optimisation and Operating Envelopes. A combination of heuristics and optimisation is proposed.

- Chapter 4: designs of multiproduct processes which must manufacture several product grades in a repeated sequence are considered. The first system under consideration comprises purely of a reactor and four product tanks. The second is an example commonly found in control structure selection literature, that of a reactor/separator system which must produce two product grades.

- Chapter 5: application of the full decomposed two-stage design procedure to synthesise an instant response reactor network. An "automatic startup and shutdown" product throughput profile is used, reflecting the intermittent nature of the customer demand.

- Chapter 6: a summary of the thesis is made and conclusions are drawn. Scope for future work is identified, both in terms of new techniques and potential case study examples.

Chapter 2

Literature review

Historically, conceptual design of chemical processes has been based almost entirely upon steady state analysis; the dynamic behaviour/operability aspects of these processes have not been incorporated into design procedures until much later. Indeed, many authors have commented that the cause of most control problems is poor design practice that is based solely on steady state operation. Luyben et al. (1999) comment upon this staged approach resulting in missed opportunities due to the close connection between process design and controllability not being exploited:

"How a process is designed fundamentally determines its inherent controllability, which means qualitatively how well the process rejects disturbances and how easily it moves from one operating condition to another. In an ideal project system, dynamics and control strategies would be considered during the process synthesis and design activities."

In view of these concerns, both 'traditional' methods and those that taken a more 'integrated' approach are discussed. This review commences with a description of the **sequential** procedures conventionally employed by engineers when

designing a chemical plant: steady state process sythesis, dynamic operability studies, and control structure selection. It continues by covering an alternative series of works that have instead considered process dynamics directly at the synthesis stage. This developing methodology, labelled integrated design, deals with operability issues together with economic ones: design and synthesis decisions are made **simultaneously** within a common framework.

2.1. Traditional design of chemical processes

The traditional, sequential approach given below was taken from Hashimoto and Zafiriou (1995).

Synthesis

- Assume steady state operation of a continuous plant
- Formulate as a static optimisation problem with economics as the performance index
  - if the flowsheet has already been decided, parametric optimisation is used to accomplish the process design
  - if the flowsheet is not yet present, a combinatorial optimisation problem is solved
- The process flow diagram (PFD) may now be obtained
- Design a control system that can ensure steady state operation

Operability

- Consider unsteady state operation, e.g. startup/shutdown

- Add additional equipment such as pumps, tanks, pipes, control system ensuring unsteady state operation to the PFD: the piping and instrumentation diagram (PID) is now obtained

- Consider uncertainty
  - within the process, e.g. reaction rates, catalytic activity, heat transfer coefficients, equilibrium relationships, etc.
  - outside the process, e.g. product specifications, feed stocks, utilities, etc.

- Consider disturbances entering the process

As one proceeds from the steady state process design towards the design of the control systems, one needs more and more information on the process dynamics. Unfortunately, steady state synthesis and dynamic operability studies have been somewhat segregated. Walsh and Perkins (1996) comment that a process may be designed by one group of people, while another design group is handed the process flowsheet and required to add on a control scheme to ensure that it operates successfully.

2.1.1. Synthesis

Synthesis (which literally means 'combining') traditionally comprises two separate tasks: determining the type and topology of the various unit operations needed to achieve pre-defined plant objectives at some optimal steady state, and then selecting an appropriate control system to ensure that the plant remains at this desired steady state. Two distinct ways of solving the synthesis problem have been developed:

- Hierarchical decomposition and the use of heuristics
- Mathematical programming and the use of mixed integer optimization

2.1.1.1. Process synthesis

Conceptual process design, or process synthesis, is concerned with the discovery of an optimal process flowsheet to perform a given task, such as the transformation of raw materials into products. A precise definition or description of the synthesis activity is somewhat difficult to formulate. One of the most widely used descriptions is one made by Grossmann (1990):

"Given are specifications of inputs (that typically correspond to raw materials) and outputs (that correspond to desired products that are to be produced). The problem then consists of integrating a process flowsheet that will convert the inputs into desired outputs so as to meet desired specifications while optimising a given objective or goal function."

Despite its generality, this definition is already somewhat restrictive as it assumes that the inputs and outputs are fixed, which may not always be the case, e.g. in the presence of uncertainty. Another difficulty in process synthesis is that, due to the large number of alternatives, the problem tends to become combinatorially explosive:

"The activity of process synthesis may be thought of as generating design alternatives and selecting the better ones based on incomplete information."

Westerberg (1989)

Douglas (1988) suggests that there are typically between 10^4 and 10^9 alternative ways to achieve the same goal for a typical chemical processing problem. Understandably, the key to this enormous synthesis challenge is the use of systematic procedures for generating and quickly screening many process alternatives.

Reviews on the developments in the area of process synthesis can be found in Hendry et al. (1973), Hlavacek (1978), Nishida et al. (1981), Westerberg (1980; 1989), Gundersen (1991) and Grossmann and Daichendt (1996).

Hierarchical Approaches The problem is decomposed in order to limit the number of design alternatives being generated at any one time. As one rises through these levels of decomposition, more detail is added to the problem description. Heuristics, based on engineering experience, guide the search direction. The application of such heuristics to perform process synthesis essentially relies upon intuition, engineering knowledge and physical principles. The Douglas hierarchy (Douglas and Stephanopoulos, 1995), shown in table 2.1, is an example of this type of approach.

The *economic potential* of candidate flowsheets is examined after the completion of each decision stage, and unpromising flowsheet alternatives are eliminated. This hopefully results in most flowsheets being rejected without the need for their detailed design. Eventually, a small number of *near-optimal* flowsheets which warrant a full design are identified. It is important to note that the Douglas approach must be carried out iteratively to produce good flowsheets, and is limited to the class of continuous plants that handle only gases and liquids. Some work was later carried out which attempted to cover solids (see end of this section).

Linnhoff et al. (1982) represent graphically their decomposition approach to the process synthesis procedure by means of an "onion" diagram (see figure 2.1). Here, the steps of the procedure are the design of the reactor subsystem followed by the separation system and finally, the heat exchangers and other utilities.

Although heuristic-driven approaches often lead to good designs, they cannot *rigorously* produce an optimal design, and in some cases result in ones that are

Level	*Title*	*Description*
0	Input information	Collect data on chemistry to be used, feed-stocks available, product specifications (amount, purity), economic constraints (e.g. maximum product unit cost, minimum ROI, maximum allocatable capital) and prevailing regulations (environmental, health, safety).
1	Number of plants	For multistep reactions (Douglas, 1990)
2	Input-output structure	Specify the feed, exit and recycle streams for each 'plant', specify the interconnections between plants for multistep reaction processes and estimate the annualised capital and operating costs associated with the feed and exit streams.
3	Recycle structure	For each 'plant' specify the reactor recycle streams and flows, the type of reactor(s), as well as the reactor size and cost, the gas recycle compressor's and feed dissolver's (for solid raw materials) sizes and cost, if any.
4	Separation system (Douglas, 1992b)	4a: General separation structure. 4b: Vapour recovery system. 4c: Solid recovery system. 4d: Liquid separation system(s). 4e: Gas separation system. 4f: Solid separation system. 4g: Combine the separation systems for multiple plants.
5	Energy integration	Specify a heat exchanger network to accomplish heat and power integration.
6	Process alternatives	Create and evaluate alternative designs

Table 2.1: The Douglas hierarchy

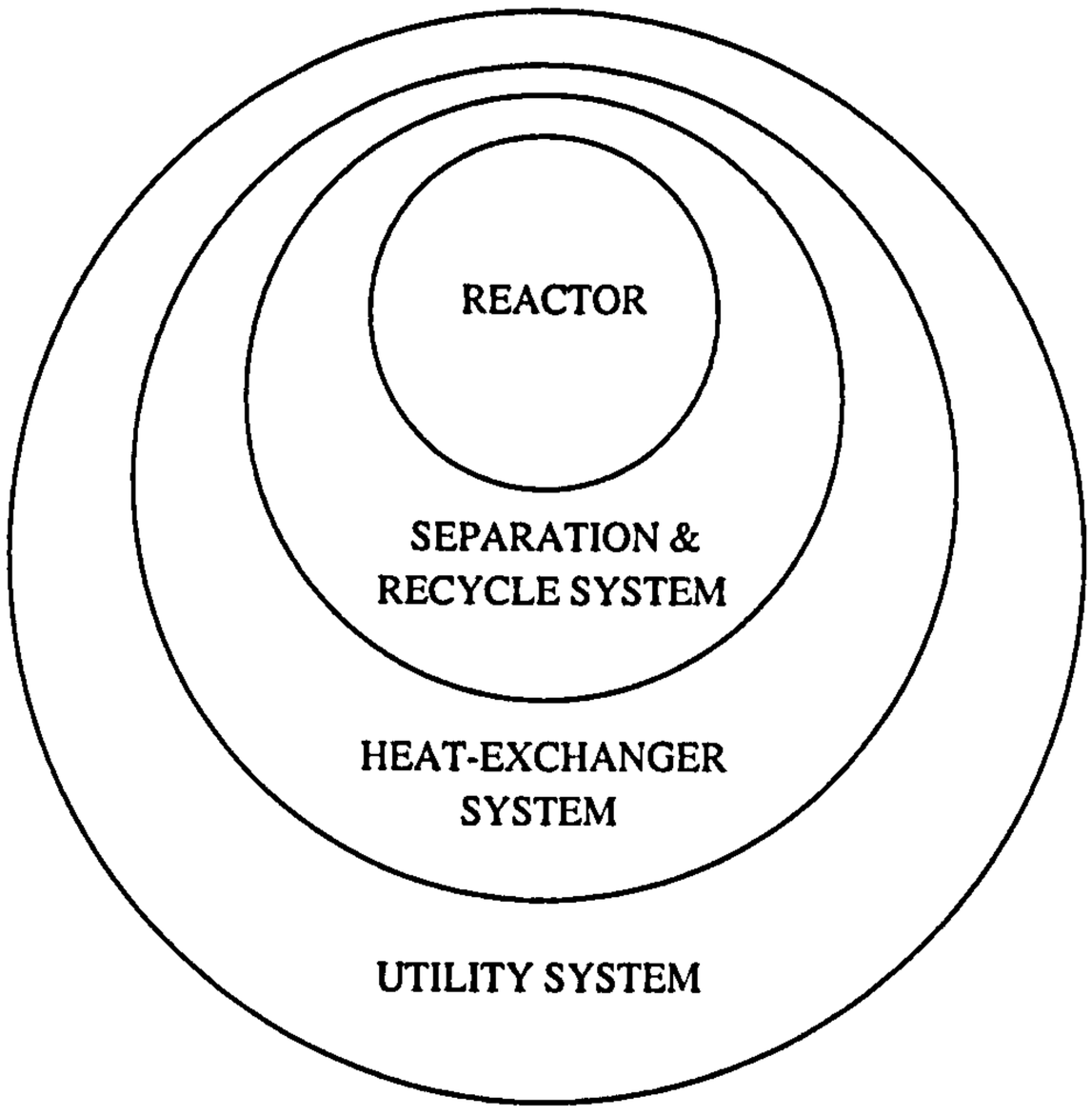


Figure 2.1: Onion diagram representation of the Process Synthesis procedure

fallible (see Papoulias and Grossmann, 1983; Colmenares and Seider, 1987; Fonyo and Mizsey, 1990). Furthermore, due to the *sequential* nature of the flowsheet synthesis, interactions among the design variables at the various decision levels may not be properly accounted for, as it is necessary to solve for them *simultaneously.* For instance, Duran and Grossmann (1986) and later Lang et al. (1988) have shown that simultaneous optimisation and heat integration of flowsheets generally produces significant improvements in profit.

Despite these shortcomings, hierarchical decomposition provides a useful approach for generating an initial flowsheet and alternatives (i.e. a base-case design and superstructure), whilst the use of heuristics provides a low-effort means by which these alternatives can be screened. This framework for decomposing the synthesis problem into a hierarchy of detailed and aggregated models is then simpler to carry out, compared to optimising the entire flowsheet, while still reflecting the presence of downstream tasks. On-going changes may also be incorporated

into the problem definition, thus fully exploiting the experience of the design engineer.

Some of the areas in which research has been undertaken in order to expand the knowledge base are:

- Synthesis of processes containing solid phases (Douglas and Ng, 1988; Ng, 1991; Rajagopal et al. 1991; Dye et al. 1995).
- Synthesis of plants containing multiple reaction systems (Douglas, 1990; Schembecker et al. 1995).
- The interactions between decisions made at different levels of the hierarchical design (Fonyo and Mizsey, 1988; Mizsey and Fonyo, 1990).
- Design of biochemical processes (Siletti and Stephanopoulos, 1992).
- Design of batch processess (Linninger et al. 1999).

Later work in this area by Douglas and Stephanopoulos (1995) is based around the application of many of the ideas used in the hierarchical design framework within a computer based system.

Mathematical Programming Approaches Process synthesis involves both continuous decisions (e.g. operating conditions) and discrete decisions (e.g. flow-sheet topology). Mathematically this can give rise to a *Mixed Integer Non-Linear Programming* (MINLP) formulation of the general form:

$$\begin{aligned} & \min_{y,x} \Phi(y,x) \\ s.t. \quad & h(y,x) = 0 \\ & g(y,x) \leq 0 \\ & y \in \{0,1\}^m, x \in \Re^n \end{aligned}$$

where y is the vector of 0-1 variables that typically denote the potential existence of units and the connections between them, while x represents the vector of continuous variables. $\Phi(y,x)$ is the objective/goal function, most often corresponding to some form of economic performance indicator. The equality constraints $h(.)$ mostly arise from steady state mass and energy balances around process units whereas the inequality constraints $g(.)$ typically represent operating constraints and design specifications. The solution of the resulting problem yields the optimal design.

A single pass solution to the MINLP would enable structural and continuous design decisions to be optimised simultaneously, and hence may offer potentially superior solutions (when compared to those produced by hierarchical methods). It must be noted however that as large MINLP problems are prone to combinatorial explosion, employing these algorithmic methods tends to be computationally intensive and could potentially limit the design engineer's function in an on-going synthesis procedure. Extensive reviews on the methods used to solve MINLPs and research carried out using them may be found in Grossmann (1985; 1990), Floquet et al. (1988), Grossmann et al. (1987), Floudas and Grossmann (1995) and Grossmann and Daichendt (1996).

Having noted little progress in automated design techniques for complete processes based on mathematical programming, Smith (1996) proposes a new methodology for the optimal design of continuous processes operating at steady state. He shows that the synthesis problem could be formulated mathematically in a completely automatic fashion from data on:

- candidate raw materials
- required end products
- available processing technology.

The solution to the resulting problem not only involves selection and connectivity of the unit operations present in the flowsheet, but also *simultaneously* considers their detailed design. The key to this new methodology was the use of detailed "multipurpose" models to characterise the behaviour of the unit operations considered for inclusion in the optimal flowsheet. Not only does this ensure the production of physically realisable designs, it also has the effect of reducing the problems of combinatorial complexity often associated with the activity of process design. To overcome the non-convexity present in this mathematical representation of the process design problem, global optimisation techniques were employed. However, despite the development of a novel algorithm aiding the generation of convex under- and over-estimators used in such techniques, only relatively simple process design problems (e.g. HENs and pump configurations) were optimised. Although larger, more realistic problems could, in principle, be dealt with using the proposed design methodology, they fall outside the practical capabilities of currently available global optimisation techniques.

2.1.2. Operability

Any designed process and its corresponding control system should be capable of overcoming uncertainties and disturbances and ensure sound operation of the process. In order to realise such a system, the concepts of flexibility, operability, and controllability have been introduced. Figure 2.2 attempts to show their relationships in a chart that positions different elements of the design task (namely static design, synthesis, parameter tuning and control structure selection) within a plot of the features that a design tool requires to solve these problems: parametric/structural decisions (shown on the horizontal axis) and steady/dynamic behaviour (vertical axis).

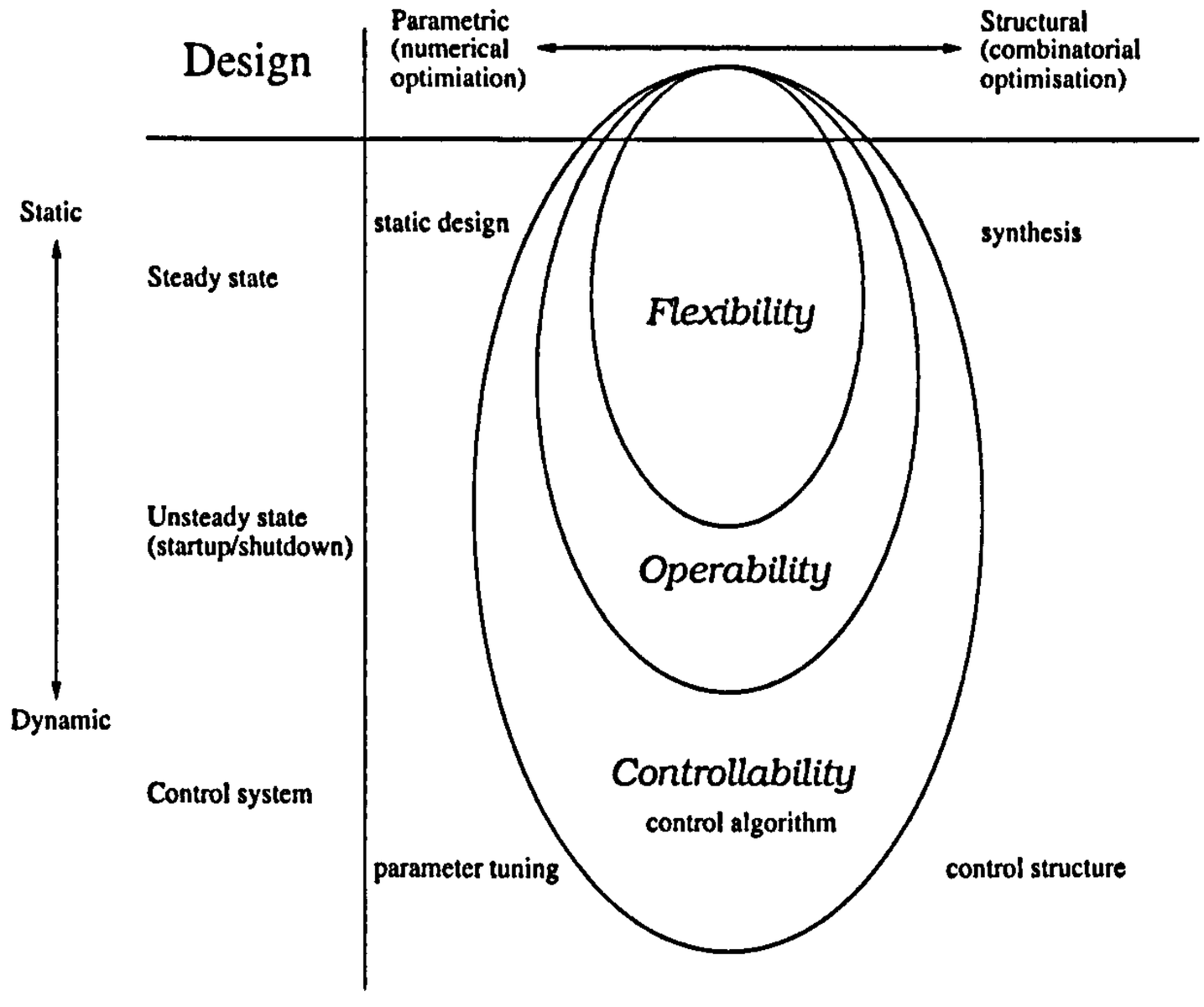


Figure 2.2: Relationships between flexibility, operability and controllability

2.1.2.1. Flexibility

"The capability that a design has of having feasible steady state operation for a range of uncertain conditions."

Biegler et al. (1997)

A successful process design must be able to operate to specification in the presence of uncertainty and variability, e.g. feed specification changes, measurement errors, process parameter changes. Design margins based upon worst/best case scenarios have traditionally been incorporated, thus creating the desired operating envelope able to deal with this variability, e.g. oversizing of distillation columns. Note that although Biegler et al. define flexibility earlier as a steady state concept, some authors, e.g. Dimitriadis and Pistikopoulos (1995) extend it to dynamic systems where the uncertainty space changes with time. It is however

also worth noting that the heat exchanger systems considered by Dimitriadis and Pistikopoulos typically have very slow dynamics.

Analysis of flexibility There are two basic analysis problems for flexibility: whether a design is feasible for a fixed range of uncertainty (the test problem) and a metric to actually quantify flexibility (the index problem).

The feasibility of operation of a design d operating at a given value of the uncertain parameters θ is determined by establishing whether by proper adjustment of the control variables z each inequality $f_j(d, z, \theta)$, $j \in J$ is less than or equal to zero. From this definition of feasibility, and noting that θ is bounded within some region T, $\theta \in T = \{\theta \mid \theta^N - \Delta\theta^- \leq \theta \leq \theta^N + \Delta\theta^+\}$, where θ^N is a nominal parameter value and $\Delta\theta^-$, $\Delta\theta^+$ are the expected deviations, Halemane and Grossmann (1983) give the general mathematical formulation of the flexibility test measure χ:

$$\chi(d) = \max_{\theta \in T} \min_{z} \max_{j \in J} f_j(d, z, \theta)$$

If $\chi(d) \leq 0$, feasible operation can be attained over the entire uncertain parameter range T. If $\chi(d) \geq 0$, feasible operation cannot be acheived, at least for part of this range.

The flexibility test problem only determines whether a design does or does not have the flexibility to operate over the specifies parameter range T. A flexibility index would *quantify* how much flexibility can actually be achieved in the given design. Initially a *variable* parameter range is defined:

$$T(\delta) = \{\theta \mid \theta^N - \delta\Delta\theta^- \leq \theta \leq \theta^N + \delta\Delta\theta^+\}$$

where δ is a non-negative scalar variable corresponding to the scaled deviation of any of the expected uncertain parameter deviations, $\Delta\theta^-$, $\Delta\theta^+$.

- $\delta = 1$: $T(1) = T$, i.e. $T(\delta)$ becomes identical to the original specified parameter range T.
- $\delta < 1$: $T(\delta) \subset T$.
- $\delta > 1$: $T(\delta) \supset T$.

Swaney and Grossmann (1985) define the flexibility index, F, as the largest value of δ such that the inequalities $f_j(d, z, \theta)$, $j \in J$ hold over the parameter range $T(F)$ (i.e. $\chi(d) \leq 0$ for $T(F)$):

$$\begin{aligned} F &= max\ \delta \\ s.t. \chi(d) &= \max_{\theta \in T} \min_{z} \max_{j \in J} f_j(d, z, \theta) \end{aligned}$$

- $F = 1$: design has *exactly* the flexibility to satisfy the constraints over the set T.
- $F < 1$: only a fraction of the expected parameter deviations can be tolerated.
- $F > 1$: design *exceeds* the flexibility requirements.

More recent methods for flexibility analysis include the branch and bound method by Kabatek and Swaney (1992), and the sensitivity based method by Varvarezos et al. (1995).

Only a deterministic (and not probabilistic) description of the uncertain parameters have been considered in the methods described above. The treatment and definition of a stochastic flexibility index is given in Pistikopoulos and Mazzuchi (1990) and Straub and Grossmann (1991). Bansal (2000) presents a new

framework based on parametric programming which unifies the various flexibility analyses and design optimisation problems that arise from non-linear, steady state systems. Algorithms are developed for the flexibility test and index problems in systems with deterministic parameters, and for the stochastic and expected stochastic flexibility evaluation problems.

Comprehensive general reviews on flexibility analysis can be found in Grossmann and Floudas (1987), Grossmann and Straub (1991), chapter 21 of Biegler et al. (1997) and Bansal (2000).

Design for flexibility The flexibility of heat exchanger networks has been considered by a number of authors. Papalexandri and Pistikopoulos (1993) present an MINLP model for the retrofit of a heat exchanger network in order to improve its ability to operate optimally over a range of conditions. The problem is solved using a multi-period MINLP technique where the periods of operation are defined by a specified set of operating conditions; the objective function is then minimised over all periods. However, the only combinations considered were the lower and upper bounds of the uncertain parameters. This work has since been extended to the design of heat exchanger networks that are cost optimal and flexible (Papalexandri and Pistikopoulos, 1994a). It is covered later in this review, in section 2.1.2.2.

The flexibility of heat integrated distillation sequences is the subject of another paper (Paules and Floudas, 1992). They use stochastic programming to deal with uncertainties in the specification of the plant to enable the development of a design that can handle these uncertainties. The result from this method is a flowsheet which is optimal over a range of feed flowrates and compositions since these are assumed to be the uncertainties in the problem. In the procedure, variables are divided into two groups

- Structural variables: fixed regardless of the values of the uncertain param-

eters

- Periodic variables: varied to optimise a flowsheet for a given set of uncertain parameters

Once again, this technique merely considers the condition that the plant should operate with a discrete set of design requirements. The method uses the MINLP heat integrated distillation sequence model from a previous paper by the same authors (Floudas and Paules, 1988).

2.1.2.2. Controllability

"A controllability analysis is concerned with determining the limitations on achievable dynamic performance. The information provided by this analysis can be used to screen initial designs and to make structural decisions while avoiding the expense of designing a control scheme."

Chenery (1997)

Analysis of controllability Perkins (1989) separates controllability analysis into three areas:

- Assessment: calculation of a numerical controllability index to rank alternatives and highlight shortcomings
- Diagnosis: explanation of the shortcomings encountered in the analysis phase
- Design: modification of the process to improve the previously mentioned controllability index and/or remove shortcomings

Mohideen et al. (1996) comment that while a good deal of research on the assessment of controllability has been performed, relatively little work has been carried out on the diagnosis and design problems.

The attainability of perfect plant control is limited by factors that prevent physically realisable inversions of the plant transfer function. Most controllability assessment studies involve identifying these factors; they may be divided into two approaches: dynamic resilience as proposed by Morari (1983a) , and functional controllability as proposed by Rosenbrock (1970) and Perkins and Wong (1985).

Resilient process plants Lenhoff and Morari (1982) describes the ability of a plant to move quickly and smoothly from one operating condition to another (including startup/shutdown) and deal with disturbances as *"resilience"*. It enabled the design engineer to screen out alternatives that were economically attractive yet unacceptably inoperable at an early stage, although Morari later indicated that assigning it any economic value was difficult. Resilience was found to be independent of the control scheme; instead it is a function of (a) non-minimum phase elements (i.e. time delays and right-half plane zeros), (b) constraints on the control action, and (c) uncertainty (i.e. plant-model mismatch). Note that the linear frequency based techniques developed for assessing resilience are based upon a process model linearised about a nominal steady-state. Their effectiveness would probably decrease if more extreme changes in operational mode were to be analysed, e.g. plant startup.

Weitz and Lewin (1996) present a simple procedure for assessing the controllability and resiliency of process flowsheets. The approach relies on a simplified modelling strategy, whereby an approximate linear dynamic model of the process is derived from steady state flowsheet information. The resulting linear approximation is then analysed using a variety of linear controllability and resiliency measures. The procedure was tested on alternative heat-integrated distillation

column schemes to screen for disturbance resiliency. The results confirmed the conclusions obtained by others, who largely relied upon closed-loop simulations using rigorous dynamic models. The overall methodology appears to be promising as a short-cut diagnostic and screening tool, which could possibly be integrated into commercial flowsheeting packages.

Functional controllability Rosenbrock (1970) defined this concept by considering a linear state space system and considered the effects of right half plane (RHP) zeros. It is shown that a linear system must be invertible to be functionally controllable. However several process attributes result in physically unrealisable inverses. Functional controllability analysis considers the effects which prevents this physical attainability.

Perkins and Wong (1985) characterised the effect of time delays with parameter Δ_{min}, which is the minimum time before a trajectory for any output can be specified. Calculation of Δ_{min} is based upon the magnitudes and locations of delays in the process transfer function. The singular value decomposition (SVD) analysis of the transfer function can be used to characterise the effects of manipulated variable constraints and model uncertainty through evaluating the process condition number $\gamma(\omega)$ and the minimum singular value, σ_{min}:

$$\gamma(\omega) = \frac{\sigma_{max}}{\sigma_{min}}$$

The desirable properties for a plant to be controllable is a small value of condition number, γ, and a large value of σ_{min}. This is because the singular values are an indication of the plant's gain. A large value of σ_{min} is said to be advantageous since it reduces the effect of manipulated variable constraints by giving a larger gain between the manipulated and controlled variables, and a small value of γ minimises the effect of possible plant/model mismatch on the control

performance.

These controllability indicators provide a *qualitative* indication of how difficult a plant will be to control; they cannot, however, easily be related to the economics and operating constraints for a particular problem. This means that the controllability indicators cannot be weighed on the same scales as the process economics. Within this framework, a choice between a plant with good steady state economics but poor controllability indicators and one with inferior steady state economics but better controllability indicators cannot be made without a *judgment* of how much the better indications of controllability are worth. In addition, there are multiple controllability indicators which are likely to give contradictory messages for any given plant. They are also usually based on linear models of the system to be controlled, leaving the effect of the nonlinearity of the true plant and process unexamined. Controllability indicators can play a useful role in highlighting when difficulties might arise, but they cannot be used for the rigorous elimination of design options. Their main use in this context is to provide the designer with information that be of assistance in finding a base-case solution quickly. A review of controllability indicators is presented by Morari (1992).

Some work has been done in the area of assessing controllability in a way that gives an economic indication of the benefit of the control (Narraway et al. , 1991; Narraway, 1992; Heath et al. , 1996; Heath, 1997). Hashimoto and Zafiriou (1995) report that in developing these *quantitative* indices which incorporate economic information, they maximise performance indices over possible disturbances, thus resulting in an explicit dependence on the designer's disturbance estimates. Even indices that do not have such explicit dependence can often be interpreted only in relation to expected disturbances, e.g. indices that predict poor performance for certain disturbance directionalities.

Design for controllability A series of authors incorporate controllability into their steady state synthesis procedure to develop economic (steady state) designs

which also possess favourable dynamic characteristics, i.e. flowsheets that are inherently easy to control.

> *"The view that the process is essentially at steady-state, but must be operable about this steady-state."*
>
> Rowe (1997)

Hierarchical approaches Although most synthesis procedures which have taken dynamic considerations into account have been limited to the more well defined tasks, such as HENS or separation sequencing, Padley (1991) develops and presents a hierarchical procedure based on that of Douglas (1988) which incorporates controllability considerations. The same approach is summarised in Barton et al. (1992). In short, the approach consists of a Douglas type synthesis procedure in which the potential disturbances to the process are identified and characterised. Conventional steady state design is then used to generate starting points whose properties are analysed using flexibility and controllability analyses to screen for undesirable behaviour or economic effects based on the identified disturbances. If undesirable characteristics are identified, the process is backed off from these characteristics by redesign of the process and its operating conditions; economic effects are assessed allowing comparison of alternative flowsheet structures. Due to the emphasis on 'local' rather than plant wide control, large scale structural changes or novel solutions may be overlooked in favour of rendering the perceived optimal steady state flowsheet controllable. The methodology does however provide a manageable approach to the inclusion of controllability with a synthesis framework.

Multi-objective approaches A number of authors have attempted to consider the steady state process economics simultaneously with some indicator of plant controllability. Morari (1983b) suggests this approach is more appropriate than the single scalar objective function (used so far).

Palazoglu and Arkun (1986) incorporate a robustness index (process condition number) into their design methodology in an attempt to satisfy dynamic operability requirements:

$$\begin{aligned} & \min_d J_0(d),\ J_1(d,\gamma(\omega)) && \forall\ \omega \in \Omega \\ s.t. \quad & c_1(d) = 0.0 \\ & c_2(d) \leq 0.0 \end{aligned}$$

where d are the design variables, $\gamma(\omega)$ is the condition number, Ω is the frequency range of interest, J_0 and J_1 are the economic and robustness objectives, c_1 are the equality constraints (process model) and c_2 are the inequality constraints (process constraints).

The multi-objective optimisation problem is reformulated to yield a scalar optimisation problem (referred to as the ϵ method) which can be solved using standard algorithms. Basically the steady-state cost is minimised subject to a sufficiently small condition number (as defined by ϵ):

$$\begin{aligned} & \min_d J_0(d) \\ s.t. \quad & J_1(d,\gamma(\omega)) \leq \epsilon && \forall\ \omega \in \Omega \\ & c_1(d) = 0.0 \\ & c_2(d) \leq 0.0 \end{aligned}$$

The resulting set of solutions are known as the pareto-optimal solutions, or the non-inferior surface. They correspond to solution points at which no one objective function can be improved without a subsequent degradation in the other (competing) objectives. A test problem consisting of three stirred tanks reactors in series was considered; the following points are made as possible drawbacks:

- Only continuous design parameters are optimised, i.e. no structural decisions are made using numerical programming.
- Controllability indicators (such as process condition number) do not relate directly and unambiguously to real performance requirements.

Abbas et al. (1993) solves a similar (one CSTR) problem using Multiple Criteria Decision Analysis (MCDA) theory. This is essentially a systematic procedure for generating the non-inferior set for the multi-objective optimisation problem. However in this case, the open loop damping ratio is used as the controllability indicator. Comments upon the drawbacks associated with this method are as given for the work of Palazoglu and Arkun, with the added disadvantage of the restrictive size of presented example.

Luyben and Floudas (1994) outline their synthesis approach:

- To incorporate steady-state controllability measures simultaneously with the mathematical programming approach to process synthesis.
- To find a best-compromise solution among economic and control objectives by applying a multi-objective optimisation algorithm which considers qualitative trade-offs.

The ϵ method is again employed to formulate a mixed-integer non-linear program (MINLP) which solves a process superstructure including all possible alternative process structures, the controlled, manipulated and measured variables

together with the transfer functions which relate them. A number of steady-state controllability criteria (RGA, minimum singular value and steady-state condition number) are used in this work.

A cutting plane algorithm is then used to determine the best-compromise solution with the trade-off information derived from the gradients of the non-inferior set. The proposed algorithm is demonstrated on a number of examples: distillation column with fixed control structure and variable number of trays; heat integrated distillation column with fixed control structure (structural decisions limited to the inclusion of high pressure column, low pressure column or both); reactor separator system with recycle. In the examples, feasibility of the designs was verified by subjecting simulations of the closed loop response of the systems to both setpoint changes and disturbances. An obvious step forward was made in this work in that structural decisions may now be included in the design and control problem. The key limitation however was the use of steady-state controllability indicators; implementational difficulties prohibited any dynamic controllability analysis.

Switchable process plants Howell (1984) and White (1995) investigate the "switchability" of distillation columns, i.e. how easily transitions between different operating states in a process may occur. Initially, Howell poses a dynamic optimisation problem to solve for optimal switching, where the index used to measure switchability is the extra cost incurred in moving from one steady-state to another. Only the *process controls* are considered as design variables. White extends this work to consider the effect of *process design* on switchability. He also uses dynamic optimisation in two case studies which show that *design changes* can improve switchability (measured by an integral squared error (ISE) objective). His second case study revisits the Nishida and Ichikawa (1975) reactor-separator example (see section 2.2), and incorporates a more rigorous dynamic separator model which enables optimisation of the number of stages through the use of

a lumped column model. The operating cost during a switch in product grade is used as the objective function. This concept of switchability is covered more thoroughly in chapter 3.

Heat exchanger network synthesis (HENS) and controllability The frequently posed HEN design problem may be stated thus: given n_h hot streams to be cooled and n_c cold streams to be heated from specified supply temperatures to specified target temperatures, design the network of heat exchangers, heaters and coolers accomplishing this task at the least cost. If a plant is energy integrated it is often harder to control, i.e. control loops are more likely to interact. Hence it is worth noting that two levels of complexity exist: (a) the impact of a HEN on *total* system controllability, and (b) the HEN controllability itself. Although "pinch" techniques are well established, optimisation based approaches are becoming more successful in finding their way into industrial application.

Marselle et al. (1982) determined that it was not sufficient to concentrate on the steady-state efficient management of energy, but that the energy recovery system should also be resilient. Here resilience is defined by the network's ability to recover maximum energy for a specified disturbance range in the input flows and temperatures. They developed a semi-heuristic design technique and coupled it with a rigorous controller design method for resilient exchanger networks. This could create designs of networks capable of handling a small number of well selected extreme operating conditions (worst case scenarios, e.g. maximum exchanger capacity, minimum ΔT). Morari (1992) observed that this method has drawbacks and commented that applying Marselle's approach often led to non-economical network designs. The authors themselves hinted in their conclusions that dynamic interaction within the network had not been adequately addressed.

Georgiou and Floudas (1990) solve the following problem:

"Given the process synthesis goal of designing an economically optimal HEN

and a process control goal to keep a certain set of control objectives at their set points in the presence of a given set of disturbances: determine whether a system exists that has low capital cost and features total disturbance rejection. If the cost is high, or total disturbance rejection cannot be achieved, determine the system which minimises disturbance propagation."

The concept of disturbability (if a path exists between the disturbance and a controlled output, it is a "disturbable" variable) is incorporated into a process superstructure in order to satisfy the total disturbance rejection criterion. Solution of the resulting mixed-integer linear program (MILP) determines a restricted superstructure (featuring total disturbance rejection) which is then optimised by solution of a non-linear program (NLP) to find minimum investment cost. A possible weakness of this approach is that total disturbance rejection is assumed as being the most appropriate (and *only*) controllability criterion to be applied, and that since this is based upon structural controllability, the system's dynamic behaviour is still not adequately addressed.

Papalexandri and Pistikopoulos (1994a) also consider the design of structurally controllable heat exchanger networks. Again, the work stems from the observation that sequential consideration of process synthesis and operability can lead to suboptimal solutions, e.g. when retrofit design of the original flowsheet is required to remove bottlenecks. The aims of this paper are similar to those of Georgiou and Floudas (1990) described above: to develop a network with the minimum annualised cost which is flexible within a range of uncertain parameters (inlet flowrates, inlet temperatures, heat transfer coefficients etc.) and is structurally controllable (i.e. features total disturbance rejection). The solution method formulates the problem as a multiperiod hyperstructure in which all possible structural and operational alternatives are taken into account. Again, the procedure does not address the dynamic behaviour of the process. The issue of dynamics and control structure consideration is, however, discussed in a sequel paper by the same au-

thors (Papalexandri and Pistikopoulos, 1994b). In this work, the hyperstructure used previously is extended to allow exploration of control structure alternatives. Process dynamics are incorporated by the use of a variable delay matrix and variable gain matrix derived from a linear dynamic model of the process. The issue of process controllability is thoroughly examined and several controllability criteria are incorporated into the synthesis problem as constraints to drive the MINLP solution to a network with favourable controllability characteristics.

2.1.2.3. Reliability, safety and the environment

Reliability is concerned with the probability of normal operation given that mechanical and electrical failures can occur; safety is concerned with the hazards that are the consequences of these failures. Environmental impact minimisation has also emerged as an important issue in the last couple of decades. The initial choice of chemistry is often crucial in such matters, as the production of damaging intermediates and byproducts can often be avoided. Cano-Ruiz and McRae (1998) present an excellent review entitled "Environmentally conscious chemical process design". Relevant research in these various areas includes:

- Characterisation of risk in combustion processes (Gorss and Kinosz, 1995).
- Flexibility, reliability and maintenance in process design (Thomaidis and Pistikopoulos, 1995).
- Waste minimisation in process design and synthesis (Douglas, 1992; Manousiouthakis and Allen, 1995).
- Life cycle analysis (Bounstead, 1995).
- Minimising the environmental impact of process plants: a process systems approach (Stefanis et al., 1995).

- Reaction path synthesis for environmental impact minimisation (Buxton et al., 1997).

2.1.3. Plantwide control structure selection

Control structure selection is defined as the *structural decisions* involved in control system design, including the following tasks:

1. *Selection of controlled outputs* (variables with setpoints)
2. *Selection of manipulated inputs* (degrees of freedom)
3. *Selection of measurements* (for control purposes including stabilisation)
4. *Selection of control* **configuration** (a structure interconnecting measurements and manipulated variables)
5. *Selection of controller type* (control law specification, e.g. proportional integral derivative)

(Foss, 1973; Morari, 1982; Skogestad and Postlethwaite, 1996)

2.1.3.1. The unit operations, i.e. not plantwide, approach

In the past, process control systems for chemical plants have been designed using a "unit operations" approach (Umeda et al., 1978; Stephanopoulos, 1983), utilising the engineering knowledge available concerning single process units such as heat exchangers, chemical reactors and distillation columns. "Plantwide" control structures were then synthesised by adding these individual control components, and then resolving any resulting conflicts/interactions through heuristics. The implicit assumption made was that both the steady state and dynamic behaviour of the unit operations were independent of the manner in which they were connected. It was only reasonable to make this assumption as long as sufficient

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intermediate storage existed between units, so that the buffering would effectively decouple the dynamics of the process equipment. Today, due to the incorporation of multiple recycle streams, higher levels of energy integration and the increasing focus on production flexibility (which is inhibited by excessive inventory), the basic assumption that underlies the unit operations approach no longer holds; the stand-alone control system of a single unit is no longer adequate when combined with the rest of the plant.

2.1.3.2. Mathematical programming approaches

Foss hinted as early as 1973 that an optimisation based approach was more appropriate in tackling this problem:

"the task is to make decisions about control structure and it is here where optimisation can be most intelligently used. Such optimisation problems are, however, a new breed. The standard formulation of optimal control will seldom suffice; it is decisions that are needed, not programming trajectories,"

Georgiou and Floudas (1989) proposed MILP formulations in order to generate a superstructure of alternative control configurations for large scale systems. Turkay et al. (1993), using a slight modification to this method, applied it to an industrial case study of a styrene production plant. Up to this point, however, the objective function being optimised was some controllability index, which was used to rank competing structures. These controllability indicators provide a *numerical* indication of how difficult a plant will be to control; they cannot, however, easily be related to the economics and operating constraints for a particular problem. This means that the controllability indicators cannot be weighed on the same scales as the process economics. Within this framework, a choice between a plant with good steady state economics but poor controllability indicators and one

with inferior steady state economics but better controllability indicators cannot be made without a *judgment* of how much the better indications of controllability are worth. In addition, there are multiple controllability indicators which are likely to give contradictory messages for any given plant. They are also usually based on linear models of the system to be controlled, leaving the effect of the nonlinearity of the true plant and process unexamined.

In view of this difficulty in unambiguously associating controllability with real plant performance, Perkins et al. (1989b) proposed a method of estimating the economic penalty, using Lagrange multipliers, for disturbances entering the system

- in open loop operation,
- under closed loop conditions using perfect control.

Narraway et al. (1991) further developed this idea to calculate the economic penalties due to disturbances acting on pre-specified frequencies for a linearised system and objective function under perfect control. Narraway and Perkins (1993) then used integer variables to represent alternative control configurations. Later Heath et al. (1996) used decentralised PI controllers to eliminate the assumption of perfect control, making the method applicable to a wider range of "single input single output" (SISO) problem. Kookos (2001) continues this work on control structure selection of a "multiple input multiple output" (MIMO) problem based on economics.

A single pass solution to the MILP would enable structural and continuous design decisions to be optimised simultaneously, and hence may offer rigorous, optimal solutions. It must be noted however that as large MILP problems are prone to combinatorial explosion, employing these algorithmic methods tends to be computationally intensive and may limit the control engineer's function in an on-going selection procedure.

2.1.3.3. Heuristic approaches

Buckley (1964) proposed a plantwide control structure philosophy consisting of two stages:

- material balance control
- product quality control

He only considered feed and product streams as potential *throughput manipulators* (the actuators linked to the product flowrate measurement). The inventory controls are then configured around the throughput manipulator such that the resulting material balance structure is *self-consistent* (able to propagate a production rate change throughout the process).

Price and Georgakis (1993) extend this work by adding extra tiers and sublevels as they propose a "plantwide regulatory control design procedure using a tiered framework". The primary process path concept is developed as the basis for the systematic selection of the material balance control structure: the primary process path is defined as the stream that flows from the major feeds to the major products. The throughput manipulator is then constrained to be located on the primary process path.

1. Production rate control
    - identify the primary process path
    - evaluate options for measuring and manipulating the production rate
    - consider stoichiometry and conversion
    - list candidates for use as throughput manipulators (TPMs)
    - eliminate candidate TPMs which are "small"
    - use process internal flows as TPMs
2. Inventory control

- identify those inventories which may require control
- use proximity rules to identify the manipulators suitable for adjusting each inventory
- the choice of a TPM will demarcate at least two subsystems of the process, one upstream and one downstream of the TPM
- use a test of the "degrees of dynamic freedom" (Tyreus, 1992) to help determine how many inventories associated with each unit operation may be controlled
- considering each inventory along the primary process path, construct a consistent chain of controls
- once the inventory controls along the primary process path are completed, construct inventory controls for side chains
- construct additional inventory control chains along any recycle path
    - if additional inlet or outlet flows remain uncontrolled, assign control loops to govern them
    - check component entrance and exit points

3. Product specification control

4. Equipment and operating constraints

5. Economic performance enhancement

They developed a number of insightful heuristics and concepts by applying their framework to a reactor/separator with recycle example, e.g.

"The control structure implementations which produce the smallest integral absolute error (IAE) in response to disturbances in either throughput or feed composition are those whose inventory control structure is self-consistent and directed

along the primary process path."

Unfortunately this approach is not systematic beyond the selection of the material balance control and no quantitative analysis is performed to motivate the development of the control structure. The material balance control philosophy does not include all feasible alternatives. However, this staged approach does analyse the whole process at every tier and hence is more "plantwide" that the modular (unit operation based) approach.

Luyben et al. (1997) proposed their own version of a plantwide control structure selection procedure:

1. establish control objectives
2. determine control degrees of freedom
3. establish energy management system
4. set production rate
5. control product quality and handle safety, operational and environmental constraints
6. control inventories and fix a flow in every recycle loop
7. check component balances
8. control individual unit operations
9. optimise economics or improve dynamic controllability

They also state a set of goals that an effective plantwide control system should achieve; however these objectives are not ranked in any discernible order of priority - indeed, the authors fail to mention which objectives are achieved by applying

their procedure.

Ponton and Laing (1993) present a procedure for addressing the plant wide control structure selection problem. They propose a hierarchy for control structure selection to run in parallel with the Douglas methodology. It does not make use of any typical controllability indicators but rather proceeds through engineering understanding of the effects that potential manipulated and measured variables have on the system behaviour. This allows the basis of a control system to be established at the very earliest stages of the design, simply by looking at the functions and interrelationships of the streams, while the process flowsheet is still very simplified. Additional advantages include an absence of any large numerically intensive tasks such as simulation or optimisation, and that the control structure evolves with the process structure.

More recently Zheng et al. (1999) propose a hierarchical procedure for systematically synthesising a plantwide control system, again motivated by Douglas' conceptual design framework. Their procedure is used to decompose a simple reactor-separator-recycle system into a hierarchy of decisions. These decisions are made using economics quantified by a controllability index ν based on surge volume introduced by Zheng and Mahajanam (1999). The authors describe this index as the cost of dynamic controllability - it is mentioned again in the next chapter when discussing a possible index for responsiveness.

The major advantage of these heuristic driven approaches is that no detailed evaluation need be undertaken, saving engineering effort and computational time for the solution to large scale problems. Obviously these methods are not as rigorous as the mathematical programming approaches detailed in section 2.1.3.2, and any solution obtained *solely* through heuristics cannot even loosely be considered "optimal". Furthermore, conflicts between these rules of thumb based upon engi-

neering knowledge are more likely to arise as the problem size increases. Indeed, Luyben et al. (1999) later admit that the application of their procedure does not result in either a unique or optimal solution.

2.2. The Integrated Design Problem

Bansal (2000) states that any methodology used to tackle the integrated design and control problem must:

- be applicable to **non-linear, dynamic** systems;
- adequately treat and ensure feasibility in the presence of both time-varying **disturbances** and time-invariant (or relatively slowly varying) **uncertainties**;
- involve **selection of the best process design**, taking into account both discrete and continuous decisions; and
- involve **selection of the best control scheme**, again incorporating structural and continuous decisions.

The short review which follows covers works that have considered dynamic process behaviour explicitly in the design procedures employed. A far more extensive and comprehensive review, listing many more references and also outlining future trends in this rapidly expanding research area, is given in van Schijndel and Pistikopoulos (2000).

Nishida and Ichikawa (1975) observe that process systems are inherently dynamic and state that this should be considered explicitly at the process synthesis stage, instead of designing a system around a nominal steady-state and compensating for dynamic behaviour via process control. Their pioneering work aims to

develop a method of synthesising a process system where dynamic operation is recognised as inevitable, and even preferable in certain cases.

Ordinary differential equations are used to describe the dynamics of the process system. An economic objective is then minimised by optimising a set of time-invariant design parameters (e.g. vessel volumes), a set of time-varying piecewise controls (e.g. feed flowrates), and a set of "structural" parameters which establish the process flowsheet (analogous to the structural integer variables used in MINLP approaches to process synthesis). The authors develop a necessary condition for an optimal dynamic system using variational calculus. The resulting optimal control problem is solved using a gradient method which determines the optimal design and control parameter values for a restricted structure. The values of the structural parameters are then updated and the system is re-optimised. Iterations continue until the change in objective function falls within a sufficiently small tolerance.

Two semi-batch reactor-separator networks are used to demonstrate this design procedure, one with the irreversible reaction $A \rightarrow B$, the other with the reversible reaction $A \rightleftharpoons B$. The dynamics in the ideal separator models are neglected as the reactor dynamics are assumed to be dominant. A restricted "superstructure" is used with the reactor holdups as design variables and the temperature profiles as the controls.

Nishida et al. (1976) extend this work by considering uncertainty. A similar approach to the previous paper is taken, except that the design and operation which minimises the effect of worst variations in selected uncertain parameters is found. These uncertain parameters significantly influenced the process dynamics and ultimately the final flowsheet structure. The observation that heuristics could have been usefully applied to restrict the initial set of process structure alternatives in the optimisation was also made.

Given the lack of computational power which existed over twenty years ago, this work was a tremendous achievement. Indeed aspects of this work (uncertainty, appropriate combination of heuristics and numerical programming) continue to pose immense challenges in the present day, as can be seen in the concluding papers in this review.

Walsh (1993) and Walsh and Perkins (1996) consider the integrated design of chemical wastewater treatment systems. They state that the aim of their design procedure is to maximise return on investment while ensuring that performance constraints are satisfied for all possible plant parameters and disturbances, and the pose the following questions: (a) can a particular design be made to satisfy its operating constraints despite uncertainty and disturbances? (b) what is the impact of this on the process economics? Their approach considers operability issues on a par with economic ones: *"performance requirements can be represented either by constraints or by a contribution to the economic objective"*. This is significantly different to the multi-objective approach adopted by a variety of previous authors, as they could not get around the difficulty of measuring dynamic resilience in terms of economics. The work of Walsh and Perkins uses heuristics and bounding properties to narrow the search domain by efficient elimination of large numbers of non-viable options and deepens the search by picking good candidate processes. As viable candidate processes are found, more sophisticated optimisation techniques using refined models are employed for full performance evaluation of the design.

Cantrell and Luyben (1996) propose a design algorithm which considers the dynamics of transition in the design of continuous multiproduct processes. The method uses

- cycle time modelling (operational research)

- process control theory
- heuristic process design

in order to incorporate the dynamics of transitions into the multiproduct plant design and cyclic schedule. Dynamic simulation is also used to examine the feasibility of the proposed process operation, i.e. to see whether any constraints were violated. See appendix B for details of this algorithm.

Mohideen et al. (1996) consider the optimal design of dynamic systems under uncertainty. Design under uncertainty is a complex problem since a feasibility test must be incorporated into the optimisation framework. Finding the feasibility function is itself a multi-level optimisation problem, and when combined with the design problem, the overall formulation becomes practically intractable. Mohideen (1996) developed a decomposition algorithm that splits the problem into a design subtask (that uses multiperiod optimisation), and a feasibility subtask (that uses the active search method, Grossmann and Floudas, 1987); information is then iteratively transferred between the two stages. The main steps of this iterative multiperiod design algorithm are summarised below:

1. Select an initial uncertainty scenario that comprises a profile and a set of points for each time dependent and time invariant uncertain variable, respectively.

2. For the current group of accumulated critical scenarios solve the multiperiod optimisation problem to determine the optimum process design and control.

3. Solve the feasibility problem, letting the free control variables vary, but keeping the structure and design obtained from *Step 2* fixed. If the design and control policy that was evaluated in *Step 2* satisfies all the constraints (feasibility test) for the whole range of uncertainties then the algorithm

terminates. Otherwise, a new set of critical uncertainty values and profiles are obtained and supplied to *Step 1*.

Mixed Integer Dynamic Optimisation (MIDO) problems must be solved in *Step 2* (generated by the embedded dynamic model and the binary variables representing the synthesis, design and control decisions) and *Step 3* (generated by the inter programming formulation of the active search method to solve the feasibility problem) of the algorithm. MIDO is covered in more detail in the next chapter. Mohideen et al. (1996) apply the framework to a simplified distillation case study involving feed rate, feed composition, cooling water temperature uncertainties; the number of trays is optimised using a column superstructure. The use of the new algorithm gives a 15% improvement in capital costs at the expense of a 7% increase in operating costs when compared to a column designed in the traditional sequential manner. A larger, more rigorous example is provided by Bansal et al. (2000a). Sakizlis (2001) reports that a serious shortcoming of the decompostion algorithm is that since the uncertainty is not addressed in the most direct or systematic manner, significant numbers of iterations between *Steps 2/3* must be performed (both of which contain potentially large MIDO problems) in order to explore the entire uncertainty space.

Bansal et al. (2000b) overcomes the iterative nature of the 'Mohideen' decomposition algorithm for **steady state problems** by finding (a) the critical values of the uncertainties that limit flexibility (Pistikopoulos and Grossmann, 1989a,b), and (b) explicit expressions of the system feasibility measures with respect to the design variables using parametric programming (Gal and Nedoma, 1972). Hence system feasibility can be identified by simple function evaluations for different designs. The extention of this work to dynamic systems would require the existence of parametric programming methods in dynamic optimisation (mp-DO) - theory and techniques for tackling this class of problem are currently under investigation

(Sakizlis, 2001).

Rowe (1997) addresses the integrated design issue very directly in his approach to designing distributed chemical manufacturing facilities. He proposes to decompose the problem into two stages:

- Develop a viable process structure through heuristics.
- Establish the design and operation of this structure through dynamic optimisation.

The objective in the preliminary design stage in the "decomposition" method is to produce a viable candidate structure for more detailed analysis. It is anticipated that the amount of data required for this level may be significantly less than is needed for the complete design. When the candidate structure is deemed feasible, it provides an upper bound on the achievable economic performance of the process. Only when a viable candidate is in place are more computationally intensive design tools employed to determine the optimal design and operation of the process. In order to incorporate the process operation into this second design stage, Rowe then makes the simplifying requirement of **cyclic** operation so that cycle time may then be used as an optimisation variable. The use of a cycle provides a way of collapsing the infinite operating horizon into a finite, but infinitely repeatable operation. Dynamic optimisation is chosen as the most appropriate technique of simultaneously determining the design and operation of processes. A discussion of dynamic optimisation methods and the specific implementation details of the feasible path dynamic optimisation code which Rowe uses to carry out his case studies are also described. The major drawbacks in this work are: (a) control system design is not adequately addressed, (b) uncertainty is not rigorously dealt with, and (c) the optimal design parameters are not validated in multiple cycle simulations - this "infinitely repeatable finite time horizon" assumption is not tested.

2.3. Conclusions

This chapter first outlines the traditional, sequential process synthesis methods and then progresses onto those which have considered process design and operation simultaneously.

Koolen (1994) believes that manufacturing will have to move from largely steady state operation to an **intentionally** dynamic one in his projection of future plant operation. Backx et al. (1998) also talk about intentional dynamics in supply chain conscious process operations:

"Dynamics has to be accepted as an additional opportunity rather than considered as a strange, undesired and even dangerous phenomenon outside the scope of normal process operations."

The operational nature of a responsive plant is likely to be characterised by frequent changes in operating point which may lie anywhere within the plant's design envelope. In the extreme (but plausible) case when demand is intermittent, the plant must be capable of automatically starting up to deliver product and shutting down when demand ceases. Given the further requirement that the plant's response to customer demands should be instantaneous, a level of stored product may be required. A responsive plant is inherently dynamic, and this dynamic behaviour must be considered explicitly within the process design of such plants. Hence for the purposes of this work, an integrated design approach based upon dynamic operation is opted for.

The works presented by Mohideen (1996) and Bansal (2000) are based entirely upon numerical programming. As computer technology is continuously improving, such solution methods become increasingly more practical. On the other hand,

both Walsh's (1993, 1996) and Rowe's (1997) approaches reinforce the claim of Floudas and Grossmann (1995) that the best design approaches will be combinations of heuristics and numerical programming. Indeed Walsh observes:

"The challenge to integrated design is to find a good division of labour between human and computer and an appropriate way of interfacing the two."

Chapter 3

Procedures and tools for design

3.1. Introduction

This chapter describes the development of an integrated approach used to tackle responsive process design problems. Brief introductions are subsequently given to the tools which may potentially be used in any proposed methodology for this work, namely worst-case design under uncertainty, Operating Envelopes and dynamic optimisation. It concludes with some consideration of responsiveness, the associated topic of switchability and discussion of an appropriate metric.

3.2. A choice of design procedure

Walsh (1993) states that the aim of the integrated design problem is to obtain minimum cost designs while ensuring that safety and performance constraints are satisfied for all possible plant parameters and disturbances. This is generally achieved by finding the economically optimal combination of:

1. unit/controller type and topology

2. unit/controller design

3. plant operation

Integrated design considers dynamics and control issues as early as possible in the design process. Hence it is generally accepted that parts 2 and 3 of the above, plant design and operation will be determined by dynamic optimisation. However, whether to incorporate the choice of unit/controller type and topology into the same optimisation framework is the question that must be resolved.

3.2.1. Mixed integer dynamic optimisation

In determining the optimal design and operation of a responsive plant, three distinct sets of decision variables will generally be involved. The choice of these variables will determine the ultimate design and operation of the process:

- discrete decisions (i.e. does a unit or controller appear in the final flowsheet) which determine the plant structure,
- continuous, time-invariant parameters (e.g. controller tuning, vessel sizes) which determine the unit and controller design,
- continuous, time-varying control profiles which may determine the plant operation.

Use of mixed integer dynamic optimisation (MIDO) enables all of these decision variables to be simultaneously manipulated to optimise some objective function (usually economic) subject to the constraints imposed on the problem. In other words, mixed integer dynamic optimisation enables the process structure, design and operation to be determined at one level. Indeed MIDO has been used by many authors in their works on the interactions of design and control: Narraway (1992), Balakrishna and Biegler (1993) , Mohideen et al. (1996), Schweiger

and Floudas (1997), Bahri et al. (1997), Sharif (1999), Avraam (2000), Bansal et al. (2000a), Fraga et al. (2000), Kookos and Perkins (2001). Note some authors discretise the time horizon entirely and solve a MINLP, while others, such as Bansal, solve a dynamic optimisation subproblem (i.e. only the control vectors are parameterised). For this work MIDO suffers a number of drawbacks and was rejected for the following reasons:

1. The design problem must be **completely** specified *a priori* before solution may begin. Any changes in problem definition during the design process would require the MIDO problem to be reformulated and solved.

2. A process superstructure covering all possible design options (process **and** control structures) can be difficult to generate, especially where numerous unit types and control strategies are possible.

3. Solution of MIDO problems can be computationally intensive for medium to large scale problems. Reliable MIDO algorithms have only recently been developed (e.g. Bansal, 2000).

3.2.2. Decomposed design procedure

The design procedure used in this work does not attempt to simultaneously solve the combined structural and operational problem described above. The alternative approach taken here is to decompose the problem into two stages which separates determination of the process structure from the process unit design and operation. The two stages of the procedure that Rowe (1997) proposed are:

- develop a candidate process structure

- determine the corresponding (optimal) operation and design for that candidate

The contribution this work hopes to make is to (a) add control structure selection to the first part, i.e. consider the complete *p*lant structure, (b) refine the optimal control techniques in the second stage using the *gPROMS/gOPT* environment, and (c) adding a third and final verification phase that ensures operational feasibility under uncertainty using Operating Envelopes. The underlying philosophy of the design procedure may be described as a "design funnel" and is shown figure 3.1.

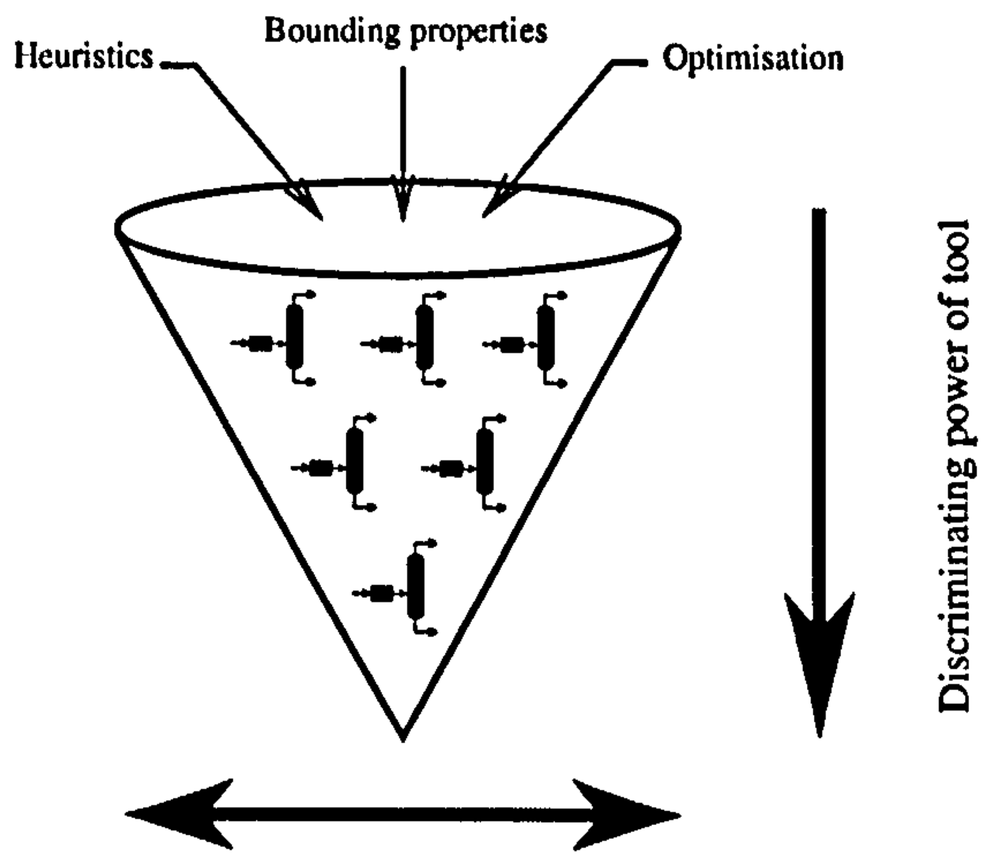


Figure 3.1: "Design funnel" to be used in process design procedure

In applying the procedure, a tradeoff is sought between the cost of using a given design tool and the level of input it requires, and the discriminating power of the tool. At the higher levels where many design options exist, discrimination between structures may be entirely heuristic. For example, at the top level nominal unit operations to be used in the process may be specified based upon past engineering experience. The objective of the preliminary design stage, which corresponds to the upper levels in the design funnel, is to produce a viable candidate design for more detailed analysis. It is anticipated that the required amount of data for this level may be significantly less than is needed for the complete design. Where the candidate structure is feasible, it provides an upper bound on the achievable economic performance of the process. Only when a viable candidate is in place are

more computationally intensive design tools employed to determine the optimal design and operation of the process; this presents a much more tractable problem than the MIDO formulation discussed previously. Indeed, robust and well-tested numerical solution methods do exist and will be discussed in section 3.3.3.

3.3. Tools for integrated design

3.3.1. Fundamentals of worst case design

Many design parameters of a chemical plant may be uncertain, e.g. heat transfer coefficients, compressor efficiencies, temperatures and flowrates of certain streams etc. A general review of process design and operations under uncertainty is given by Pistikopoulos (1995). Walsh and Perkins (1996) and Walsh (1993) present a design example of an effluent treatment plant that uses a worst case design solving a min/max problem that ensures that the dynamic constraints of a plant are satisfied for all uncertain parameter realisations within the specified bounds. The method tries to find the worst case combination of these parameters by maximising constraint violation(s), and does not assume that this will be a combination of variables at their lower and upper bounds (vertices). The following assumptions are initially made:

- the uncertainty is defined by **bounds** on model characteristics or parameters
- the constraints must be satisfied at all times

Hence all solution techniques must ensure feasibility for **all** possible realisations of the uncertainty (worst-case design).

The problem is how to tackle the infinite number of uncertain parameter realisations that must be considered. As the constraints must be satisfied for each and

every point in the uncertainty space, effectively an infinite number of constraints must be dealt with. The approach adopted is to approximate the continuous uncertain parameter space $v \in V$, by a discrete set $v \in V_i$ and to update the set until it yields a design for which no constraint violation can be found. This is known as an outer-approximation algorithm, as the constraints associated with $v \in V_i$ map an outer approximation to the feasible region associated with $v \in V$. This is illustrated in figure 3.2, which shows how three scenarios of the uncertain parameters give an approximation to the true feasible region.

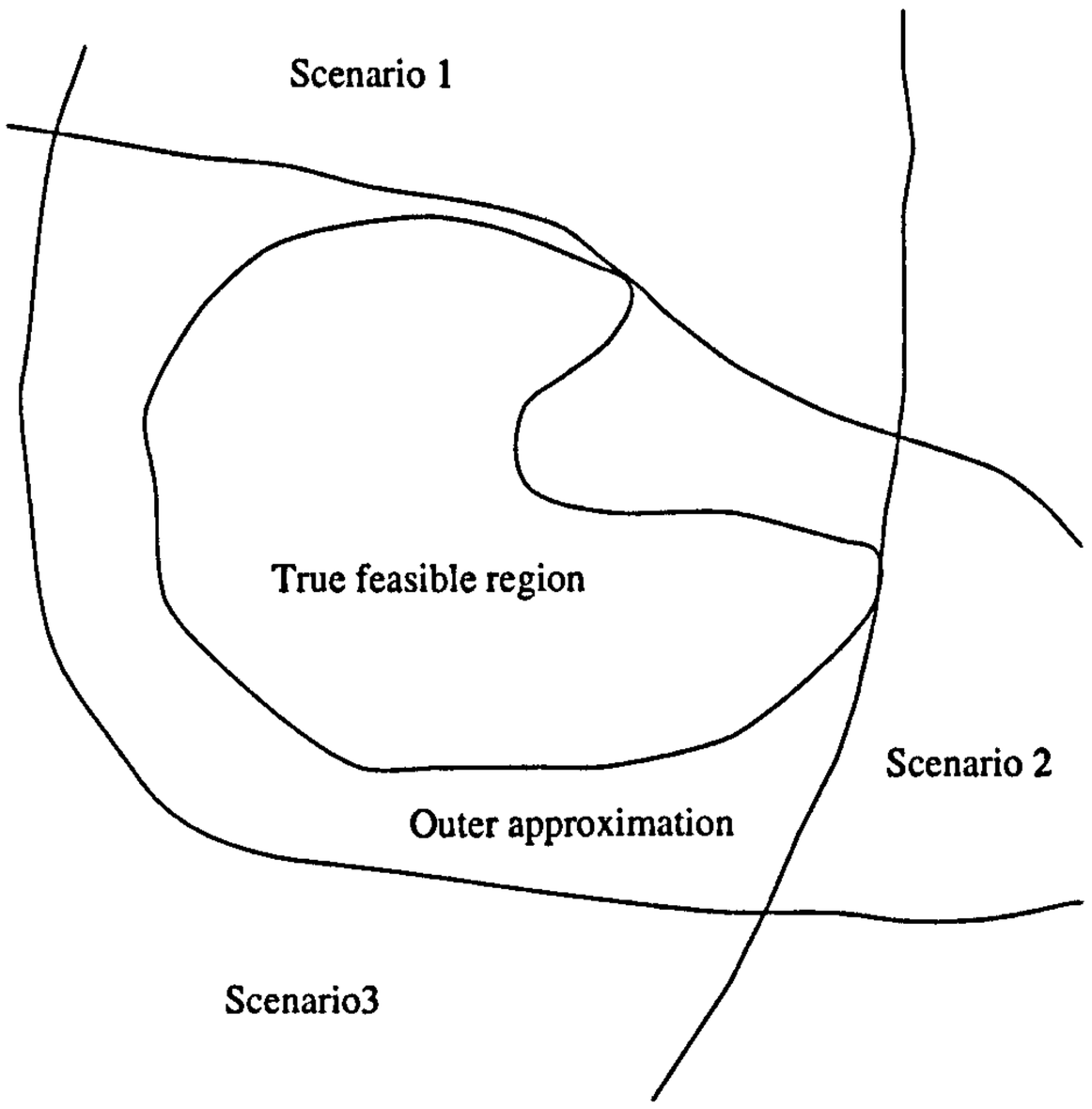


Figure 3.2: Effect of outer approximation

Two approaches to worst-case design have been applied successfully to engineering design problems with uncertainty:

- One attempts to reduce the problem size by assuming the worst-case uncertain parameters lie at the vertices of the parameter space V.

  - Grossman et al. (1983), Swaney and Grossmann (1985), Grossmann and Floudas (1987)

- The other applies general global optimisation techniques, such as grid enumeration, which assumes that checking whether or not an element v is within V is computationally cheap.

  - Polak (1982), Tits (1985), Polak and Stimler (1988), Mayne et al. (1990)

Walsh and Perkins (1996) present a new algorithm for design with uncertainty which does not differ radically from previous approaches, but rather represents a new *variant* of the basic outer approximation algorithm. The key feature is the use of approximate global optimisation to give reduced reliance on convexity properties compared to the algorithm of Grossmann et al. discussed above, while not relying on the rigorous but expensive global optimisation methods favoured by Polak et al. The algorithm seeks to reduce computational cost by relying on local search methods in initial iterations and by introducing a systematic overdesign factor to the design iterations. The optimisation formulation is:

$$\min_{p}\{J(p,V) \mid \max_{v\in V}\max_{k\in K}[c_k(p,v)] \leq 0\}$$

where J is the objective function to be minimised, p is a vector of design variables, v is a vector of uncertain variables, and c_k is the k^{th} element of the constraint vector c. The reader is pointed to the work of Walsh and Perkins (1996) for a more thorough treatment.

Although this approach could have been adopted as part of the proposed integrated design framework, solving a min/max problem to attempt to find global

optima was deemed too computational intensive for the purposes of this work. At this stage if good (i.e. locally optimal) feasible solutions can be produced, global optimality is not a requirement. This, combined with the difficulty in implementing this technique in the *gPROMS/gOPT* (the modelling/optimisation environment used in this work) led to a similar, simpler approach being taken, described in the next section. Note that Cobden (1998) attempted to implement Walsh's worst case algorithm in the *gPROMS/gOPT* environment using MATLAB.

3.3.2. Operating Envelopes

Operating Envelopes specify process operation in terms of a region within which the operating variables should be maintained to guarantee feasible and profitable operation of the process. The approach is to determine a range of operating policies (the union of which is called the *Operating Envelope*) during the design stage over which the plant performance can be guaranteed to meet certain targets. The formulation is based on a typical optimal control and design formulation and is taken directly from the paper written by Samsatli et al. (2001). For the solution procedure and examples, please refer to the paper.

$$\max_{\boldsymbol{u},\boldsymbol{\nu},\tau} \Phi(\dot{\boldsymbol{x}}(\tau), \boldsymbol{x}(\tau), \boldsymbol{y}(\tau), \boldsymbol{u}(\tau), \boldsymbol{\nu}, \tau)$$

subject to

$$\begin{aligned} &\boldsymbol{J}_0(\dot{\boldsymbol{x}}(0), \boldsymbol{x}(0), \boldsymbol{y}(0), \boldsymbol{u}(0), \boldsymbol{\nu}) = \mathbf{0}, \\ &\boldsymbol{h}(\dot{\boldsymbol{x}}, \boldsymbol{x}, \boldsymbol{y}, \boldsymbol{u}, \boldsymbol{\nu}, t) = \mathbf{0} \qquad \forall\, t \in (0, \tau], \\ &\boldsymbol{g}(\dot{\boldsymbol{x}}, \boldsymbol{x}, \boldsymbol{y}, \boldsymbol{u}, \boldsymbol{\nu}, t) \leq \mathbf{0} \qquad \forall\, t \in [0, \tau]. \end{aligned} \tag{M1}$$

In the above model (M1), $\boldsymbol{x}$ are differential state variables, $\dot{\boldsymbol{x}}$ their derivatives with respect to time, $\boldsymbol{y}$ are algebraic state variables, $\boldsymbol{u}$ are time-varying controls,

$\boldsymbol{\nu}$ are time-invariant optimisation parameters (that may represent both design decision variables and time-invariant control variables), τ is the processing time, and t time. The constraints $\boldsymbol{J}_0$ represent the initial conditions of the system, whereas $\boldsymbol{h}$ and $\boldsymbol{g}$ are, respectively, general equality and inequality constraints. The latter may be active throughout the time horizon (path constraints) or at any particular point in time (point constraints). The majority of the equality constraints are due to the model equations; a small number will be used for various product and process specifications. The inequality constraints typically represent constraints on safety, operational limitations, product specifications, etc.

The solution of the above problem results in unique profiles and values for $\boldsymbol{u}$ and $\boldsymbol{\nu}$, respectively, that maximise the objective function, Φ. However, since it will not be possible to control some of the $\boldsymbol{u}$ and $\boldsymbol{\nu}$ variables perfectly throughout the operation of the process, or from batch to batch, the approach is to find the largest region in which the process may be operated such that a certain level of performance can be guaranteed (i.e. Φ is always above a certain value). If this region in the $\boldsymbol{u}$ and $\boldsymbol{\nu}$ variables is denoted by E (the "Operational Envelope"), then the formulation can be written as follows.

$$
\begin{aligned}
&\max f(E) \\
&\text{subject to} \\
&\quad \boldsymbol{J}_0(\dot{\boldsymbol{x}}(0), \boldsymbol{x}(0), \boldsymbol{y}(0), \boldsymbol{u}(0), \boldsymbol{\nu}) = \boldsymbol{0} \quad \forall\, \boldsymbol{u}, \boldsymbol{\nu}, \tau \in E, \\
&\quad \boldsymbol{h}(\dot{\boldsymbol{x}}, \boldsymbol{x}, \boldsymbol{y}, \boldsymbol{u}, \boldsymbol{\nu}, t) = \boldsymbol{0} \quad \forall\, \boldsymbol{u}, \boldsymbol{\nu}, \tau \in E,\ t \in (0, \tau], \\
&\quad \boldsymbol{g}(\dot{\boldsymbol{x}}, \boldsymbol{x}, \boldsymbol{y}, \boldsymbol{u}, \boldsymbol{\nu}, t) \leq \boldsymbol{0} \quad \forall\, \boldsymbol{u}, \boldsymbol{\nu}, \tau \in E,\ t \in [0, \tau], \\
&\quad \Phi(\dot{\boldsymbol{x}}(\tau), \boldsymbol{x}(\tau), \boldsymbol{y}(\tau), \boldsymbol{u}(\tau), \boldsymbol{\nu}, \tau) \geq \Phi^{\star} \quad \forall\, \boldsymbol{u}, \boldsymbol{\nu}, \tau \in E; \qquad \text{(M2)}
\end{aligned}
$$

where $\Phi^{\star}$ is a specified lower bound.

In order to solve this problem, it is necessary to specify a geometric form for the Envelope. Before doing this, it is more convenient to partition the optimisation parameters into those that cannot be controlled adequately and those that can. The former will be denoted by the vector $\boldsymbol{b}$ and will be called "bounded" variables (*i.e.* one wishes to have some operational bounds for flexibility in these); the latter will be denoted by the vector $\boldsymbol{a}$ and will be called "non-bounded" variables (*i.e.* point values for these will suffice). To make the formulation less cumbersome, the $\boldsymbol{a}$ and $\boldsymbol{b}$ variables will represent both time-invariant optimisation parameters and time-dependent controls (it makes little difference since $\boldsymbol{\nu}$ is just a special case of $\boldsymbol{u}$). The Envelope, E, will therefore be a region only in the space of the $\boldsymbol{b}$ variables.

A simple hyper-rectangular form for the Envelope is chosen for a variety of reasons. From a numerical point of view, this form of Envelope means that the solution of the above model will be relatively easy. From a physical perspective, it makes sense to specify the Envelope in terms of allowable ranges of the bounded parameters; anything more complex would be difficult to implement in practice and would require unusually complicated operating policies. A significant benefit of selecting a hyper-rectangular form is that it decouples the control objectives, so that each parameter can be individually maintained within its bounds without knowing precisely what the values of the other parameters are. As a consequence, for multi-stage processes, each stage may be operated without any information about how previous stages were operated (except that they were operated within their Envelopes); no adaptation of the operating policy (or, rather, Envelope) will be required.

The Envelope is now represented by the region $E \equiv \{\boldsymbol{b} | \boldsymbol{b}^{\min} \leq \boldsymbol{b} \leq \boldsymbol{b}^{\max}\}$, where $\boldsymbol{b}^{\min}$ and $\boldsymbol{b}^{\max}$ are vectors of the lower and upper limits of the ranges of the "bounded" parameters. The function $f(E)$ may then be written as a function of these lower and upper bounds. A suitable function may be:

$$f(E) \equiv \prod_{i=1}^{N_b} (b_i^{\max} - b_i^{\min})$$

where N_b is the number of bounded variables (= dim($\boldsymbol{b}$)). However, this function is a highly non-convex volume and could easily be substituted with a linear one. Defining Δb_i as $\Delta b_i \triangleq b_i^{\max} - b_i^{\min}$, the objective function may be replaced by:

$$f(E) \equiv \frac{1}{N_b} \sum_{i=1}^{N_b} \frac{\Delta b_i - \Delta b_i^{\min}}{\Delta b_i^{\max} - \Delta b_i^{\min}}$$

where $\Delta b_i^{\min}$ and $\Delta b_i^{\max}$ are lower and upper bounds on the sizes of the ranges. The function therefore takes the form of a scaled perimeter. Lower bounds on the sizes of the ranges are required because, in practice, there will be a limit to how tightly an operating variable can be controllable. Upper bounds are applied to prevent the solution containing unrealistically large ranges. If there is a level of control (*i.e.* the width of the range) that is trivial to maintain, this is set as the upper bound on a range so that any remaining flexibility in the process can be used to widen the range of a different parameter.

The above objective function therefore lies in the range [0,1], with 0 representing the smallest possible Envelope (*i.e.* $\Delta b_i = \Delta b_i^{\min} \quad \forall i$) and 1 representing the largest ($\Delta b_i = \Delta b_i^{\max} \quad \forall i$). The overall formulation is shown in the model below (M3).

$$\max_{a,b^{\min},b^{\max}} f \equiv \frac{1}{N_b}\sum_{i=1}^{N_b}\frac{\Delta b_i - \Delta b_i^{\min}}{\Delta b_i^{\max} - \Delta b_i^{\min}}$$

subject to

$$\begin{aligned}
&J_0(\dot{x}(0), x(0), y(0), a(0), b(0)) = 0 \quad \forall\, b \in [b^{\min}, b^{\max}],\\
&h(\dot{x}, x, y, a, b, t) = 0 \quad \forall\, b \in [b^{\min}, b^{\max}],\ t \in (0, \tau], \tau \in b,\\
&g(\dot{x}, x, y, a, b, t) \leq 0 \quad \forall\, b \in [b^{\min}, b^{\max}],\ t \in [0, \tau], \tau \in b,\\
&\Phi(\dot{x}(\tau), x(\tau), y(\tau), a(\tau), b(\tau), \tau) \geq \Phi^{\star} \quad \forall\, b \in [b^{\min}, b^{\max}], \tau \in b,\\
&\Delta b = b^{\max} - b^{\min},\\
&\Delta b^{\min} \leq \Delta b \leq \Delta b^{\max}.
\end{aligned} \tag{M3}$$

Note that τ may be a member of the Envelope variables, b; i.e. we may be interested in finding lower and upper bounds on the processing time (e.g. how important it may be to quench a reaction at a precise time).

3.3.2.1. Solution procedure

The solution of this problem requires the evaluation of the constraints (both equality and inequality) over the whole of the Envelope. This means that an essentially infinite number of constraints need to be satisfied and as such the problem will be intractable. However, if the feasible region is convex one need only consider the vertices of the Envelope to ensure that operation is feasible within. Further, it is often the case that only a relatively small number of vertices limit the size of the Envelope (so-called critical vertices). This leads naturally to a simple solution procedure, where these critical vertices are identified (by simulation) and only these vertices are used in the optimisations.

1. Solve the optimisation problem considering two opposite vertices of the Envelope (more vertices may be considered in subsequent iterations).

2. Simulate the resulting Envelope for all vertices and identify potentially critical vertices (those with the highest constraint violation).

3. Update the vertices used in the optimisation in step 1 by adding or replacing with critical ones, then return to step 1. If there are no constraint violations in any vertex, then terminate.

The solution procedure can be extended to cover the possibility of non-convex feasible regions by posing an optimisation problem that maximises the constraint violation *within* the Envelope. If no violation can be found, then the solution procedure is terminated; otherwise, the point with the worst violation is added as an additional critical scenario in step 1 above.

3.3.3. Dynamic optimisation

3.3.3.1. Background

"Optimal control may be defined as the determination of time-dependent **controls** *and time invariant* **parameters** *that optimise some performance index of a dynamic system."*

Vassiliadis (1993)

Dynamic optimisation, or optimal control, examines the performance of a system operating under transient conditions. Complex process plants with tight operating constraints and inherently dynamic, e.g. batch processes are two applications where dynamic optimisation could be usefully applied. Edgar and Himmelbrau (1989) give some examples of process tasks that may be solved by optimal control:

- Maximise production of a key component
- Minimise yield of a contaminant

- Process start-up and shut-down
- Moving from one steady state to another.

Indeed the responsive plant problem may be viewed as a combination of the last two tasks listed above. Reviews of this area may be found in Biegler (1990), Vassiliadis (1993) and Bell and Sargent (2000).

Approaches to optimal control may be distinguished by

1. whether the dynamic equations are solved by conventional simulation techniques (feasible path) or as part of the optimisation using approximation techniques to transform the problem to algebraic constraints (infeasible path)
2. how constraints on values of variables over time (state of path constraints) are dealt with (see below)
3. how gradients are calculated, e.g. adjoint equations or sensitivity equations
4. the optimisation methods applied, e.g. SQP, iterative dynamic programming

The view of Walsh and Perkins (1996) was that feasible path methods, in which the solution of the model equations with time is carried out by extensively developed and refined integration software, are at present more reliable than infeasible path methods. Feasible path methods are also easier to implement as the size of the optimisation problem is much smaller. Hence, they are chosen despite evidence that infeasible path methods may be more efficient on some problems.

3.3.3.2. Mathematical statement and solution method

The mathematical description of a typical dynamic optimisation problem has already been given given in this chapter (M1). The controls $\boldsymbol{u}(t)$ can be replaced by additional time-invariant parameters by parameterising the variation of time

explicitly. The time horizon is split into a finite number of elements. In each time element, the control variables are defined as low-order polynomial functions (e.g. piecewise linear functions of time). The optimiser then selects the desired control profile by manipulating both the time interval lengths and the parameters which describe the nature of the polynomial. Hence this infinite dimensional optimisation problem may be converted into a finite dimensional one.

The solution of the discretised problem obtained above follows a two stage iterative procedure:

1. Integration of the process system equations for fixed values of the optimisation parameters to obtain constraint residual information and/or gradients

2. Solution of a non-linear programming problem to determine new values for the optimisation parameters which improve feasibility and/or the objective function

3.3.3.3. Implementation

In all of the case studies presented, the mathematical model describing the process is first developed in the *gPROMS* (Barton, 1992) modelling environment, which uses DASOLV (Jarvis and Pantelides, 1992) to integrate the DAE system and calculate the sensitivity values, which may be done in parallel with the forward integration. The specific optimisation problem to be solved is then specified in an accompanying *gOPT* input file containing the following information:

- definition of the objective function
- definitions of problem constraints (both equality and inequality constraints)
- specification of optimisation parameters
  - the set of parameters which are available to be optimised

– optimisation parameter bounds

gOPT is basically a front-end interfacing DAEOPT (Vassiliadis, 1993) to *gPROMS*. Unfortunately it does not possess the full generality of the original DAEOPT, in particular, not permitting independent control interval structures. Refer to appendix A for details of how control profiles may be implemented **directly** in the *gPROMS* environment. For more information about the *gPROMS/gOPT* modelling/optimisation environment please go to www.psenterprise.com.

3.4. Responiveness

The concept of "responsiveness" has not often appeared in the literature, although there is some coverage of the associated topic of switchability. Sargent and Sullivan (1979) used optimal control techniques to determine the optimal trajectories of the controls for a refinery distillation train during feed changeovers. A similar technique was used in the determination of the optimum switching policy for a plant with a given structure, studied by Howell (1984) in which the subject of switchability was first addressed. Here the method used was that of determining the optimum switching policy by solving an optimal control problem where the objective was to minimise the extra cost in moving from one steady state to another.

Using Howell's technique, Perkins et al. (1989a) studied the switchability characteristics of three integration schemes for a crude oil distillation column. This allows the structures to be ranked in order of the optimal switch cost. This is somewhat like the controllability indicators mentioned in chapter 2, in that the capital cost of the plant is not explicitly considered, although it could be incorporated into the cost of the switch.

Switchability describes how readily a process switches between different operating states. It is concerned with the dynamic behaviour of systems which

experience major disturbances in operating conditions, often exceeding the limits which have been set for flexibility. Under these conditions, the range of control action will be constrained by physical and safety factors. Emphasis must be made of the nonlinear features of the system response, so analysis based upon linear models is deemed inappropriate: the essential difference between controllability and switchability.

Howell's work gives a direct measure of the cost of a particular switch in economic terms, in contrast to an abstract measure of controllability. White (1995) believes that optimal switching, i.e. the determination of a plant's switchability by applying optimal control techniques to nonlinear models, is the best way of assessing the switchability of a plant design. He observes that columns operating under higher reflux ratio, but with fewer stages are more "switchable". Reducing tray weir heights reduces the integral squared error (ISE) switchability objective.

Switchability may be seen as a subset of a more general concept of responsiveness. The design of responsive plants which frequently switch operating points in reaction to customer demands must consider process operation as a degree of freedom. But how does one incorporate this responsive element into an integrated design procedure? How does one assess the degree of responsiveness of a particular design? Two approaches may be taken in dealing with this issue:

- Formulate a quantitative index of responsiveness, similar to that of controllability and switchability
- Impose responsive design requirements on process alternatives, to ensure feasibility in operation, i.e. non-violation of "responsive" constraints

The second approach was taken by Rowe et al. (1997), where responsive plants are mentioned in the literature for the first time. They propose a plant capable of instant response to a customer's intermittent demand for product. The case study was intended to represent a "worst-case" demand profile, since the process must

be capable of automatic startup/shutdown. It illustrates a potential situation where environmental regulations on chemical storage make production *in situ* using a responsive plant the only viable means to satisfy a customer's raw material requirements.

Recently Zheng and Mahajanam (1999) proposed a simple quantitative index of controllability, ν, the minimum *additional* surge capacity required for a given flowsheet and corresponding control structure to satisfy *dynamically* all the objectives and constraints for all expected disturbances. This allows different plant alternatives to be quantitatively compared. A binary distillation column is presented as a simple example. Zheng et al. (1999) apply the index to a reactor-reactor-recycle case study. Although a one-product, continuous chemical process is assumed, it is conceivable to incorporate this index into the responsive plant problem, in order to rank control schemes.

3.5. Conclusions

This chapter has considered the concept of responsiveness and approaches for the integrated design and operation of responsive chemical manufacturing facilities. A decomposed problem which first develops a viable process structure and then establishes the design and Operational Envelope of this structure is proposed.

The heuristics employed in the first phase of the decomposed design strategy must take the responsive nature required by the plant into account. Initially, the location and type of output variability must be determined by the customer. All candidate structures which are then generated must be capable of satisfying this instant response criterion.

In the following dynamic optimisation stage, the plant operation must be a degree of freedom to satisfy the responsive requirement imposed upon the design in the first stage. Several scenarios could also be accounted for by the use of a

variety of objective functions, e.g. economics, stability, minimum waste etc. The trade-offs between different, sometimes conflicting, objectives are often not clear and hence unambiguous problem statements are essential. Additionally, the physical changes within the plant that result from the design parameters being changed during the optimisation must be considered in order to verify that they are both feasible and reasonable. An Operating Envelope technique specifying operation in terms of a region within which the operating variables should be maintained to guarantee feasible process operation was deemed appropriate to handle this uncertainty.

This combined approach was chosen mainly due to the fact that computationally intensive techniques are only employed after a candidate structure has been selected through heuristics, bounding properties and screening tools.

Chapter 4

Multiproduct processes

4.1. Introduction

"In a multiproduct plant all the products follow essentially the same path and use the same equipment. Normally such plants produce one product at a time, a range of products being achieved by carrying out production runs or campaigns for each product in turn. A multiproduct plant is usually designed for a fixed set of products."

Sparrow et al. (1975)

Continuous multiproduct plants arise frequently in the chemical process industries when a number of products with different specifications must be manufactured by a continuously operating system. When it is uneconomic to purchase a processing system for each product, multiple products must be scheduled on a much smaller set of processing units. Typical industrial examples of this type of process are

- petroleum refineries

- polymer and speciality chemicals plants
- paper production
- cement milling.

Pinto and Grossmann (1994) have identified a current trend in the chemical industries towards continuous flexible multiproduct plants that can respond more quickly to demand changes and to the processing of a variety of products. The desire for "green" designs will also make multiproduct plants the wave of the future, as new processes will tend to share waste handling systems to spread infrastructure costs (Blau, 1995).

Cantrell and Luyben (1996) state that the key to designing such plants is to tackle the problem in terms of optimising steady states: each steady state operating period would correspond to the single product being produced at that time. Once the optimal steady state plant has been designed, a control scheme is added which is both capable of moving the plant between the operating points required for each product and holding the plant at those operating points. Finally, a production schedule (the order in which products are generated) may be determined to meet supply requirements while minimising inventory and transition costs (the cost incurred while changing operating points). This schedule is typically cyclic; not only does this provide repeatable and easily implementable operating strategies, but also enables process operation to be incorporated into the design problem. The authors observe that as the production tasks need to be performed in sequence, rather than in parallel, the transitions that occur in switching from one steady state to another need to be incorporated into this scheduling problem.

Sahinidis and Grossmann (1991) agree that product transitions and transition times are of critical importance to the scheduling problem. They also note that

although products may not be manufactured simultaneously, product demand will be continuous; proper modelling should therefore also take inventory considerations into account. In fact the major trade-off which must be considered is the one between transition frequency vs. inventory cost. A production schedule consisting of a minimum number of transitions (i.e. long cycle time) may at first seem preferable, since transition generates offspec material. However, minimum number of transitions corresponds to the plant operating at each of its steady-states for maximum time, generating large amounts of product in one campaign. As the demand for a given product is fixed, continuous, and must be met from stored inventory during all production periods (including those in which that product is *not* being generated), the size of the storage vessels will be suboptimally large. Sahinidis and Grossmann conclude that *an optimal cycle time which is determined largely by the inventory and transition costs* must exist.

Cantrell and Luyben give a useful insight into the three main historical perspectives that have been taken in the design and operation of multiproduct plants. A short summary of this review is given below:

- process design
  - Sparrow et al. (1975) developed a production time cycle model in order to study multiproduct equipment design for batch processes, given that the product demand schedule is known.
- product scheduling
  - Some of the more recent work in the scheduling of multiproduct plants can be found in Shah and Pantelides (1992), Sahinidis and Grossmann (1991), and Voudouris and Grossmann (1993).
- product transition dynamics and control

– The general theory of transitional control (Ray, 1981) is developed more explicitly for this kind of situation in the paper by Farber and Laurence (1986) for a CSTR undergoing a change in operating state. McAuley and MacGregor (1992) studied the optimisation of product changeovers of a polyethylene reactor, where the objective function included penalties for transition time and the offspec product produced during transition.

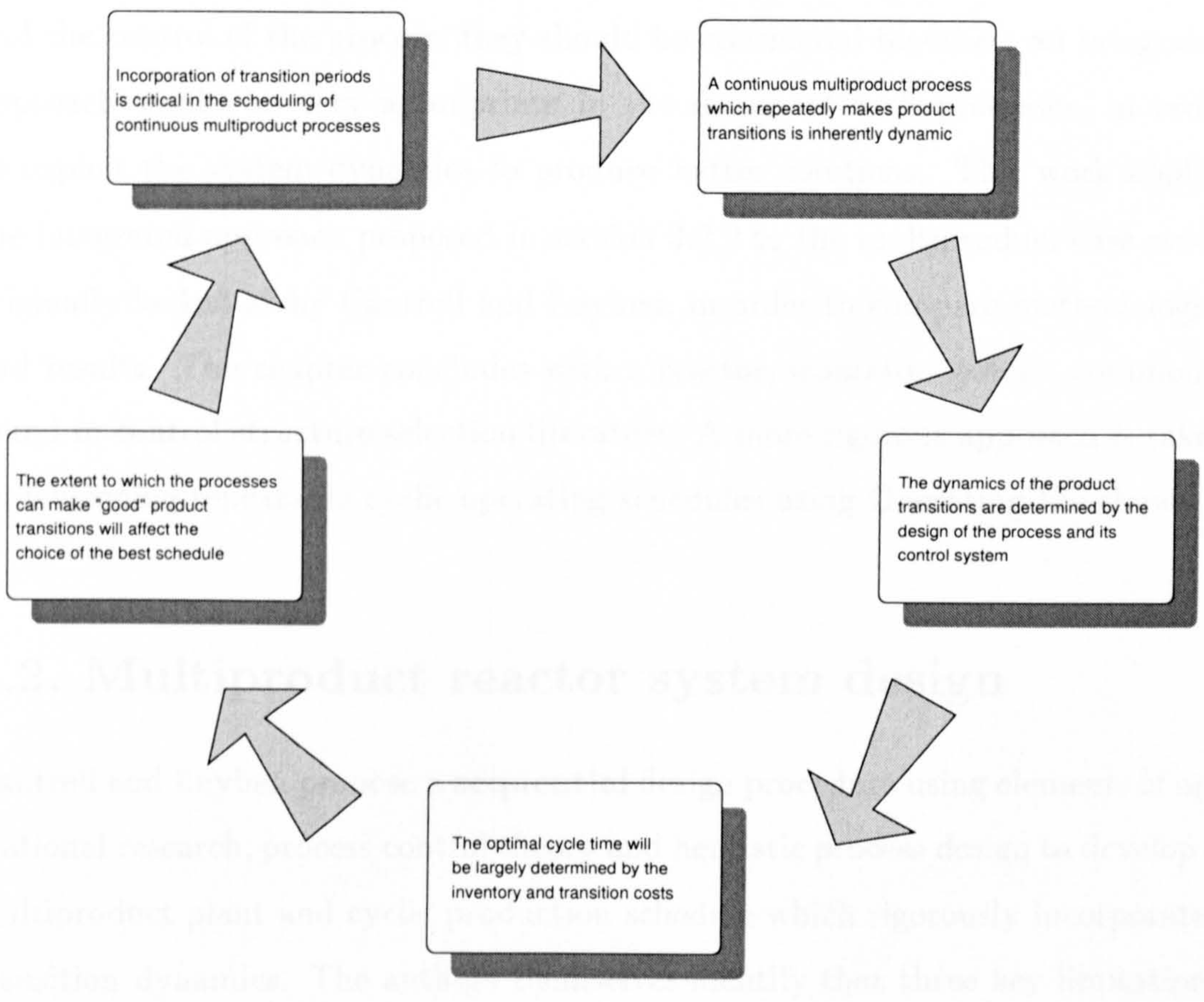


Figure 4.1: Interactions between process design, product transitions and production schedule

Rowe (1997) summarises this discussion in figure 4.1. In addition to this fundamental work on the design and operation of such processes, many extensions to these basic problems have been discussed in the literature: energy integration

(Lee and Reklaitis, 1995), combined maintenance and product scheduling (Dedopoulos and Shah, 1995), supply chain management (Wilkinson et al. 1996) etc. For reference, more comprehensive reviews in the area may be found in Rippin (1983, 1993) and Reklaitis (1990).

To summarise, continuous multiproduct processes are intrinsically dynamic, and product transitions are of critical importance in determining an economically optimum schedule. As product transitions are determined by both the design and the control of the process, they should be considered together: an integrated approach would be very appropriate in the design of such processes, in order to exploit the system dynamics to produce better solutions. This work applies the integrated approach proposed in section 3.2.2 to the multiproduct case study originally looked at by Cantrell and Luyben, in order to compare methodologies and results. The chapter concludes with a reactor/separator system, commonly found in control structure selection literature. A more rigorous approach is taken in developing repeatable cyclic operating schedules using Operating Envelopes.

4.2. Multiproduct reactor system design

Cantrell and Luyben propose a **sequential** design procedure using elements of operational research, process control theory and heuristic process design to develop a multiproduct plant and cyclic production schedule which rigorously incorporates transition dynamics. The authors themselves identify that three key limitations of the procedure presented are: a high simulation load to determine transition times; a potentially large set of possible schedules to enumerate; optimisation of the design variables.

However, since the nature of the product transitions is determined largely by

the design and control of the process, development of a truly optimal process operation should consider the process design as a degree of freedom. In other words, the process design and operation (i.e. schedule) should be considered together. The aim of Rowe's (1997) optimisation based methodology is to determine not only the design but also the *cyclic schedule* of this multiproduct process. The major assumption made at the outset is that if the cyclic constraints are rigorously enforced, operation may be based upon a single, optimal, infinitely repeatable production cycle. One important difference is that Rowe considered the dynamics of *both* the reactor *and* the storage vessels. This contrasts with the previous work which dealt only with the reactor dynamics and appended the storage behaviour based on a simplified cycle time model.

The work in this section revisits the example above in order to incorporate gain scheduling for the controllers and to investigate this "infinitely repeatable cycle" assumption. Multiple cycle simulations are then undertaken to test this assumption. A more robust approach in guaranteeing infinite repeatability, incorporating critical *ranges* rather than *points*, is then established and employed in the following section containing a multiproduct reactor/separator example.

4.2.1. Problem statement

A continuous stirred tank reactor (CSTR) is to be used to manufacture four grades of product B through the reaction scheme:

$$A \rightarrow B \qquad \Delta H \ -ve \tag{4.1}$$

Specifications on the four products ($B_1 \rightarrow B_4$) are summarised as follows. Purity requirements are given in terms of concentration of A:

Specification	B_1	B_2	B_3	B_4
continuous demand ($lbmol/hr$)	25	25	25	25
nominal purity ($lbmol/ft^3$)	0.0500	0.0375	0.0250	0.0125
purity upper bound ($lbmol/ft^3$)	0.0501	0.0376	0.0251	0.0126
purity lower bound ($lbmol/ft^3$)	0.0499	0.0374	0.0249	0.0124

Table 4.1: Product demand rates and purity requirements

- The CSTR produces one product grade at a time; continuous demand for all grades is met by storage of each product.
- The CSTR's operation is cyclic; each product is produced one per cycle.
- Any offspec material produced has an associated cost implication and *cannot* enter any product storage.
- A proportional-integral controller is used in the primary composition loop which controls the reactor composition within the reactor by adjusting the setpoint on a secondary temperature controller. This loop has a five minute deadtime.
- A proportional controller is used for the secondary temperature loop which receives its setpoint from the primary composition controller described above. It controls the temperature by manipulating the cooling water flow through the CSTR jacket. This loop has a one minute lag. The maximum allowable cooling water flow is set at four times the steady-state value.
- The CSTR is assumed as having constant volume; A feed rate determines the product rate for each product.

The objective is to minimise total annualised cost and meet product supply requirements by finding the optimal process design, controller settings and cyclic scheduling of the products. Unfortunately the problem as stated in the Cantrell and Luyben paper was in imperial units and due to missing critical information

(see section 4.2.3.5) conversion to S.I. was not possible.

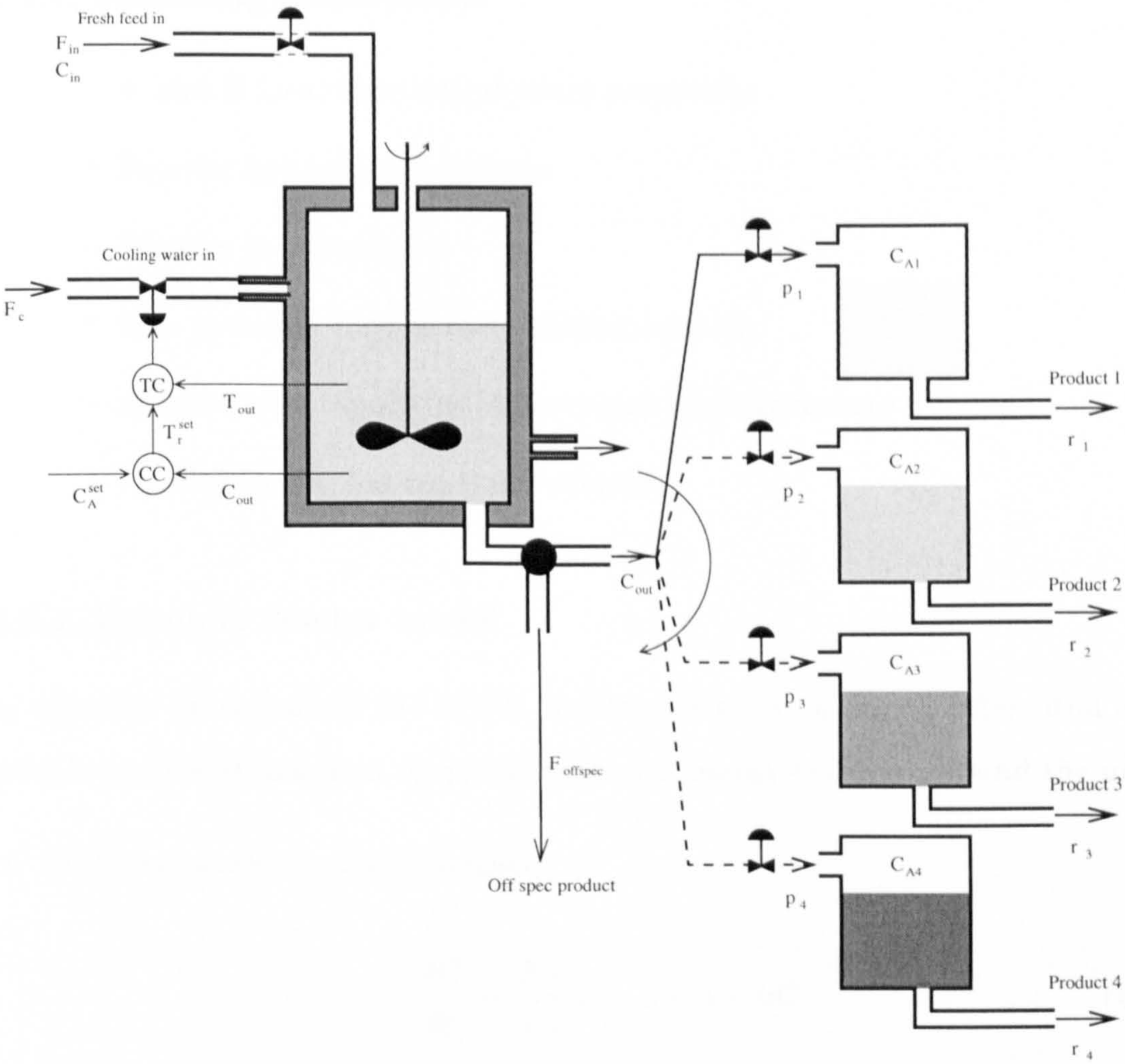


Figure 4.2: Multiproduct reactor case study

4.2.2. Process model

4.2.2.1. Modelling assumptions

- A and B have identical physical properties
- Reactor has constant volume
- Reactor is well stirred
- Low pressure, high density (liquid) system.
- Specific heat capacities independent of temperature
- Cooling jacket has constant volume

4.2.2.2. Dynamic reactor model

The dynamic behaviour of the CSTR is represented by a set of differential and algebraic equations based on dynamic mass and energy balances around the unit.

1. feed component material balance,

$$\frac{dC}{dt} = \frac{F_{in}}{V_R}(C_{in} - C) - kC \tag{4.2}$$

where C is concentration of A in the reactor ($lbmol/ft^3$), F_{in} is the volumetric feed rate (ft^3/hr), V_R is the reactor volume (ft^3), C_{in} is the concentration of A in the feed and k is the reaction rate (hr^{-1}).

2. reactor energy balance,

$$\frac{dT}{dt} = \frac{F_{in}}{V_R}(T_{in} - T) - \frac{\Delta H_R k C}{\rho C_p} + \frac{U A_H}{\rho V_R C_p}(T_c - T) \tag{4.3}$$

where T is the reactor temperature (^oF), T_{in} is the temperature of the feed, T_c is the jacket temperature (^oF).

3. jacket energy balance,

$$\frac{dT_c}{dt} = \frac{F_c}{V_J}(T_{c,in} - T_c) - \frac{UA_H}{\rho_J V_J C_{p,J}}(T_c - T) \tag{4.4}$$

where V_J is the jacket volume, F_c is the volumetric feed rate to the jacket and $T_{c,in}$ is the cooling water feed temperature.

The reaction rate, k, is defined by:

$$k = k_0 e^{\left\{\frac{-E_a}{R(460+T)}\right\}} \tag{4.5}$$

values for the reaction rate parameters and physical constants used in the above equations are given in table 4.2. The heat transfer area (in ft^2) between jacket and reactor , A_H, is defined by:

$$A_H = 4.65 V_R^{\frac{2}{3}} \tag{4.6}$$

The jacket volume (in ft^3) is defined in terms of the reactor volume as follows:

$$V_J = 1.55 V_R^{\frac{2}{3}} \tag{4.7}$$

Parameter	*Value*	*Parameter*	*Value*
k_0	$8.16 \times 10^{10}\, hr^{-1}$	E_a	$30000\, BTU/lbmol$
ΔH_R	$-30000\, BTU/lbmol$	R	$1.99 BTU/(lbmol\,^\circ R)$
C_{in}	$0.50\, lbmol/ft^3$	U	$150\, BTU/(hr\, ft^2\,^\circ R)$
T_{in}	$70\,^0 F$	$T_{c,in}$	$70\,^\circ F$
C_p	$0.75\, BTU/(lb_m\,^\circ R)$	$C_{p,J}$	$1.0\, BTU/(lb_m\,^\circ R)$
ρ	$50\, lb_m/ft^3$	ρ_J	$62.3\, lb_m/ft^3$

Table 4.2: Model parameters for multiproduct reactor case study

4.2.2.3. Controller modelling

As stated earlier, control over the reactor composition is achieved through two control loops: a primary proportional-integral composition controller and a secondary proportional temperature controller.

Composition controller Recall that there is a deadtime associated with the composition controller. In the model this is approximated by a number, n, of first order lags in series:

$$\left(\frac{t_d}{n}\right)\frac{dC_{dt,1}}{dt} = (C - C_{dt,1})$$
$$\left(\frac{t_d}{n}\right)\frac{dC_{dt,i}}{dt} = (C_{dt,i-1} - C_{dt,i}) \quad i = 2,..,n \tag{4.8}$$

where t_d is the deadtime and $C_{dt,i}$ is the outlet concentration of the ith lag block.

The proportional integral controller is then defined by the equations:

$$\xi_C = C_A^{sp} - C_{dt,n} \tag{4.9}$$
$$\tau_I \frac{d\xi_I}{dt} = K_c^C \xi_C \tag{4.10}$$
$$T_R^{sp} = K_c^C \xi_C + \xi_I \tag{4.11}$$

where ξ_C is the composition controller error, C_A^{sp} is the composition setpoint, ξ_I is the integral error, τ_I is the controller reset time, T_R^{sp} is the setpoint on the temperature controller and K_c^C is the composition controller gain. In the work which follows a value for n of twenty was used to give a reasonable approximation to the deadtime while avoiding an excessive increase in the size of the model.

Temperature controller The 1 minute lag in the temperature control loop is incorporated by the simple first order lag model:

$$\tau_T \frac{dT_R^{meas}}{dt} = (T - T_R^{meas}) \tag{4.12}$$

where τ_T is the lag time in the control loop and T_R^{meas} is the lagging temperature measurement. The cooling water flow rate F_c^c is calculated as follows:

$$\xi_T = T_R^{sp} - T_R^{meas} \tag{4.13}$$

$$F_c^c = K_c^T \xi_T \tag{4.14}$$

where ξ_T is the temperature controller error. Recall that the coolant flow is bounded between zero and four times the steady-state flow. The temperature controller output must therefore be clipped to ensure that it remains within these bounds. In doing this it is important to avoid introducing significant discontinuities into the model which can cause numerical problems during integration, such as failures when evaluating sensitivities or cycling across the discontinuity. Walsh (1993) presents integral freezing in a controller as an example of this. Smoothed clipping functions with continuous first derivatives (based upon those suggested by Walsh for achieving smooth integral desaturation) are therefore implemented:

$$K_{clip} = \begin{cases} 1 & \text{if } \epsilon < F_c^s < 1 - \epsilon \\ \frac{3}{\epsilon^2}(F_c^s)^2 - \frac{2}{\epsilon^3}(F_c^s)^3 & \text{if } 0 \leq F_c^s \leq \epsilon \\ \frac{3}{\epsilon^2}(1 - F_c^s)^2 - \frac{2}{\epsilon^3}(1 - F_c^s)^3 & \text{if } 1 - \epsilon \leq F_c^s \leq 1 \\ 0 & \text{otherwise} \end{cases}$$

where ϵ typically between 0.01 and 0.05 and F_c^s is the scaled controller output :

$$F_c^s = \frac{F_c^c}{F_c^{range}} \tag{4.15}$$

F_c^{range} is the cooling water flow range (four times the steady-state value). The actual cooling water flow to the process is therefore given by:

$$F_c = \begin{cases} F_c^{range} F_c^s K_{clip} & \text{if } F_c^s < 0.5 \\ F_c^{range} (1 - (1 - F_c^s) K_{clip}) & \text{otherwise} \end{cases} \tag{4.16}$$

4.2.2.4. Dynamic storage model

As mentioned previously, because the multiproduct plant problem is to be solved within a dynamic optimisation framework, the dynamics of the storage vessels must be considered explicitly in the model of the process.

The storage tanks are assumed to be well mixed, with no transfer of energy taking place. Storage tank i is therefore described by the following dynamic overall and component mass balances:

$$\left.\begin{aligned} \frac{dV_{store_i}}{dt} &= p_i - r_i \\ V_{store_i} \frac{dC_{A_i}}{dt} + C_{A_i} \frac{dV_{store_i}}{dt} &= Cp_i - C_{A_i} r_i \end{aligned}\right\} \quad i = 1 \text{ to } 4 \tag{4.17}$$

where V_{store_i} is the volume of storage tank i, p_i is the volumetric flow of feed to storage tank i from the reactor, r_i is the volumetric flow of product from storage tank i, C is the composition of product leaving the reactor and C_{A_i} is the composition of product leaving storage tank i.

The flowrate of off specification material, $F_{offspec}$, may then be calculated from an overall material balance on the reactor:

$$F_{offspec}(t) = F_{in} - \sum_{i=1}^{4} p_i(t) \tag{4.18}$$

The amount of off specification material generated during a cycle, $V_{offspec}$, is then

given by:

$$V_{offspec} = \int_0^{t_{cycle}} F_{offspec}(t)\, dt \tag{4.19}$$

where t_{cycle} is the cycle time of the process. It should be noted that for the purposes of this case study, the physical properties of the feed and product streams are assumed to be equal.

Addition of a make-up controller for numerical robustness The major tool for carrying out the integrated design and operation in this work will be dynamic optimisation. A model which accurately represents the physical system and is robust during the course of a dynamic simulation will not necessarily be numerically robust in the regions that may be entered during solution of the dynamic optimisation problem. It is therefore important to ensure that process models to be used for optimisation purposes are as numerically reliable as possible. As an aside, it should be noted that placing "soft constraints" (e.g. inequality constraints) on some variables such that numerically problematic regions lie outside the feasible region will not overcome the problem since such constraints are satisfied *only* at the solution. The storage model described above must therefore be reconsidered in this light.

As has already been stated, the profile over time for the exit flow is fixed and cannot be manipulated. During the course of a dynamic optimisation it is possible for a situation to arise in which the storage tank empties *and* product continues to be drawn at a rate which is greater than the flow of feed to the storage tank. This will cause the holdup to violate variable bounds and the equations describing the storage tank concentration to become physically meaningless. The dynamic storage may be made robust by the addition of a proportional level controller on

the tank as follows.

$$Q_{mu} = \begin{cases} 0.0 & \text{if } V_{store_i} > V_{store_{i,l}} \\ K_c(V_{store_{i,l}} - V_{store_i}) & \text{otherwise} \end{cases} \tag{4.20}$$

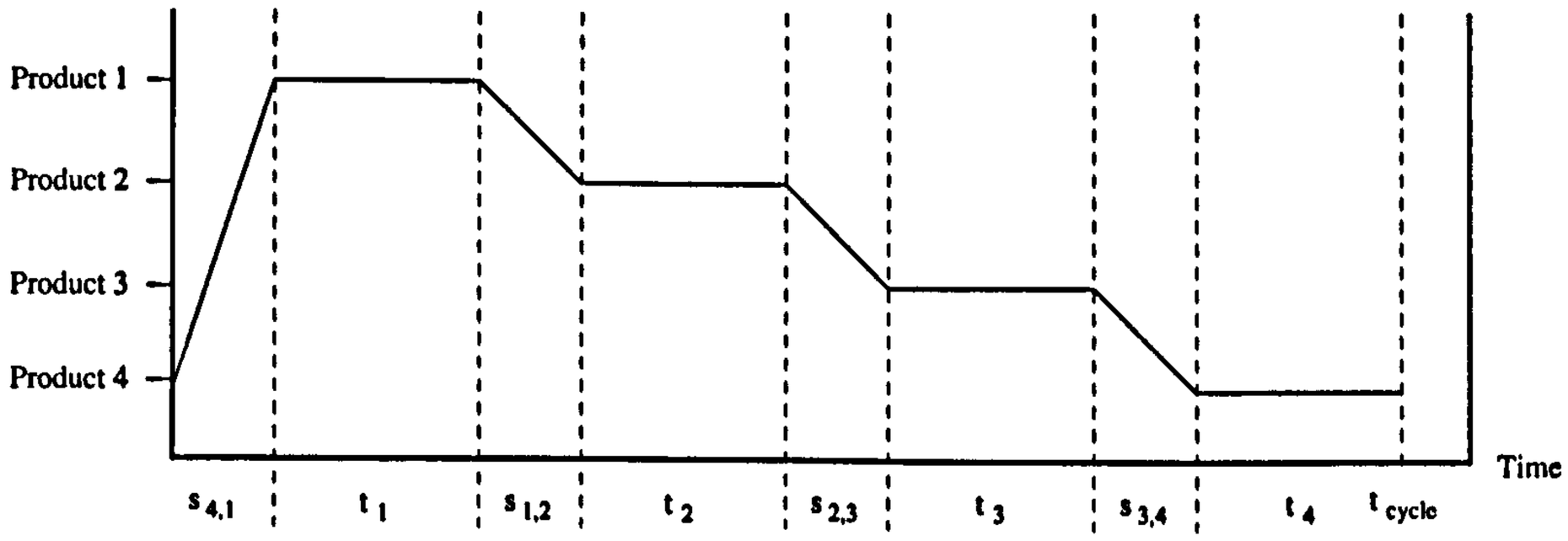
where K_c is a controller gain and $V_{store_{i,l}}$ is a lower limit on the storage tank holdup. The level controller ensures that the holdup is at least at $V_{store_{i,l}}$ during the optimisation by using a "dummy" make–up stream (Q_{mu}) with a concentration equal to that of the tank itself. A path constraint on the tank holdup ensures that the proportional controller does not operate at the solution point.

4.2.3. Optimisation

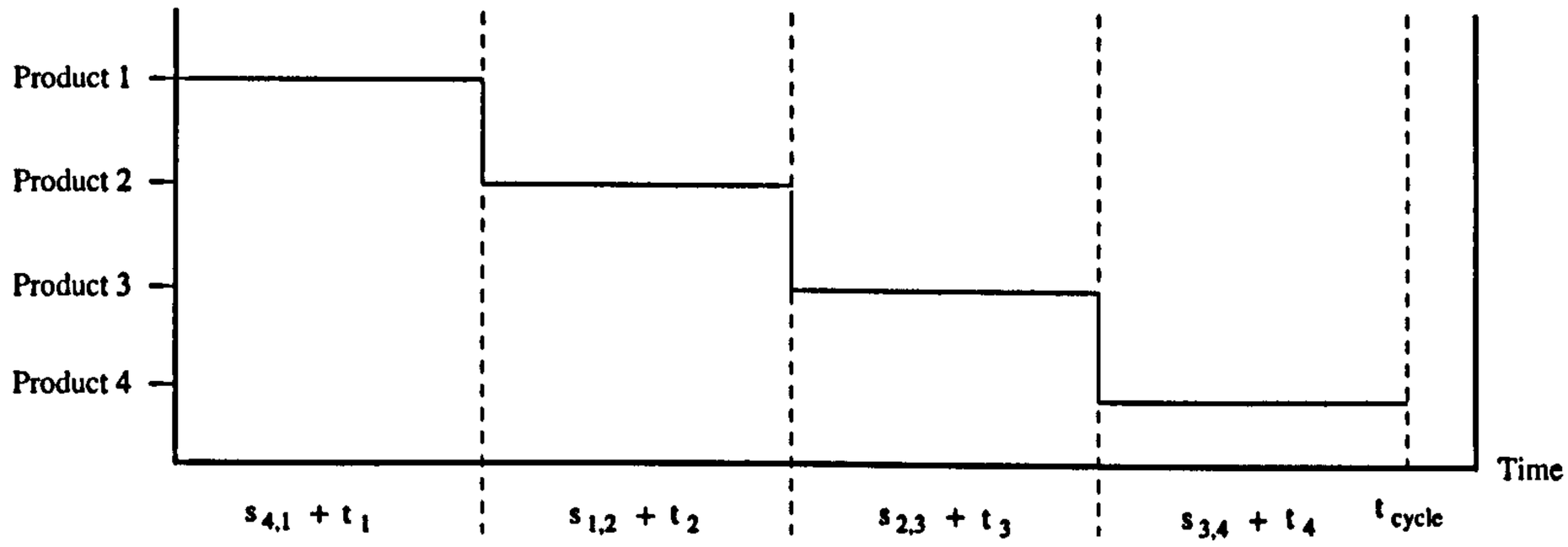
Recall that the operation of the process is cyclic and that each product is manufactured once per cycle. If a product is not in production, then a transition period must be occurring.

Graph (a) in figure 4.3 shows a typical time profile for the four product case study as described by the cycle time model used by Cantrell and Luyben. t_i indicates the length of production period i and s_{ij} indicates the transition period from product i to j.

Graph (b) of figure 4.3 shows the corresponding composition setpoint profile. Each of the products to be manufactured has an appropriate setpoint value. Since four products are to be produced, the operating cycle may be split into four periods, each with the setpoint required to make one of the products. In this case study it is therefore the *sequence in which the composition setpoint changes which determines the process schedule.* By including the setpoint profile for the cycle as a time varying optimisation variable (control), the optimal production schedule may be integrated into the optimisation problem.



(a) process cycle showing product transitions



(b) process cycle showing underlying setpoint changes

Figure 4.3: Time profile for system described by cycle time model

The optimisation framework must also be flexible enough to enable:

- on-spec products to be fed to storage for any production schedule,
- offspec material produced during transitions to be diverted as waste.

This is achieved by subdividing each of the four periods shown in graph (b) figure 4.3 into a transition period and a production period (as shown in graph (a) of the same figure). Manipulating the product flows (p_i) allows the storage tanks to be filled with on-spec product during the production periods. Offspec product produced in the transitions may be prevented from entering the storage tanks by setting the product flows to zero.

The framework set out above will form the basis of the dynamic optimisation problem to be solved.

4.2.3.1. Objective function

The objective function for this case study is to minimise the annualised cost of the process. This cost is made up of three constituent parts:

1. capital cost of the reactor,

2. capital cost of the storage,

3. transition costs.

Capital costs The costs of the reactor and the storage vessels are related to the total molar holdups in the respective vessels according to the following relationships:

$$C_{CSTR} = 2523 \left(H_{reactor}\right)^{0.62} \tag{4.21}$$

$$C_{store_i} = 2523 \left(H^{max}_{store_i}\right)^{0.62} \quad i = 1 \; to \; 4 \tag{4.22}$$

where C_{CSTR} is the reactor cost in dollars, C_{store_i} is the cost in dollars of the ith storage tank, $H_{reactor}$ and $H^{max}_{store_i}$ are the maximum molar holdups in the reactor and ith storage vessel respectively. It may seem odd that reactors and storage vessels are costed identically — in fact Cantrell and Luyben comment that in many processes a reactor will cost more than a storage tank of the same size due to more expensive materials of construction, heat transfer, and mixing equipment and instrumentation. Indeed they adjust the original objective function later on in their paper accordingly; however for purposes of comparison with Rowe (1997), the unadjusted version is used here. The total annualised capital cost of the process is therefore given by:

$$C_{capital} = \frac{1}{3}\left\{\sum_{i=1}^{4} C_{store_i} + C_{CSTR}\right\} \tag{4.23}$$

The $\frac{1}{3}$ pre-multiplier is a capital charge factor (CCF) which approximates "*the time value of money*" (Douglas, 1988), and converts the total capital cost to an annualised equivalent.

Transition costs In addition to the capital cost of the processing equipment and storage vessels, there is a cost incurred for any material produced during the operating cycle which is not to specification. This cost represents a devaluation for being offspec or a cost for extra processing required to achieve the desired product. The offspec is charged at $\$0.1/lbmol$. The amount of offspec produced per cycle, $V_{offspec}$, was defined earlier in equation 4.19. If it is assumed that the process operates continuously, in a given year there will be:

$$N_{cycle} = \frac{365 \times 24}{t_{cycle}} \tag{4.24}$$

cycles, where t_{cycle} is the cycle time of the process in hours. The total cost per year for offspec is therefore equal to:

$$C_{transition} = 0.1\, V_{offspec}\, \rho_m \left(\frac{8760}{t_{cycle}}\right) \tag{4.25}$$

where ρ_m is the molar density of the product.

The total annualised cost for the process is the sum of transition and capital costs and the objective function is given by:

$$\min_{u(t),d} \{C_{capital} + C_{transition}\} \tag{4.26}$$

4.2.3.2. Optimisation parameters

The aim in solving this optimisation problem is to determine the optimal process design and cyclic production schedule. This is achieved by manipulation of a number of degrees of freedom in the process:

- Time varying parameters (controls).
  - Reactor setpoint profile
  - Product flowrate profiles to each of the storage tanks
  - Gains for the composition and temperature controllers
- Process initial state.
  - Initial state of the storage system only (reactor initial state is determined by the composition setpoint at t_0).
- Time invariant optimisation parameters.
  - Reactor feed
  - Reactor residence time
  - Reset time for the composition controller
- Equipment dimensions.
  - Reactor size
  - Storage vessel size

4.2.3.3. Time discretisation and control parameterisation

Independent interval structures (see appendix A) exist for the piecewise linear reactor setpoint and product flowrate profiles. As can be seen from figure 4.4,

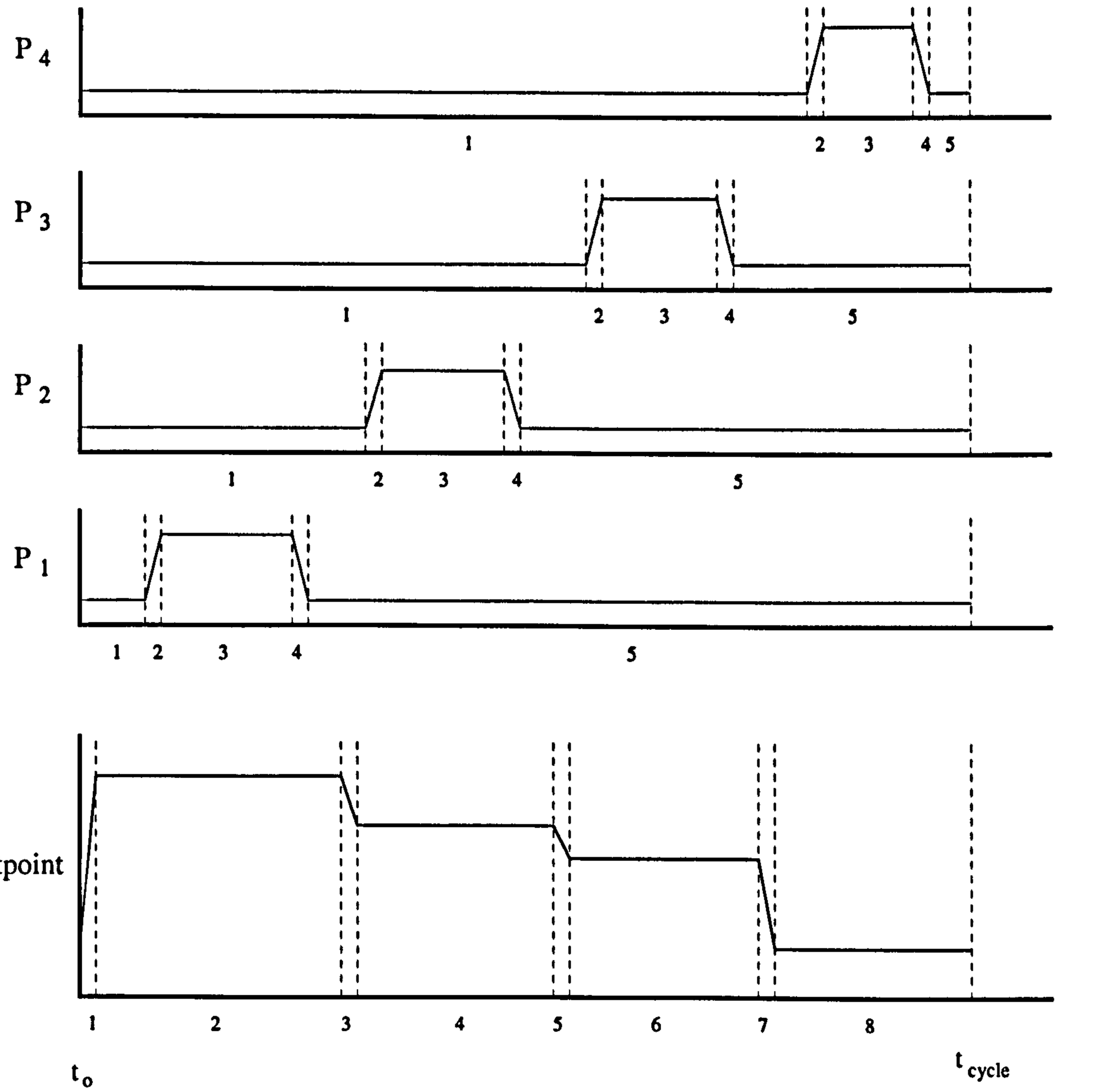


Figure 4.4: Time discretisation used in the multiproduct plant case study

the cycle time is discretised into eight intervals for the setpoint point profile, and five for each of the product flowrate profiles. The discretisation essentially consists of variable length intervals required for the transition-production form of the cycle, each of which is separated by fixed intervals of length 10^{-3} hours. These shorter intervals approximate valve dynamics. Four intervals, coinciding with the production periods, are used for the piecewise constant gain profiles.

Note that as the demand and ramping intervals are known, the area under the trapezium in each product profile may be calculated directly in the model.

The optimiser then need only find the initial switching time (the first point on the trapezium) and leaves the model to determine the other three corresponding times. Obviously, if the demand and ramping times are uncertain, the optimiser will have to find these extra points.

4.2.3.4. Constraints

Purity constraints These must be enforced for all times in the cycle and are therefore implemented as inequality path constraints. It is important to note that these constraints are applied to the products flowing *from the storage tanks.* In their approach, Cantrell and Luyben apply these constraints to the reactor effluent because the storage dynamics are not considered. This may prove to be conservative, as potentially beneficial product buffering resulting from the storage holdup is not considered.

$$
\begin{aligned}
0.0499 &\leq C_{A_1} \leq 0.0501 \\
0.0374 &\leq C_{A_2} \leq 0.0376 \\
0.0249 &\leq C_{A_3} \leq 0.0251 \\
0.0124 &\leq C_{A_4} \leq 0.0126
\end{aligned} \tag{4.27}
$$

Storage volume constraints A path constraint is applied to the volumes in the storage tanks ensuring that they never run dry. Hence the continuous demand for each product may be met over all time.

$$V_{store,i} \geq 0 \qquad \forall i = 1..4 \tag{4.28}$$

This constraint also ensures that the "dummy make up stream", Q_{mu}, remains inactive at the solution point.

Offspec flow constraint A similar path constraint must be applied to the flow of offspec material to prevent the storage tanks from drawing material from the offspec "sink".

$$F_{offspec}(t) \geq 0 \tag{4.29}$$

Cyclic Constraints The following equality endpoint constraints ensure cyclic operation by forcing the compositions and volumes in the storage vessels to be equal at the beginning and end of each cycle:

$$\left.\begin{array}{l} C_{A_i}(t_0) = C_{A_i}(t_{cycle}) \\ V_{store_i}(t_0) = V_{store_i}(t_{cycle}) \end{array}\right\} \quad i = 1 \; to \; 4 \tag{4.30}$$

To avoid introducing excessive complexity into the problem by specifying cyclic endpoints on each of the twenty-six states that exist in the reactor, the simplification is made that the reactor is at steady state at time zero in the cycle.

Implicit constraints A variable $interval_i$ is defined in the model as the times between the production campaigns of each of the products. It is bounded in the optimiser as being greater than zero, ensuring that two products cannot be produced as the same time. A similar method is used to ensure that product cannot flow to two storage tanks simultaneously.

4.2.3.5. Optimisation results and discussion

Details of the implementation and solution of the dynamic optimisation problem is given in section 3.3.3. The values obtained for the design parameters in the reactor and its control systems are shown in table 4.3. The piecewise constant gain profiles are given in Figure 4.5. Figures 4.6 and 4.7 show the profiles over time of the reactor concentration and product storage compositions. Figure 4.8 shows the variation of the four product storage volumes over time.

Parameter	*Value*
τ_I	$0.026\,hr$
F_{in}	$41.15\,ft^3/hr$
τ_R	$1.03\,hr$
t_{cycle}	$5.02\,hr$

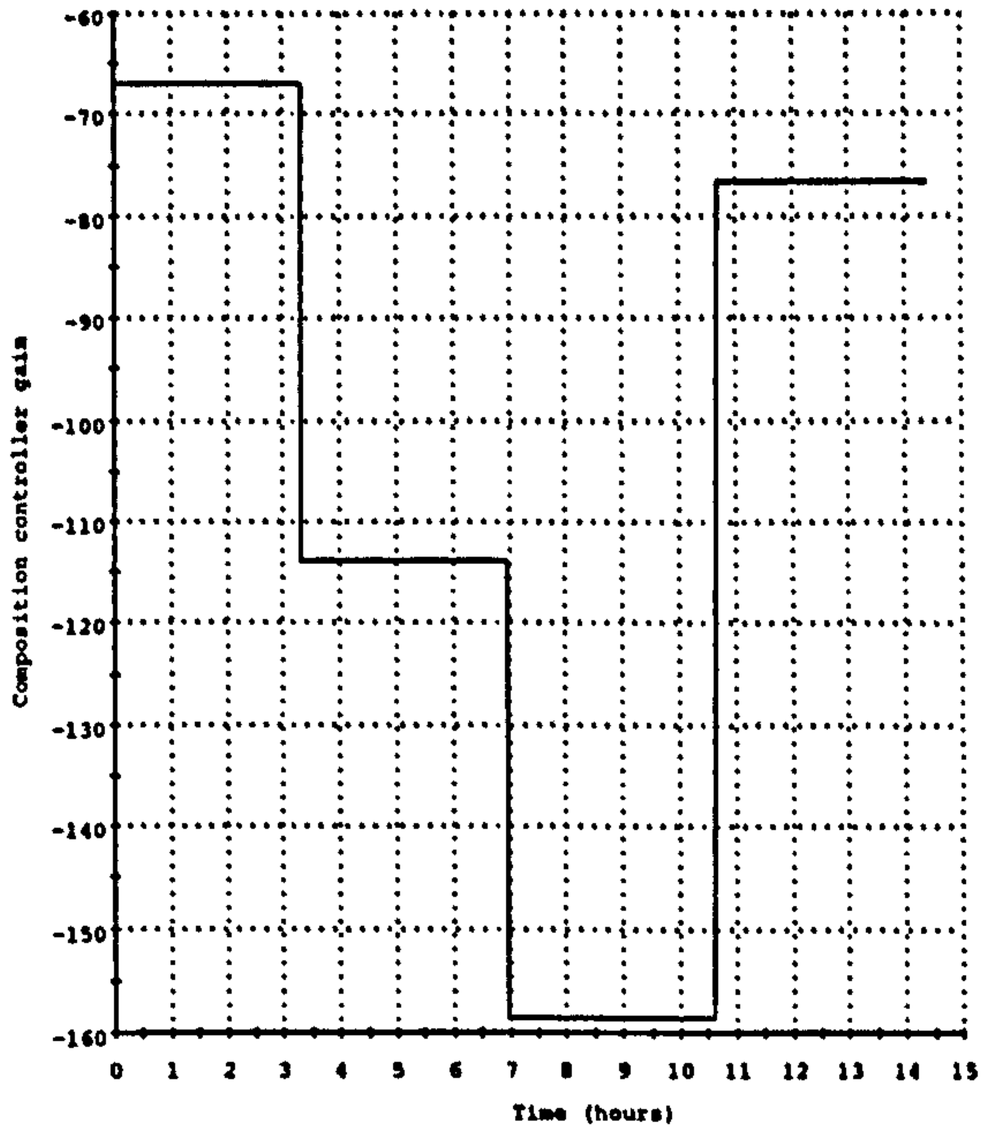
Table 4.3: Reactor parameters for economically optimal multiproduct plant

The optimum annualised cost for the system was found to be **$138446**. The cycle time for the schedule was $5.0183\,hours$. The product schedule chosen, shown in figure 4.6, was $B_2 \rightarrow B_4 \rightarrow B_3 \rightarrow B_1$. Due to a lack of a molar density value in the Cantrell and Luyben paper, a value of $3.0\,lbmol/ft^3$ was used in equation 4.25 so as to obtain comparable results. Assuming the molar density to be approximately correct, the cost obtained through optimisation is some 44% lower than that found by the sequential procedure.

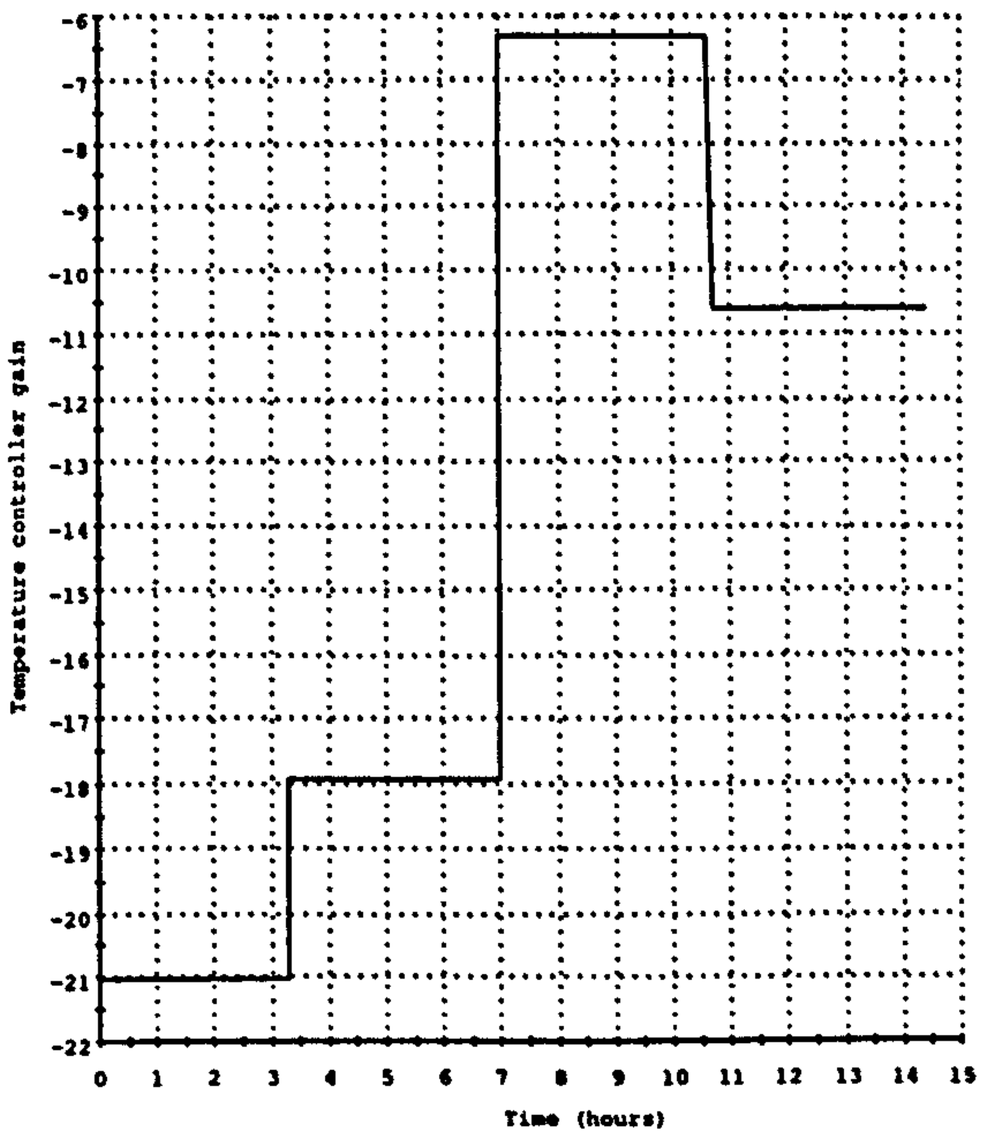
Cantrell and Luyben do not fully exploit the inherently dynamic nature of this problem, as can be seen by the application of purity constraints on the reactor effluent. In their paper, they wait for the reactor effluent composition to converge to a steady state before "starting" a production period, thus unnecessarily increasing transition times. In the optimisation based approach, the reactor composition is always oscillating as shown in figure 4.6, and it is consequently easier to change operating point (i.e. more responsive) which helps reduce the cycle time and therefore lower storage costs, reinforcing the comments made by Backx et al. (1998) in section 2.3 concerning *intentional dynamics*.

In figure 4.8, the minimum value of stored product for all four products (which occurs when each of tanks is about to be re-charged from the reactor) is somewhat greater than zero. Although it might initially seem that this would lead to larger storage volumes, much shorter transition times are possible because of the additional attenuation of deviations in the reactor composition. The overall result of this is to reduce the total operating cycle length and enable smaller storage tanks to be used. The dynamic nature of the storage tanks is also taken into account, thus utilising the buffering effect they provide: a great deal of oscillation in the effluent composition may be tolerated without violating the purity constraints being enforced in the storage vessels. Thus, the residence and cycle times are further reduced: the residence time for Cantrell and Luyben's reactor was 6 *hours*, compared to 1.03 *hours* for the proposed approach. On another note, the ratio between reactor volume and storage tank volume may be regarded as a metric of responsiveness with respect to that product grade: the lower the buffer volume in each tank, the less time the system needs to reach the appropriate production steady state. This is discussed again in section 5.4.1.1.

Cantrell and Luyben dynamically simulate this process for a range of residence times/reactor sizes, and tune the controllers accordingly, using Ziegler-Nichols tuning methods. The schedule with the minimum transition time is chosen as the optimum. The heuristics used in this approach often lead to erroneous decisions being made, e.g. the optimum schedule chosen was excluded by Cantrell and Luyben based on the heuristic that it will, "...require moving back and forth through the same operating region more than once." The schedule does however avoid the large jump from operating point B_4 to B_1 required by the Cantrell and Luyben schedule.



(a) Composition controller gain



(b) Temperature controller gain

Figure 4.5: Gain Scheduling

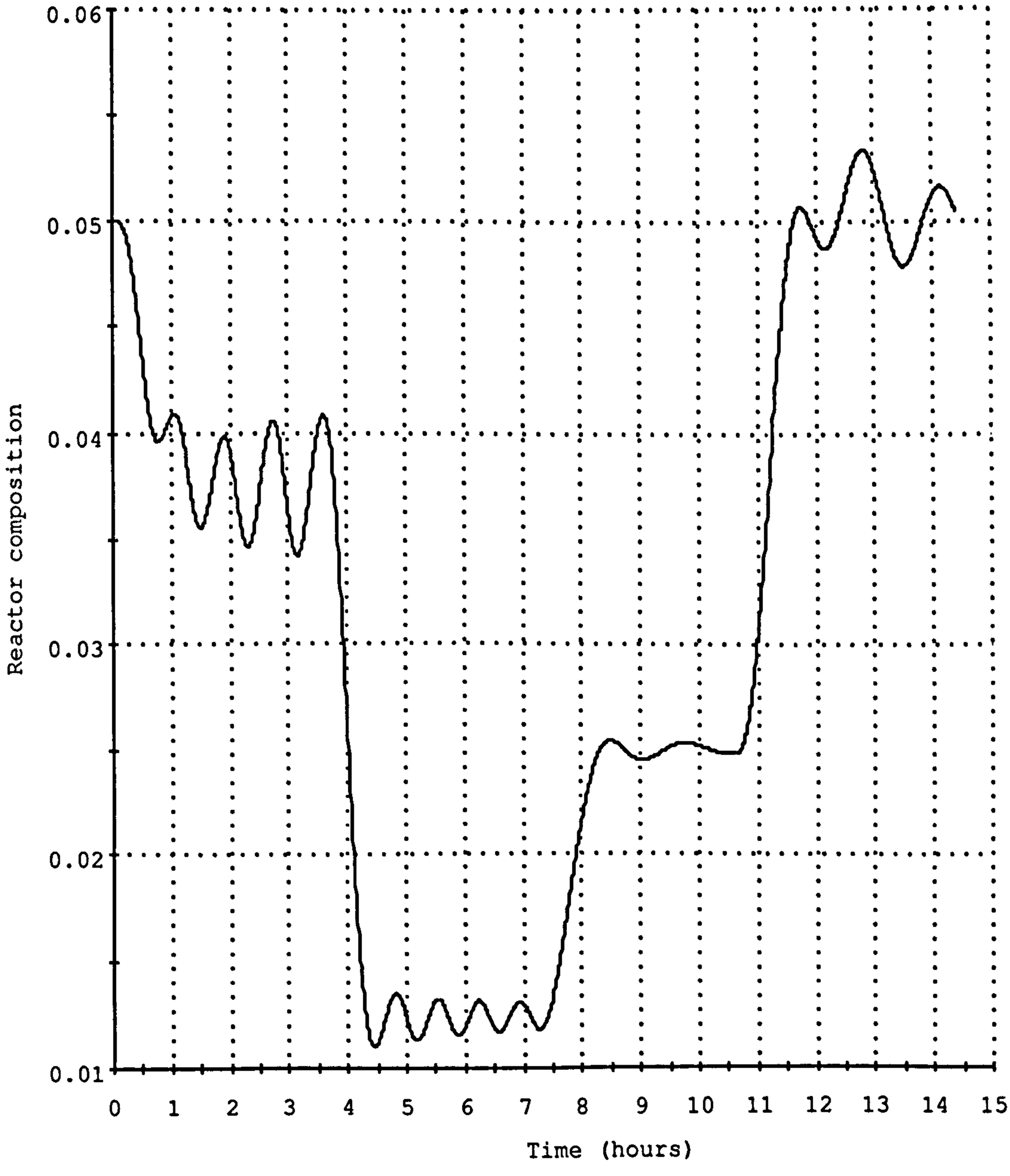
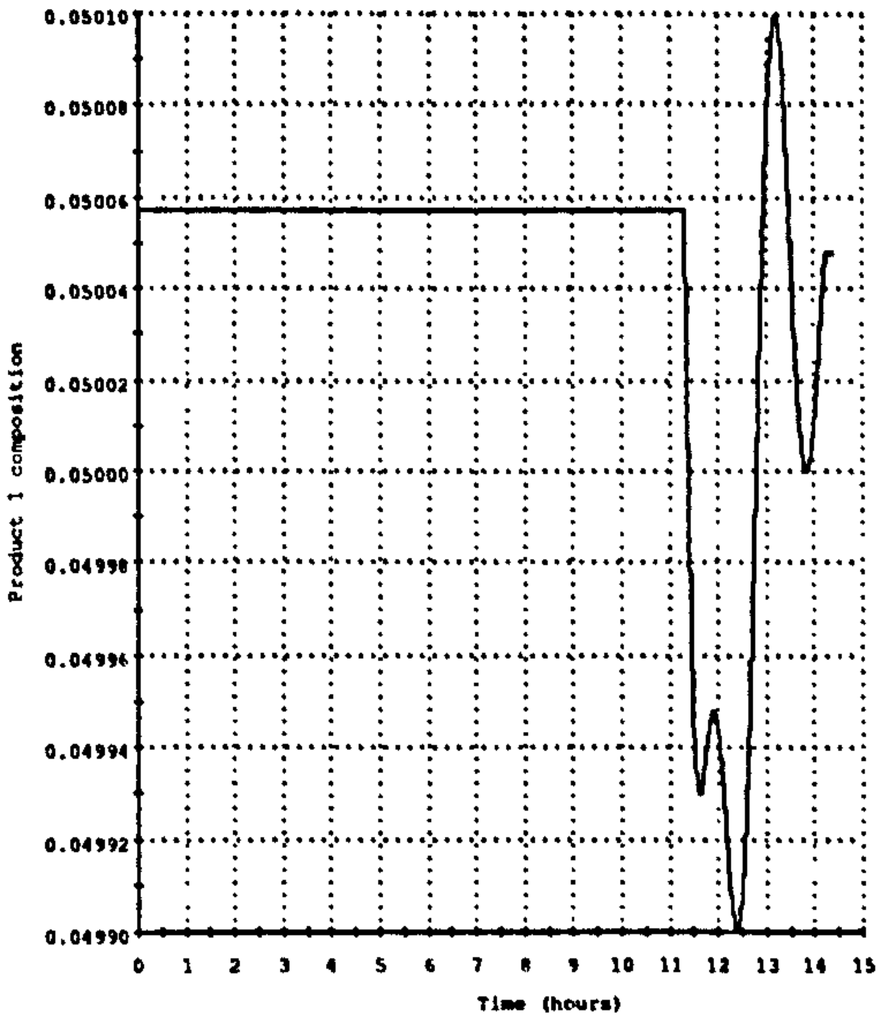
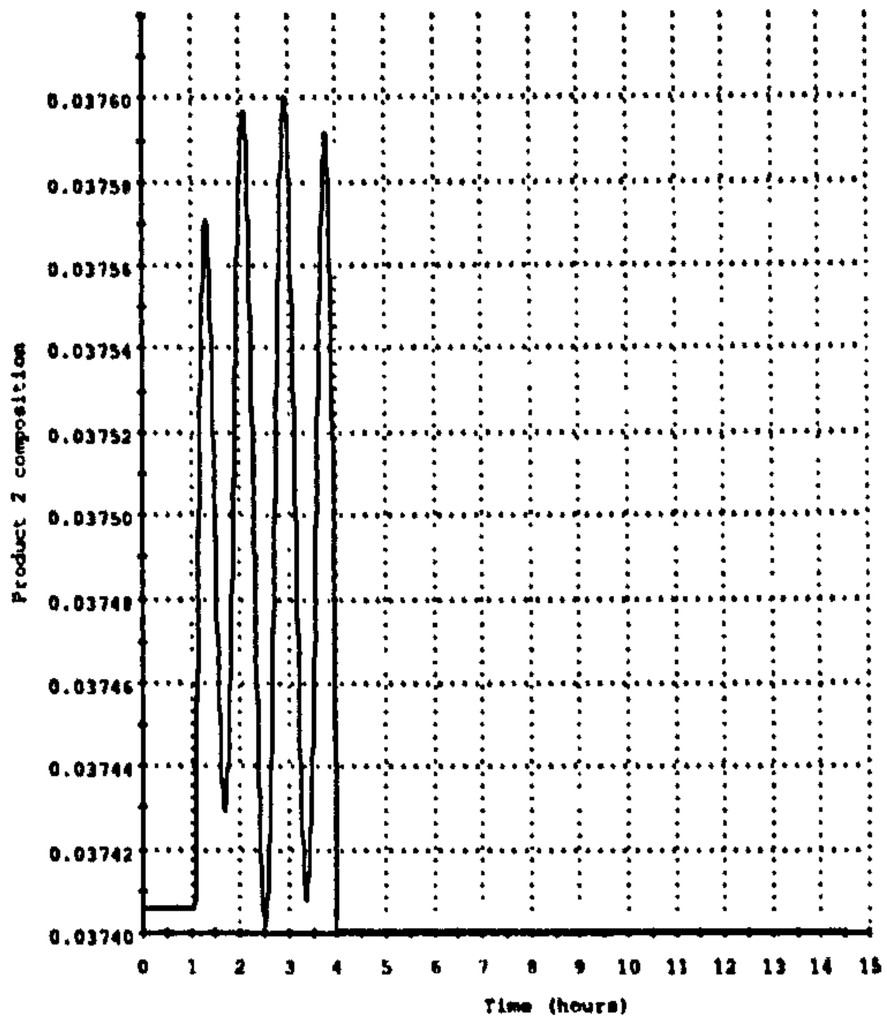


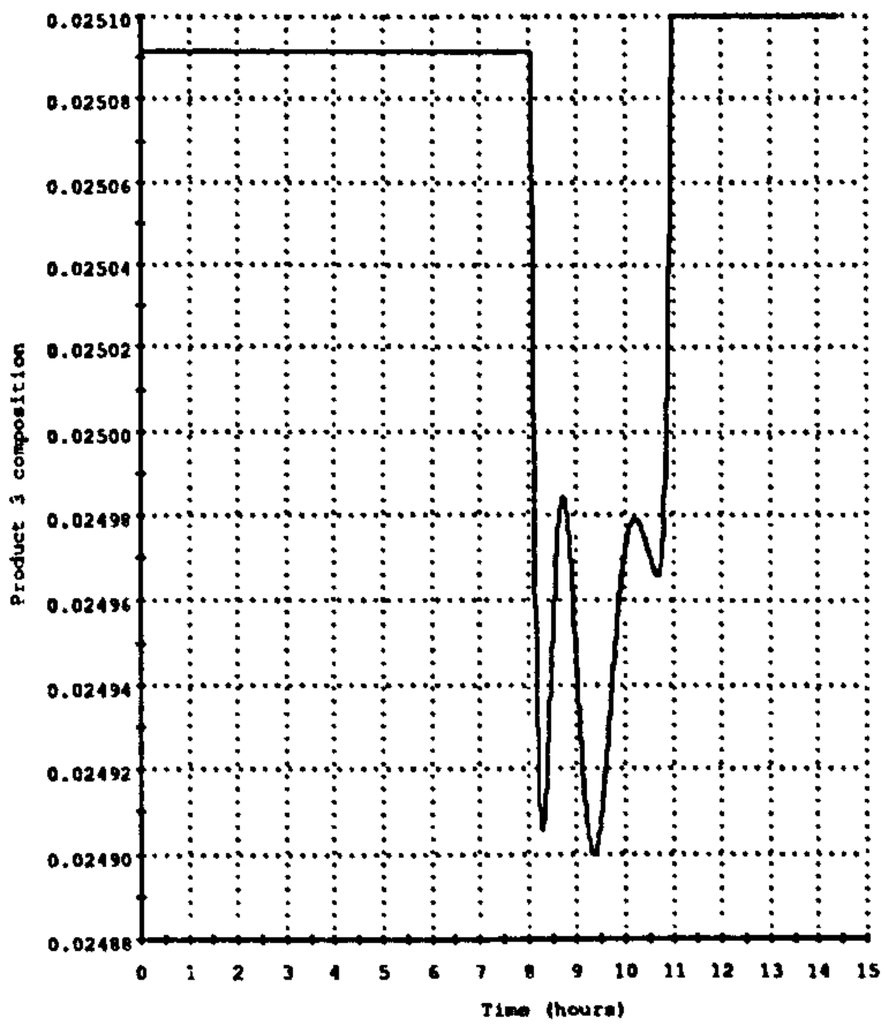
Figure 4.6: Reactor composition



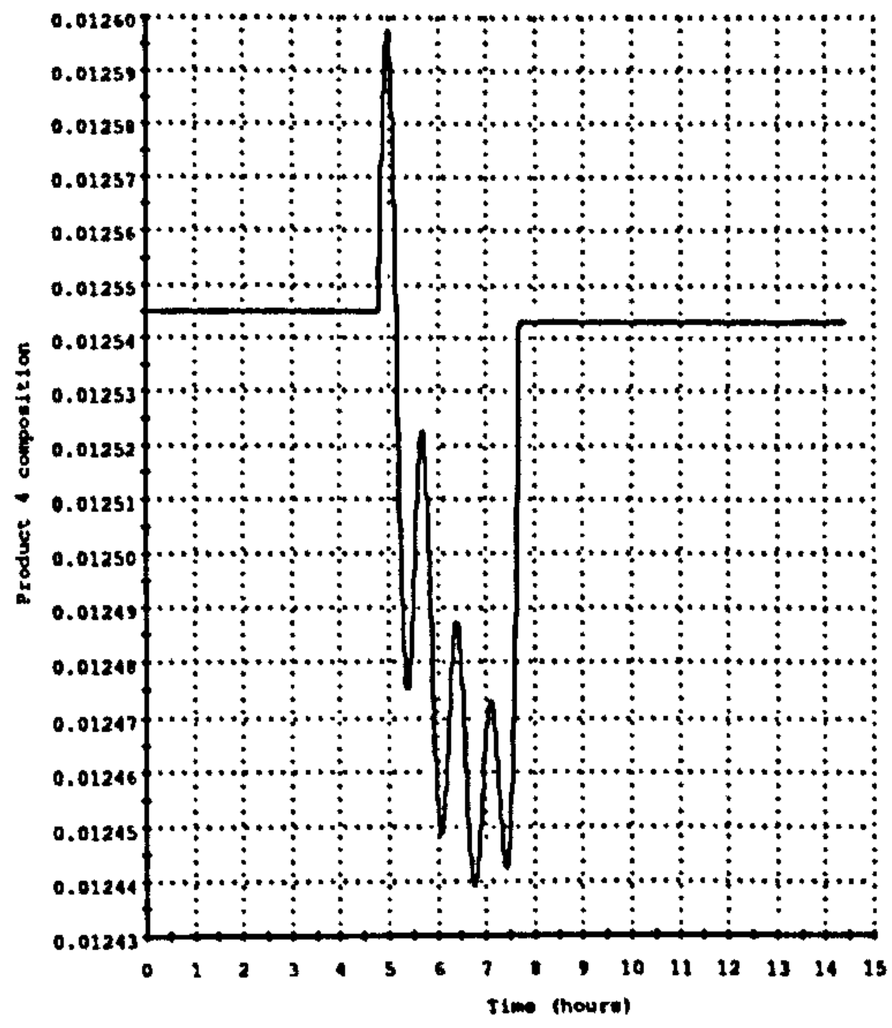
(a) Storage tank 1 composition



(b) Storage tank 2 composition

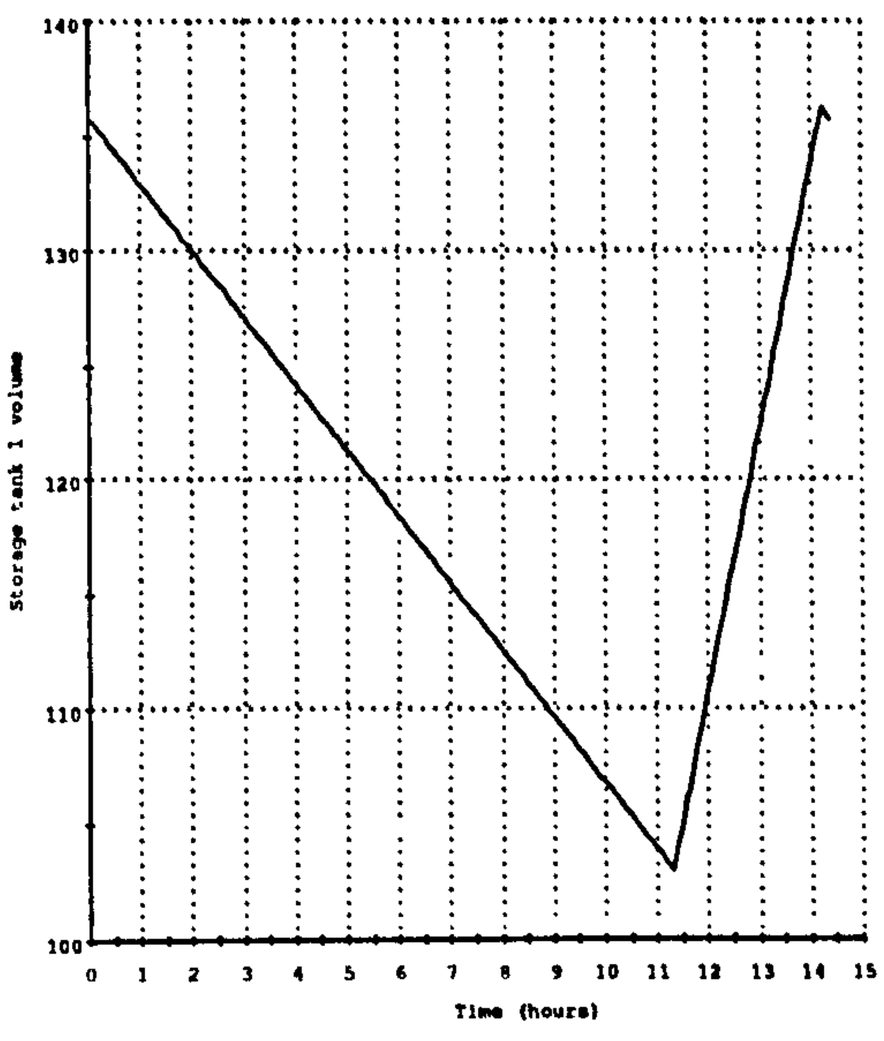


(c) Storage tank 3 composition

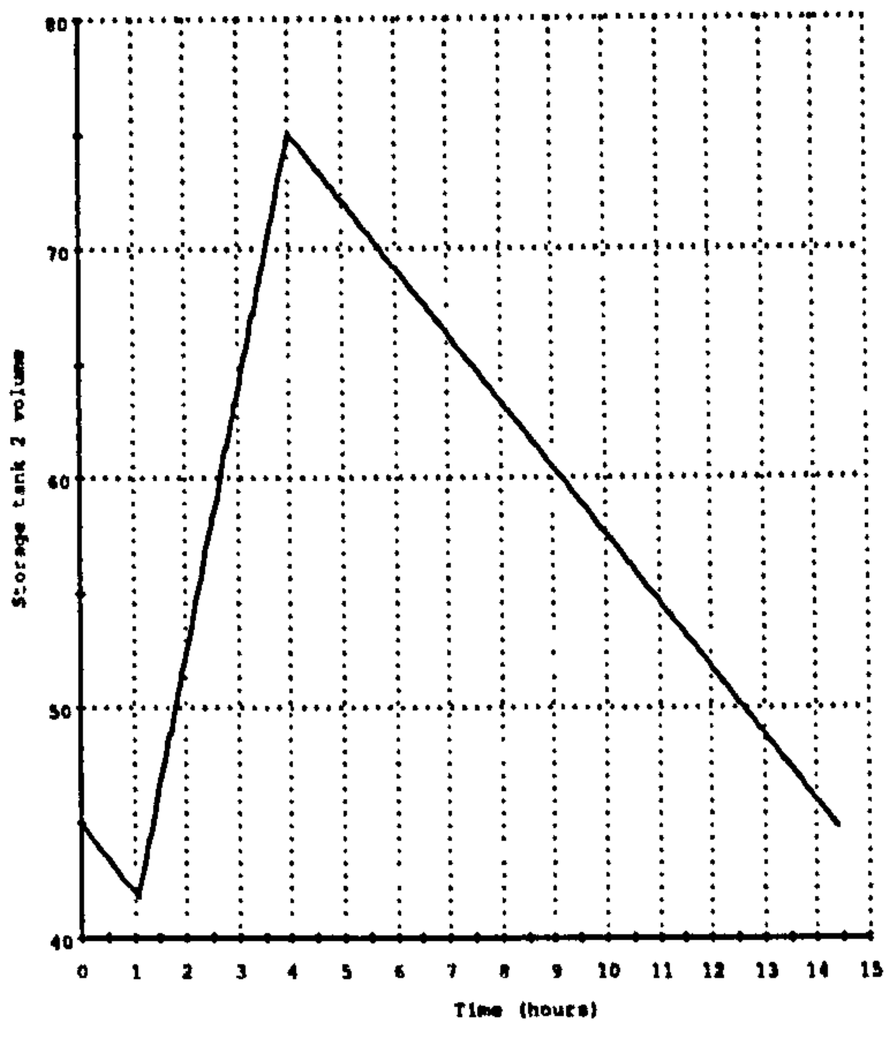


(d) Storage tank 4 composition

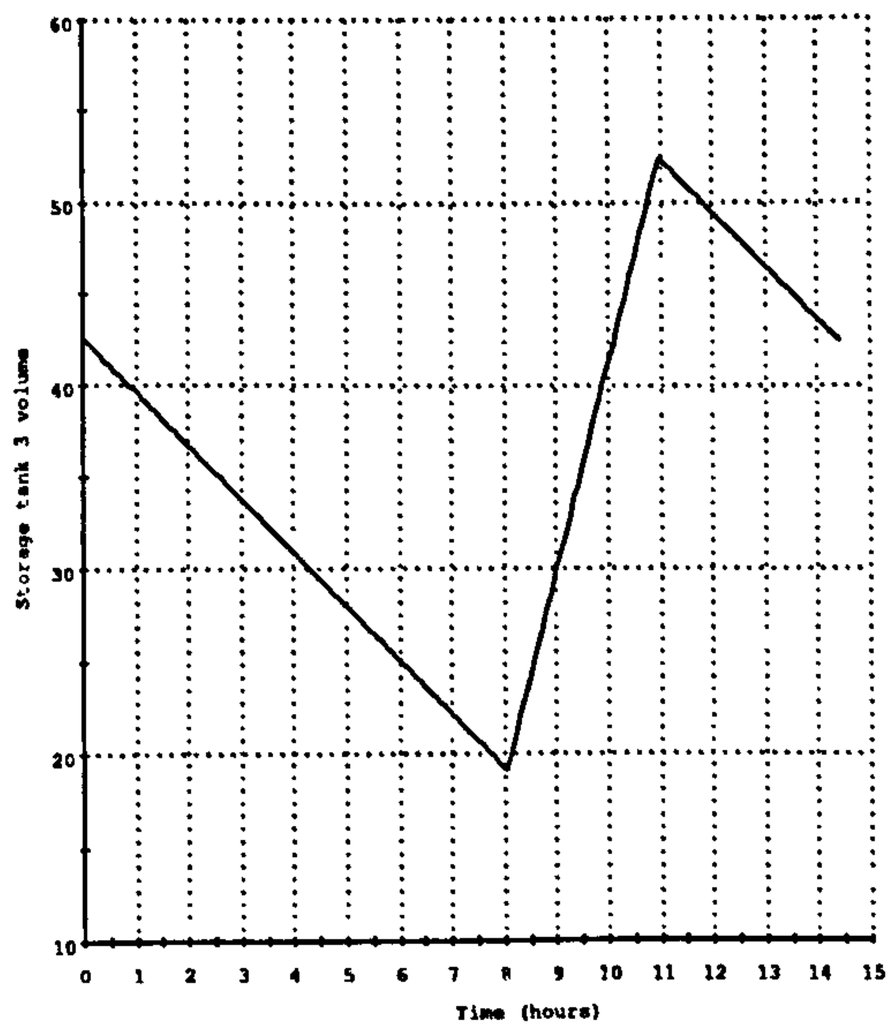
Figure 4.7: Storage compositions



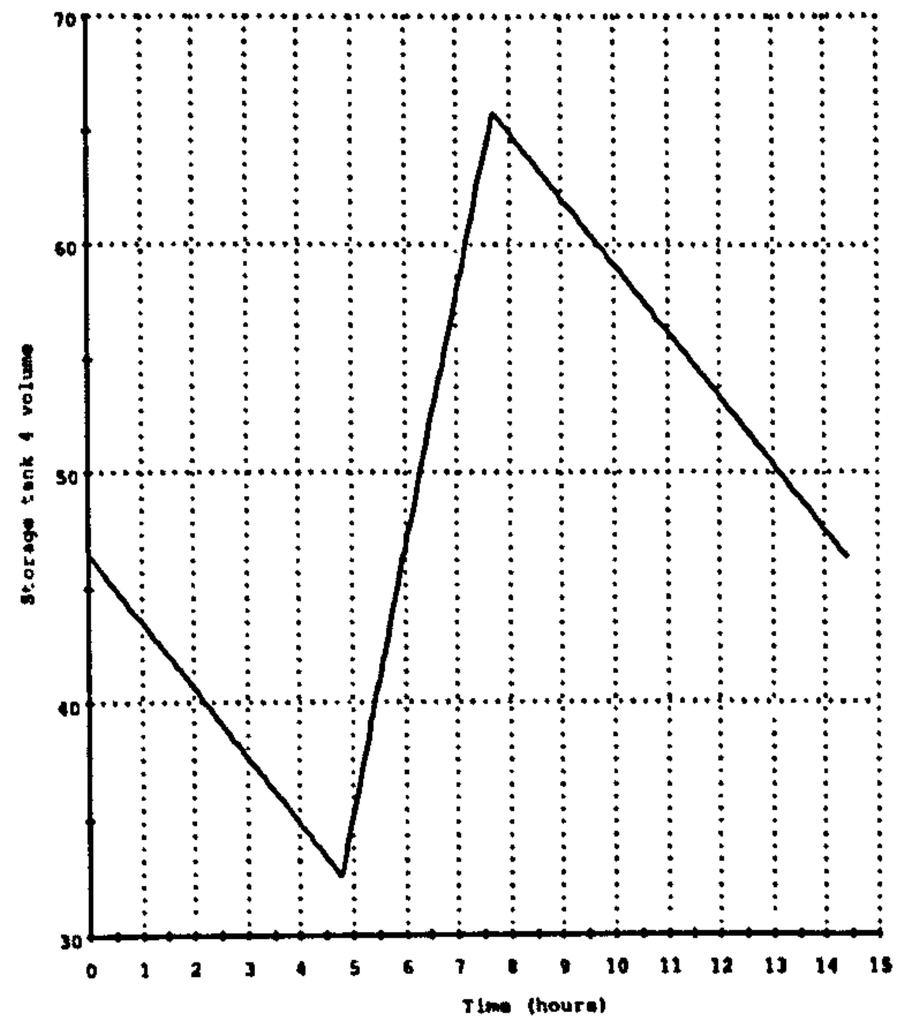
(a) Storage tank 1 volume



(b) Storage tank 2 volume



(c) Storage tank 3 volume



(d) Storage tank 4 volume

Figure 4.8: Storage volumes

4.2.4. Remarks

The optimisation framework for cyclic operation developed in section 4.2.3 has been successfully applied. A number of key qualitative differences between the solution obtained by optimisation and by Cantrell and Luyben were discussed, the most important being that consideration of only the reactor dynamics and subsequent application of purity constraints on the reactor product is highly conservative. In this instance, the production phase for a given product can only begin when the reactor concentration reaches and remains within the specifications given for that product. By comparing the responses of reactors with different residence times, Cantrell and Luyben determined that a larger reactor which made smooth transitions to the setpoint was most suitable. Imposing this form on the reactor response would seem greatly to reduce the scope for further optimisation of the design. A problem formulation which simultaneously considered both the storage and reactor dynamics has allowed economically superior solutions to be found.

One possible criticism of the optimisation based solution is that stability seems to have been sacrificed for a better economic optimum. Since Cantrell and Luyben wait for smooth transitions to steady states, stability is almost guaranteed as a result of the slightly different nature of their problem definition. As no stability criteria are included in this optimisation based approach, it is unsurprising that solutions contain unstable regions. There are two ways of tackling this problem. The first is explicitly to introduce some kind of condition for stability, as a constraint, into the optimisation. The other is to treat this economic optimum as the maximum achievable limit, adding some sort of tolerance (say a 10% increase) as a constraint, and then re-optimising using a stability based objective function, e.g. Integral Square Error (see Figure 4.9).

A major assumption made in this work is that all material generated during a transition period, approximately a fifth of everything produced by the plant, is labelled "offspec" and diverted to waste. Perhaps one could recycle it to the reactor or even allow some offspec material to enter the product storage tanks, carefully monitoring for any purity path constraint violations. Thus, by allowing the optimiser to decide this tolerable amount, the buffering effect of the storage tanks is fully exploited. Yet another objective function to be considered is the minimisation of the amount of offspec produced. Just before a transition is due to occur, the reactor could be emptied to some minimum inventory level by cutting its feed. As soon as the reactor composition satisfies a purity constraint (which could be an optimisation parameter), fresh reactant feed to the reactor could be resumed and product of the appropriate composition would enter storage.

The major flaw in the formulation was found to be the cyclic constraints on the storage states. Recall, it is assumed that if these are satisfied, the cycle is guaranteed to be infinitely repeatable. However, parametric sensitivity was found when the design variables were verified by the dynamic simulator. As shown by figure 4.7, the compositions do not return *precisely* to their original values, which causes doubt as to whether the results are valid for multiple cycles. Hence the design variables reported in table 4.3 and figure 4.5, minimising the cost, were tested in a simulation running for 2 cycles. The purity path constraints *and* the cyclic composition constraints were rapidly violated in the second cycle. Even a 2 cycle run using optimisation results from minimising the ISE suffered from similar (but smaller) violations. In light of these findings, the problematic cyclic constraints were removed and a single set of optimisation parameters were sought minimising both cost and ISE over multiple cycles. The idea being employed here was that the greater the number of cycles over which the path constraints were not violated, the more reliable this set of design variables would be over the plant's operational period. Indeed, it may be argued that if feasible operation was ensured for a sufficiently large number of cycles, the elapsed time is of an adequate

length before plant shutdown is required, perhaps for maintenance reasons. In any case, optimisation results minimising cost for 2 and 5 cycles, and minimising ISE for 2, 5 and 10 cycles may be found in appendix C. The final set of profiles in this appendix correspond to the design variables from the 10 cycle case working for 100 cycles without violating the purity path constraints. Two trends are observed from these results:

- from figure C.5, the reactor composition is *completely* dynamic, not adhering to the setpoint profile in any way, promoting the belief that the optimal solution does in fact lie in regions dominated by dynamic behaviour.

- from figures C.10 and C.12 the compositions certainly seem to converge to an optimum steady state operating point, albeit non-cyclic, encouraging us to accept that our choice of design variables is reliable enough to carry the plant though a number of cycles greater than 100.

In an attempt to provide a more robust solution a "range" requirement should be imposed. As can be seen from figure 4.10, the storage state starts and finishes within a certain range which will (hopefully) be fixed by the optimiser. This formulation is more likely to guarantee infinite repeatability, although runs must again be performed for a sufficient number of cycles to check this. The assumption being made here is that if the profiles starting at the extremes of the allowable range return to within it at the end of the time horizon, 'internal' profiles, will also converge to within the range. This approach is adopted in the second case study in this chapter where Operating Envelopes are used to find the allowable ranges that the critical plant states can take just before transitions.

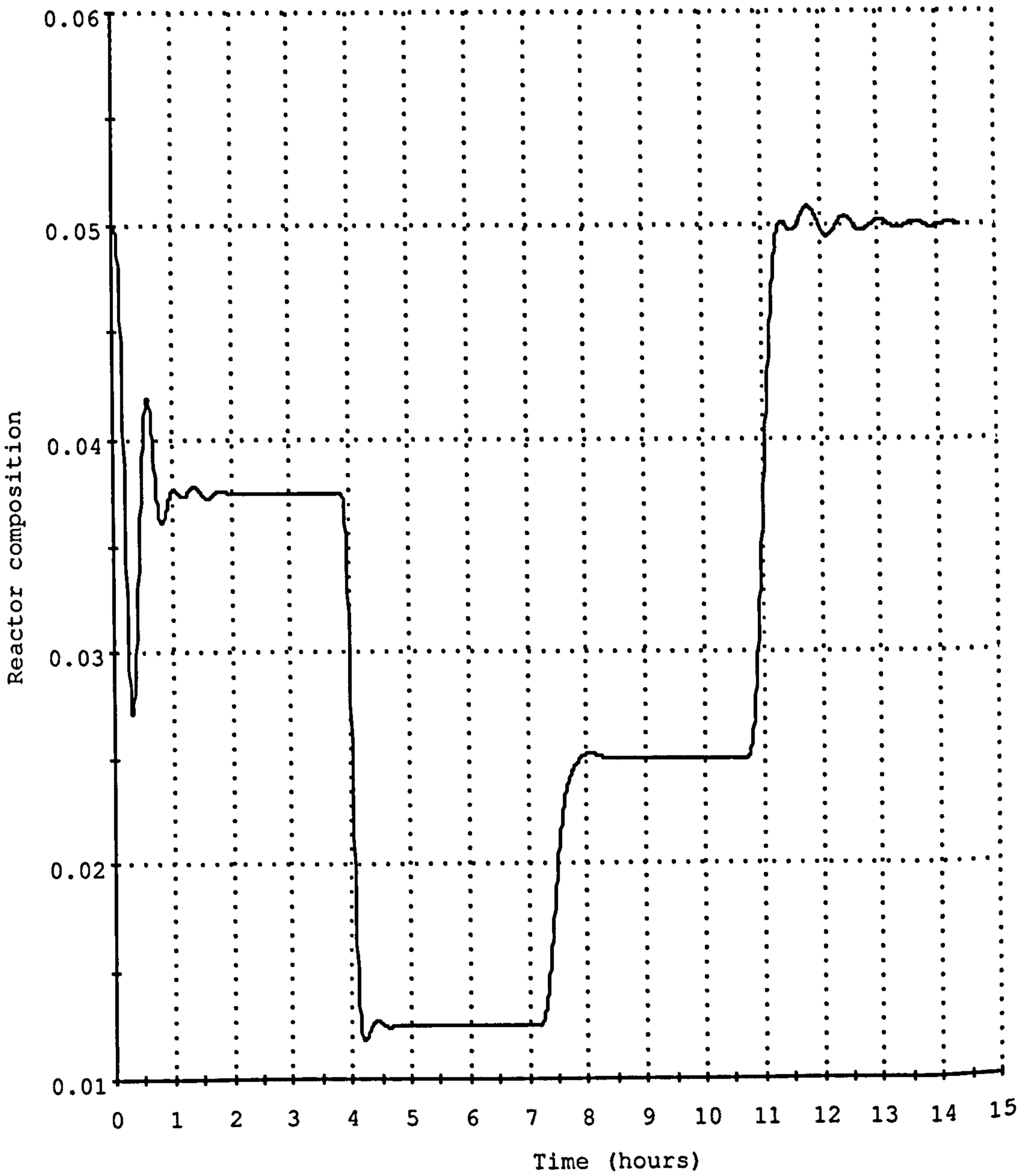


Figure 4.9: Damped reactor response produced by minimising ISE

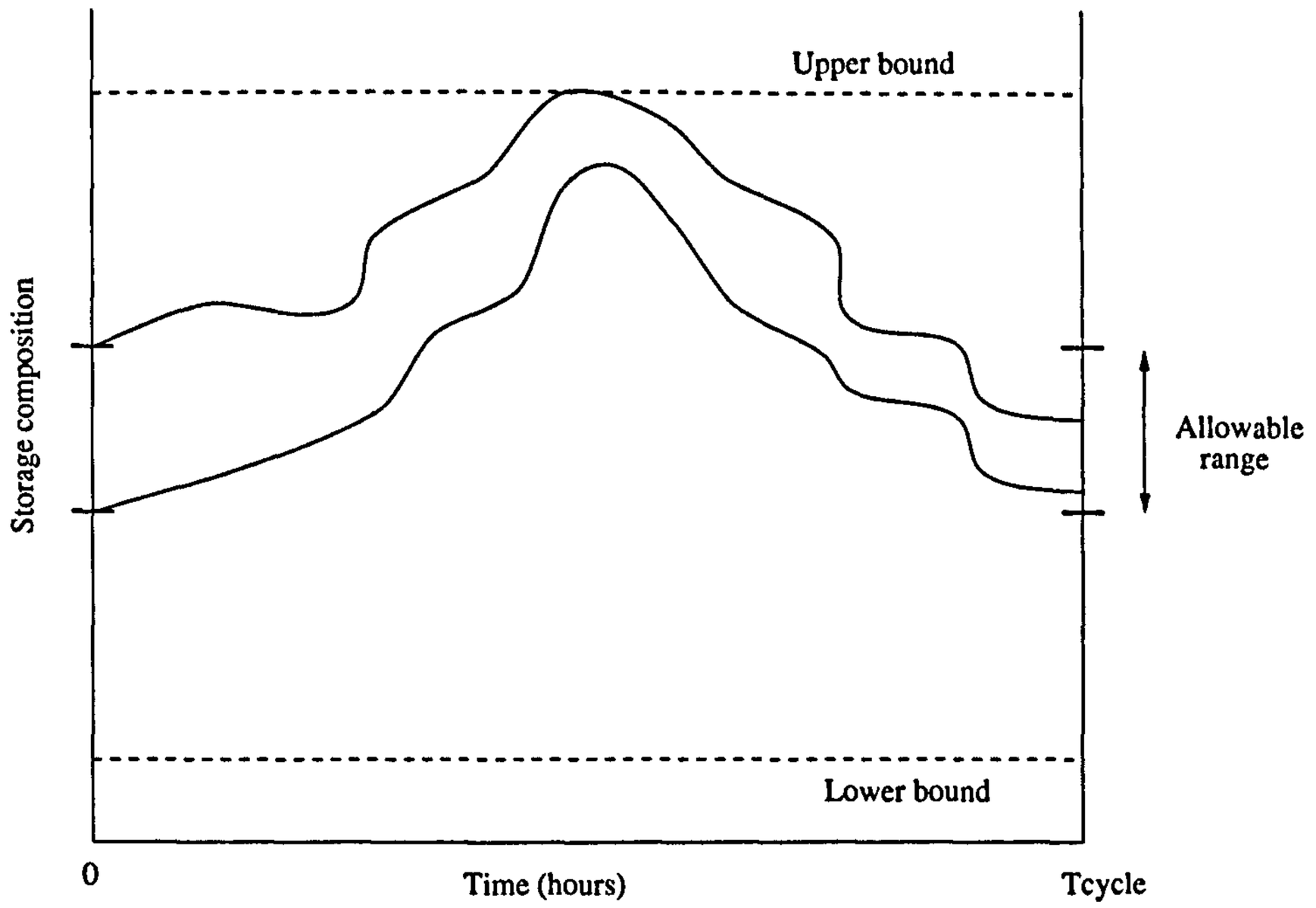


Figure 4.10: "Range" requirement proposed to replace cyclic constraint

4.3. Multiproduct reactor/separator system

4.3.1. Problem description

This example has been adapted from one used by Belanger and Luyben (1998). The reactor/separator process is shown in Figure 4.11. The fresh feed consists of component A and a first order irreversible reaction $A \rightarrow B$ takes place in the single phase CSTR. The output of the CSTR, containing both components, is fed into a distillation column where the unreacted A is separated from the product and recycled back to the reactor. The bottoms from the column is then routed to either one of two product tanks (depending on the grade) or an offspec tank. Perfect control over the reactor temperature is assumed in order to manipulate system compositions for the necessary grade changes.

Specification	B_1	B_2
continuous demand $(lbmol/hr)$	100	100
nominal purity $(lbmol/ft^3)$	0.97	0.90
purity upper bound $(lbmol/ft^3)$	0.975	0.91
purity lower bound $(lbmol/ft^3)$	0.965	0.89

Table 4.4: Product demand rates and purity requirements

For the purposes of finding ranges for critical states that ensure feasible grade changes, the system is assumed to be operating open-loop. Clearly a control system must be incorporated for setpoint tracking and disturbance rejection in reality, but as only the concept of feasible "cyclic" grade changes using Operating Envelopes is being scrutinised, open-loop operation is deemed sufficient. The case study in the next chapter applies Operating Envelopes to a closed loop system.

The objective is to minimise offspec material production and meet the continuous product supply requirements by determining the optimal cyclic operation of the plant. Operating Envelopes will be used to find ranges that critical initial system

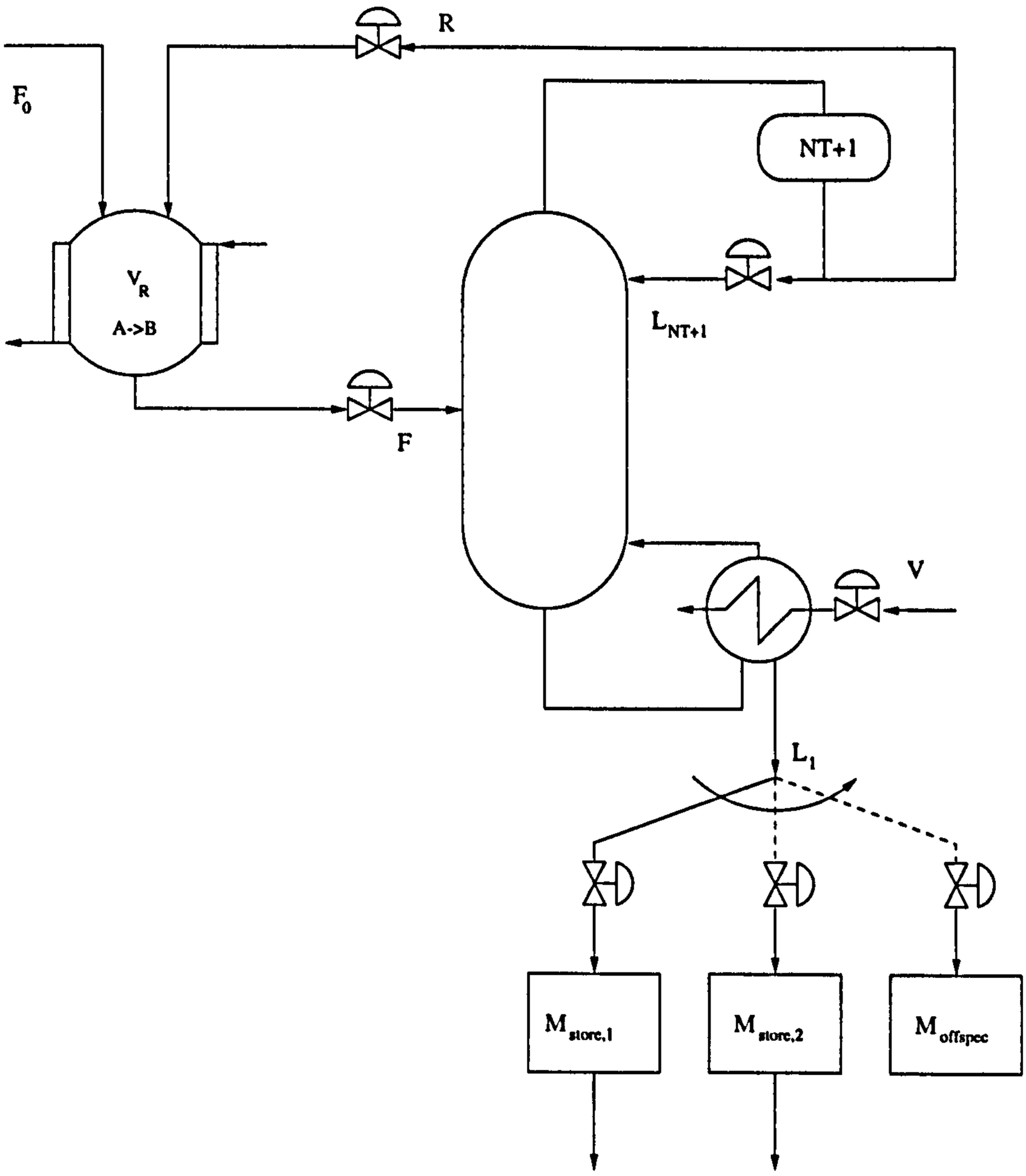


Figure 4.11: The reactor/separator process

states can take *and* return within at the end of the time horizon, ideally producing an infinitely repeatable operating cycle.

4.3.2. Model equations and assumptions

In order to develop a relatively simple mathematical model of the reactor/separator process the following assumptions are used

- the liquid on trays and in the reactor is perfectly mixed and has uniform properties
- thermodynamic equilibrium on all trays is achieved
- the pressure drop in the column is negligible
- constant relative volatility and equimolar overflow
- the vapour hold-up is negligible
- the reflux is saturated liquid
- the liquid hydraulics are described by the Francis weir formula
- constant holdup on each tray

All mole fractions are given in terms of the reactant A:

main distillation column excluding feed tray: $j = 2 \ldots NT; \quad j \neq NF$

$$\frac{d}{dt}(M_j) = L_{j+1} - L_j - V_j + V_{j-1} \tag{4.31}$$

$$M_j \frac{d}{dt}(x_j) = L_{j+1}(x_{j+1} - x_j) - V_j(y_j - x_j) + V_{j-1}(y_{j-1} - x_j) \tag{4.32}$$

$$y_j = \frac{\alpha}{1 + (\alpha - 1)x_j} \tag{4.33}$$

$$V_j = V_{j-1} \tag{4.34}$$

$$L_j = c_0 \rho_L \ell_W \left(h_{OW_j}\right)^{3/2} \tag{4.35}$$

$$M_j = (h_W + h_{OW}) \frac{\pi D_C^2}{4} \frac{\rho_L}{M_{WL}} \tag{4.36}$$

feed tray: $j = NF$

$$\frac{d}{dt}(M_j) = L_{j+1} - L_j - V_j + V_{j-1} + F \tag{4.37}$$

$$M_j \frac{d}{dt}(x_j) = L_{j+1}(x_{j+1} - x_j) - V_j(y_j - x_j) + V_{j-1}(y_{j-1} - x_j) + F(x_R - x_j) \tag{4.38}$$

$$y_j = \frac{\alpha}{1 + (\alpha - 1)x_j} \tag{4.39}$$

$$V_j = V_{j-1} + (1 - q_v)F \tag{4.40}$$

$$L_j = c_0 \rho_L \ell_W (h_{OW_j})^{3/2} \tag{4.41}$$

$$M_j = (h_W + h_{OW}) \frac{\pi D_C^2}{4} \frac{\rho_L}{M_{WL}} \tag{4.42}$$

column base: $j = 1$

$$\frac{d}{dt}(M_j) = L_{j+1} - L_j - V_j \tag{4.43}$$

$$M_j \frac{d}{dt}(x_j) = L_{j+1}(x_{j+1} - x_j) - V_j(y_j - x_j) \tag{4.44}$$

$$y_j = \frac{\alpha}{1 + (\alpha - 1)x_j} \tag{4.45}$$

$$L_j = C_V^b K_V^b \sqrt{M_j} \tag{4.46}$$

column condenser and reflux drum: $j = NT + 1$

$$\frac{d}{dt}(M_j) = V_{j-1} - (L_j + R) \tag{4.47}$$

$$M_j \frac{d}{dt}(x_j) = V_{j-1}(y_{j-1} - x_j) \tag{4.48}$$

$$L_j = C_V^l K_V^l \sqrt{M_j} \tag{4.49}$$

$$R = C_V^r K_V^r \sqrt{M_j} \tag{4.50}$$

reactor:

$$\frac{d}{dt}(M_R) = F_0 + R - F \tag{4.51}$$

$$M_R\frac{d}{dt}(x_R) = F_0(x_0 - x_R) + R(x_{NT+1} - x_R) + M_R x_R k_0 exp\left(\frac{-E}{R_g(T+T_0)}\right) \tag{4.52}$$

$$F = C_V^{rr} K_V^{rr} \sqrt{M_R} \tag{4.53}$$

product storage and offspec tanks:

$$\frac{d}{dt}(M_{store,1}) = s_1 L_1 - F_{prod,1} \tag{4.54}$$

$$M_{store,1}\frac{d}{dt}(x_{store,1}) = s_1 L_1(x_1 - x_{store,1}) \tag{4.55}$$

$$\frac{d}{dt}(M_{store,2}) = s_2 L_1 - F_{prod,2} \tag{4.56}$$

$$M_{store,2}\frac{d}{dt}(x_{store,2}) = s_2 L_1(x_1 - x_{store,2}) \tag{4.57}$$

$$\frac{d}{dt}(M_{offspec}) = L_1(1 - s_1 - s_2) \tag{4.58}$$

where M is the liquid holdup; L and V are the liquid and vapour flows; x and y are the molar fractions in liquid and vapour phase respectively; R_g is the universal gas constant; E the activation energy; T is the temperature; D_C is the column diameter; ℓ_W is the length of the weir and h_W its height; h_{OW} is the height of the liquid over the weir; c_0 is a Francis weir constant; ΔH^{VAP} is the heat of vaporisation; Q_R is the heat input in the reboiler; ρ_L is the liquid density and M_{WL} is the molar weight. The subscript j denotes the tray number (1 to $NT+1$) and the split fraction s determines which product/offspec tank is being filled at any given time. The value of the parameter q_v (quality of the distillation feed) is set to one. The parameters of the model are given in table 4.5 and can also be found in Belanger and Luyben (1998).

Parameter	*Value*	*Parameter*	*Value*
F_0	$348.4\,lbmol/hr$	q_v	1
x_0	1.0	T_0	460
k_0	$2.7759 \times 10^{10}\,hr^{-1}$	E	$30000\,BTU/lbmol$
NT	$31\,trays$	α	2
D_C	$5.43\,ft$	h_W	$1\,inch$
C_V^r	50	C_V^l	120
C_V^b	80	C_V^{rr}	20
C_0	9600	R_g	$1.99 BTU/(lbmol\,^\circ R)$
M_{WL}	$50.0\,lb_m/lbmol$	ρ_L	$50.0\,lb_m/ft^3$

Table 4.5: Model parameters for reactor/separator case study

4.3.3. Optimisation

4.3.3.1. Interval structure specification

The operating cycle is divided into six intervals as described below and shown in figure 4.12.

1. Reaction at the high temperature; feeding to product tank 1.

2. Reaction temperature switched to low; feeding to product tank 1. (Some offspec product may be fed to the product tank, provided that the purity is maintained on specification in the product tank. This will minimise the offspec produced.)

3. Reaction at the low temperature; feeding to offspec tank while the grade change is taking place.

4. Reaction at the low temperature; feeding to product tank 2.

5. Reaction switched to high; feeding to product tank 2. (Analogous to interval 2.)

6. Reaction at the high temperature; feeding to offspec tank while the grade is changing back to product 1.

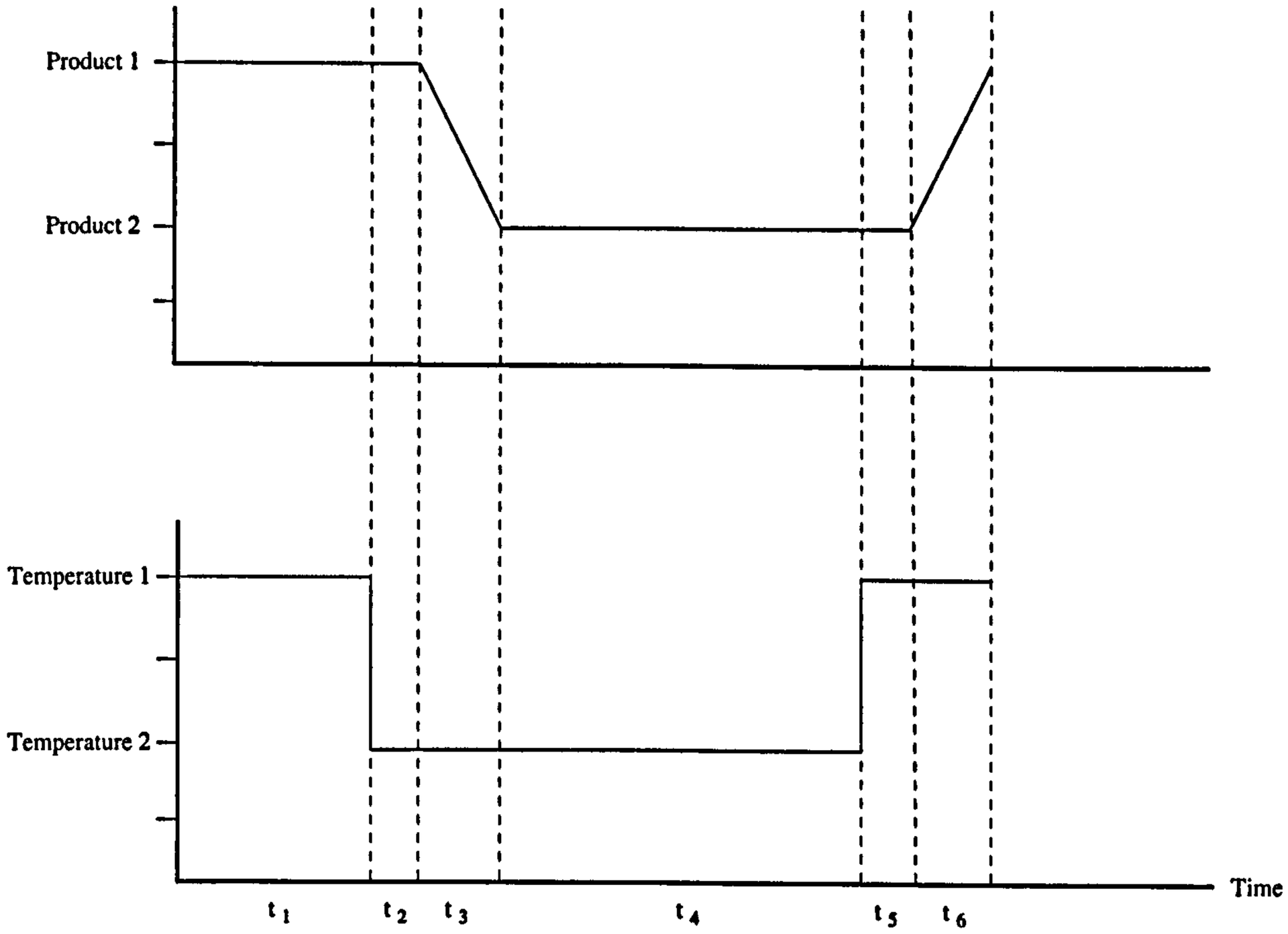


Figure 4.12: Interval structure for dynamic optimisation.

4.3.3.2. Objective function

$$\min_{u(t),d} \{M_{offspec}(t_f)\} \tag{4.59}$$

4.3.3.3. Optimisation parameters and constraints

- Controls:
  - Reactor temperature profile. Note this profile must follow the structure in figure 4.12; the interval durations and the high and low temperature levels are the actual degrees of freedom.
  - Product flowrate profiles to each of the storage and offspec tanks. The flow from the distillation column is split between the product tanks

and offspec tank as described above and in figure 4.12; the magnitude being equal to the bottoms flowrate of the distillation column.

- Process initial state:
  - Initial compositions in the storage tanks.
  - Initial composition in the reactor.
  - Initial composition in the column base.
- Time invariant optimisation parameters:
  - Product storage vessel sizes.
- Path constraints:
  - on the storage tank volumes, so they remain positive.
  - on the storage tank purities, so they remain within specification.
- Cyclic constraints — the final value must be within the specified Envelope:
  - on the storage tank compositions.
  - on the reactor composition.
  - on the column base composition.

4.3.4. Results and discussion

A base case (non-cyclic) optimal process design and product scheduling was established through dynamic optimisation, minimising the molar holdup of the offspec produced, rather than total cost as in the previous example. Figure 4.13 and table 4.6 show the key results of this optimisation.

Operational Envelopes are then used to pose the cyclic optimisation problem, by specifying the initial conditions of critical states as dimensions of the Envelope

Interval duration	*value (hours)*
t_1	1.43
t_2	1.44
t_3	1.39
t_4	1.75
t_5	1.12
t_6	2.87
Temperature	*value* (^oF)
High	135.7
Low	129.6

Table 4.6: Key results from the base case optimisation

and constraining the states to return to within the Envelope at the end of the time horizon. By guaranteeing that the process will return to its starting range for all possible vertices of the initial range, we assume that any initial state *within* the Envelope will also do so. Here, we fix the size of the Envelope to a suitable tolerance and allow the optimisation to determine its position. This procedure differs from the traditional approach of optimising the initial conditions and forcing the states to return to their exact initial values, where any discrepancy between the initial and final values may become exacerbated in subsequent cycles, possibly resulting in a completely different cyclic steady state.

In this example, we consider the initial purities in each of the storage tanks and the compositions in the column base and reactor as dimensions of the Envelope, forming a 4-dimensional hypercube, with 16 vertices to examine in the solution procedure. Note that although all tray compositions should ideally be considered as Envelope dimensions, this would make the problem intractably large.

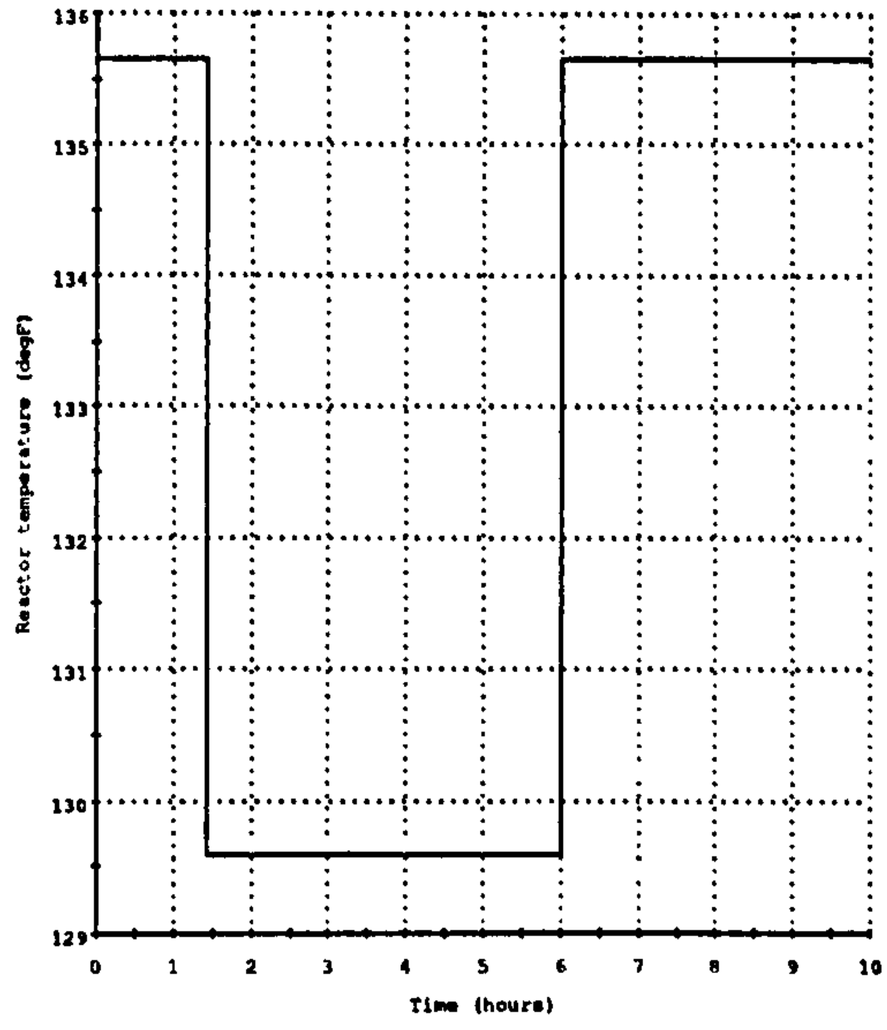
The overall problem took 2 iterations to converge. The first iteration consisted of a cyclic optimisation using scenarios 1 and 16 (all initial values on their lower bound and the opposite vertex). After simulating all 16 scenarios using the solution of the optimisation, constraint violations were identified and are shown in figure 4.14. Scenarios 4 and 13 were chosen for the second iteration, based on the cyclic constraint violations in the column base composition and storage tank

purities, and on the purity path constraint in product storage tank 1. The result of this second optimisation, with scenarios 4 and 13, was again simulated for all 16 scenarios. Figure 4.15 show that none of these scenarios were violated beyond the the tolerance of the optimisation, hence the construction of the Envelope is complete. The Envelope is displayed in table 4.7: any initial condition within these ranges is expected to converge on a cyclic steady state also within this Envelope; see figure 4.10. The cyclic behaviour can be seen in figure 4.16 where the critical states from all 16 scenarios are plotted; note that they all converge to a point or points within the initial range at the end of the cycle. This is deemed a more robust method for guaranteeing infinitely repeatable operational cycles than the the cyclic point constraints used in the original problem formulation of the multiproduct reactor example examined earlier in this chapter.

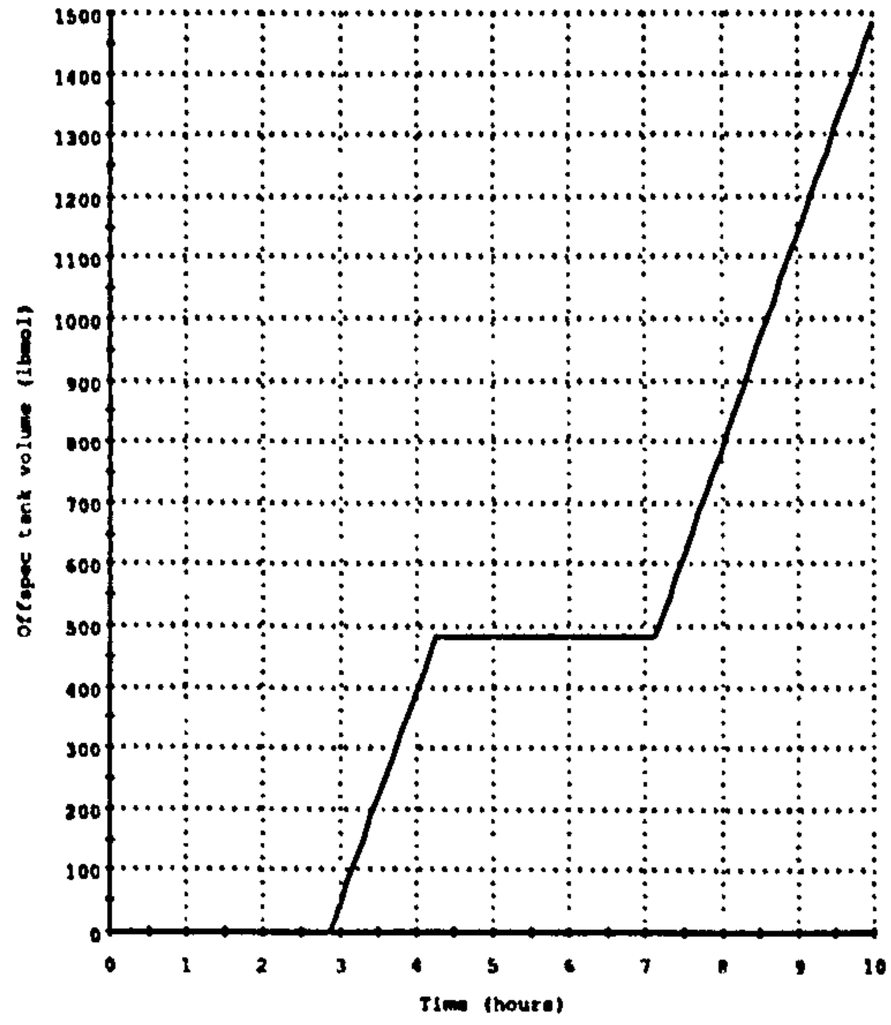
Envelope dimension	*Lower limit*	*Upper limit*
Storage tank 1 initial mole fraction of B	0.965	0.970
Storage tank 2 initial mole fraction of B	0.903	0.908
Reactor initial mole fraction of A	0.391	0.396
Column tray 1 initial mole fraction of A	0.035	0.040

Table 4.7: Cyclic Envelopes

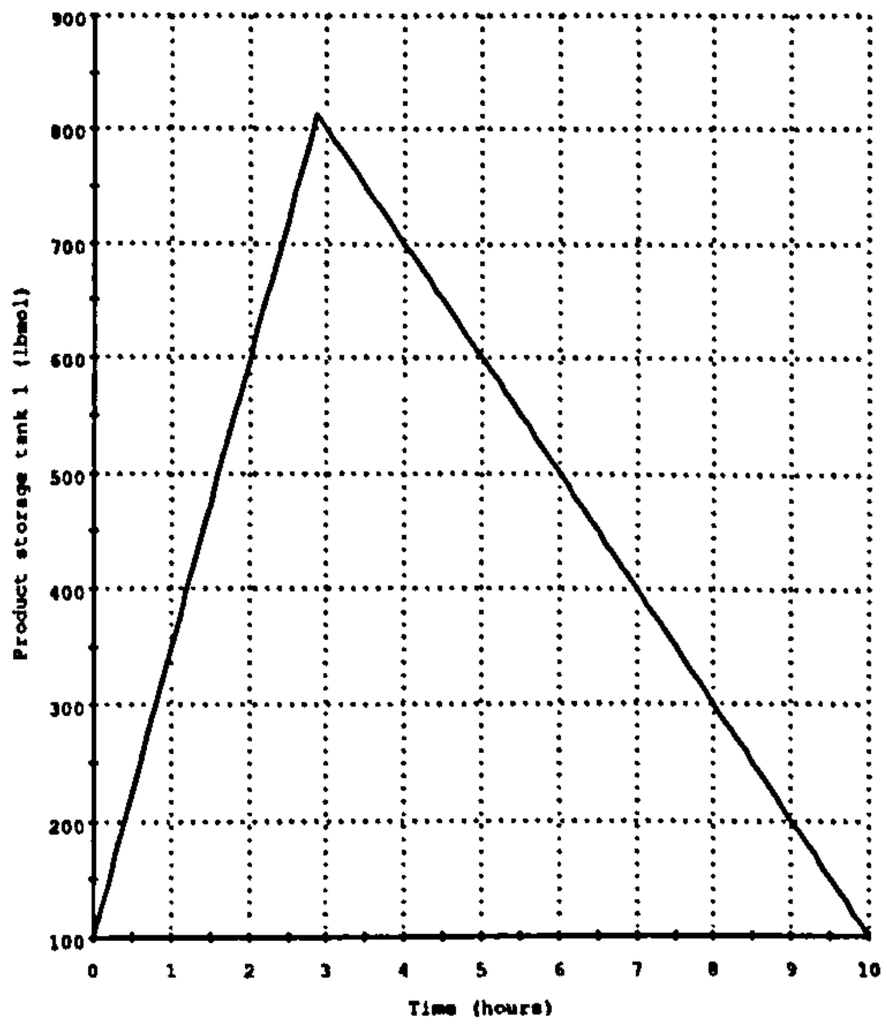
Note that the larger tolerated violations seen in figure 4.15(c) (the four points *above* the x-axis) correspond to the profile of storage tank 2 mole fraction visibly failing to return to within its Envelope, as shown in figure 4.16(c).



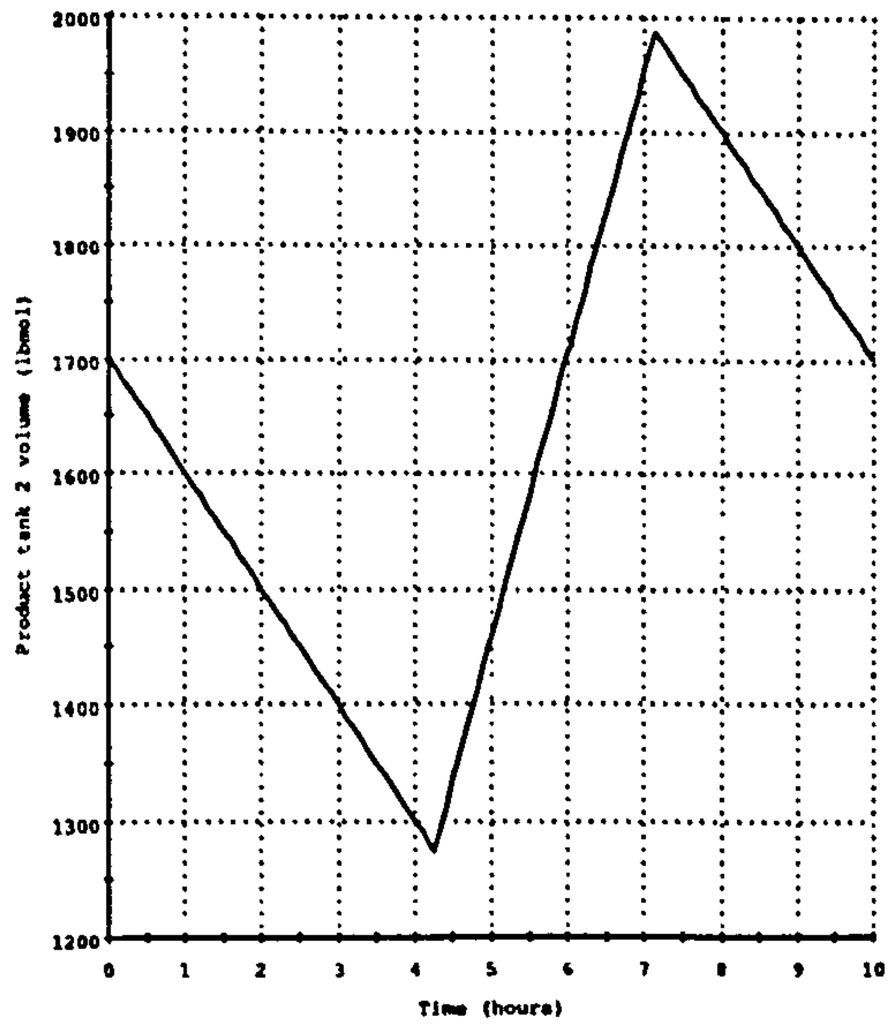
(a) Reactor temperature



(b) Offspec tank volume

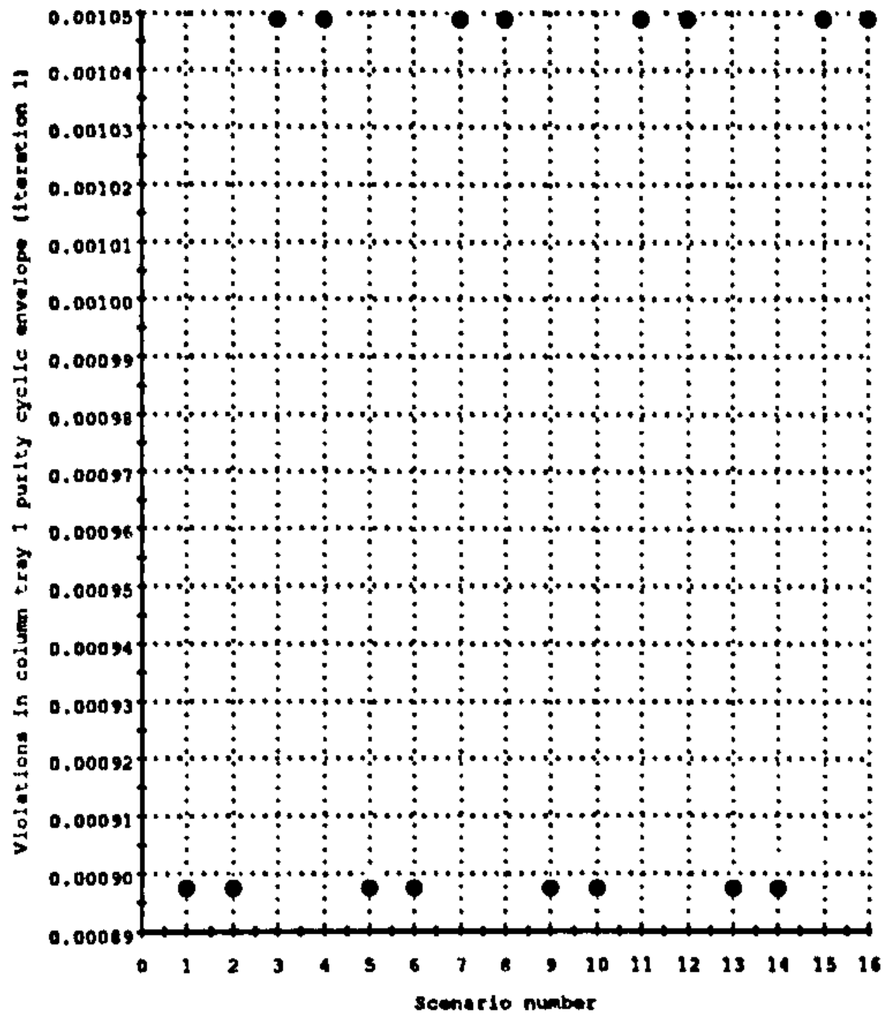


(c) Product tank 1 volume

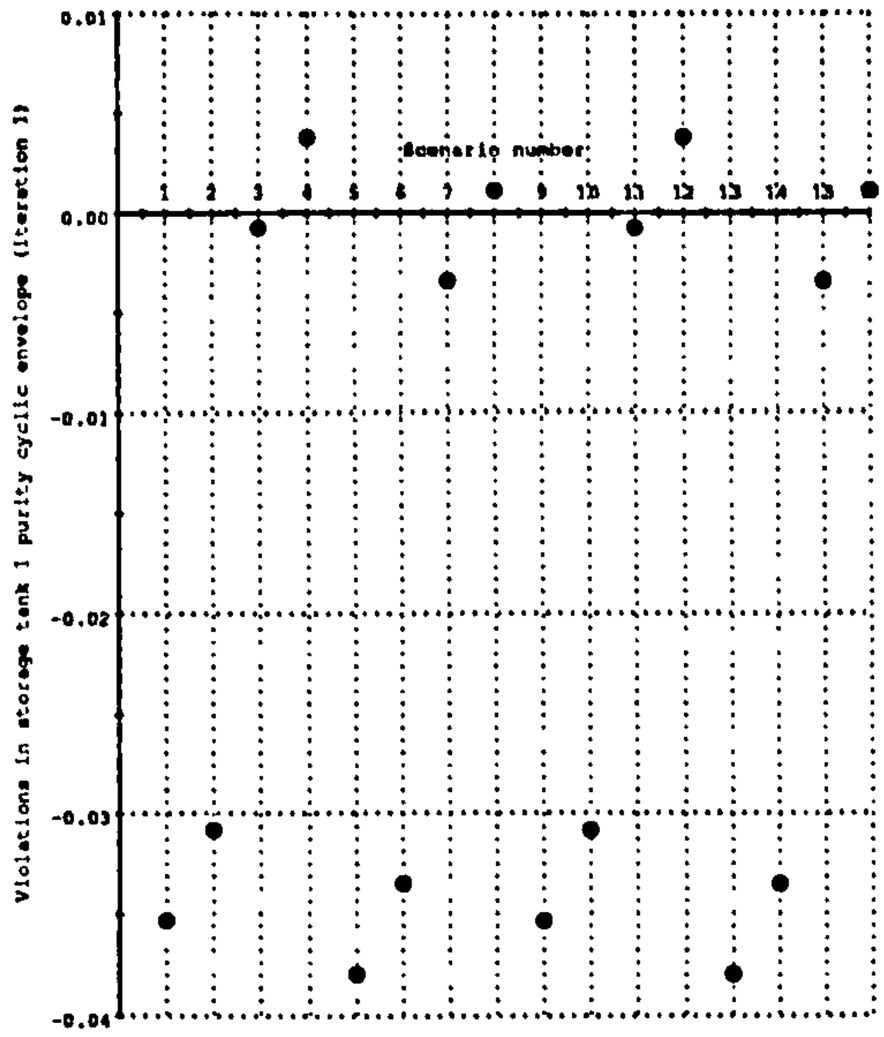


(d) Product tank 2 volume

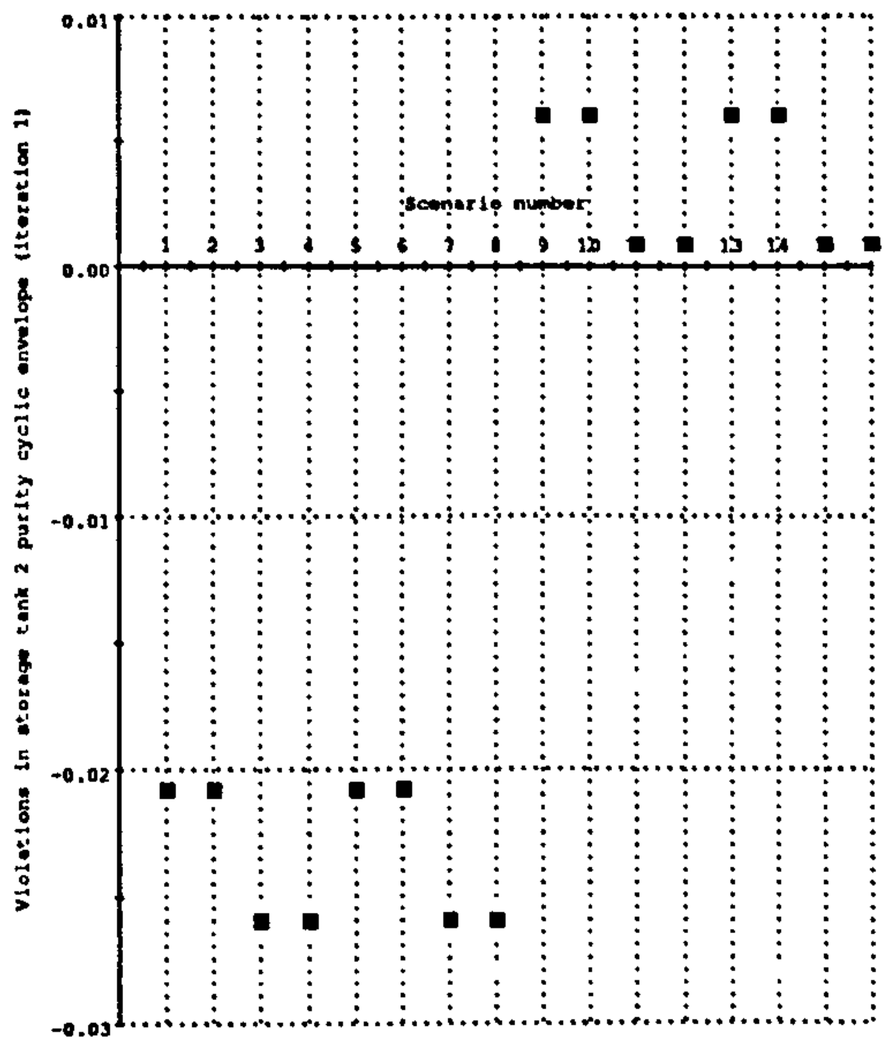
Figure 4.13: Some typical system profiles



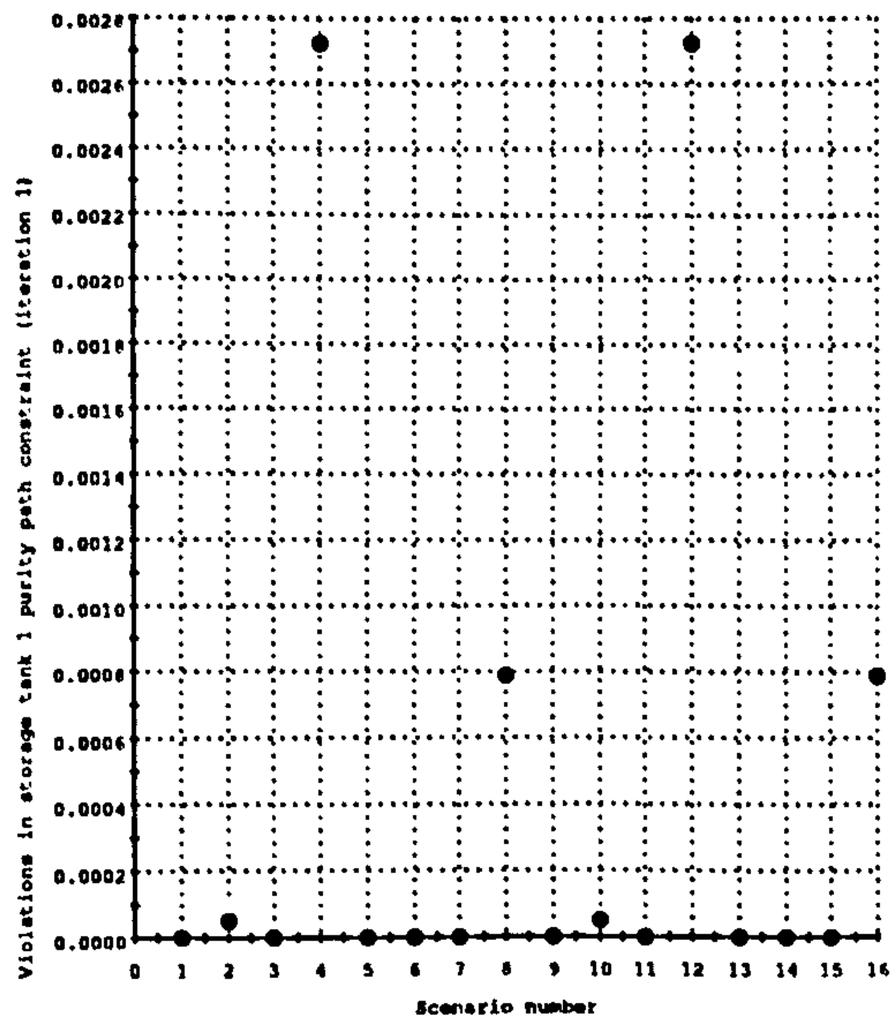
(a) Violations in column tray 1 purity cyclic constraints for each scenario



(b) Violations in storage tank 1 purity cyclic constraints for each scenario

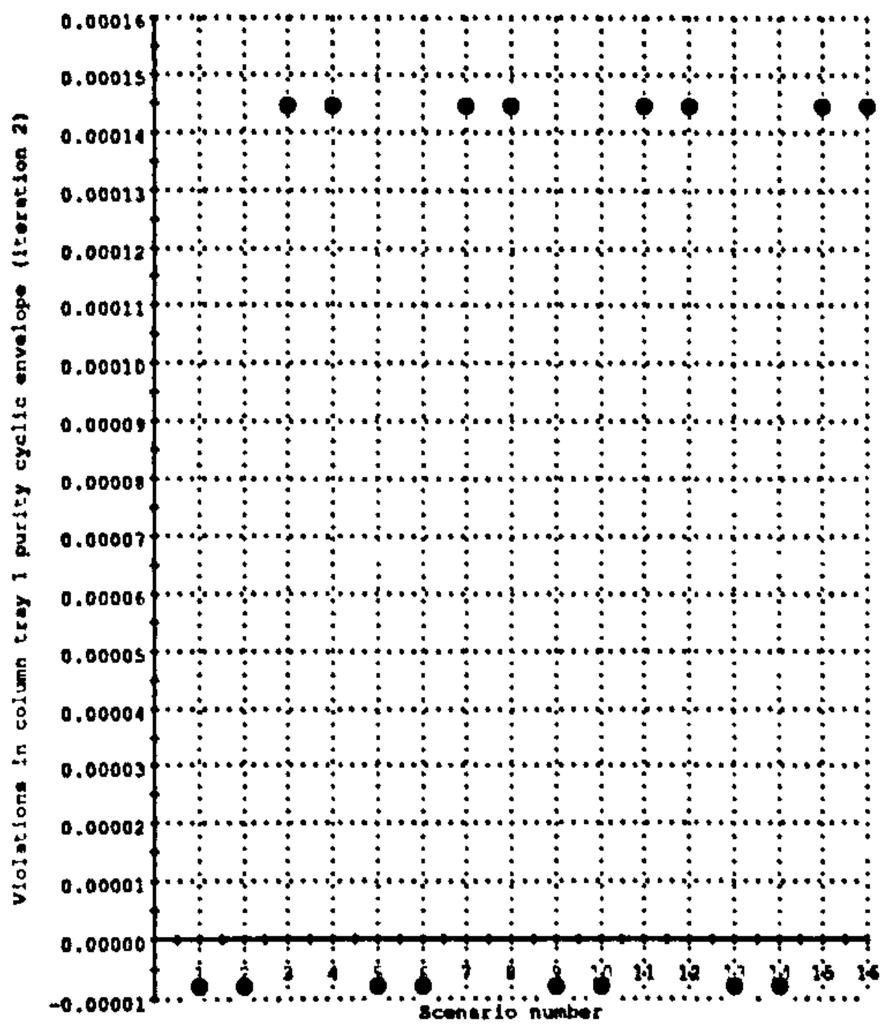


(c) Violations in storage tank 2 purity cyclic constraints for each scenario

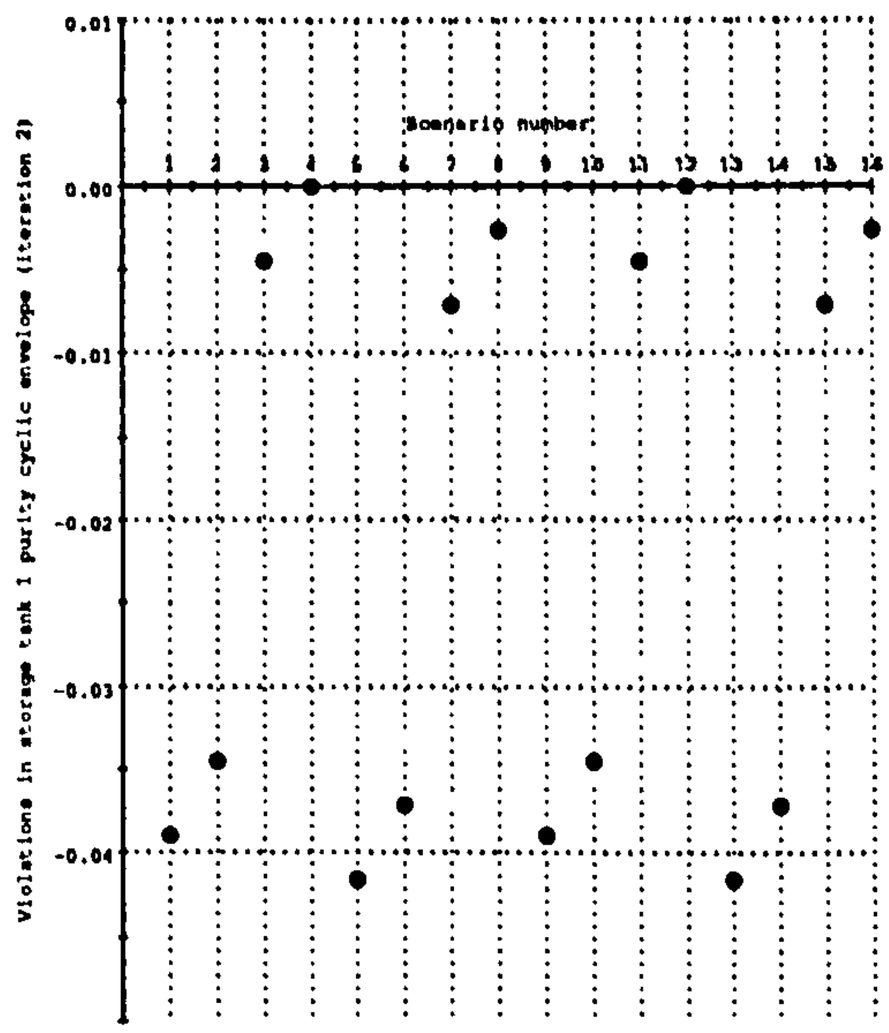


(d) Violations in storage tank 1 purity for each scenario

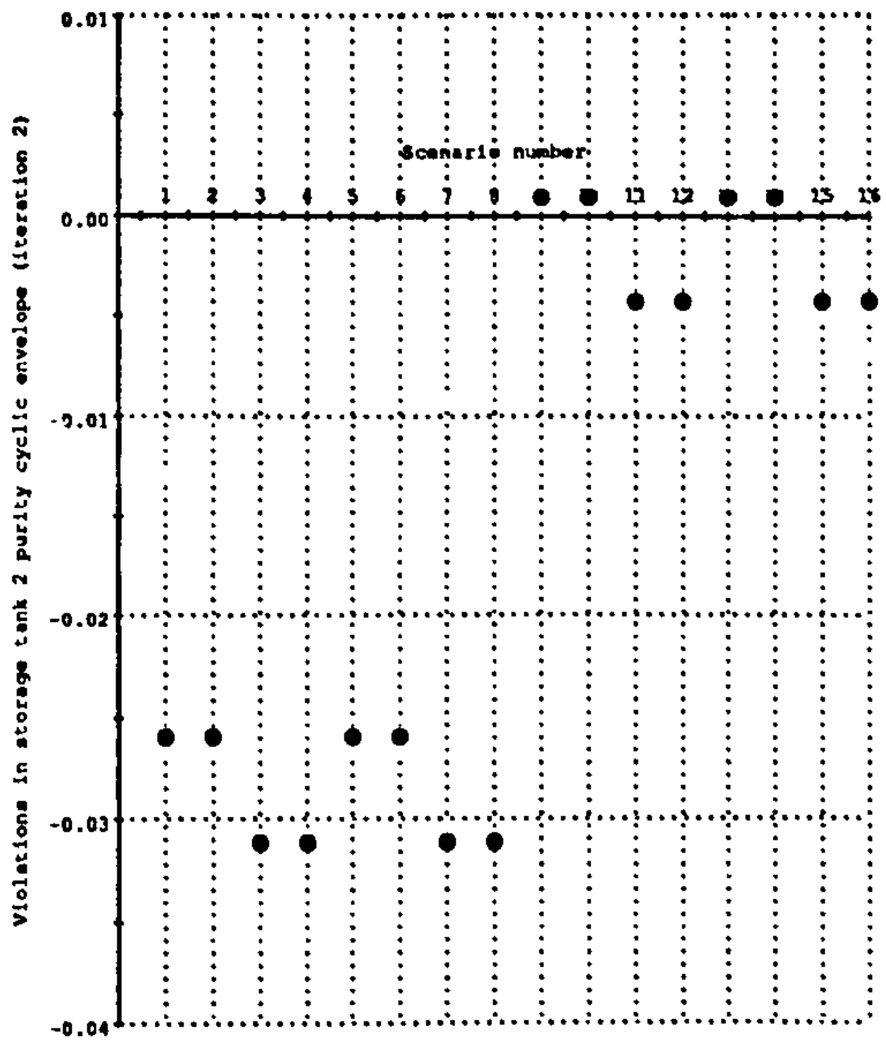
Figure 4.14: Cyclic and path constraint violations for the first iteration



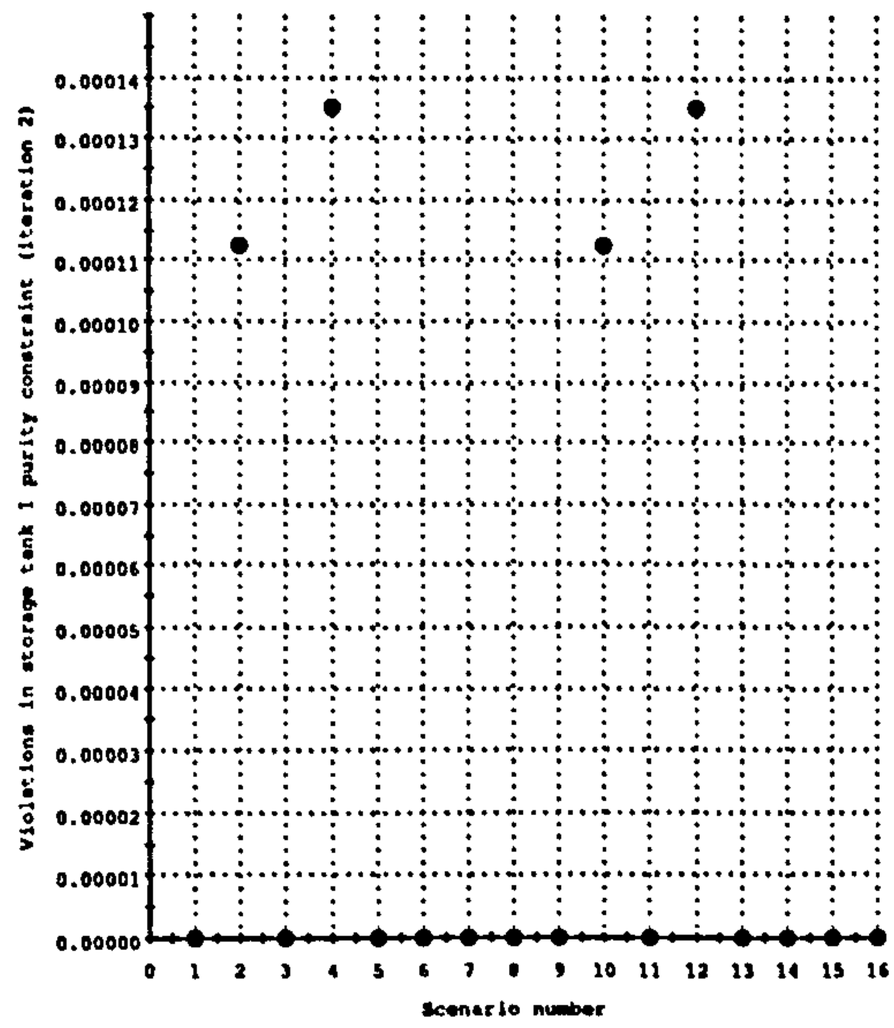
(a) Violations in column tray 1 purity cyclic constraints for each scenario



(b) Violations in storage tank 1 purity cyclic constraints for each scenario

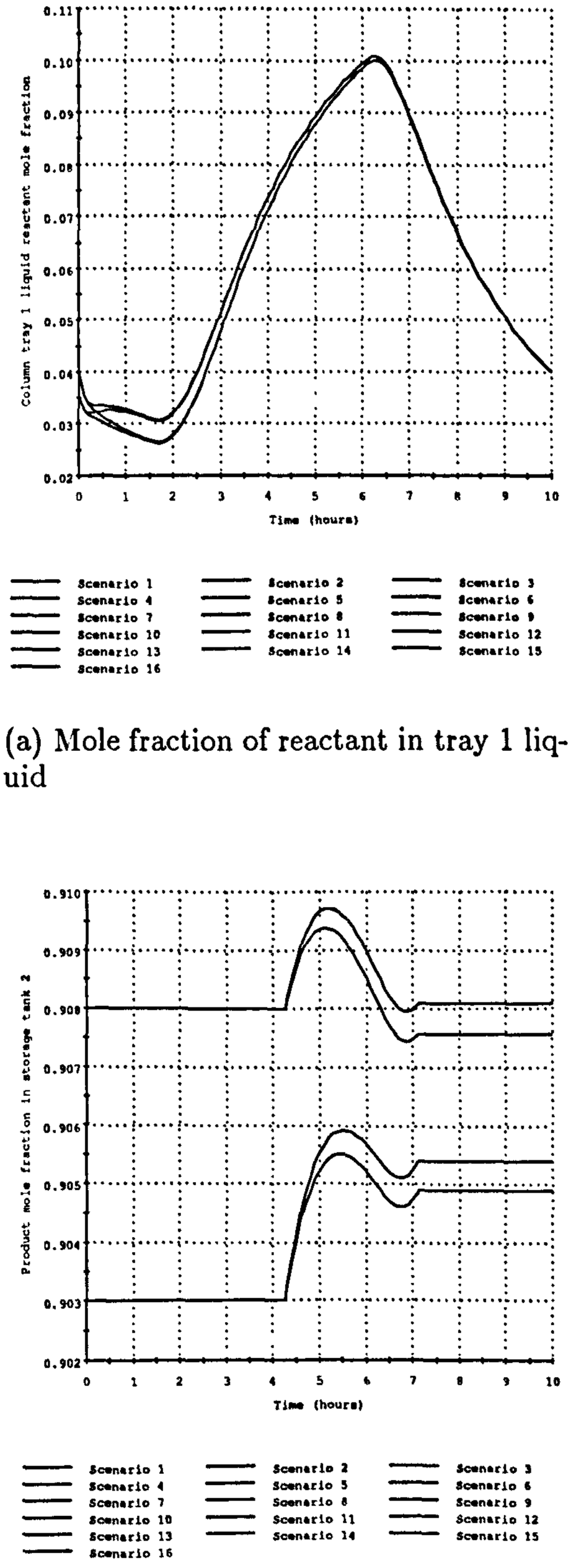


(c) Violations in storage tank 2 purity cyclic constraints for each scenario

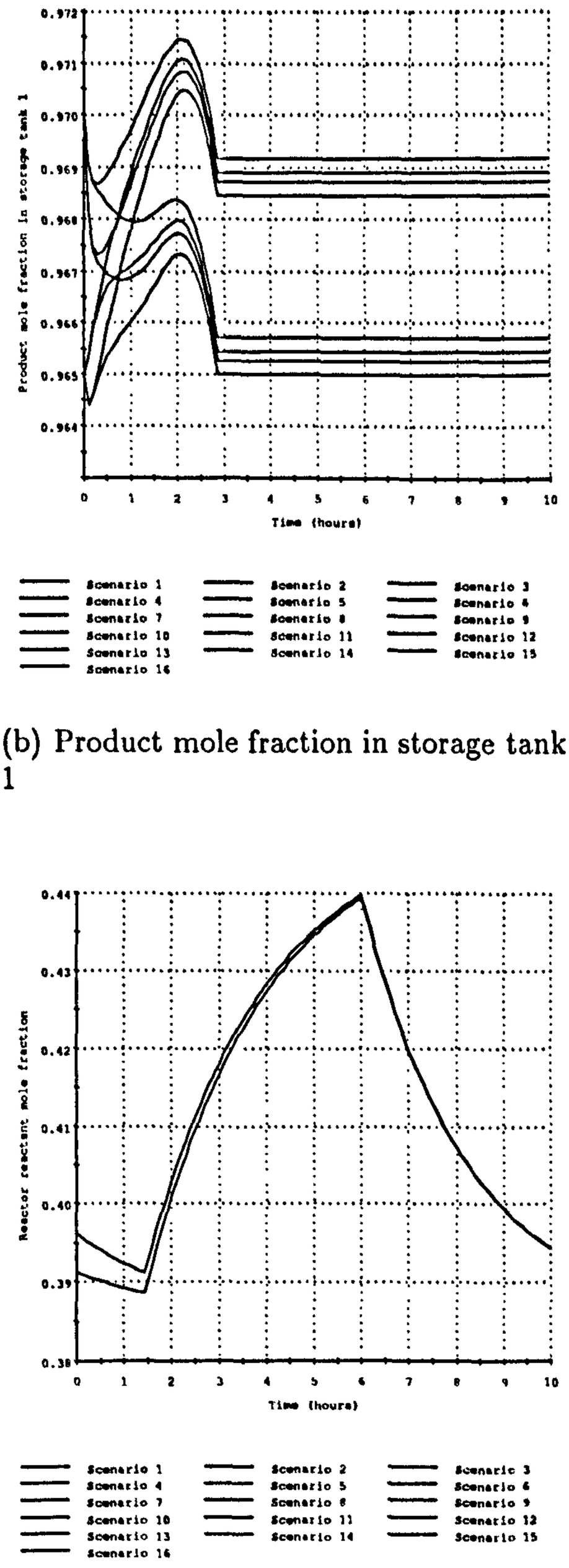


(d) Violations in storage tank 1 purity for each scenario

Figure 4.15: Cyclic and path constraint violations for the second iteration



(a) Mole fraction of reactant in tray 1 liquid



(b) Product mole fraction in storage tank 1

(c) Product mole fraction in storage tank 2

(d) Reactant mole fraction in the reactor

Figure 4.16: Profiles of critical states for all scenarios

4.4. Summary and conclusions

At the beginning of this chapter, an insight into multiproduct processes was given followed by a short review. The works of Cantrell and Luyben (1996) and Rowe (1997) were critically assessed and improvements to the revisited multiproduct reactor system outlined. Although one strength of Rowe's methodology was the variety of objective functions that could be used (ecomomics, stability and offspec production have all been used in the multiproduct examples), the main weakness was the inability to guarantee an infinitely repeatable production cycle. This arose from the incorporation of cyclic *point* constraints that were impossible to enforce rigorously within the dynamic optimisation framework. In view of this, a second multiproduct reactor/separator example was undertaken to employ a novel approach to gurantee cyclic operation. This involved using Operational Envelopes to define *ranges* in the state variables from which the system is guaranteed to return to within the same range after one cycle and hence the cycle is infinitely repeatable.

Chapter 5

Responsive reactor networks

5.1. Introduction

The aim of this case study is to find the optimal design and operation of a responsive process plant capable of instant response to a customer's intermittent product demand and startup and shutdown automatically as required. The two-stage design procedure discussed in section 3.2.2 is employed in

1. selecting the optimal number of reactors through heuristics and bounding properties,

2. determining the optimal network design and *open-loop* operation through dynamic optimisation.

This work is then extended by

- selecting a control scheme through heuristics and proximity rules to investigate the *closed-loop* behaviour of the reactor network, supplementing stage one above,

- solving a multiperiod dynamic optimisation to determine Operating Envelopes in the presence of uncertainty, supplementing stage two above.

5.2. Problem Statement

The customer is replacing one of his/her feedstocks with material B, which must be made onsite. Requirement for B will be 12.3 litres/min and intermittent, but it must be available on demand and lie within purity limits of 81.69% to 84.90% (w/w). The plant operates for 365 days per year with one twelve hour shift per day, under the following constraints:

1. B degrades if stored for long periods. This degradation accelerates with increased temperature, and is considered significant above $48.89°C$ ($120°F$).

2. Environmental regulations dictate a maximum inventory of B of 50 litres per vessel, as this is the largest tolerable discharge in the event of a vessel failure.

3. Total inventory of B for the entire site is limited to 400 litres, which could maintain demand for just more than 30 minutes.

Material B may be generated from an environmentally acceptable feed component A via the exothermic, catalytic liquid phase reaction:

$$A \rightarrow B \qquad \Delta H \ -ve \tag{5.1}$$

As previous work (de Hennin, 1994; de Hennin et. al., 1994) using this first order reaction scheme has concentrated on continuous stirred tank reactors, a CSTR based design will be developed and evaluated. Each CSTR unit will have one outlet and two inlets: one for fresh feed of A and the other for connection to the preceding upstream unit. Each CSTR possesses a cooling jacket that uses water as the cooling medium. Cooling water is available at $20°C$ and has a maximum allowable discharge temperature of $48.89°C$ ($120°F$). Fresh feed of A is available

at $20°C$ and at 1 kg/litre. The reaction is catalytically driven; the catalyst may be introduced into or removed from the reacting liquid within each CSTR to achieve effective control over the reaction. It is assumed that the instantaneous response demanded by the customer can be achieved by simply storing the required product. Given that product storage buffering may be needed, a product storage tank must be allowed for in the system.

5.3. Dynamic modelling

5.3.1. Modelling assumptions

- A and B have identical physical properties
- Reactor is well stirred
- Low pressure, high density (liquid) system
- Specific heat capacities independent of temperature
- Constant cooling water volume in reactor jacket

5.3.2. Reactor

The dynamic behaviour of a CSTR is represented by a set of differential and algebraic equations based on dynamic mass and energy balances around the unit. In the notation below the superscript 2 refers to the properties (flow, concentration and temperature) of the fresh feed and the superscript 1 to the stream from the upstream unit.

Overall material balance,

$$\frac{dV_R}{dt} = F_{in}^1 + F_{in}^2 - F_{out} \tag{5.2}$$

Feed component material balance,

$$V_R\frac{dC_A}{dt} + C_A\frac{dV_R}{dt} = F_{in}^1 C_{in,A}^1 + F_{in}^2 C_{in,A}^2 - F_{out}C_A - kV_R C_A \tag{5.3}$$

Energy balance,

$$V_R\frac{dT}{dt} + T\frac{dV_R}{dt} = F_{in}^1 T_{in}^1 + F_{in}^2 T_{in}^2 - F_{out}T - kV_R C_A\frac{(\Delta H_R)}{\rho C_p} - \frac{Q}{\rho C_p} \tag{5.4}$$

The rate constant, k, is defined as,

$$k = \left[k_0 e^{\frac{-E_a}{R(460+T)}}\right] Catalyst_{control} \tag{5.5}$$

where the variable $Catalyst_{control}$ denotes the level of catalyst in the reactor and is bounded from zero to one in the steady state optimisation. A $Catalyst_{control}$ value of zero effectively quenches the reaction. Note that in the dynamic case it was necessary to increase the upper bound on this variable to 1.2 — see section 5.5 for a full explanation. The rate of heat removal, Q, is given by,

$$Q = UA_H(T - T_c) \tag{5.6}$$

where the temperature of the cooling water, T_c, is defined as the mean of the inlet

and outlet cooling water temperatures,

$$T_c = \frac{1}{2}(T_{c,out} + T_{c,in}) \tag{5.7}$$

An energy balance on the jacket side of the reactor gives,

$$Q = \rho C_p F_c (T_{c,out} - T_{c,in}) \tag{5.8}$$

where F_c is the volumetric flow of cooling water. Finally, the flow leaving the reactor is proportional to the square root of the reactor holdup,

$$F_{out} = C_v k_v V_R^{0.5} \tag{5.9}$$

where C_v is a valve constant and k_v is the valve position. In experiments using exothermic reactors to teach control techniques Brisk observed that,

> *under all relevant operating conditions the exit cooling water temperature approached the reactor temperature to* $0.5 \pm 0.1°F$.

This assumption is used to simplify the expression for the rate of heat removal, Q,

$$Q = \rho C_p F_c (T_{out} - T_{c,in} - 0.5) \tag{5.10}$$

which enables equations 5.6, 5.7 and 5.8 to be removed from the system. Note that in this expression the temperature units used are $°F$. Table 5.1 summarises the kinetic parameters and input information used in the model.

K_0	$5.4 \times 10^{11}\ min^{-1}$
E_a/R	$16000\ ^\circ R$
ΔH_R	$-6089\ kJ/kmol$
C_{in}	$1\ kg/litre$
T_{in}	$20\ ^\circ C$
$T_{c,in}$	$20\ ^\circ C$

Table 5.1: Reactor parameters and input information

5.3.3. Storage tank

It is assumed that no energy transfer or reaction occur in the storage tank. The storage tank may then be modelled simply by dynamic overall and component mass balances in a similar fashion to the CSTR model above:

Overall material balance,

$$\frac{dV_{store}}{dt} = F_{in} - F_{product} \tag{5.11}$$

Feed component material balance,

$$V_{store}\frac{dC_{product,A}}{dt} + C_{product,A}\frac{dV_{store}}{dt} = F_{in}C_{in,A} - F_{product}C_{product,A} \tag{5.12}$$

The product flow rate from the storage tank is determined solely by the customer and is assumed to be a pumped flow.

5.4. Open-loop design procedure

The aim of this design is to find the economically optimal combination of:

1. unit type and topology

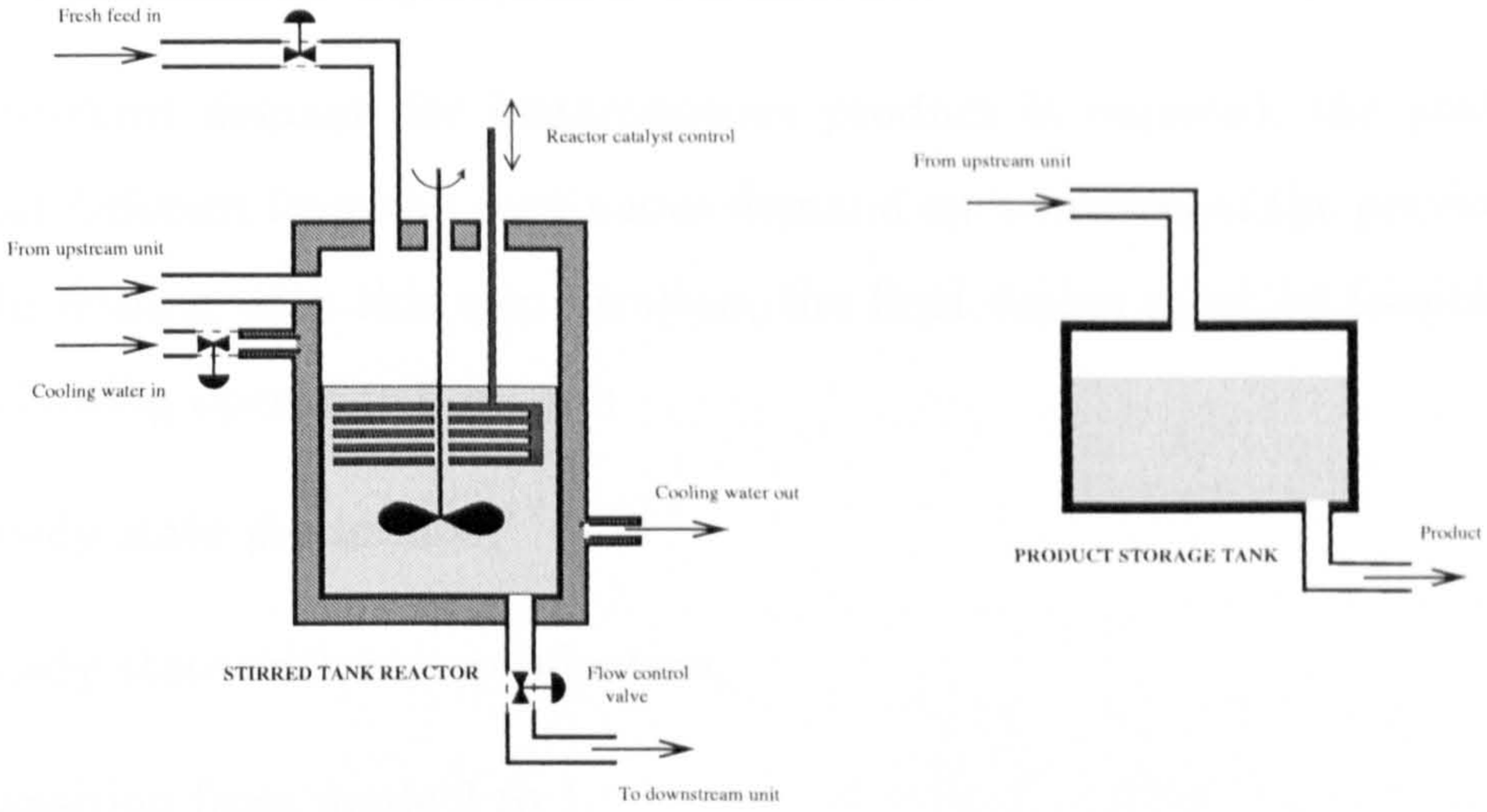


Figure 5.1: Process building blocks

2. unit design

3. cyclic process operation

As indicated in section 3.2.2, no attempt is made to solve the structural and operational problems simultaneously. Instead, the two stage, decomposed, design procedure will be employed:

- develop a candidate process structure
- apply a dynamic optimisation formulation to determine the corresponding optimal operating cycle and and design for that candidate

By decomposing the problem into these two levels, potentially expensive dynamic optimisation techniques are applied only when a viable process structure is in place. The resulting second stage optimisation problem is greatly simplified by the absence of combinatorially explosive integer decisions used to determine the process structure.

5.4.1. Candidate structure selection

As *intermittent* demand for instantaneous product is required, the problem is somewhat different from the continuous demand encountered in the previous case study. In dealing with this complication, the final design must be feasible in all of the following operational modes:

1. steady state production,
2. steady state with zero production,
3. transition from mode 2 to 1,
4. transition from mode 1 to 2.

A possible starting point in developing a candidate structure is to assess the feasibility of the design in each of these modes.

1. **feasible steady state production**

   In the presence of an appropriate set of reactor conditions (i.e. residence time, reactor temperature), production of B at the required rate and quality will be realisable.

2. **feasible steady state with zero production**

   As the product demand is zero in this state, the overall material balance dictates that all process material flows must also be zero. Observing the steady state material and energy balances in the reactor:

$$0 = kV_R C_A \tag{5.13}$$

$$0 = -kV_R C_A \frac{(\Delta H_R)}{\rho C_p} \tag{5.14}$$

k, C_A or V_R must become zero for the design to have a feasible shutdown steady state.

3. **feasible transitions from mode 1 to 2 and from mode 2 to 1**

It is assumed at this stage of the design procedure that the required transitions between operating points are feasible. Recall also that a premise in this work is that the instantaneous response demanded by the customer can be achieved by storing the required product. Since this process has a single product it is assumed here that a single storage vessel gives sufficient buffering between the customer and the process during transient modes of operation. In other words the storage will act as a source for product during operational mode 3 and a sink for product during operational mode 4. The feasibility of startup and shutdown is verified when the process is investigated closed-loop; see section 5.5 for the profiles of the base case where this is demonstrated.

5.4.1.1. Steady state bounding

If the assumptions regarding the transitional periods are valid, any process containing a series of n CSTRs and a storage tank may be considered as a feasible design alternative. A method for efficient screening such alternatives is needed, as it would be computationally inefficient to enumerate using dynamic optimisation. Consider the design of a corresponding 'steady state' process which is required to produce at the same rate and purity as the 'dynamic, responsive' process outlined in the problem statement. However, although the total annual production of both processes is identical, the steady state process will produce B over one continuous operational period.

"The optimal annualised cost of the steady state process will be less than or equal to the cost of the dynamic process."

Rowe (1997)

This heuristic will provide the optimal number of CSTRs in the successful candidate and a lower bound for the dynamic optimisation (see figure 5.2). When the bounding property described above does not hold, i.e. the cost of the dynamic process exceeds the second-best steady state solution) further analysis of the dynamic solution is required. The analysis aims to identify reasons for the increased cost and determine the extent to which these increases will propagate through to the alternative systems. One obvious extra cost of the dynamic system is that of the storage tank. Indeed the degree of responsiveness may be viewed as what needs to be "added" to the steady state system to make it dynamic. The closer the two are, either in terms of volume or cost, the more responsive the underlying system structure. This follows the comments made in the previous chapter in section 4.2.3.5 that one metric may be the ratio between the product storage volume/cost and that of the rest of the system.

5.4.1.2. Steady state optimisation

Essentially, the steady state form of the models described in section 5.3 is used to carry out the optimisation. The *gPROMS/gOPT* environment was used to carry out this phase of the design, as well as the optimal control problem in the following phase.

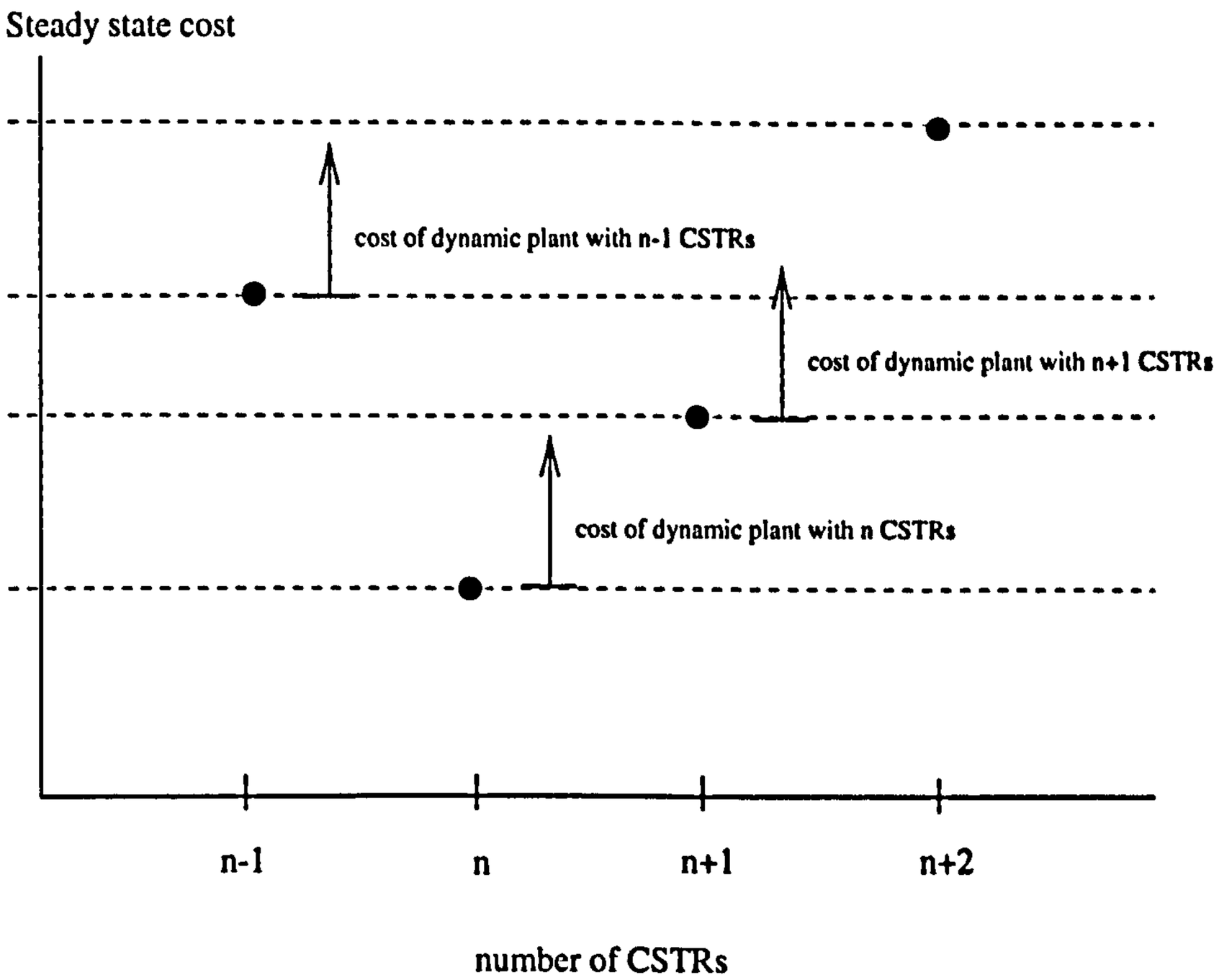


Figure 5.2: Screening dynamic process alternatives through steady state optimisation

5.4.1.3. Objective function

The objective function to be used is the minimisation of the total annualised cost of the process.

$$\min_{u,d}\{C_{op}^{ss} + C_{capital}\} \tag{5.15}$$

The individual contributions of capital and operating costs are determined as follows. The operating cost per minute of a single steady state CSTR is given by,

$$C_{CSTR_{op}}^{ss} = 7.063 \times 10^{-2} F_{in}^2 + 3.532 \times 10^{-4} F_c + 1.766 \times 10^{-2} V_R^{max} Catalyst_{control} \tag{5.16}$$

Note that the index to F_{in} denotes the second feed into the reactor, and does *not* mean squared. Recall that the total annual production of the steady state process must equal that of the dynamic process. Given this, the annualised operating cost of a system with n CSTRs will be,

$$C_{op} = N_{cycle} t_{product} \sum_{i=1}^{n} C^{ss}_{CSTR_{op_i}} \tag{5.17}$$

The nominal values used for N_{cycle} and $t_{product}$ are 365 (days/year) and 12 (hours/day) respectively.

The annualised capital cost of the process is recalculated without a term for the cost of storage, which is not required during steady operation,

$$C_{capital} = \frac{1}{3} \sum_{i=1}^{n} C_{CSTR_i} \quad cost\,units \tag{5.18}$$

where,

$$C_{CSTR_i} = 1 \times 10^5 + 9.629 \times 10^3 (V_{R_i}^{max})^{0.7} \quad cost\,units \tag{5.19}$$

5.4.1.4. Optimisation parameters

The parameters available for optimisation are the steady state values of the controls and the equipment sizes.

1. flow of fresh A to the reactor,

2. flow of cooling water through the cooling jacket,

3. position of catalyst system,

4. reactor holdup through manipulation of exit valve position.

5.4.1.5. Constraints

In addition to providing simple bounds on the optimisation variables the necessary operational constraints must be applied as inequality constraints in the steady state optimisation as follows:

$$\left.\begin{array}{l} T_i \leq 48.89 \\ V_{R_i}^{max} \leq 50 \end{array}\right\} i = 1, n \tag{5.20}$$

$$81.69 \leq C_{product} \leq 84.90 \tag{5.21}$$

Note that the maximum holdup constraint is actually enforced directly by appropriate bounding of the holdup optimisation parameter.

5.4.2. Results from steady state optimisation

Reactor networks consisting of between 1 and 5 CSTRs were optimised at steady state. The results from the first 3 optimisations are presented below in tables 5.2, 5.3 and 5.4. The total annualised costs for each system are summarised in table 5.5; the most economic solution (with a cost of is 641178 cost units) is to use two CSTRs. It is interesting to note that the solutions presented share a number of features in common:

1. all reactors are operated at the maximum allowable temperature,

2. the B produced is of the lowest allowable purity (81.69%),

3. all fresh feed required enters through the first reactor.

The choice of these operating decisions may be explained as follows.

- Operation of the reactors at the maximum allowable temperature gives the maximum conversion of A to B for a given reactor residence time.

- Producing B of increasing purity requires greater residence times and results in increasing costs.

- Feeding all the fresh A to the first reactor gives a residence time distribution for the system which is as close to a plug flow reactor as possible (Froment and Bischoff, 1990). This arrangement gives the highest conversion for a given CSTR network involving a first order rate of reaction.

Note that to obtain the required product purity in a system containing just one CSTR would require a CSTR volume of 97.15 *litres* which violates the maximum holdup limit of 50 *litres*.

Design parameter	
Fresh feed of A	$12.3\,litres.min^{-1}$
Cooling water flow	$3.43\,litres.min^{-1}$
Position of catalyst	1.00
Holdup	$97.15\,litres$
Reactor states	
Temperature	$48.89\,°C$
Concentration of B	$0.8169\,kg.l^{-1}$
Economics	
Annual operating cost	679498
Annualised capital cost	112341
Total annualised cost	791839

Table 5.2: Optimal steady state design for 1 CSTR

Design parameter	CSTR 1	CSTR 2
Fresh feed of A	$12.3\,litres.min^{-1}$	$0.00\,litres.min^{-1}$
Cooling water flow	$0.00\,litres.min^{-1}$	$3.43\,litres.min^{-1}$
Position of catalyst	1.00	1.00
Holdup	$38.73\,litres$	$21.02\,litres$
Reactor states		
Temperature	$48.89\,°C$	$48.89\,°C$
Concentration of B	$0.6401\,kg.l^{-1}$	$0.8169\,kg.l^{-1}$
Economics		
Annual operating cost	408049	97896
Annualised capital cost	74837	60396
Total annualised cost	482886	158292

Table 5.3: Optimal steady state design for 2 CSTRs

Design parameter	CSTR 1	CSTR 2	CSTR 3
Fresh feed of A	$12.3\,litres.min^{-1}$	$0.00\,litres.min^{-1}$	$0.00\,litres.min^{-1}$
Cooling water flow	$0.0\,litres.min^{-1}$	$2.00\,litres.min^{-1}$	$1.43\,litres.min^{-1}$
Position of catalyst	1.00	1.00	1.00
Holdup	$38.73\,litres$	$8.75\,litres$	$8.76\,litres$
Reactor states			
Temperature	$48.89\,°C$	$48.89\,°C$	$48.89\,°C$
Concentration of B	$0.6401\,kg.l^{-1}$	$0.7432\,kg.l^{-1}$	$0.8169\,kg.l^{-1}$
Economics			
Annual operating cost	408049	40779	40789
Annualised capital cost	74837	47980	47996
Total annualised cost	482886	88759	88785

Table 5.4: Optimal steady state design for 3 CSTRs

System	*Annualised cost*
1 CSTR	791839
2 CSTRs	641178
3 CSTRs	660430
4 CSTRs	691567
5 CSTRs	731791

Table 5.5: Summary of reactor network costs

Through the use of steady state optimisation the system with lowest annualised cost is identified as having two CSTR units. Based on the steady state bounding procedure described earlier, this system is carried forward as the candidate structure to be considered in the second stage of the overall design procedure.

5.4.3. Dynamic optimisation

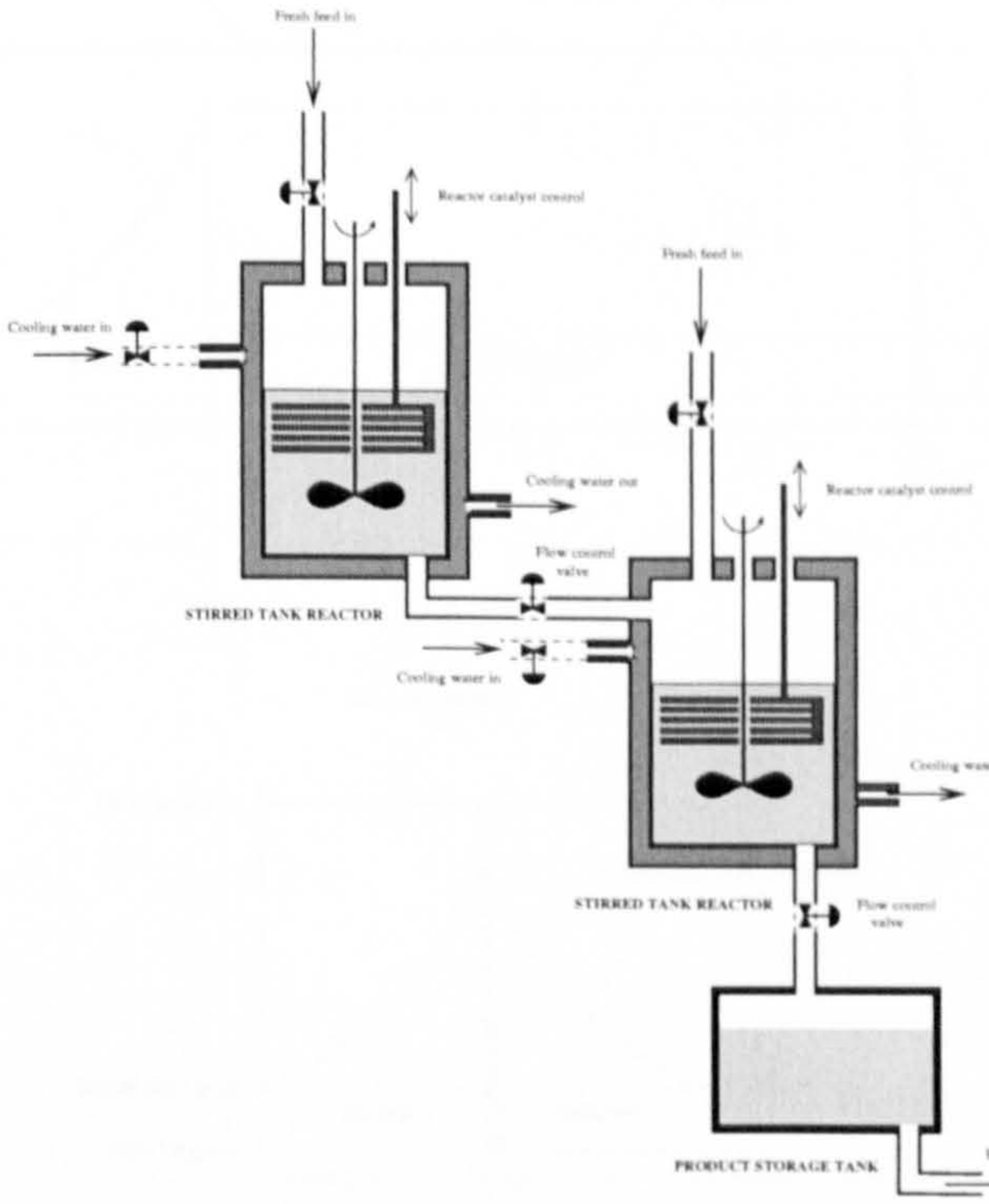
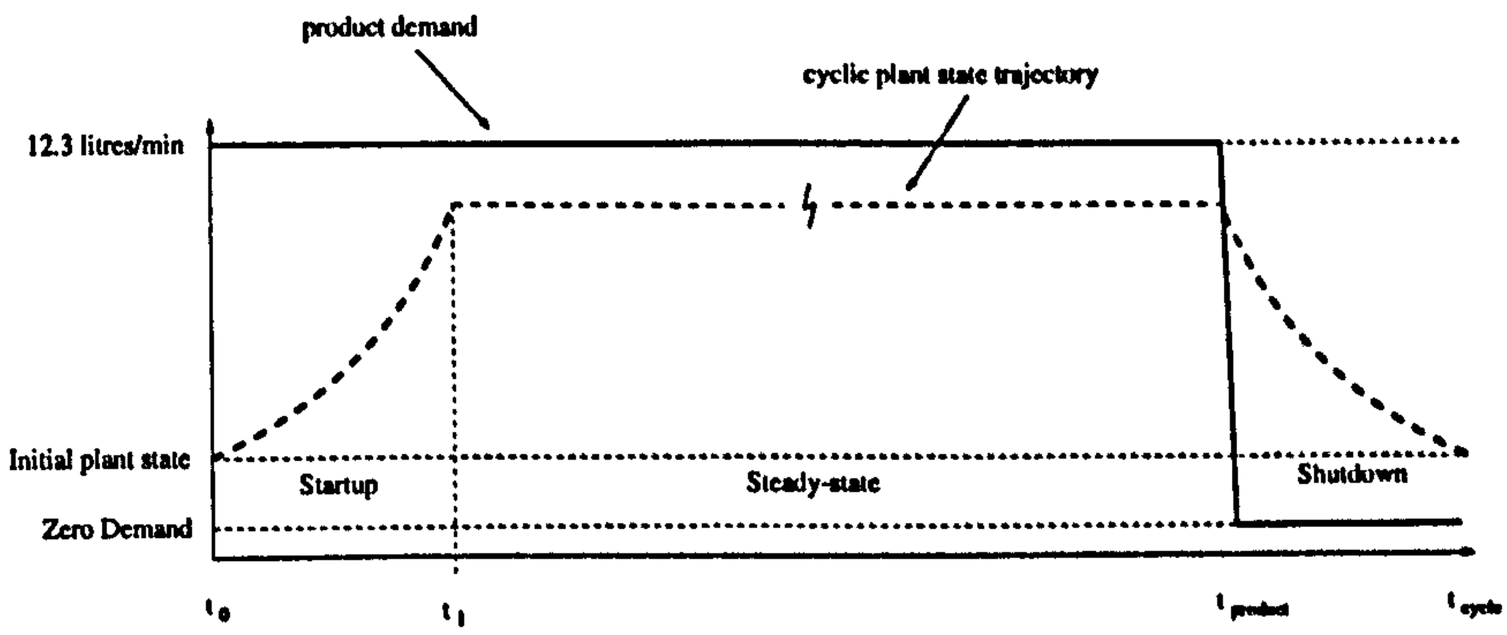


Figure 5.3: Successful candidate for the optimal control problem

Figure 5.4 shows how the problem description maps to the cyclic operation framework. At time t_0 the customer demands product. The process then moves from its shutdown state to the production steady state, which is reached at time t_1. At this time the controls are frozen. At time $t_{product}$ no further product is required and demand drops to zero. Simultaneously, the process moves back to the shutdown state defined at time t_0. This state is reached at time t_{cycle}. The cycle for the actual process is collapsed as shown in figure 5.4 to avoid unnecessary



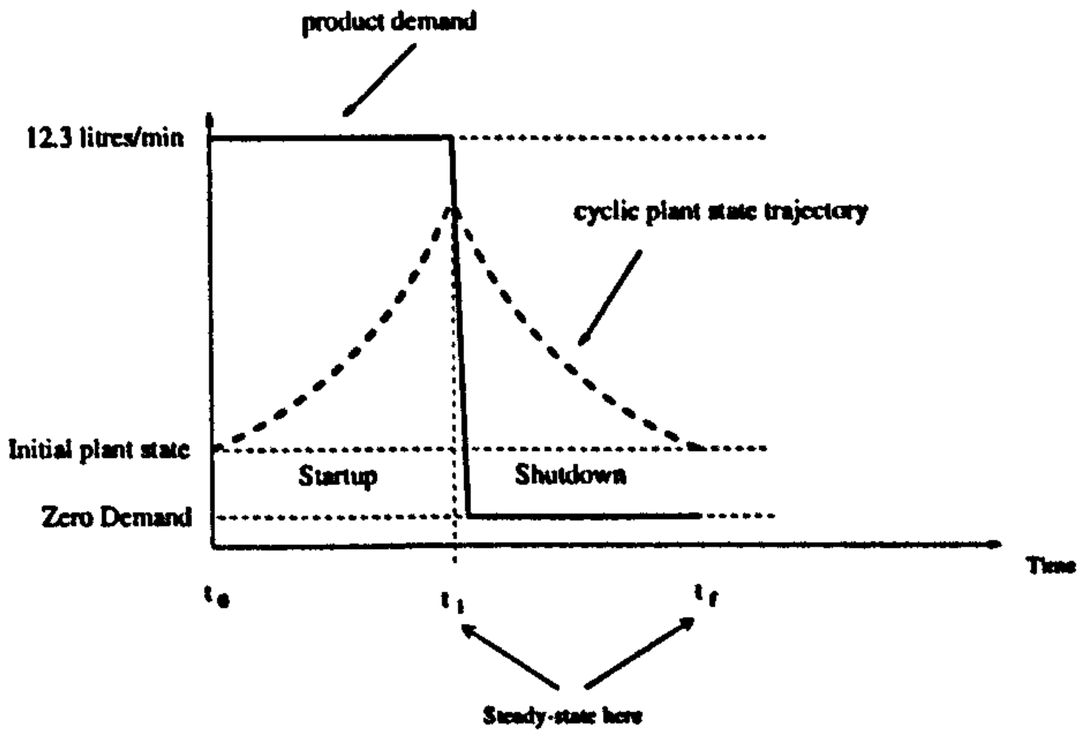


Figure 5.4: Diagram showing original operating cycle (top) and the collapsed cycle which is implemented (bottom).

and time consuming integration through the steady state period during solution. Provided that this is accounted for in the process operating cost, the two cycles are identical for the purposes of determining the optimal cyclic operation.

The disadvantage in collapsing the time horizon as described lies in the assumption that steady state is reached when the first order derivatives of the system states "equal" zero at some time t_1. Equality point contraints are imposed in the dynamic optimisation framework used in this case study (see section 5.4.7.2) that ultimately cannot guarantee convergence to steady state since the derivatives can only be contrained to being within finite tolerances. Although some rudimentary measures are taken to account for this, the only rigorous way to enforce convergence is to impose additional equality point constraints on all higher order derivatives which would make the problem intractable. This shortcoming is resolved in the second part of this chapter with the addition of a robust control scheme as closed-loop operation will guarantee a stable steady state.

5.4.4. Objective function

As discussed in section 2.2 one of the underlying requirements when posing an integrated design problem is that the problem can be *written entirely in terms of costs and constraints* (Walsh and Perkins, 1996). The motivation for solving such a problem is to find the cheapest process design satisfying the constraints on the design. The objective function for the optimisation should therefore be an economic one. The specific form of this objective will now be considered.

Recall that the customer requirements given in section 5.2 specify a production rate of $12.3\,litres.min^{-1}$. The precise profile for the demand over the course of a year is, however, uncertain. Figure 5.4 shows a nominal operating cycle for the process. The cycle consists of three phases: two transient phases (start up and shut down) and one steady phase. In any given year there will a number of such cycles, N_{cycle}. Each cycle has associated with it a period for which product is demanded,

$t_{product}$, as shown in the figure. The final design of the process will be a function of the form of this demand profile. For example, a plant which is optimally designed for a demand profile consisting of a large number of short production runs (large N_{cycle} and small $t_{product}$) will typically have different characteristics to one whose design is based upon much fewer runs of longer duration (large $t_{product}$ small N_{cycle}). Qualitatively, increasing N_{cycle} while reducing the length of $t_{product}$ would be expected to increase economic impact of the transient phases in the operating cycle. Conversely, as $t_{product}$ increases and N_{cycle} is reduced it is not unreasonable to expect the economics of the optimal production steady state to dominate. The economic objective function to be used should therefore take account of the form of the demand profile. The cost of a single operating cycle of the process may be written as:

$$operating\ cost\ per\ cycle = C_{su} + C_{sd} + (t_{product} - t_1)C_{ss}$$

where C_{su} is the cost of the startup phase, C_{sd} is the cost of the shutdown phase, C_{ss} is the cost per unit time of the steady state operation and t_1 is the time to start up (which must be less than $t_{product}$). The expected annualised operating cost of the process is therefore given by:

$$\begin{aligned} E(annual\ operating\ cost) \;=\; & E(N_{cycle})(C_{su} + C_{sd}) + \\ & E\left(N_{cycle}(t_{product} - t_1)\right) C_{ss} \end{aligned} \tag{5.22}$$

If $t_{product}$ and N_{cycle} are independent then:

$$E(N_{cycle}.t_{product}) = E(N_{cycle})E(t_{product}) \tag{5.23}$$

The *total* expected annualised cost of the process including capital costs is there-

fore given by:

$$E(annualised\ cost) = E(N_{cycle})(C_{su} + C_{sd}) + C_{capital} + E(N_{cycle})\,(E(t_{product}) - t_1)\,C_{ss} \quad (5.24)$$

where $C_{capital}$ is the annualised process capital cost.

The optimisation objective function is then:

$$J = \min_{u(t),d} E\{annualised\ cost\} \quad (5.25)$$

where $u(t)$ are the time varying controls acting on the process and d is the set of design parameters.

For this case study it is assumed that, on average, the plant will undergo one operating cycle per day with a mean product delivery time of twelve hours. This corresponds to values for $E(N_{cycle})$ and $E(t_{product})$ of 365 and 12 hours respectively.

5.4.4.1. Capital costs

As stated previously, the final design will consist of a number of CSTRs followed (potentially) by a product storage tank. The capital cost of each CSTR building block as shown in figure 5.1 is given by,

$$C_{CSTR} = 1 \times 10^5 + 9.629 \times 10^3 (V_R^{max})^{0.7} \quad cost\ units \quad (5.26)$$

where V_R^{max} is the maximum level of holdup in the CSTR (in *litres*) during the operating cycle. The cost of the storage tank building block is defined in a similar

way,

$$C_{store} = 5 \times 10^4 + 4.814 \times 10^3.(V_{store}^{max})^{0.7} \quad cost\,units \tag{5.27}$$

where V_{store}^{max} is the maximum storage requirement (in *litres*) during the operating cycle. For a system containing n CSTRs and taking the most general case when product storage is required, the total annualised capital cost may be written as,

$$C_{capital} = \frac{1}{3}\left\{\sum_{i=1}^{n} C_{CSTR_i} + C_{store}\right\} \quad cost\,units \tag{5.28}$$

The $\frac{1}{3}$ pre-multiplier is a capital charge factor (CCF) which approximates "*the time value of money*" (Douglas, 1988), and converts the total capital cost to an annualised equivalent.

5.4.4.2. Operating costs

The operating cost may also be considered for a single CSTR building block. In each CSTR the fresh feed, cooling water flow and catalyst all contribute to the operating cost. Fresh feed of A is costed at $7.063 \times 10^{-2} cost\,units.litre^{-1}$. Cooling water is costed at $3.532 \times 10^{-4} cost\,units.litre^{-1}$. During use the catalyst will become poisoned and will periodically need to be renewed. The costs associated with this are incorporated as follows: when being "consumed" by the liquid reactant, the catalyst is costed at $1.766 \times 10^{-2} cost\,units.min^{-1}.litre^{-1}$, otherwise it has zero cost. Note that in the case of catalyst cost, the unit $litre^{-1}$ is per unit reactor volume since the catalyst required is assumed to be in proportion to the maximum reactor holdup. Product pumping costs from the storage tank are neglected so that this unit has zero operating cost. The operating cost per unit

time for a single CSTR is therefore given by,

$$C_{CSTR_{op}}(t) = 7.063 \times 10^{-2} F_{in}^2(t) + 3.532 \times 10^{-4} F_c(t) + 1.766 \times 10^{-2} V_R^{max} Catalyst_{control}(t) \quad (5.29)$$

where, as before, $Catalyst_{control}$ takes a value from 0 to 1 and indicates the presence of catalyst in the reactor. The cost of an operating cycle per CSTR is then given by,

$$C_{CSTR_{op}} = 7.063 \times 10^{-2} \int_0^{t_{cycle}} F_{in}^2(t)dt + 3.532 \times 10^{-4} \int_0^{t_{cycle}} F_c(t)dt + 1.766 \times 10^{-2} V_R^{max} \int_0^{t_{cycle}} Catalyst_{control}(t)dt \quad (5.30)$$

where t_{cycle} is the length of a *process* operating cycle. As discussed earlier, the process operates at steady state for a period of time $(t_{product} - t_1)$. The optimisation cycle considered is of length t_f, where $t_f = t_{cycle} - (t_{product} - t_1)$, since (as stated previously) integration over the steady state period of operation is unnecessary. The cost of the steady state period must, however, be incorporated into the objective function. Equation 5.30 is therefore modified to reflect this.

$$\begin{aligned} C_{CSTR_{op}} = \; & \int_0^{t_f} (7.063 \times 10^{-2} F_{in}^2(t) + \\ & 3.532 \times 10^{-4} F_c(t) + 1.766 \times 10^{-2} V_R^{max} Catalyst_{control}(t))dt \\ & + (7.063 \times 10^{-2} F_{in}^2(t_1) + 3.532 \times 10^{-4} F_c(t_1) + \\ & 1.766 \times 10^{-2} V_R^{max} Catalyst_{control}(t_1))(t_{product} - t_1) \end{aligned}$$

The total annual operating cost given that the plant completes N_{cycle} per year is then:

$$C_{op} = N_{cycle} \sum_{i=1}^{n} C_{CSTR_{op_i}} \quad (5.31)$$

The objective function (5.25) to be used is therefore,

$$\min_{u(t),d}\{C_{op} + C_{capital}\} \tag{5.32}$$

It should be noted that in the objective function described above the shutdown state of the plant at time t_f is assumed to have an associated zero operating cost.

5.4.5. Plant parameters for optimisation

Determination of the best combination of plant design and operating policy is achieved by the manipulation of three distinct sets of design parameters in the process:

1. time varying optimisation parameters (*controls*),
2. the plant initial state,
3. equipment sizes.

Similarly to the discussion of the objective function earlier, analysis of the optimisation parameters to be used in this case study may be considered in terms of each process building block. Four controls are available on each CSTR (see figure 5.1) as follows:

1. flow of fresh A to the reactor,
2. flow of cooling water through the cooling jacket,
3. exit flow from reactor (through manipulation of the control valve position),
4. position of catalyst system.

In the case of the product storage tank no control variables are available. The product demand from storage is set by the customer and the feed to the storage

is supplied by the exit flow of the preceding reactor (which is a control variable for that reactor).

Each CSTR unit is described by three state variables: volume, concentration and temperature. The storage tank is described by just two state variables; volume and concentration. In the cyclic operation framework proposed the initial values for all of these states are included as optimisation parameters.

Finally, the volume of all CSTR units in the plant together with that of the storage tank are optimised. The equipment size is given by the maximum value of the holdup in a unit during the operating cycle. V_R^{max} and V_{store}^{max} are recorded directly by a lagging variable.

5.4.6. Control parameterisation and time discretisation

Recall from chapter 3 that a feasible path control parameterisation method will be used to solve the dynamic optimisation problem. This section considers the choice of both the functional form of the controls and the manner in which the time horizon is discretised.

5.4.6.1. Control parameterisation

Piecewise linear controls will be implemented as these give the highest precision while still ensuring the control variables remain correctly bounded using just simple bounds on the break points.

5.4.6.2. Time discretisation

As with the functional form of the controls, the number of discretisation intervals must also be fixed *a priori* in the problem description. Although a complete operating cycle is being optimised, it essentially consists of three phases: startup to production steady state, product ramped to zero and shutdown. The number

of variable time intervals chosen will therefore be split equally between the startup and shutdown phases of the cycle. Thirteen discretisation intervals are used, which allows 6 intervals for each of the startup and shutdown phases. This is anticipated to give sufficient flexibility during solution whilst giving an acceptable number of optimisation parameters for designs incorporating 2 CSTRs.

In addition to fixing the number of time discretisation intervals to be used, the lengths of those intervals which are optimised must also be bounded. An upper bound is arbitrarily set at 80 minutes. The lower bound on the interval length was set at 0.1 minutes. These bounds were inactive at the optimum solution. The fixed time interval over which the product flow is ramped to zero is given a value of 1×10^{-3} minutes to represent the product flow being cut off by the customer.

5.4.7. Constraints

The optimal design and operation of the reactor network is subject to a number of constraints arising from two distinct sources:

1. operational constraints on the plant itself,

2. constraints required by the cyclic optimisation framework.

In this section both sources of constraints will be discussed in relation to the context of the optimisation problem to be solved.

5.4.7.1. Path Constraints

Path constraints are those constraints on the system which must be met for all times in the optimisation. The majority of these result from operational requirements.

Recall from section 5.2 that a number of factors have led to on-site manufacture of B being considered, namely:

- limits on inventory in individual vessels and on total site inventory,

- degradation of B in storage, which becomes significant above $48.89°C$.

The implication of the inventory limits is clear, the second factor is incorporated by restricting the maximum temperature of any vessel in which B is produced to $48.89°C$. These requirements must be met at all times and may be represented as inequality path constraints in the optimisation. The constraints may be generalised for n reactor units as follows.

$$\left.\begin{array}{l} T_{out_i} \leq 48.89 \\ V_{R_i}^{max} \leq 50 \end{array}\right\} i = 1, n \tag{5.33}$$

No heating or cooling occurs in the storage unit and so only the maximum inventory constraint is applied:

$$V_{store}^{max} \leq 50 \tag{5.34}$$

The final operational constraint on the plant is that the customer requires all feedstock supplied to contain between 81.69% and 84.90% (w/w) of B. Again, this requirement must apply at all times and is written as the path constraint:

$$81.69 \leq C_{product} \leq 84.90 \tag{5.35}$$

where $C_{product}$ is the concentration of B in the product. A means of "marking" the maximum holdup in a unit is required both for costing the plant capital (as part of the objective function) and for use in the inventory constraints described above. This is achieved by using lagging variables that record the maximum value over time, as mentioned previously.

Recall that numerical robustness in the storage tank model was achieved through a proportionally controlled make-up stream (see section 4.2.2.4 for discussion). This was added to prevent negative holdup in this tank. At the solution point the make-up stream must not be active and a further path constraint must be added to ensure this.

5.4.7.2. Equality point constraints

The introduction to this section discussed the way in which this case study fits into the general framework proposed in chapter 3. The remaining constraints to be included in the problem definition are a specific requirement of this framework. Briefly, in the context of this case study the purpose of these constraints is to:

1. ensure that a steady state is reached following the startup phase,

2. ensure that a steady state is reached following the shutdown phase,

3. ensure that the complete operation is cyclic.

At time t_1 in the operating cycle the plant is required to be at a steady state capable of meeting the demand requirements of the customer. Stationary points in the state profiles at t_1 **should** ensure that this demand is sustainable *ad infinitum*. However, for verification purposes and to aid the convergence of the optimal control problem, the controls are frozen for a one minute period to check if steady state is indeed achieved. At the final time in the operating cycle, t_f, customer demand is zero and the plant must have reached a new shutdown steady state. As at time t_1, this state is sustainable indefinitely until the customer again requires product. By requiring the state of the plant at the start of the cycle, t_0, to be the same as that at t_f, the cycle itself is guaranteed to be repeatable indefinitely. These requirements are enforced as equality constraints which must be satisfied at specific points in time (t_1 and t_f) and are applied to the states and their respective time derivatives in each unit. Each CSTR unit has three states describing its temperature (T_{out}), concentration (C_{out}) and holdup (V_R). The storage tank has two states describing its concentration ($C_{product}$) and holdup (V_{store}). Since the dynamic behaviour of the process states is determined using optimal open-loop control, exact steady state requirements must be imposed at t_1 and t_f. For a system of n CSTRs and storage tank the constraints are therefore:

1. steady state constraints

$$\left.\begin{array}{l} \left.\begin{array}{rcl} \dot{T}_{out_i}(t) & = & 0 \\ \dot{C}_{out_i}(t) & = & 0 \\ \dot{V}_{R_i}(t) & = & 0 \end{array}\right\} i = 1, n \\ \begin{array}{rcl} \dot{C}_{product}(t) & = & 0 \\ \dot{V}_{store}(t) & = & 0 \end{array} \end{array}\right\} t = \{t_1, t_f\} \qquad (5.36)$$

2. cyclic operation constraints

$$\left.\begin{array}{rcl} T_{out_i}(t_0) & = & T_{out_i}(t_f) \\ C_{out_i}(t_0) & = & C_{out_i}(t_f) \\ V_{R_i}(t_0) & = & V_{R_i}(t_f) \end{array}\right\} i = 1, n$$
$$\begin{array}{rcl} C_{product}(t_0) & = & C_{product}(t_f) \\ V_{store}(t_0) & = & V_{store}(t_f) \end{array} \qquad (5.37)$$

where $\dot{C}$ denotes the time derivative of C *etc.*

5.4.8. Optimal cyclic operation of the candidate structure

This section describes how the optimal cyclic operation of the candidate structure (shown in 5.5) is determined using the dynamic optimisation formulation presented earlier. Recall the premise discussed earlier that for the dynamic system *instantaneous response is achievable through the use of product storage.* Based on this premise, a product storage tank is added to the optimal steady state solution as shown in figure 5.5.

5.4.8.1. Implementation

Model generation Using the unit models described in section 5.3 a complete model of the candidate process was developed within the *gPROMS* modelling environment. In addition to the underlying equations describing specific process units in the model, a number of ancillary equations were included. The additional equations provide information to be used by the optimiser in calculating values for the objective function and constraints. The total operating cost of the cycle (as given in equation 5.30) is determined by integrating the "per unit time" operating cost over the length of the operating cycle. To achieve this, a new state variable,

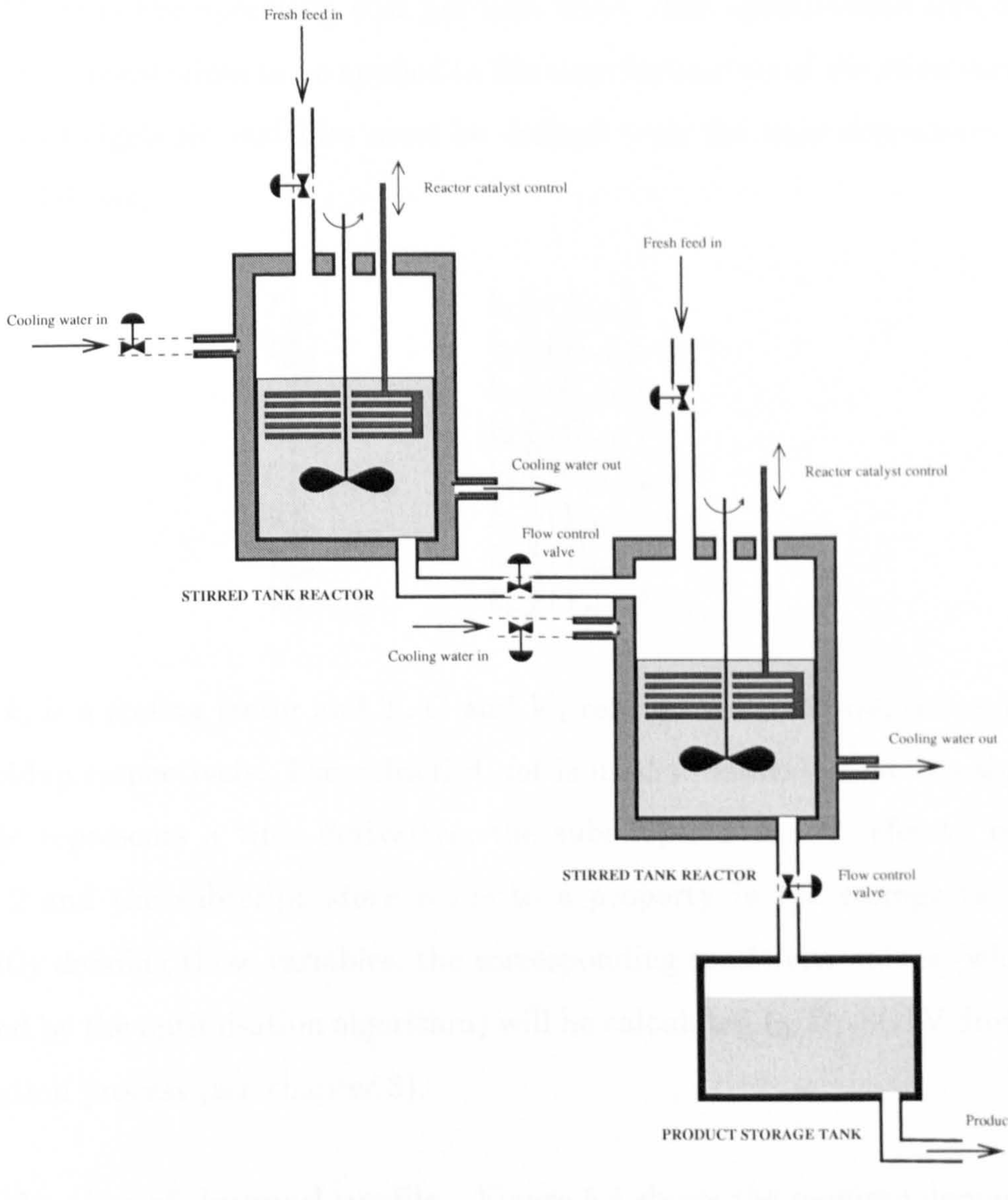


Figure 5.5: Candidate process for cyclic optimisation

C_{op}^{total} is defined in the model as follows,

$$\frac{d}{dt}(C_{op}^{total}) = C_{op}(t) \tag{5.38}$$

where $C_{op}(t)$ is the operating cost per unit time. The optimisation formulation also requires constraints to be applied to the time derivatives of the state variables.

A set of algebraic variables must be defined from the time derivatives of the states as follows,

$$\begin{aligned}
T_{dot}^1 &= k_1 \tfrac{d}{dt}(T_{out_1}) \\
T_{dot}^2 &= k_2 \tfrac{d}{dt}(T_{out_2}) \\
C_{dot}^1 &= k_3 \tfrac{d}{dt}(C_{out_1}) \\
C_{dot}^2 &= k_4 \tfrac{d}{dt}(C_{out_2}) \\
C_{product_{dot}} &= k_5 \tfrac{d}{dt}(C_{product}) \\
V_{dot}^1 &= k_6 \tfrac{d}{dt}(V_{R_1}) \\
V_{dot}^2 &= k_7 \tfrac{d}{dt}(V_{R_2}) \\
V_{store_{dot}} &= k_8 \tfrac{d}{dt}(V_{store})
\end{aligned} \tag{5.39}$$

where k_i is a scaling factor and T, C and V_R refer to temperature, concentration and holdup respectively. The subscript *dot* is used to indicate that the algebraic variable represents a time derivative, the subscripts 1 and 2 refer to reactors 1 and 2 and the subscript *store* refers to a property in the storage tank. By explicitly defining these variables, the corresponding sensitivity values (which are required by the optimisation algorithm) will be calculated by DASOLV during the integration process (see chapter 3).

Specification of demand profile Figure 5.4 shows the required demand profile for the cyclic operation. This section briefly describes the way in which this profile is incorporated in the optimisation.

The time discretisation to be used for this problem (see section 5.4.6.2 for further details) may be summarised as follows. Product is required for the first six time intervals, it is ramped down to zero over the fixed seventh time interval

and remains at zero for the final six time intervals. To achieve this, the product demand was set up in the optimisation as a time-varying control variable in the optimisation input file. The breakpoint values for this control were then defined to give the required profile, but were excluded from the set of available optimisation parameters. In doing this, the correct product demand profile was superimposed on the operating cycle.

5.4.8.2. Results and discussion for the candidate structure

The solution of the dynamic optimisation was initialised using the optimal steady state point obtained in section 5.4.2. The economic optimum for the annualised cost of the dynamic system was found to be **663223** cost units (compared to a value from the earlier steady state optimisations of 641178 for 2 CSTRs and 660430 for 3 CSTRs). This optimum will be re-assessed in the discussion which follows.

Figure 5.6 illustrate examples of the behaviour of the states. Figures 5.7 and 5.8 show the controls in the process over the course of an operating cycle. The graphs demonstrate two key points.

- The trajectories of the two states shown (concentration and holdup) are indeed cyclic.

- Time t_1, at which the product demand switches to zero occurs at $2.15 mins$. Both at this time and at the final time (t_f) the time derivative shown is zero (as are the other time derivatives). Freezing the control variables at either of these points would therefore give a steady state until such time as the control values are changed.

The optimised initial value for the storage holdup is on its lower bound ($V_{store_{min}}$), around 1.4 litres. It is inferred from this small volume that the storage serves no significant purpose in the operation of this process. Further proof that this is the

case can be seen in the state trajectory for concentration of B in the second CSTR. The purity of B in this CSTR does not violate product purity specifications at any time in the cycle. In other words, product of the required purity can feasibly be supplied at all times directly from the second CSTR. This finding concurs with the comments of Price and Georgakis (1993):

"Today's tigher focus on a customer's needs requires plants with substantial production flexibility. In such situations, excessive inventory is a liability, so designers tend to minimise the storage capacity between operations."

In the light of this analysis, the economic optimum will now be reviewed. The fixed costs associated with the storage tank are subtracted from the optimal cost obtained previously. In doing this a revised optimum of **644151** is calculated. This figure is within 0.5% of the steady state value. The optimum also lies below the steady-state optimum for 3 CSTRs. Note that if infinitesimal switching times were allowed in the control parameterisation this value would be identical to the steady-state economics. Based on the bounding properties discussed in section 5.4.1.1 no further structures need to be considered.

Figures 5.7 and 5.8 show catalyst controls, coolant flows and fresh feed flows. These graphs demonstrate two further important points:

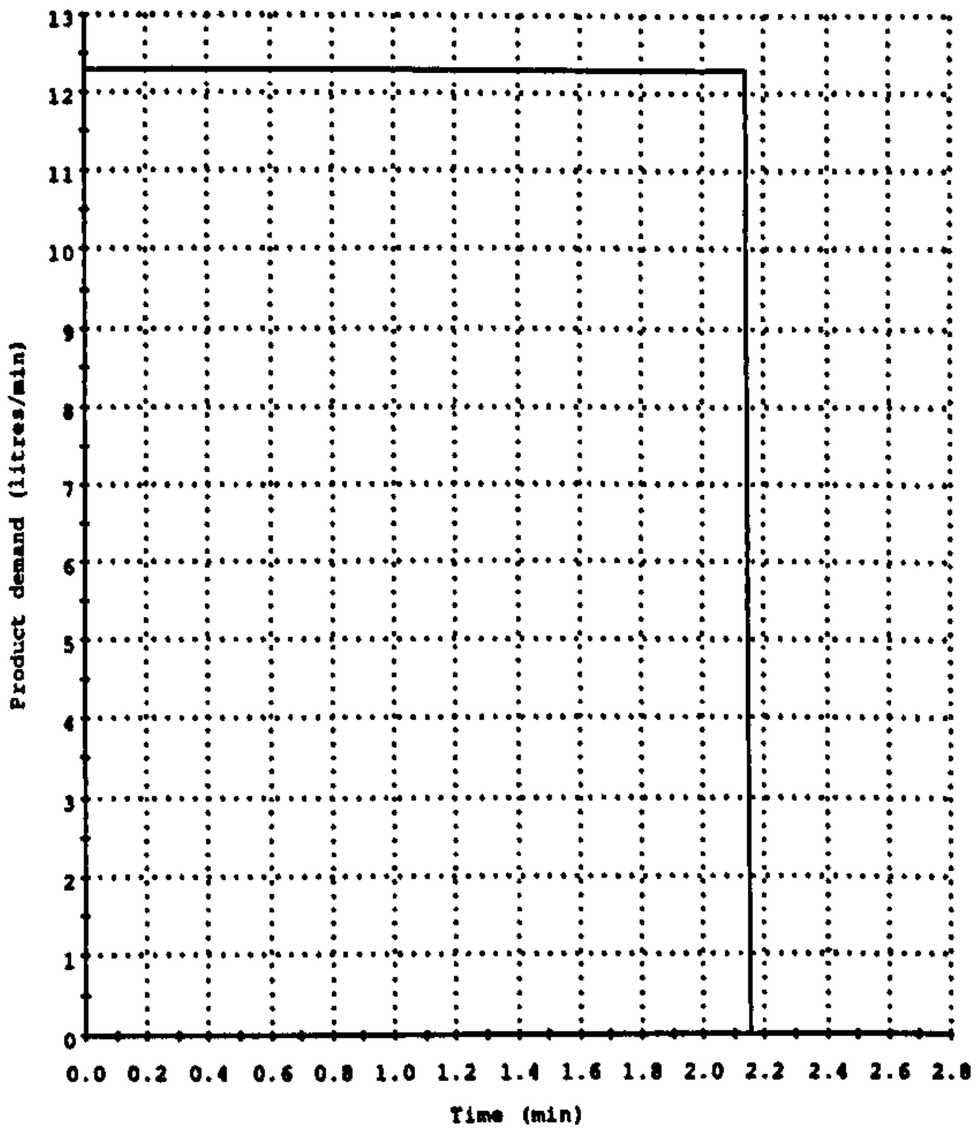
- At the final time all controls are at zero. The earlier assumption of a zero cost endpoint in the cycle is therefore validated.
- When product is required, the controls take almost identical values to those determined in the steady state optimisation. When no product is required, all controls are zero.

The peaks seen in the CSTR 1 reactant fresh feed supply and catalyst position profiles correspond to the attempt made by the optimiser to enforce the cyclic constraints.

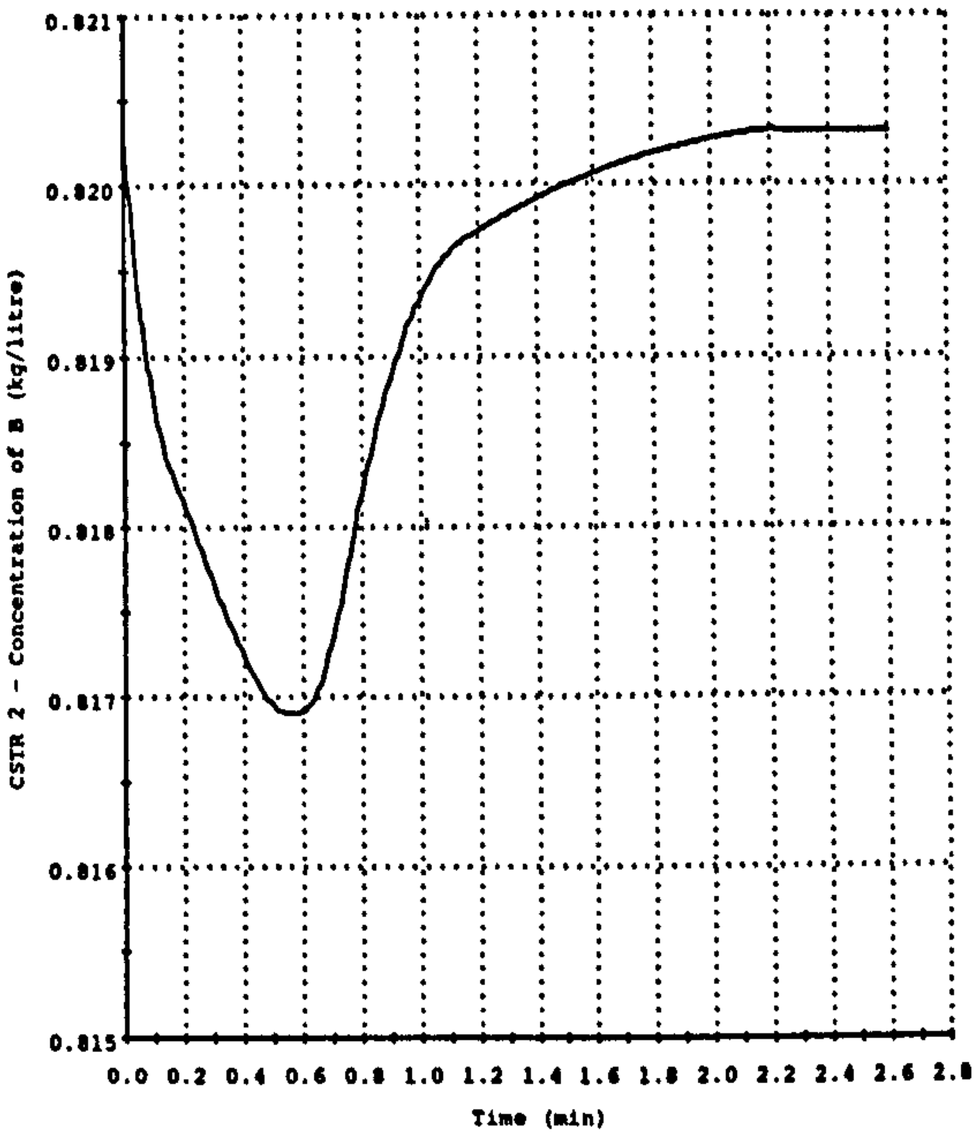
Table 5.6 summarises the values of all the states in the process at the shutdown steady state value (corresponding to the initial and final condition in the cycle) and at the production steady state. The values obtained in the steady state optimisation are presented for comparison. The difference between the states in the plant at shutdown and during steady production are clearly negligible. Transition between the two states is achieved simply by switching the controls from zero to the value required for optimum steady state production. A shutdown state with zero cost is clearly desirable given that, in theory, the plant can remain idle for an indefinite period of time. Achieving the fastest possible transitions between steady states is also an important factor in the design and will now be discussed. One of the assumptions in the operating cycle proposed in this work is that the process must complete the current transition between operating points before the reverse transition can be initiated. The implication here is that the process cannot start up during a shut down operation and, conversely, cannot shut down during a start up operation. Clearly, if the transitions are rapid the process offers greater flexibility to the customer.

state	*production state*	*shutdown state*	*steady state optimum*
CSTR 1: conc B	0.640	0.640	0.640
CSTR 2: conc B	0.817	0.823	0.817
CSTR 1: temp	48.823	48.885	48.889
CSTR 2: temp	48.885	48.835	48.889
CSTR 1:holdup	38.838	38.455	38.730
CSTR 2:holdup	21.567	21.111	21.020

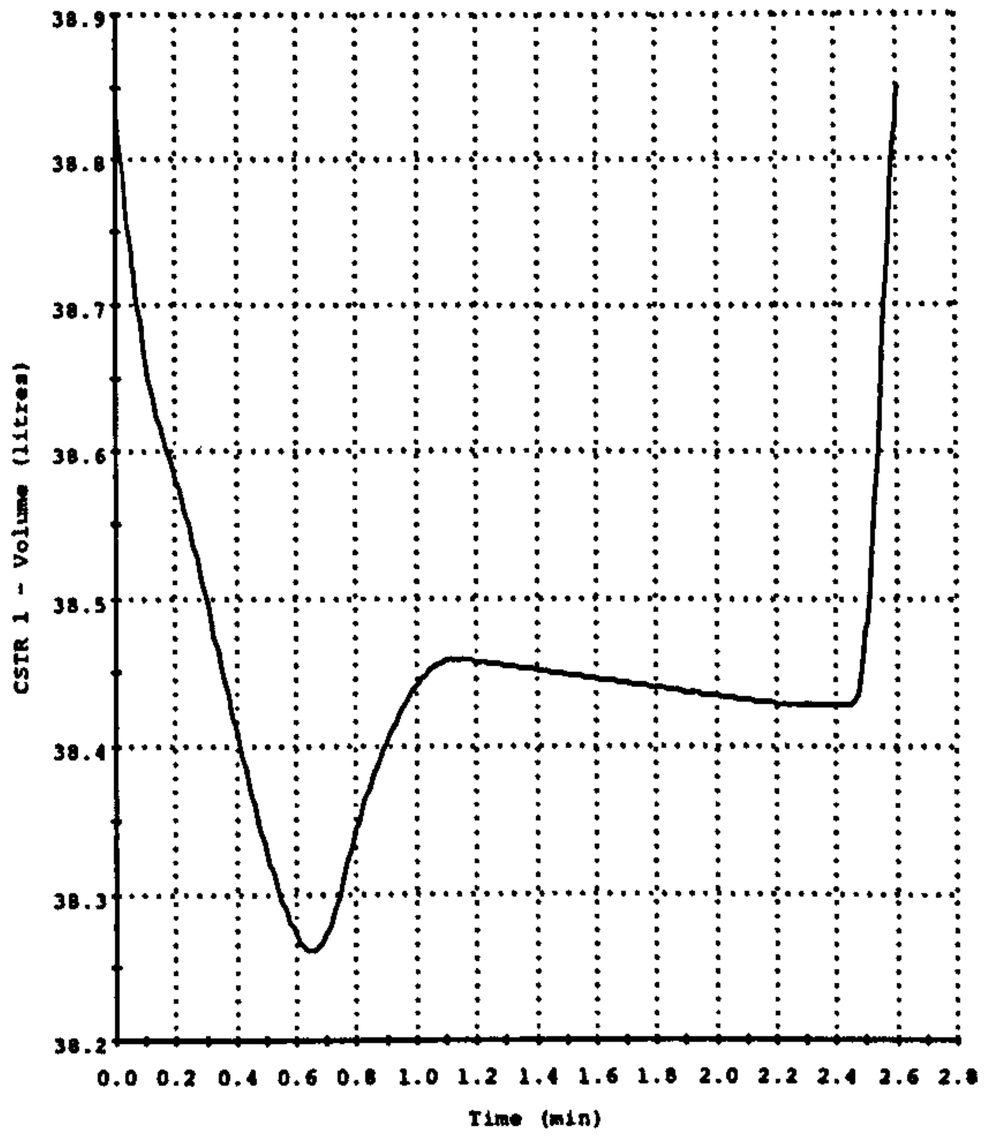
Table 5.6: State variables at shutdown and production steady states in both reactors



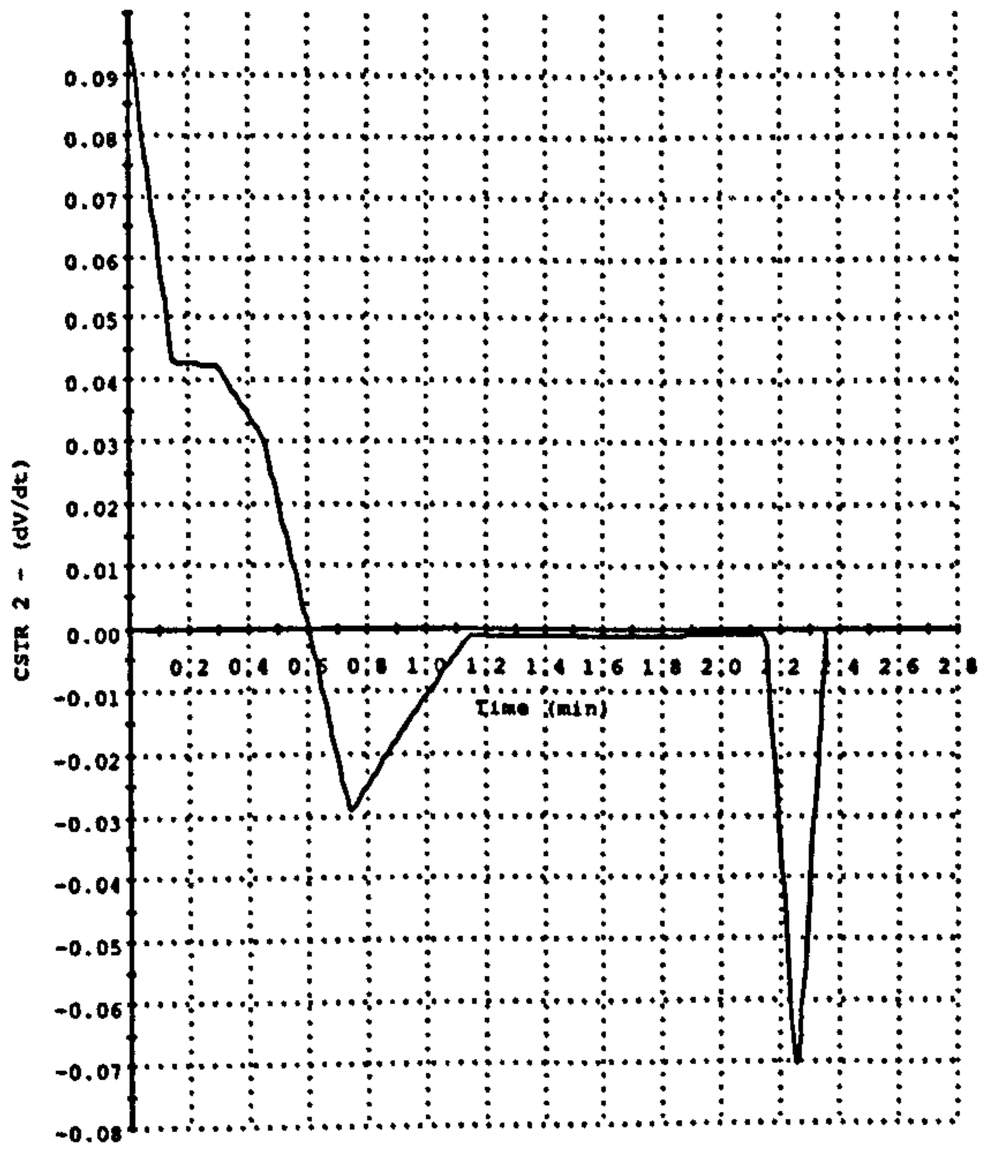
(a) Product demand



(b) CSTR 2 product concentration

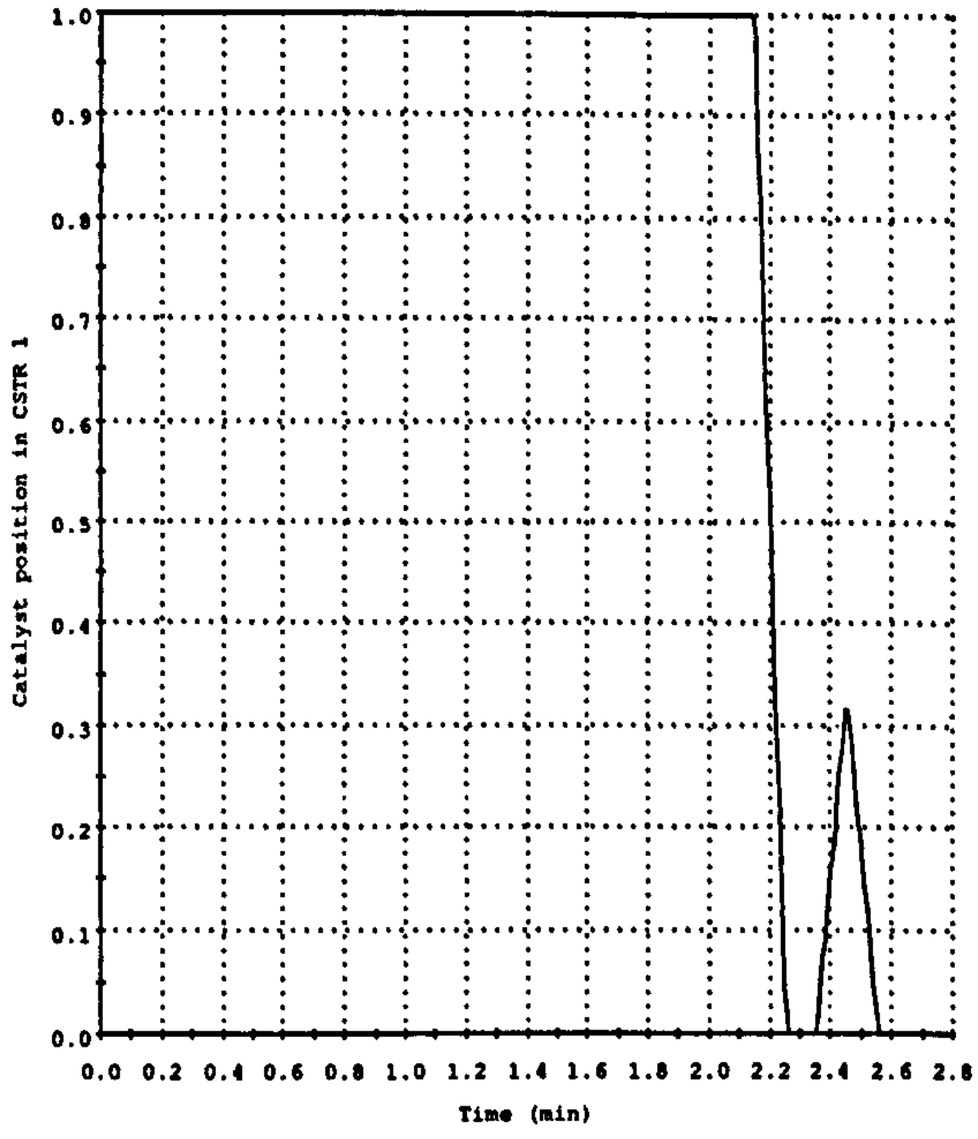


(c) CSTR 1 volume

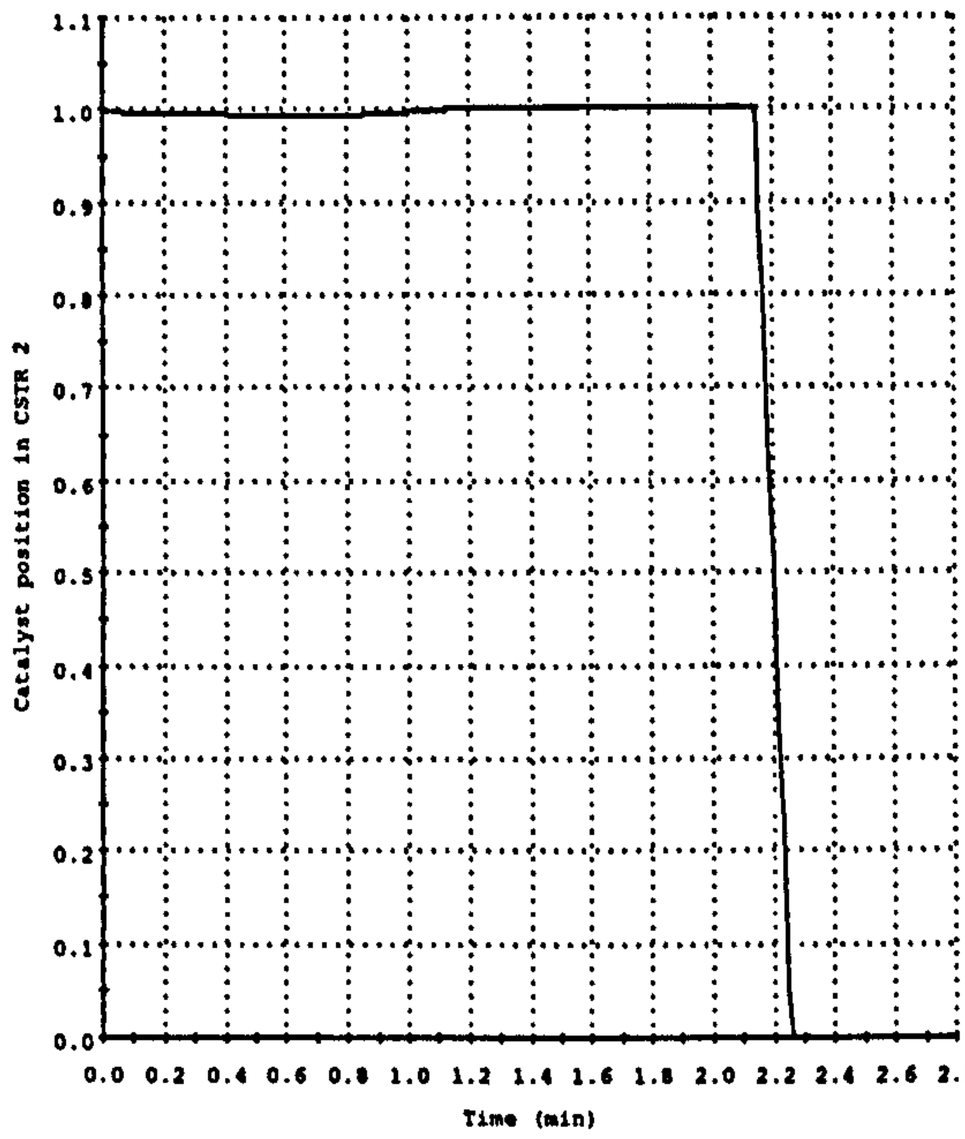


(d) Derivative of CSTR 1 volume

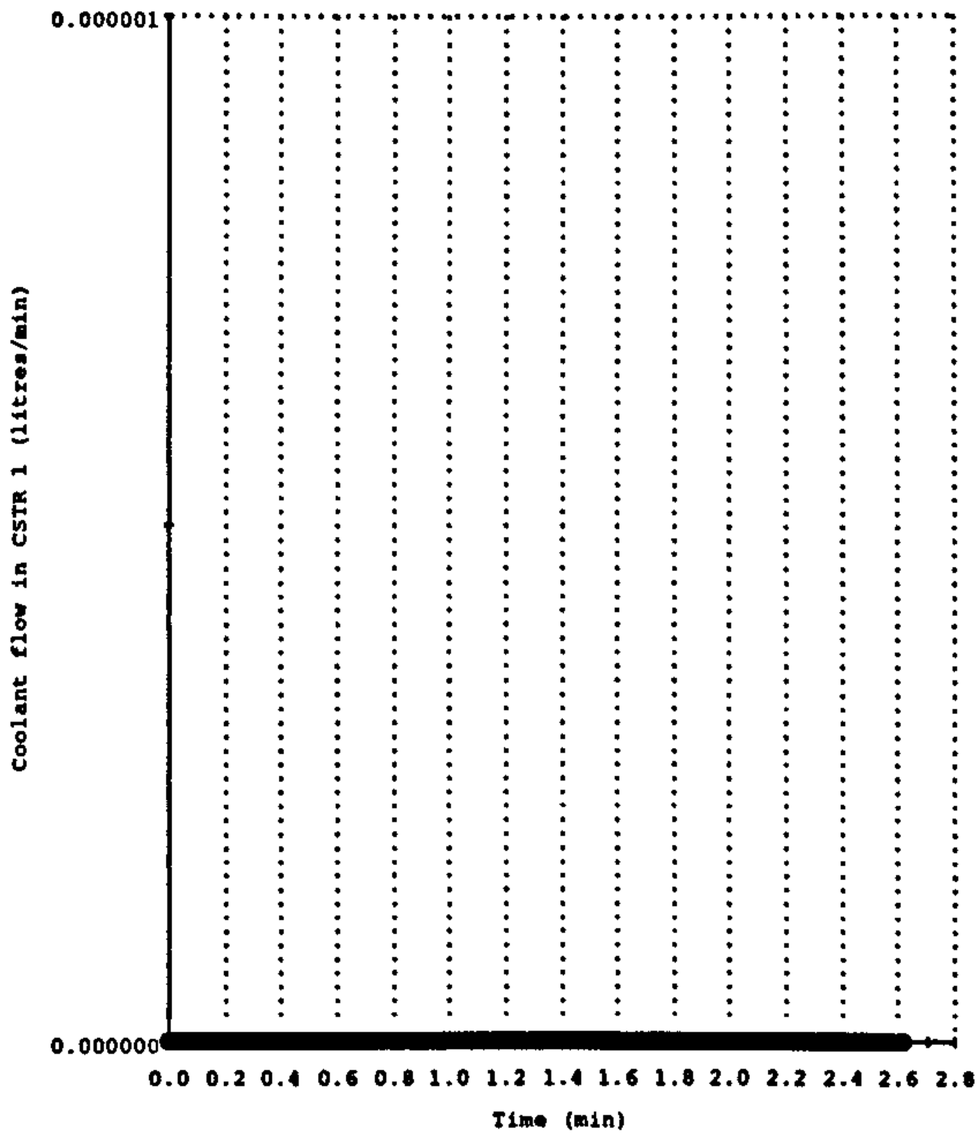
Figure 5.6: Typical state profiles from cyclic optimisation of candidate structure



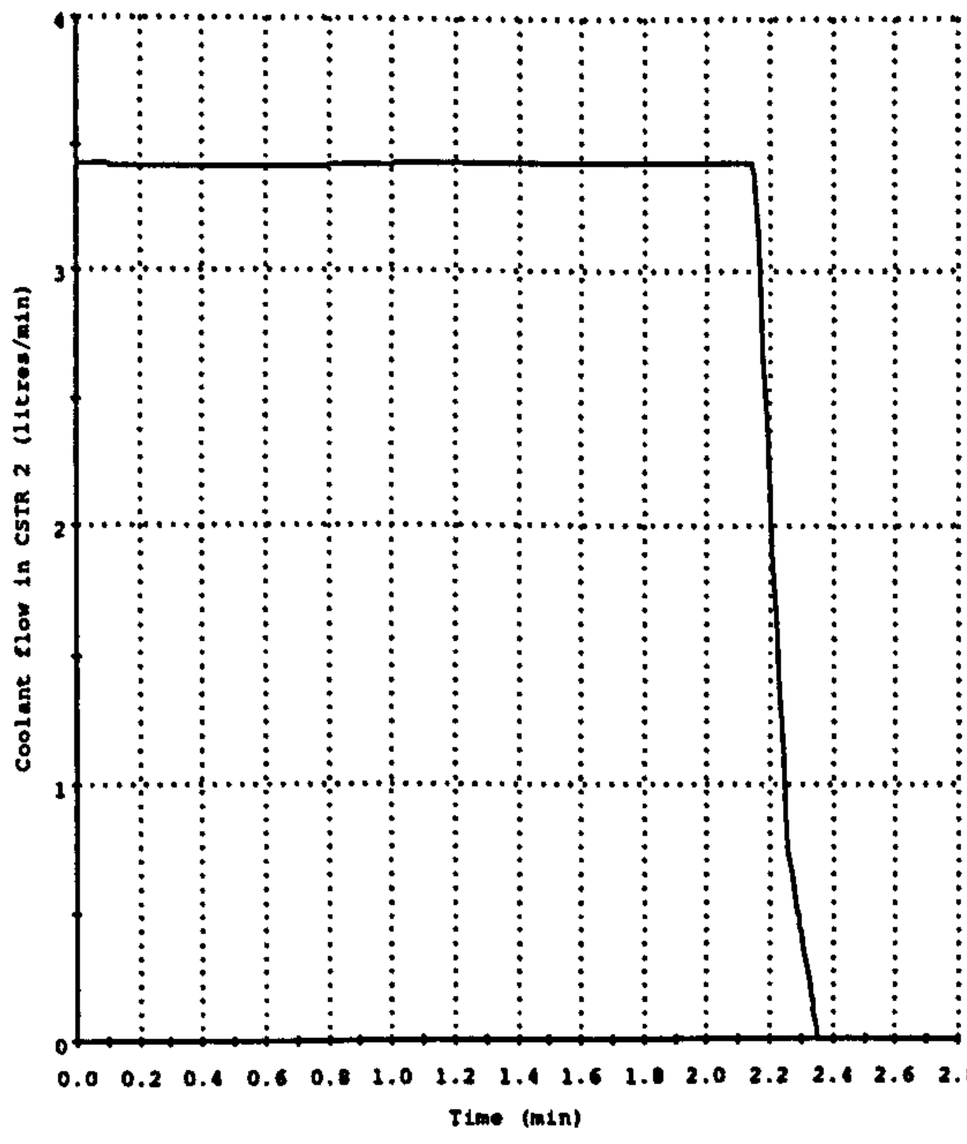
(a) CSTR 1 catalyst position



(b) CSTR 2 catalyst position

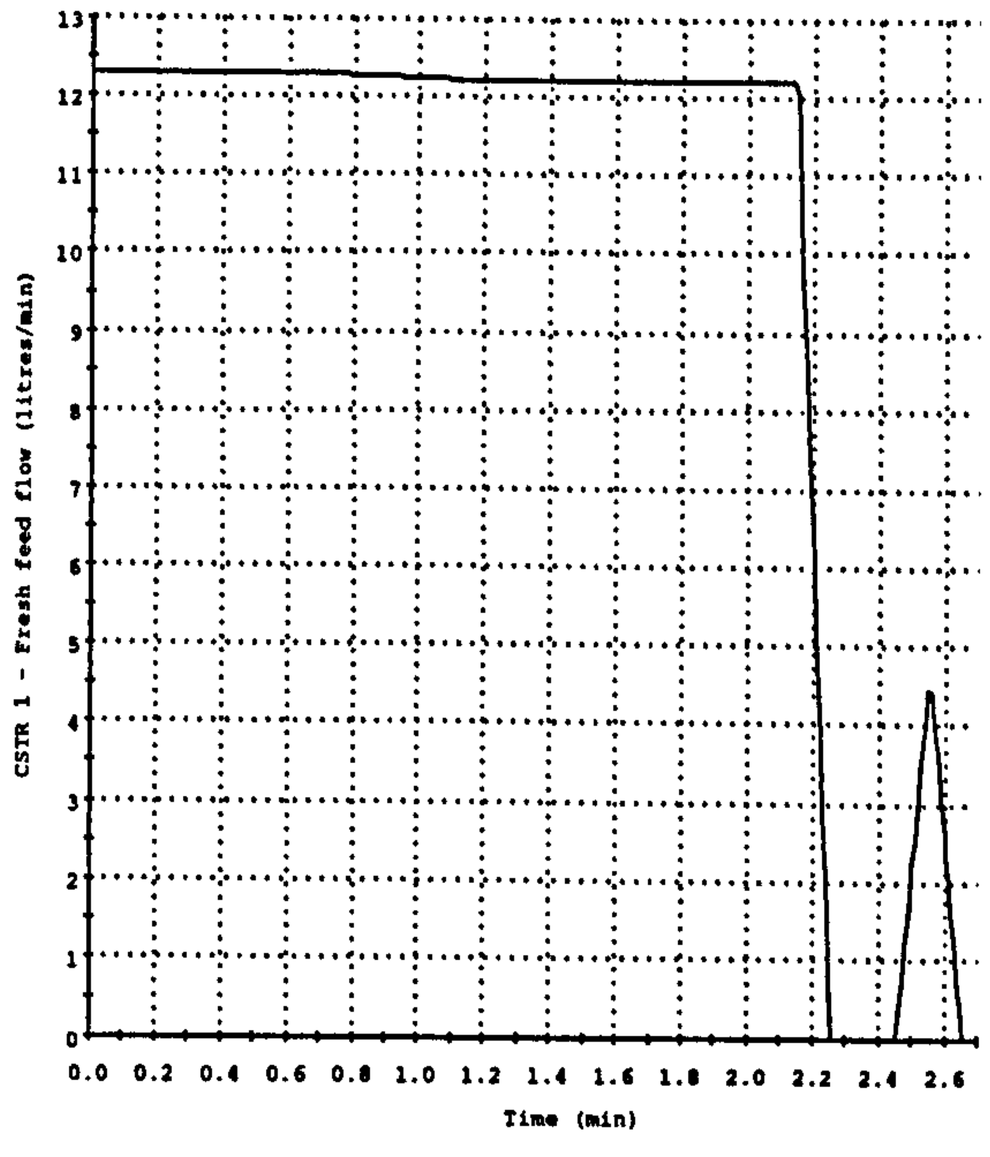


(c) CSTR 1 coolant supply

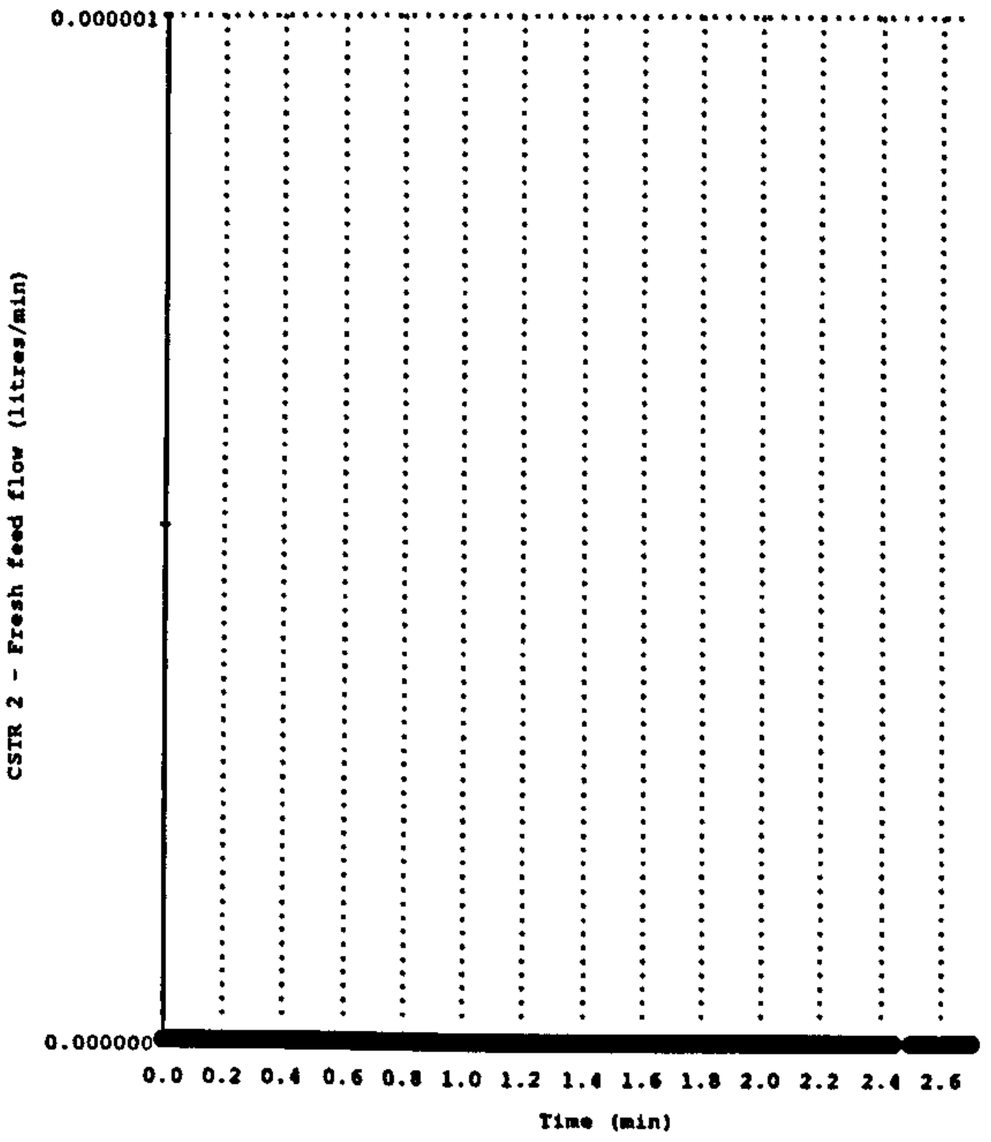


(d) CSTR 2 coolant supply

Figure 5.7: Control profiles from cyclic optimisation of candidate structure



(a) CSTR 1 reactant supply



(b) CSTR 2 reactant supply

Figure 5.8: Control profiles from cyclic optimisation of candidate structure

5.5. Closed-loop operation

Having designed the reactor network on an open-loop basis, the assumption that startup and shutdown would be feasible will now be tested by adding a decentralised control system made up of proportional integral (PI) controllers. The heuristics of Price and Georgakis (1993) are used to generate a control scheme for this system (refer to section 2.1.3.3 on control structure selection). Cohen-Coon open-loop tuning is used to provide initial guesses for the controller gains and reset times. Dynamic optimisation is then used to fix these parameters, minimising the integral squared error (ISE) produced by startup and shutdown. Although one could also use an economic objective, it is deemed unnecessary here since the operating costs are dominated by the steady state period and it is assumed that the vessel volumes will not change appreciably, leaving the capital costs unchanged. The pertinent profiles for this base case closed-loop system can be seen in figures 5.10 to 5.12; note the volumes do indeed remain relatively static.

Figure 5.11 shows that the catalyst activity in the second reactor reaches an upper bound of 1.2. It was found that if an upper bound of 1 was used for this control (as it was for the steady state optimisation) in the dynamic case, the optimal steady state, when simulating the shutdown and subsequent startup of the process, was impossible to reach. In reality an activity of 1 would indicate a catalyst immersion of around 83 %, the suitable steady state backoff. This highlights the need to establish the dynamic behaviour of the process around the solution of a steady state optimisation. Clearly, the original problem needs to be modified to enable a proper study of the shutdown and startup characteristics of the process.

5.5.1. Control structure selection

The demand profile is set by the customer, so valve (E) cannot be used as a degree of freedom. Hence the levels in the system must be controlled in the opposite sense to that of the direction of flow. Hence the level in the storage vessel is controlled by preceding valve (D), the level in the second reactor by valve (B) and the first reactor, valve (A). Note that valve (C) could have been used to control the level in the second reactor, but this would not result in a self consistent level control structure.

For the remaining loops it seems theoretically possible to use (a) the cooling water flowrate of the first reactor to control the temperature in the second reactor, and (b) the catalyst immersion in the first reactor to control the storage vessel composition, but this is unreasonable from a proximity point of view as there are "closer" controls. Hence the cooling water flowrates of both reactors are used to control their respective temperatures, and the catalyst immersion in the second reactor is used to control the product composition in the storage vessel.

One degree of freedom is left unused, the catalyst immersion in the first reactor. This is made to follow its open-loop behaviour, i.e. fully immersed during startup and steady state production, and fully withdrawn upon shutdown. The chosen control structure is given in figure 5.9.

It is assumed at this point that the customer **only** has control over valve (E). It *could* be envisaged that the customer could also have complete control over all the other degrees of freedom in the system, setting all the controls simultaneously to zero, as in the open-loop case, but this would effectively eliminate any resulting dynamic behaviour.

5.5.2. Operating Envelopes

The plant design and control structure are now fixed in order that uncertainty in the cooling water temperature and shutdown state may be taken into account.

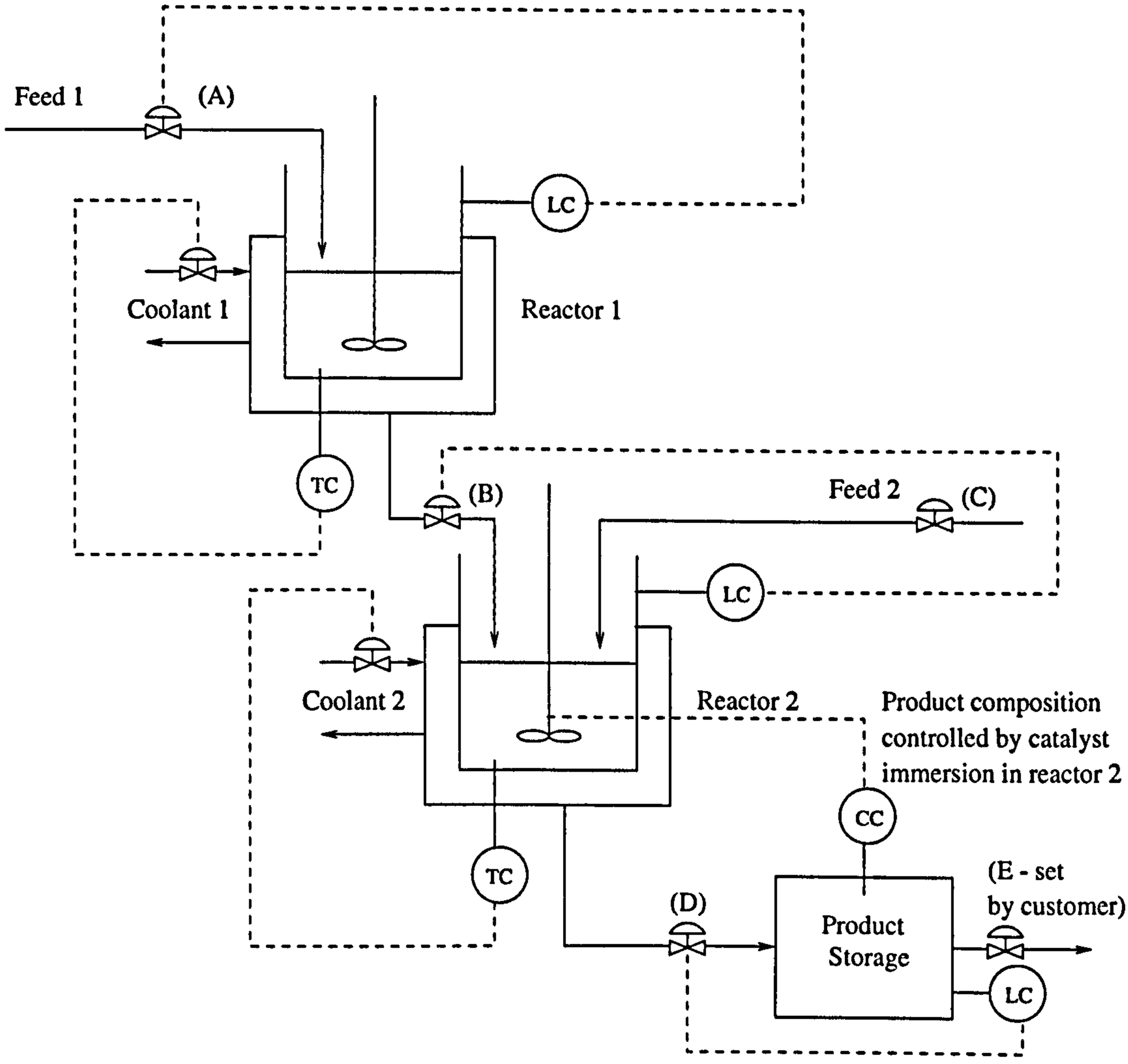
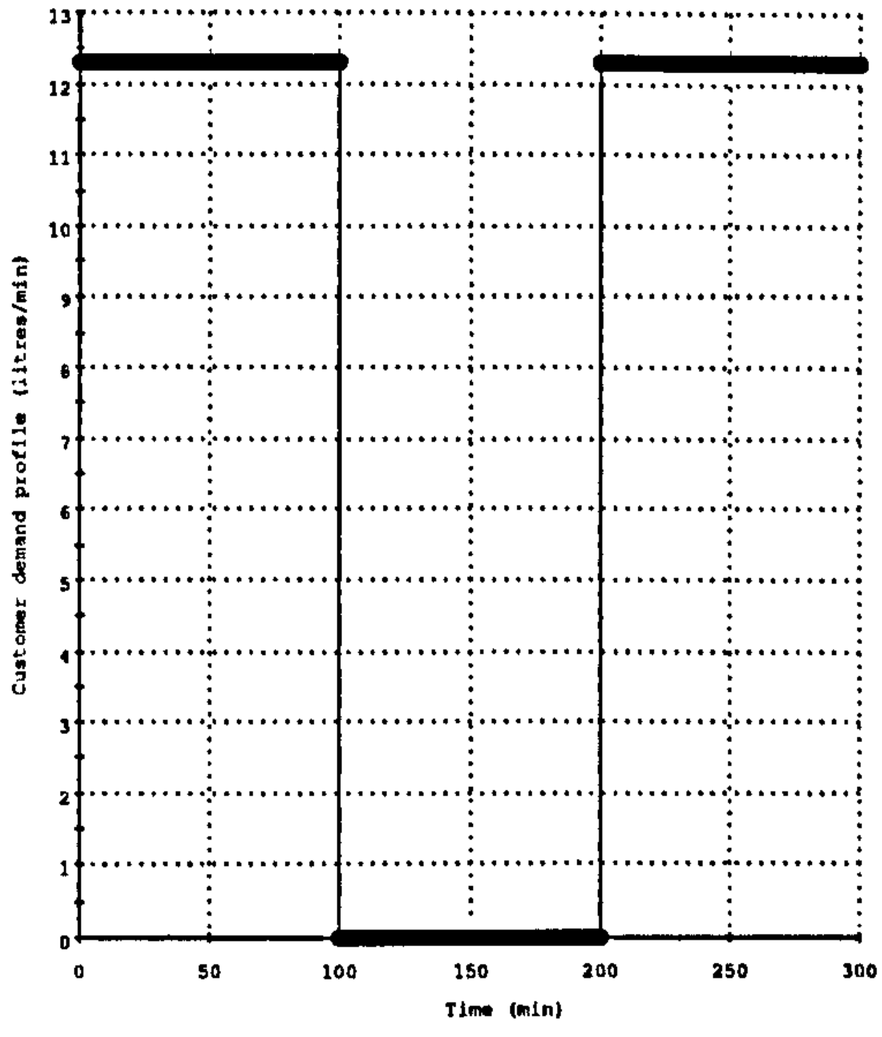


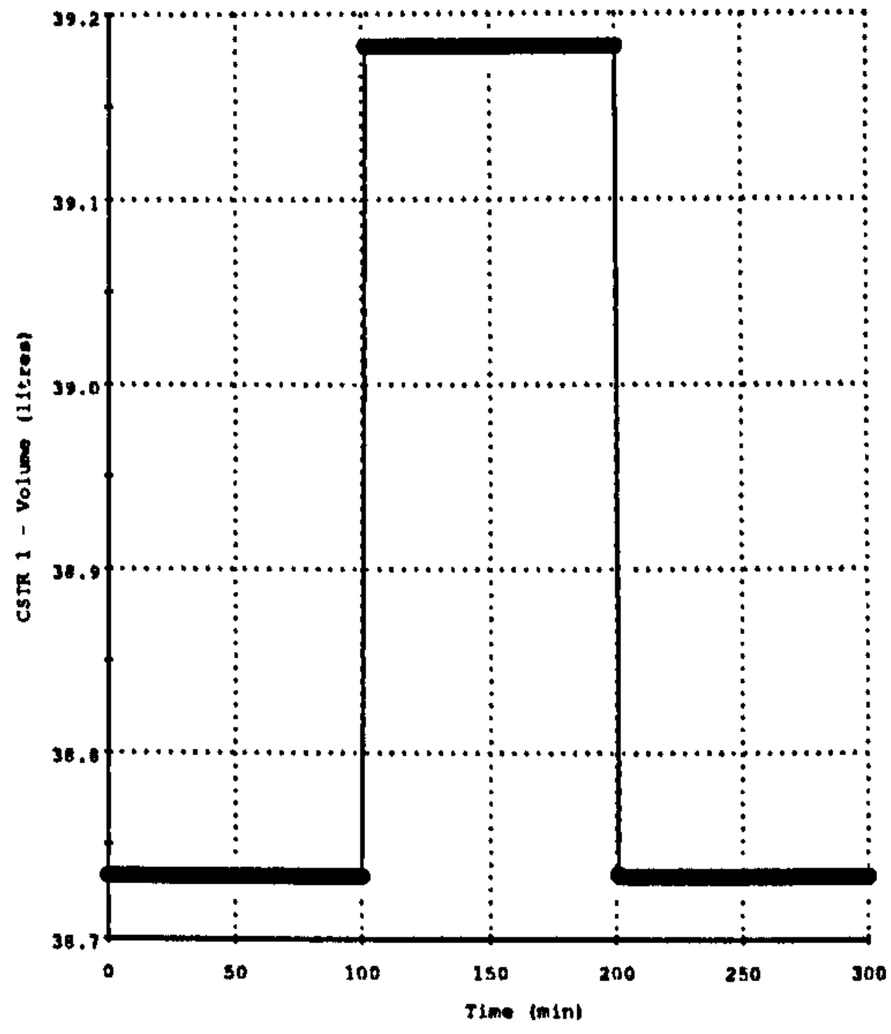
Figure 5.9: Control structure for the responsive reactor network

This is done by considering the cooling water temperature and the initial conditions of various critical system states as ranges to be optimised rather than fixed points. The Operational Envelope method provides the mechanism for doing this.

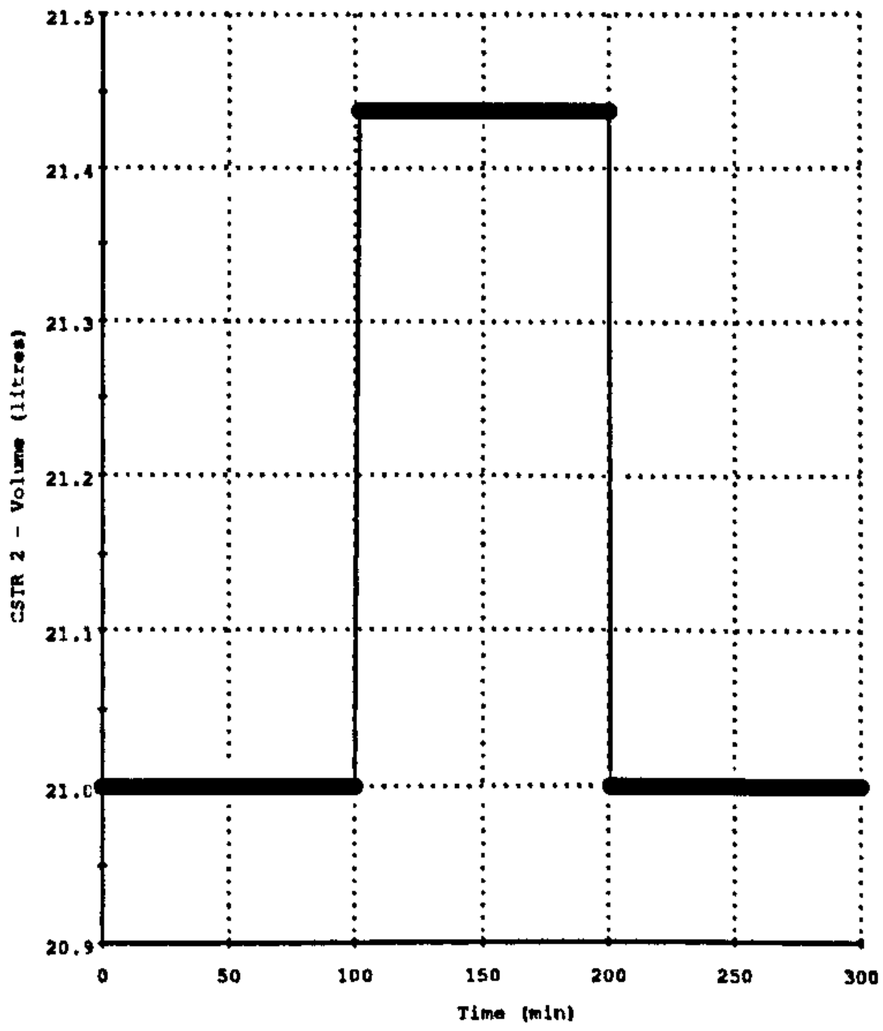
The initial concentrations in each of the vessels, the initial temperatures in the two reactors and the cooling water temperature constitute the dimensions of the Operational Envelope. Considering the holdups in each vessel was deemed unnecessary, since they remain relatively constant during the shutdown and startup periods. Thus the Envelope will form a 6-dimensional hypercube, with 64 vertices



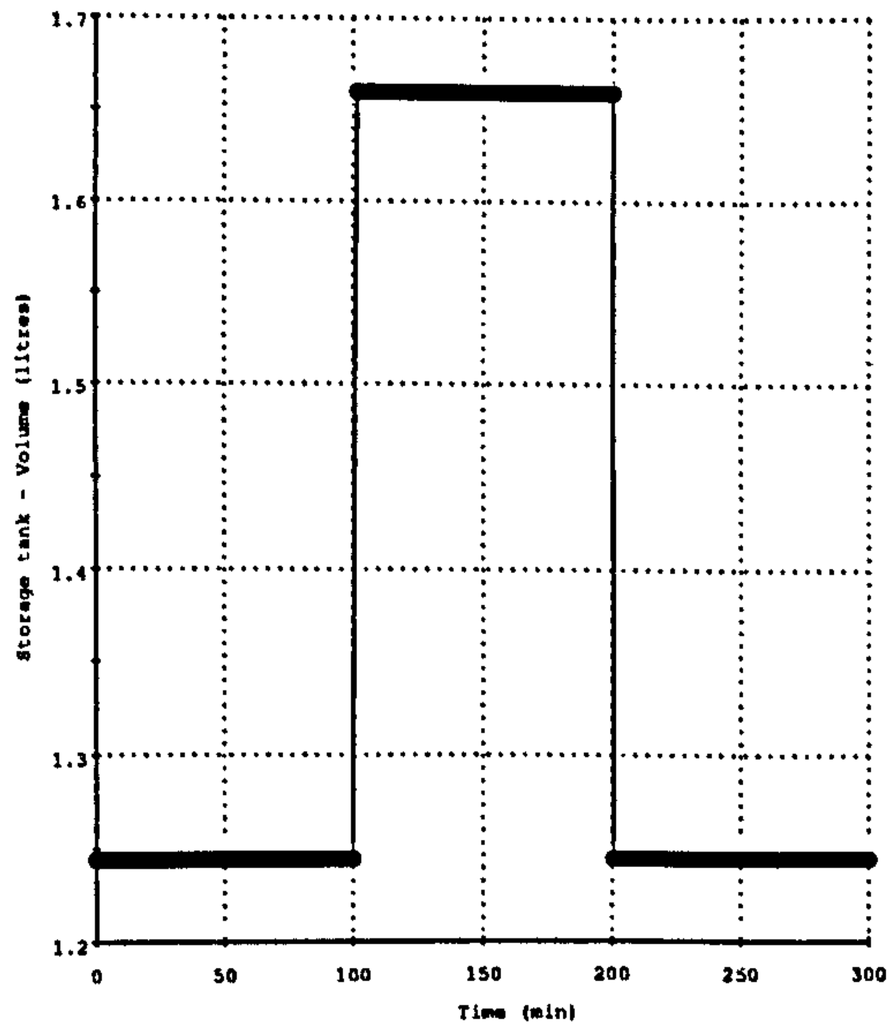
(a) Customer demand profile



(b) CSTR 1 Volume

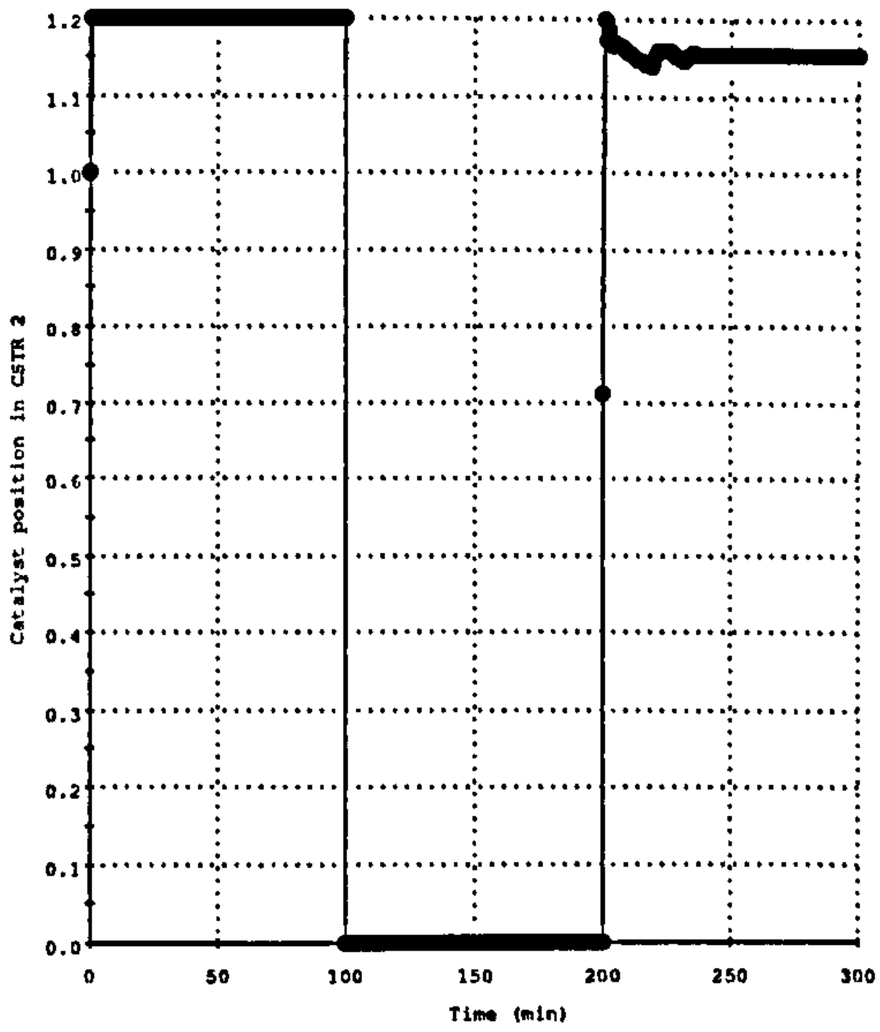


(c) CSTR 2 Volume

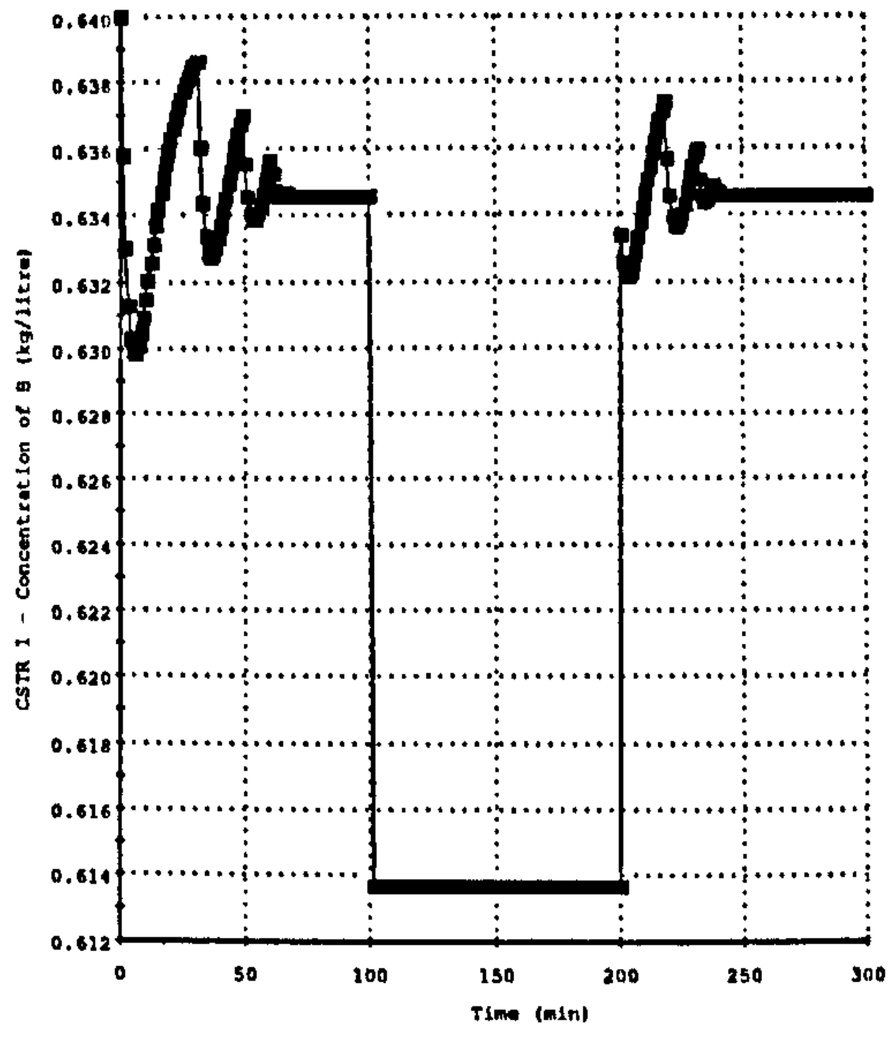


(d) Storage tank volume

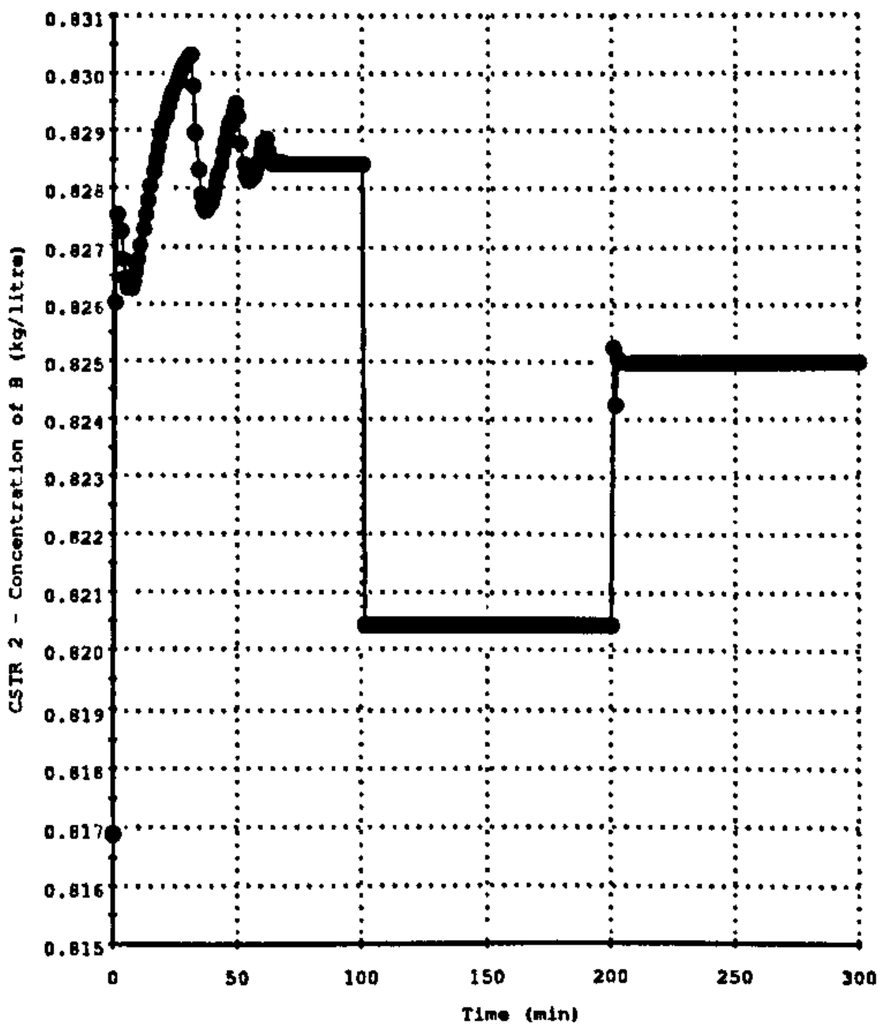
Figure 5.10: Closed-loop base case: customer demand profile and vessel volumes



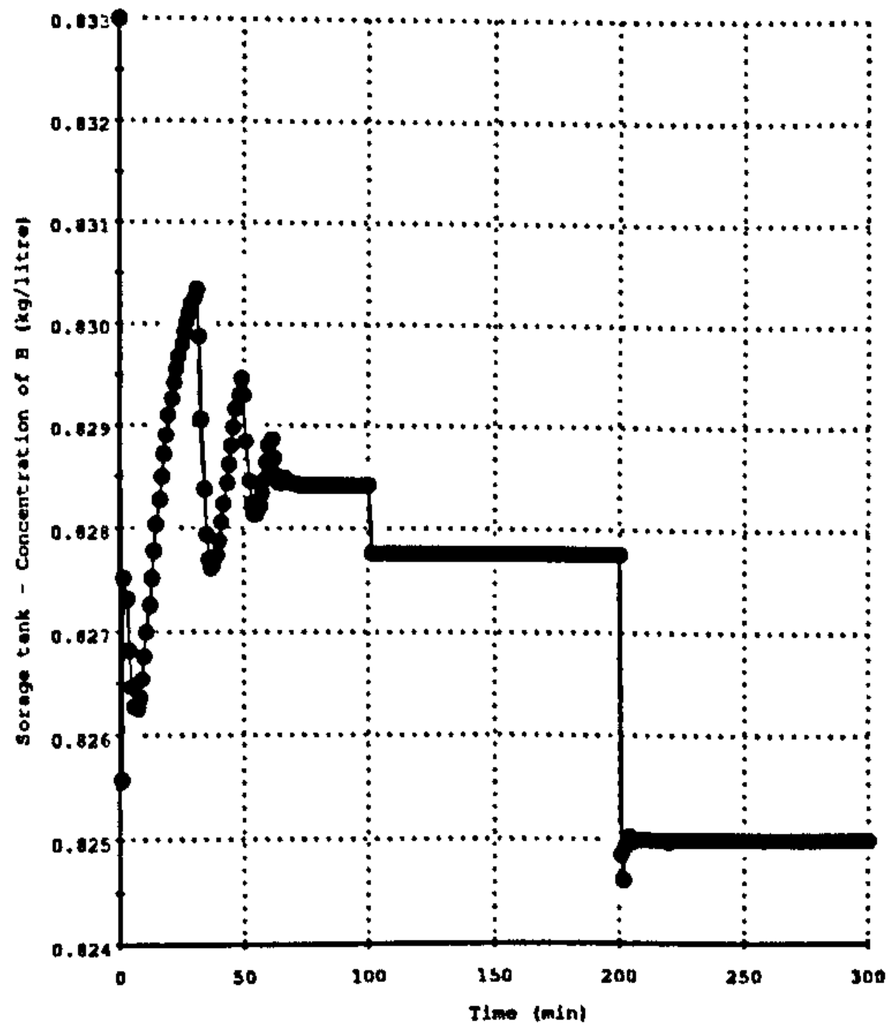
(a) CSTR 2 catalyst activity



(b) CSTR 1 product concentration

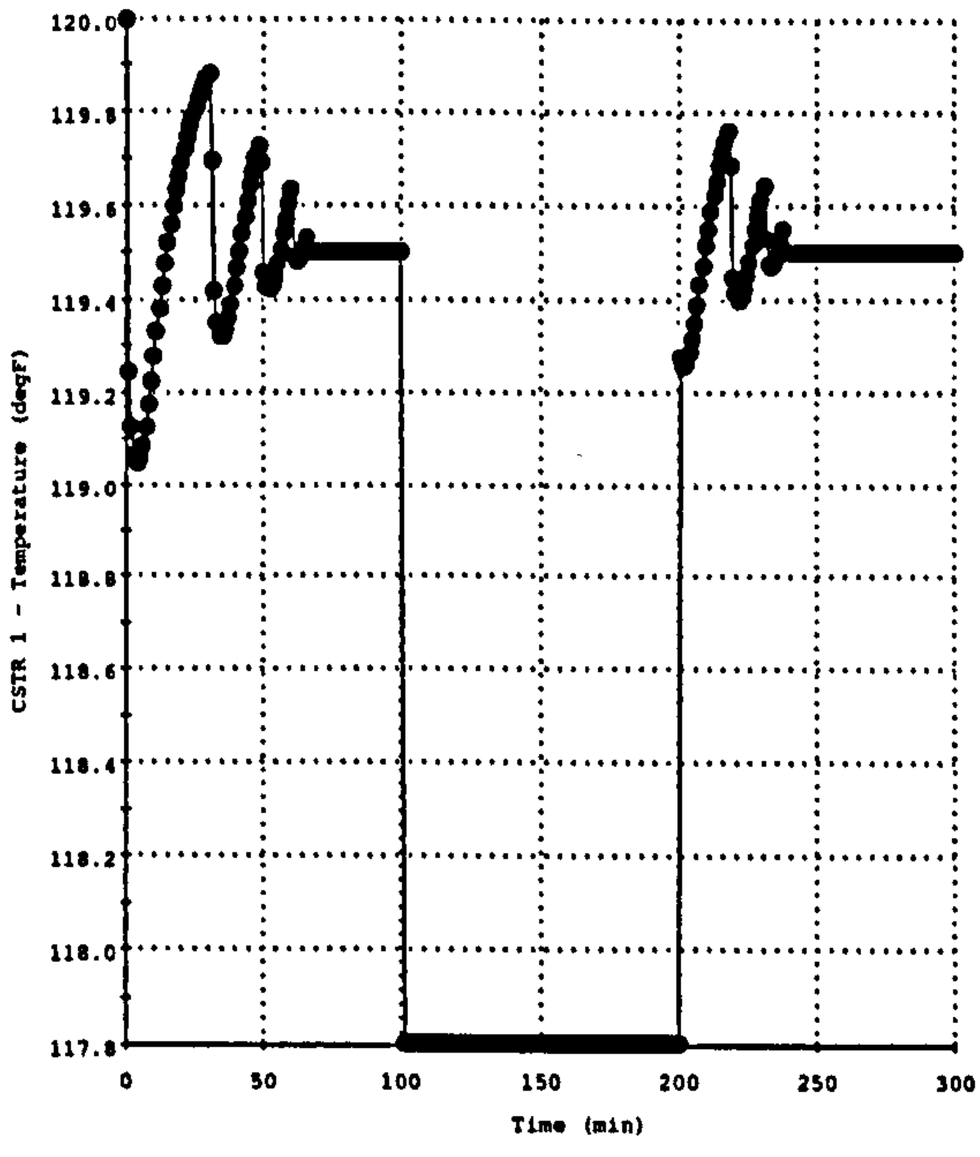


(c) CSTR 2 product concentration

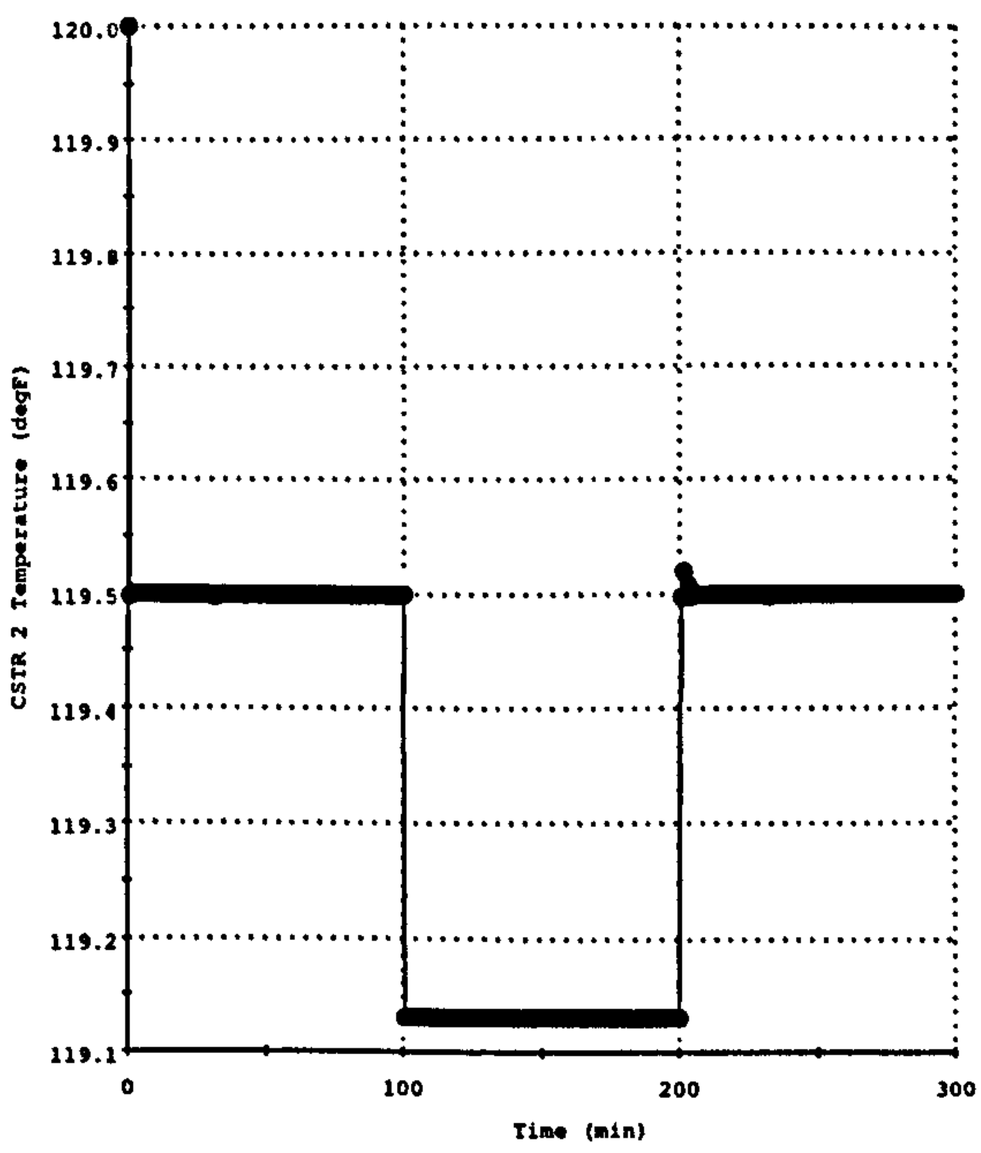


(d) Storage tank product concentration

Figure 5.11: Closed-loop base case: CSTR 2 catalyst activity and vessel product concentrations



(a) CSTR 1 temperature (^0F)



(b) CSTR 2 temperature (^0F)

Figure 5.12: Closed-loop base case: CSTR temperatures

to examine in the solution procedure.

The overall problem took 3 major iterations to converge. The pair of vertices for the first iteration was selected arbitrarily (the vertex with all bounded variables at their lower bounds, along with its opposite vertex). Only the main constraints of reactor temperature and stored product purity were considered. The number of violated vertices was determined through the simulation step. The pair of vertices for the second iteration was selected simply by considering the worst violation pairing in CSTR 1 temperature and storage product purity (CSTR 2 temperature was not violated at this iteration): these are scenarios 48 and 17, as shown in figures 5.13 (a) and (b). For the third and final iteration, another scenario was chosen (44) from figures 5.13 (c) and (d): violations in CSTR 2 temperature and storage product purity. After this third scenario was included in the optimisation, simulation of all scenarios showed that none of the vertices are violated: hence the construction of the Envelope is complete.

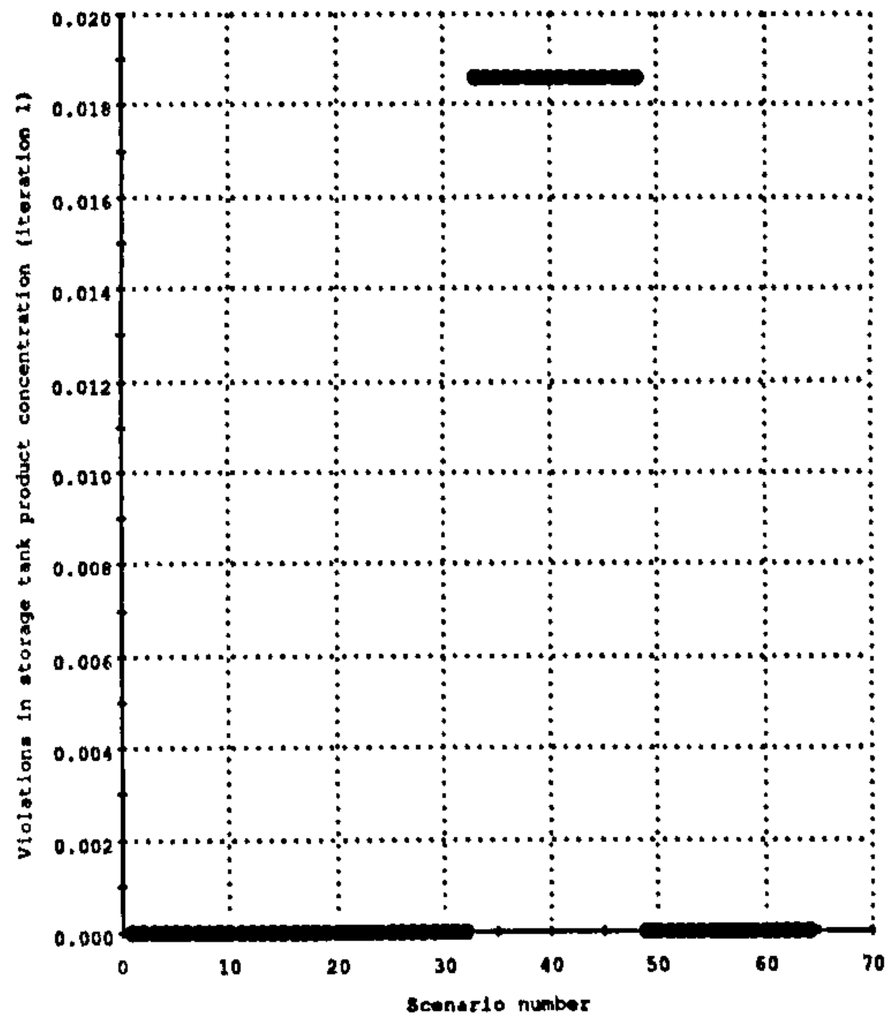
Three critical, constrained system states are shown in figure 5.14. Envelopes of the initial values of the reactor temperatures and the product purity in the storage vessel are denoted by the intervals between starting points of the solid and broken lines in each profile. Observe how the states, even though they initialise at the extremes of their Envelopes (check corresponding values with table 5.7), are made to converge to their steady state production values by the control system without violating the path constraints.

The complete Envelope can be seen in table 5.7 and in the "spider" diagrams in figure 5.15. Each dimension of the hypercube is denoted by an arm in the web-like plots. In the first diagram the solid lines joining the arms represent the actual Operating Envelope — the outer hexagon joins the upper bounds of the Envelope and the inner hexagon the lower bounds. The broken line shows where the base case optimisation results lie relative to the Envelope — the base case CSTR 1 initial B concentration lies outside the Envelope as uncertainty in the cooling water temperature was not considered when the base case was determined.

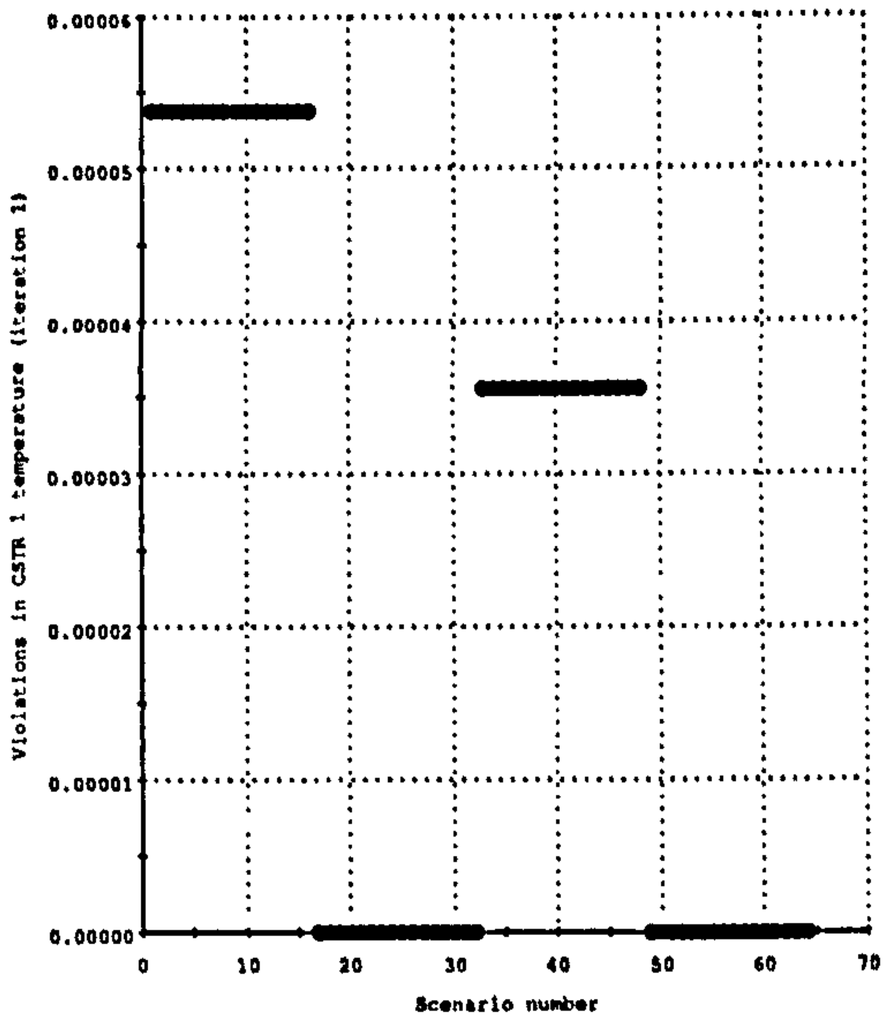
The second diagram shows the size of the Envelope in relation to the lower and upper bounds on its size. Here each arm corresponds to a scaled Δ_i ($\frac{\Delta_i - \Delta_i^{min}}{\Delta_i^{max} - \Delta_i^{min}}$) — where a value of zero indicates the Envelope dimension on its lower bound, and and a value of one indicates that it is on the upper bound. The hexagon shows the relative size of the Envelope as determined by the optmisation.

Envelope dimension	*Lower limit*	*Upper limit*	*Base case*
CSTR 1 initial B conc ($kg/litre$)	0.6135	0.6209	0.6400
CSTR 1 initial temperature (^0F)	118.41	120.00	120.00
CSTR 2 initial B conc ($kg/litre$)	0.8170	0.8247	0.8169
CSTR 2 initial temperature (^0F)	118.44	120.00	120.00
Storage tank initial B conc ($kg/litre$)	0.8172	0.8248	0.8169
Cooling water temperature (^0F)	67.965	69.605	68.000

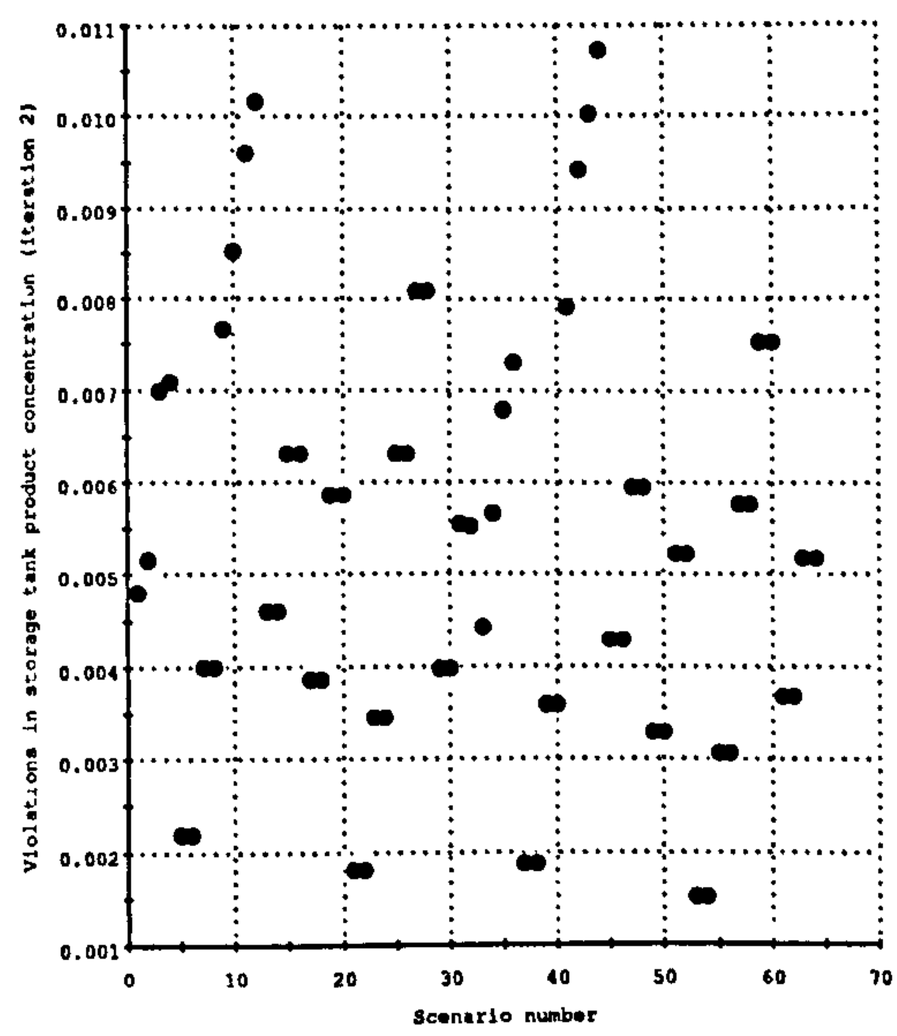
Table 5.7: Operating Envelope



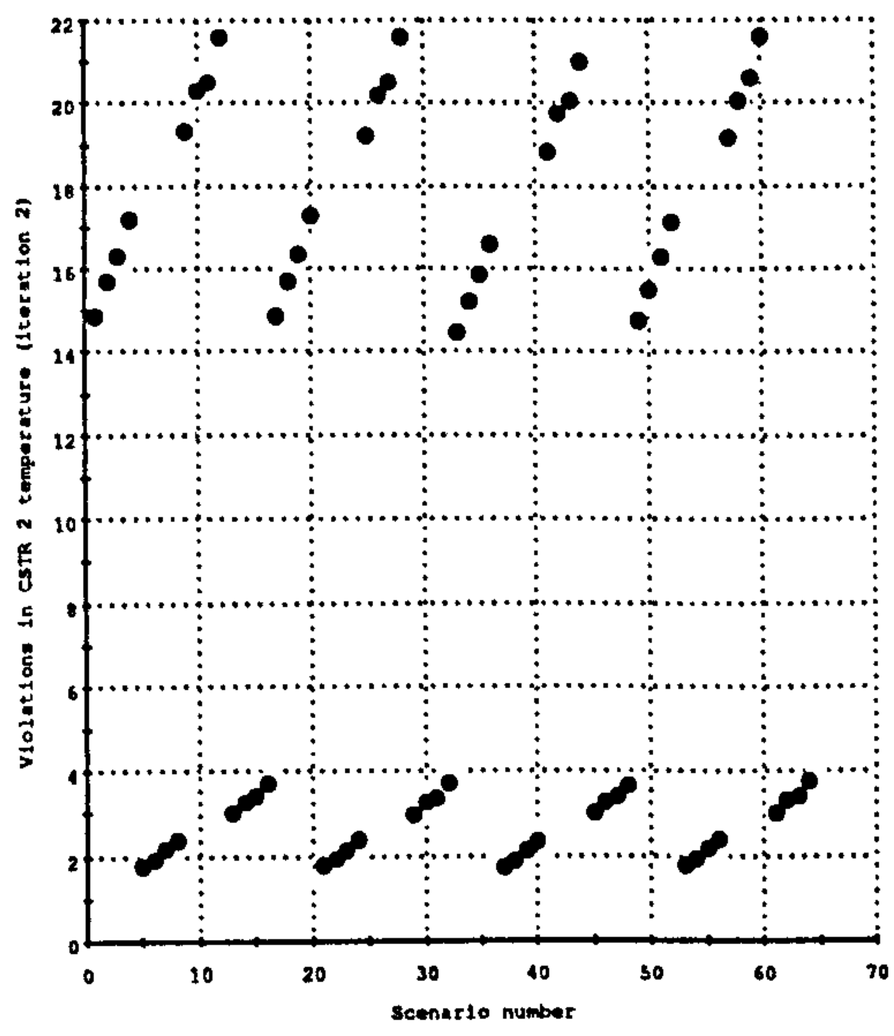
(a) Purity violations in storage tank (iteration 1)



(b) Temperature violations in CSTR 1 (iteration 1)

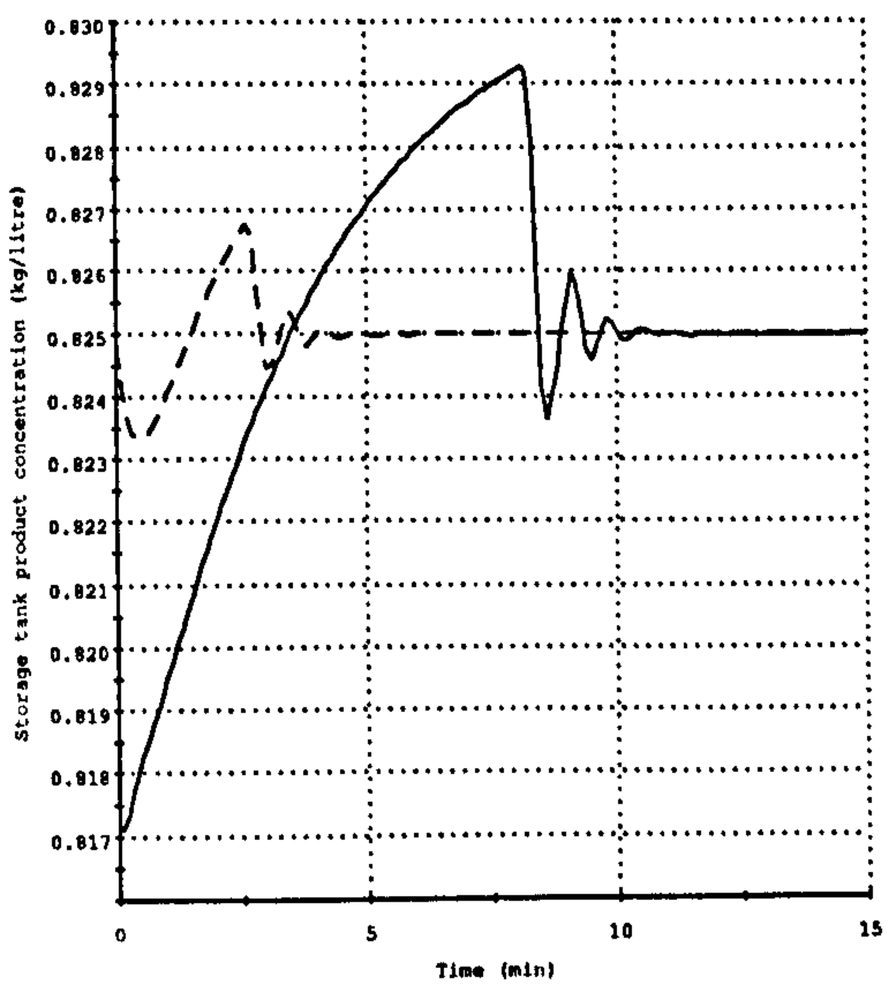


(c) Purity violations in storage tank (iteration 2)

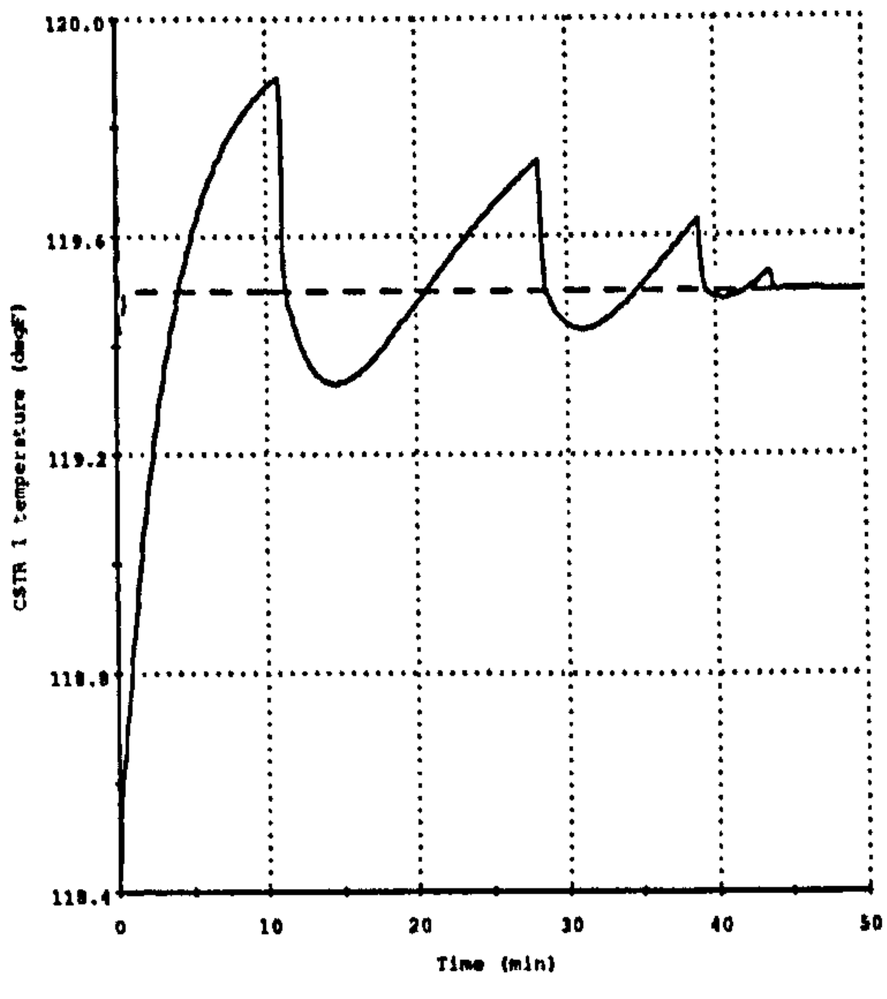


(d) Temperature violations in CSTR 2 (iteration 2)

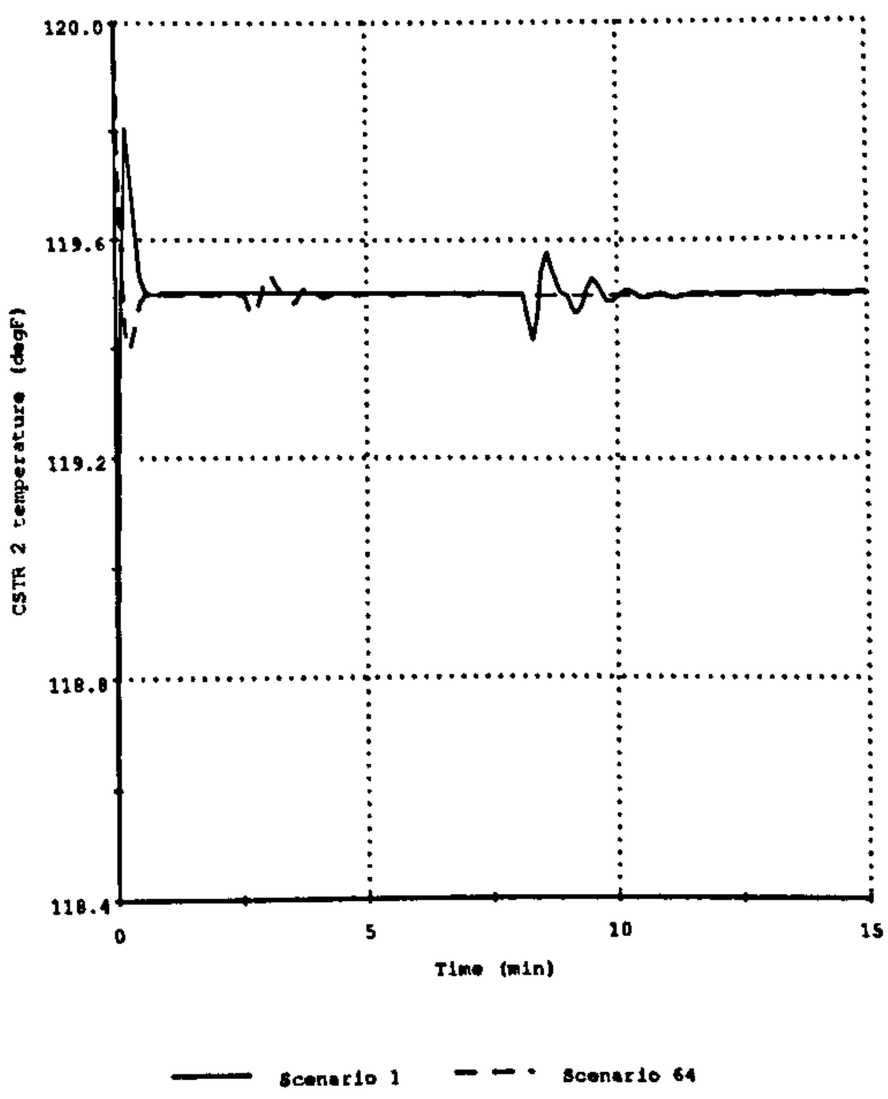
Figure 5.13: Constraint violations over 2 iterations



(a) Storage tank purity Envelope

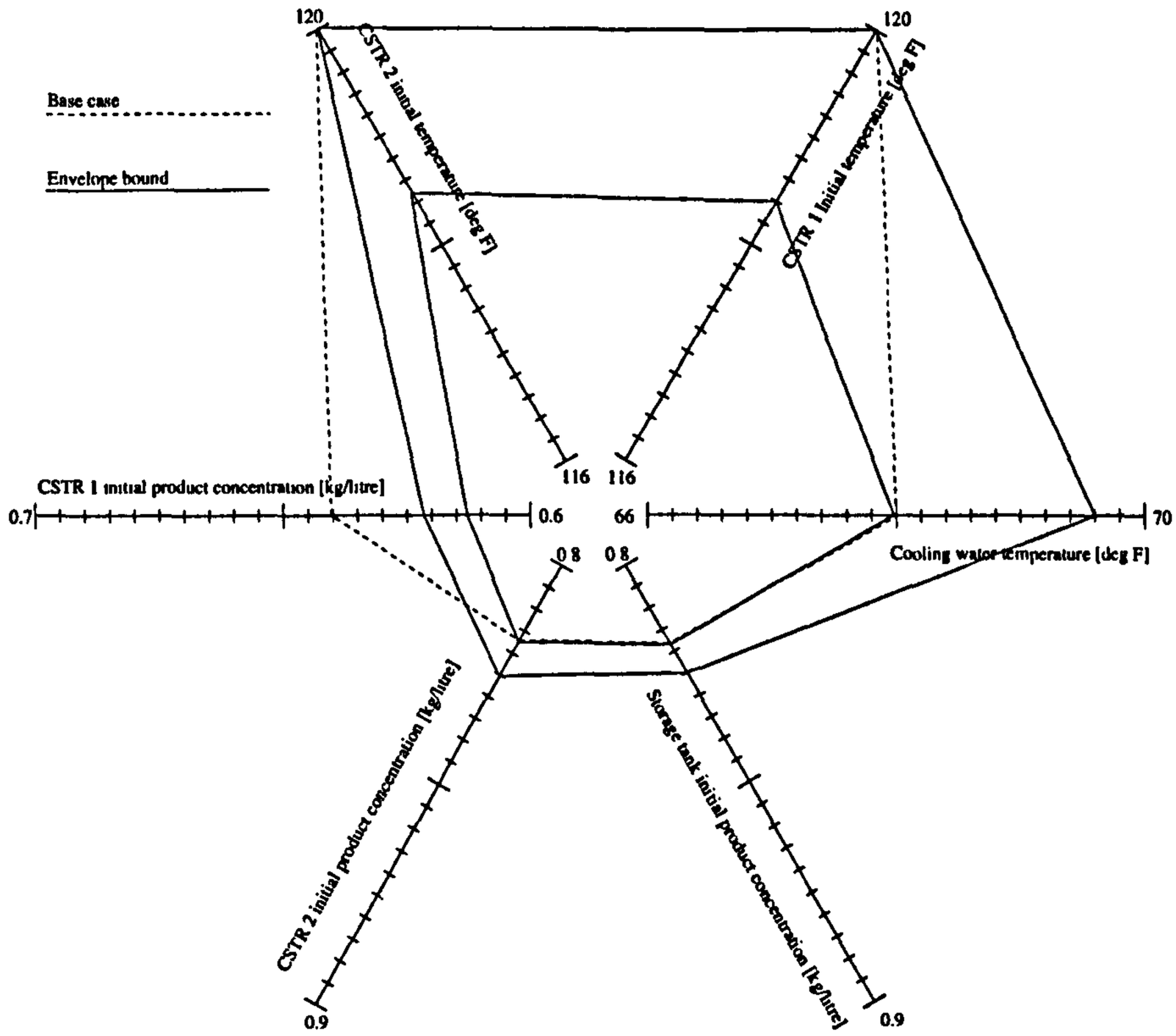


(b) CSTR 1 temperature Envelope

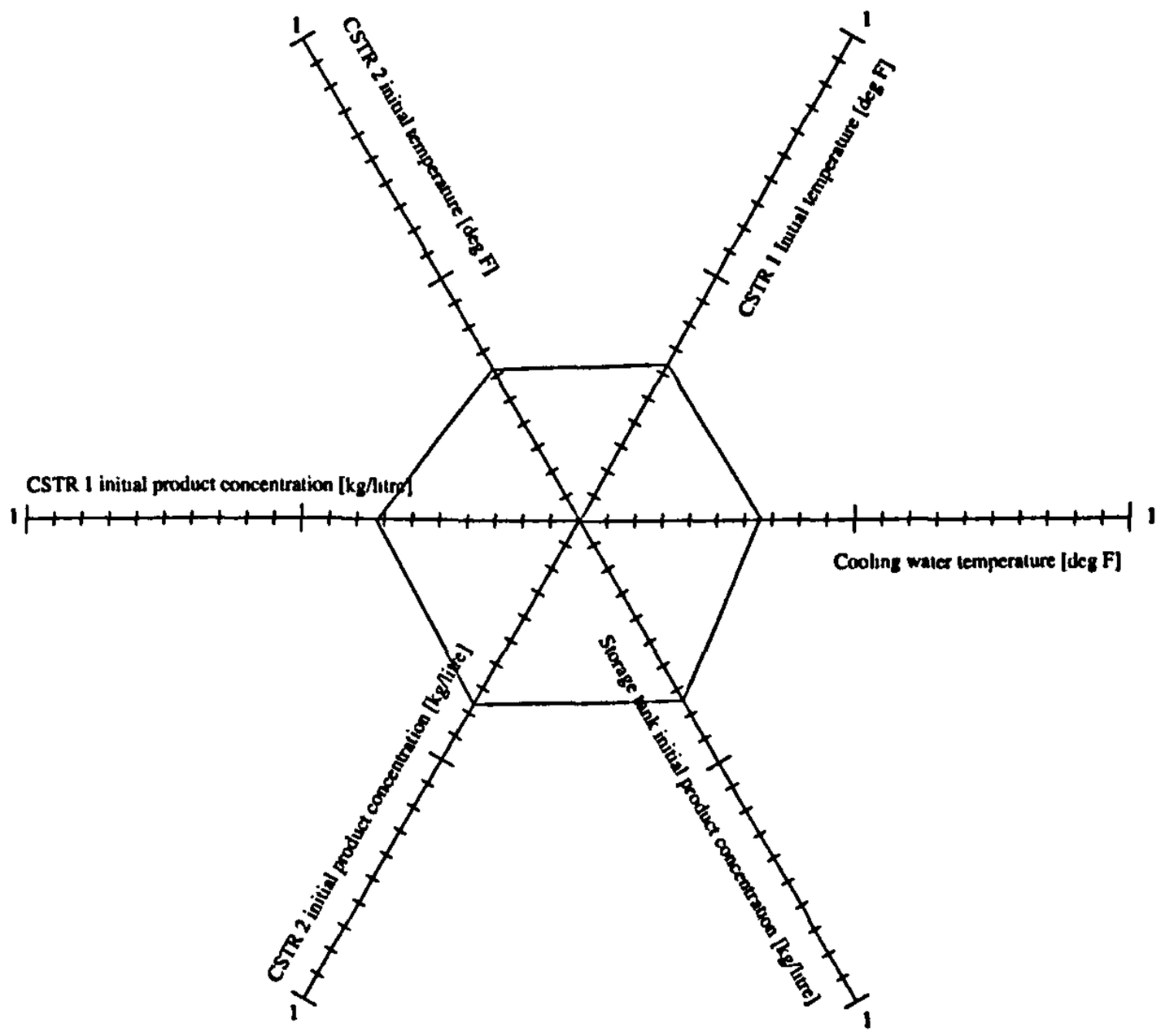


(c) CSTR 2 temperature Envelope

Figure 5.14: Operating Envelopes



(a) Actual Operating Envelope



(b) Scaled Δ_i

Figure 5.15: "Spider" diagrams for the Operating Envelope

5.6. Conclusions and summary

The work in this chapter has considered the design and optimal cyclic operation of a reactor network. The problem definition presented was based upon the customer requiring the production of B on-demand, *in situ*. A feasible solution to the problem was found using a two stage design procedure: development of a candidate structure followed by optimisation of the operating cycle and design of this structure. A steady state bounding heuristic was applied to screen design alternatives efficiently for cyclic operation through a steady state production phase:

> *The optimal annualised cost of the steady state process will be less than or equal to the cost of the dynamic process.*

The solution exploited system behaviour which essentially enabled the process to be moved instantly to the optimal steady state operating point.

At this point, a control system was added and feasible startup/shutdown investigated. After a base case was established, Operating Envelopes were constructed about this nominal point in order to determine feasible startup from a range of shutdown states, given an uncertainty in the cooling water supply temperature. In this way, the problematic cyclic constraints were eliminated from the formulation and replaced by something significantly more robust and more likely to produce infinitely repeatable operational cycles.

5.6.1. Another possible scenario

Presently, it is assumed that the reaction is automatically quenched when the catalyst is disengaged: i.e. when $Catalyst_{Control} = 0$. If disengagement does not occur cleanly, some catalyst may remain in the reaction mixture to cause some degree of background reaction. In this case a larger storage volume of on-spec product would be an advantage, as this would increase the buffering effect on the

over-spec flow coming in from the reactors. Indeed Envelopes could be used to quantify the maximum shutdown times possible before feasible startup becomes impossible. One way of doing this would be to add the background catalyst immersion at shutdown and storage volume to the hypercube dimensions. In the event of infeasible startup (the limit of which would be that both reactors would end up containing pure product) a blending policy could precede startup, where fresh reactant is fed to both CSTRs in order to dilute them; the product storage tank would only be replenished when the flow emerging from the second CSTR was on-spec. One implication of this scenario is that the shutdown state would no longer have a zero associated cost and any economic objective function would have to be adjusted appropriately.

Chapter 6

Conclusions and future work

This thesis considered an integrated approach to designing responsive chemical manufacturing facilities in the presence of uncertainty.

6.1. Summary

Chapter 1 provided the background and motivation for this work, highlighting the importance of competitive and sustainable production **on demand** in the face of variable feed stocks and product demand (both in quality and quantity).

Chapter 2 critically reviewed previous work in the traditionally segregated fields of synthesis and operability. Major contributions which attempt to consider these fields *simultaneously* were identified and discussed. Of the "integrated" approaches found in the literature, the one based explicitly upon dynamic operation was deemed most appropriate for this work, as it was the only way to ensure that process constraints have not been violated. A recent trend of combining heuristics and mathematical programming techniques was also noted and commented upon.

Chapter 3 described two possible variants of a design procedure which could potentially be used to tackle the integrated design of responsive plants. The first, a single pass MIDO approach, was regarded as impractical due to the difficulty in construction a superstructure containing all control alternatives, compounded by its overly intensive computational nature. Instead, a two stage decomposed design methodology of (a) generating feasible candidate structures through heuristics, bounding properties and screening tools followed by (b) the application of dynamic optimisation techniques in order to determine the candidates' optimal design and operation, was proposed as a more efficient method of generating a tractable design problem. Some consideration was also given to the concept of responsiveness and its contribution to design procedures. An Operating Envelope technique specifying operation in terms of a region within which the operating variables should be maintained to guarantee feasible process operation was chosen over a worst-case design methodology which posed min/max problems to handle uncertainty.

A multiproduct reactor plant example was presented as in chapter 4. Initially, a brief review of previous work in this area was given followed by a published design algorithm (Cantrell and Luyben, 1996). An optimisation based approach was then proposed, minimising the total annualised cost and finding the optimal process design, controller settings and cyclic scheduling of the products. The solution and methodology were compared favourably to Cantrell and Luyben, the key being that due to the purity path constraints being enforced less conservatively, economically superior results were obtained. After discovering that the purity path constraints were rapidly violated within 2 cycles, optimisation *w*ithout cyclic point constraints over multiple cycles was carried out, resulting a more robust set of design variables that produced no purity path constraint violations in a variety of multiple cycle simulations. Having highlighted the need to remove cyclic point constraints from the formulation, cyclic Envelopes were constructed for in a sec-

ond example, a multiproduct reactor/separator. This was deemed far more likely in producing infinitely repeatable operational cycles.

Finally, chapter 5 introduced a plant capable of instantaneously supplying an intermittent product demand. The entire decomposed design procedure proposed in chapter 3 was effectively applied in (a) selecting the optimal candidate process *and* control structure through heuristics, and (b) determining the optimal cyclic plant operation through dynamic optimisation. The final design was capable of near-instant switching from the shutdown state to the steady state optimum operating point. Feasible startup from a range of shutdown states was also investigated using Operational Envelopes, given an uncertainty in the cooling water supply temperature.

6.2. Conclusions

An integrated design approach to responsive chemical manufacturing facilities has been developed that considers operability together with economics, permitting design and synthesis decisions to be made simultaneously within a common framework. Finally, uncertainty in certain model parameters was considered to ensure operational feasibility.

6.2.1. Design algorithm

1. After generating a set of feasible process *and* control structures that fulfil the instant response criterion, use heuristics, bounding properties and screening tools to narrow down the alternatives, ideally to a single candidate.

2. Use dynamic optimisation to determine and validate the optimal design and operation of the candidate(s).

3. Use Operating Envelopes to ensure feasible operation of the candidate in the presence of uncertainty. In particular investigation of feasible grade changes and startup from a variety of shutdown states; this eliminates the need for cyclic point constraints.

6.3. Future work

6.3.1. New techniques

- The description of model parameter uncertainty/variability used in this work has been entirely deterministic. Stochastic descriptions of uncertainty could be used, employing techniques mentioned in section 2.1.2.1 on flexibility.

- Only simple de-centralised proportional integral (PI) control structures have been considered in this work. Advanced multivariable model predictive control (MPC) could be used to provide a centralised "overbrain" for this PI array, enabling the incorporation of changing external conditions such as reactant price variation, customer demand profile changes, etc.

- More complex unit operations, with their own distinctive dynamic characteristics, could be used in example case studies, e.g. membrane technology and reactive distillation. More complex kinetics and reaction mechanisms combined with possible by-product formation could also be considered. This would introduce different transient behaviour which could potentially be exploited in novel ways.

- An index of responsiveness should be investigated more thoroughly, ideally with a link to economics. A basis for this could be the observation that minimising inventory (typically used to buffer dynamics in the examples in this work) increases the responsiveness of a *system* as *less* material has to

"change state". Some parallels may be drawn with the controllability index proposed by Zheng and Mahajanam (1999).

- Worst case design involving minimising the maximum constraint violations (Walsh and Perkins, 1996) could be used in dealing with uncertainty more rigorously, possibly providing more global solutions. See section 3.3.1 for a description of worst case design under uncertainty.

6.3.2. Potential case studies

A series of example problems to test, demonstrate and assist in the further development of the integrated design procedure must be undertaken. Indeed, heuristics can only be garnered from experience. Hence the investigation of more case studies is the way forward in deriving more engineering rules of thumb. Unfortunately there is a tendency for them to be case specific, but it is hoped that certain patterns and similarities will emerge.

The following cases have identified; the last one is dealt with in more detail below:

- Nitrogen generators
- Poly-vinyl acetate (PVA) polymerisation
- Explosives manufacture
- Automobile catalytic converters
- HDA process (Douglas, 1988)
- VOC abatement

One application which may be benefitted by responsive operation can be found in the field of catalytic hydrocarbon abatement. Currently industry uses continuous catalytic oxidation units for VOCs (Volatile Organic Compounds) emitted

undesirably from processes as diverse as chemicals and electronics production, textiles manufacture, printing, metal coating, wire enamelling etc. These units are generally oversized and difficult to control and are typically used as a sink through which all effluent lines of the plant must flow. A more efficient solution may be the use of several smaller, responsive units, each dedicated to a single line. This "array" of versatile oxidizers may prove to be more economically attractive since they will only operate for as long as they are required to do so. It is envisaged that as soon as some critical concentration of VOC is reached in the buffer preceding the catalytic oxidation unit, the temperature in the reactor would be raised to the light-off point of the predominant species about to enter. As soon as the VOC concentration in this buffer falls below an acceptable level that can be released into the atmosphere, the reactor is turned off. Challenges of this design problem include:

- determining how small the preceding buffer can be,
- selecting the optimal temperature profile for operation,
- finding what the concentration in the reactor must fall to at shutdown, in order to compensate for the time delay incurred later during startup as the temperature is raised from ambient to "light-off", whilst maintaining the outlet composition constraint.

Heterogeneous catalysis of VOCs is typically carried out in **monolith** reactors. A 2-D model representing monolith channels as right cylinders of the same hydraulic diameter as the channels is deemed appropriate for the following reasons:

- mass and heat transfer coefficients may be calculated from simulation,
- the heterogeneous reaction at the wall is considered without assuming constant radial temperature, concentration and velocity profiles.

The real challenge to this case study is the incorporation of a distributed model into the integrated design framework, as only lumped models have been used so far. A more detailed proposal for this case study can be found in appendix D.

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Appendix A

Piecewise profiles

The feasible path dynamic optimiser *gOPT* only allows a single interval structure to be specified for all the time varying controls. The following fully continuous differentiable approximations (Cobden, 1998) were implemented in the *gPROMS* environment in order to provide different interval structures for the reactor composition setpoint and the flows to the storage tanks in the multiproduct reactor example from chapter 4. In chapter 5, the product demand profile and all the controls utilise these interval structures in the open-loop design.

A.1. Piecewise constant

$$\alpha_k(t) = \frac{1+\tanh[\beta(t-T_{k+1})]}{2} \qquad k = 1..N-1 \tag{A.1}$$

$$u(t) = u_1 + \sum_{k=2}^{N}(u_k - u_{k-1})\alpha_{k-1} \tag{A.2}$$

$$0 = T_1 \leq T_2 \leq T_3 \ldots \leq T_{N+1} = H \tag{A.3}$$

where N is the number of intervals, u_k are the values of the control in each interval, T_k are the times at which the control switches value, H is the time hori-

zon, $u(t)$ is the continuous differentiable control, and α_k is the 'decision' variable that enables "switching". As β increases the function tends closer and closer to an instant change until β equals infinity, at which point the function becomes discontinuous.

A.2. Piecewise linear continuous

Virtually the same equations apply as for the piecewise constant case:

$$\alpha_k(t) = \frac{1+\tanh[\beta(t-T_{k+1})]}{2} \qquad k = 1..N-1 \tag{A.4}$$

$$u(t) = u_1(t) + \sum_{k=2}^{N}(u_k(t) - u_{k-1}(t))\alpha_{k-1} \tag{A.5}$$

$$0 = T_1 < T_2 < T_3 \ldots < T_{N+1} = H \tag{A.6}$$

u_k in the second equation, however, is now a function of time, instead of a constant value:

$$u_k(t) = \frac{ul_{k+1} - ul_k}{T_{k+1} - T_k}(t - T_k) + ul_k \qquad k = 1..N \tag{A.7}$$

There are $N+1$ values of ul_k, the value of the control variable $u(t)$ at each boundary.

A.3. Piecewise linear discontinuous

The same set of equations are used, with $u_k(t)$ a different function of time again:

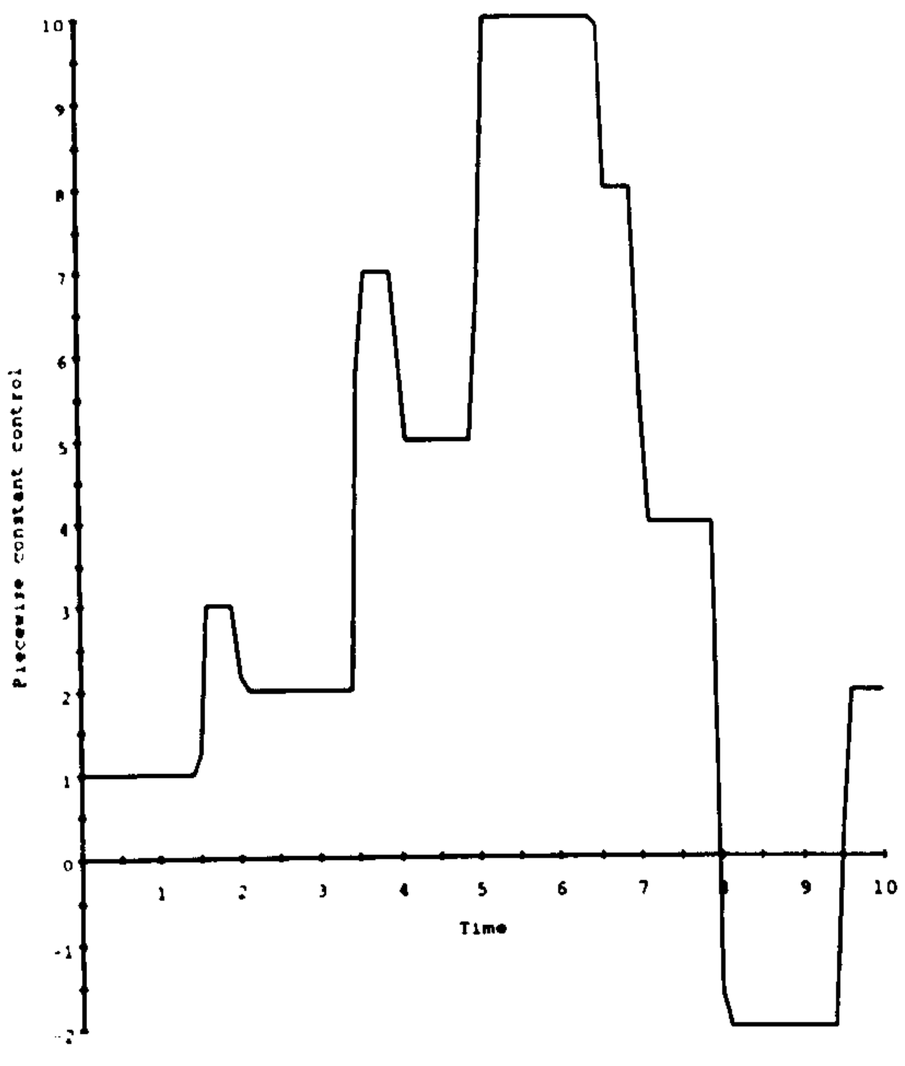
$$\alpha_k(t) = \frac{1+\tanh[\beta(t-T_{k+1})]}{2} \qquad k = 1..N-1 \tag{A.8}$$

$$u(t) = u_1(t) + \sum_{k=2}^{N}(u_k(t) - u_{k-1}(t))\alpha_{k-1} \tag{A.9}$$

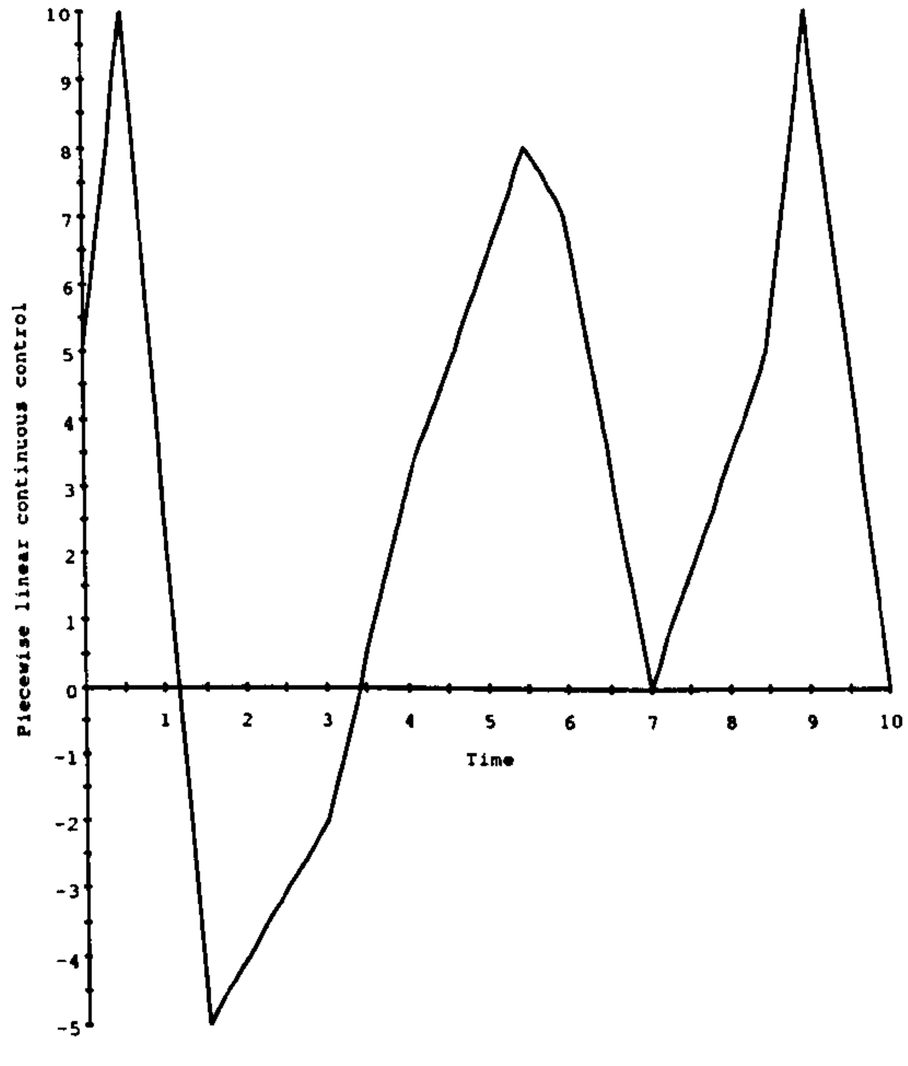
$$0 = T_1 < T_2 < T_3 \ldots < T_{N+1} = H \tag{A.10}$$

$$u_k(t) = \frac{ud_{2k} - ul_{2k-1}}{T_{k+1} - T_k}(t - T_k) + ud_{2k-1} \qquad k = 1..N \tag{A.11}$$

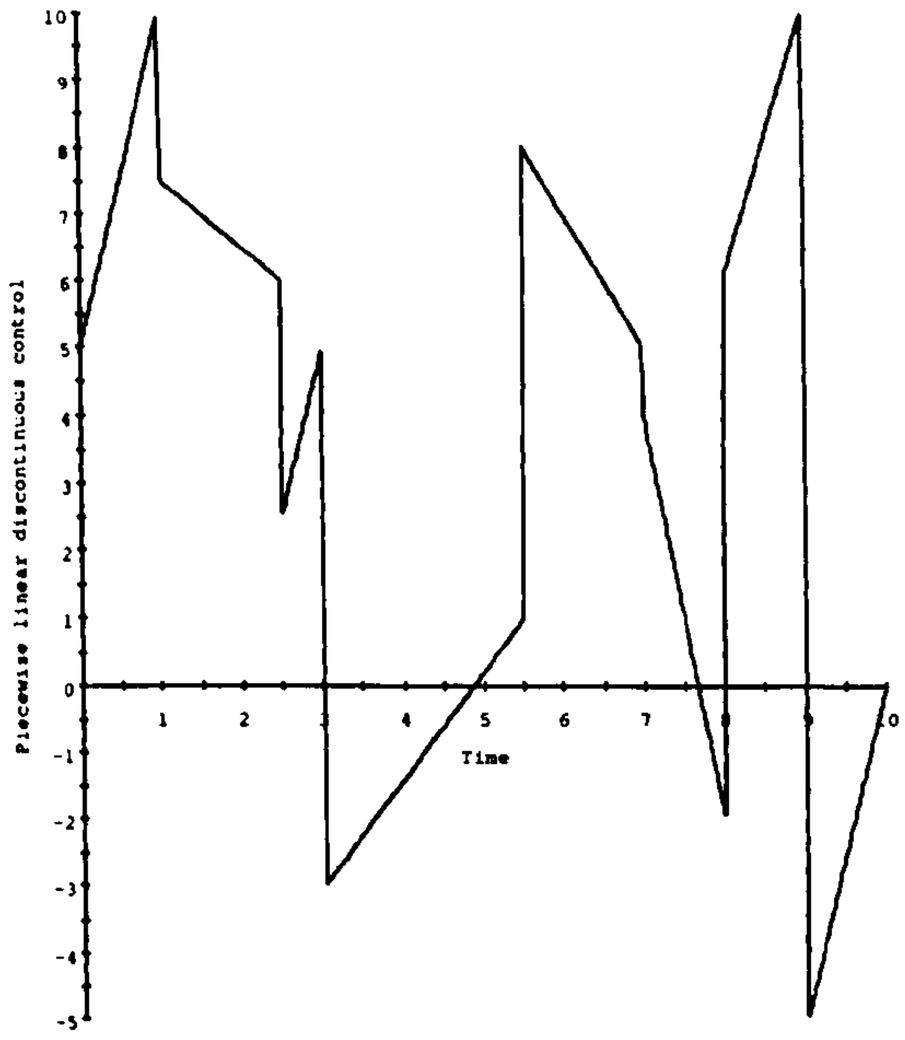
There are $2N$ values of ud_k: one at both ends of the time horizon (T_1 and T_{N+1}) and two at all intermediate boundaries ($T_2 \ldots T_N$).



(a) Piecewise constant



(b) Piecewise linear continuous



(c) Piecewise linear discontinuous

Figure A.1: Examples of piecewise control approximations

Appendix B

Published design algorithm for multiproduct processes

The cycle time model used by Cantrell and Luyben (1996) needs the

- product demand rate (fixed *a priori*),
- production rates (design variables) and
- transition times (found using dynamic simulation).

in order to calculate the necessary production times of each product within the production cycle. The actual cycle time calculation (Hanssmann, 1962) makes the following assumptions:

1. no slack time,
2. common cycle for all products (each produced once per cycle) and
3. transitions between products are sequence dependent.

Thus, a process producing n products requires n steady state periods connected by product transitions. When production of the nth product ends, the cycle is repeated.

Based on these assumptions, an expression for the lengths of the production periods may be derived using a balance on the rates of demand and production of each product over a complete cycle:

$$p_i t_i = r_i \left(\sum_{j=1}^{n} t_j + \theta \right) \qquad i = 1..n \tag{B.1}$$

where p_i is the rate of production of product i, t_i is the length of the production period for product i, r_i is the rate of demand for product i and θ is the sum of the transition times for a specified production schedule:

$$\theta = \sum_{(i,j) \in s_{trans}} s_{ij} \tag{B.2}$$

where s_{trans} is the set of transitions from product i to the next product, j, in the specified schedule and s_{ij} is the associated transition time. Note that the total cycle time is given by the following expression:

$$t_{cycle} = \sum_{i=1}^{n} t_i + \theta \tag{B.3}$$

Equation B.1 may be rearranged to give an explicit expression, in matrix form, for the n production times (t_i):

$$\boldsymbol{T} = \theta[\boldsymbol{P}^{diag} - \boldsymbol{L}\boldsymbol{R}^T]^{-1}\boldsymbol{R} \tag{B.4}$$

In equation B.4, $\boldsymbol{T}$ is an $n \times 1$ matrix of the production times (t_i), $\boldsymbol{P}^{diag}$ is an $n \times n$ diagonal matrix with the production rates (p_i) on the diagonal. $\boldsymbol{R}$ is an $n \times 1$ matrix of the product demand rates (r_i) and $\boldsymbol{L}$ is a $n \times 1$ matrix of ones.

Given the n demand and production rates, the following necessary condition must be satisfied for the cycle time to be positive and finite:

$$\sum_{i=1}^{n} \frac{r_i}{p_i} < 1 \qquad \text{(B.5)}$$

In the case where the left-hand side of inequality B.5 is exactly equal to one, i.e. undefined transition times, the cycle time is infinite and the bracketed term in equation B.4 becomes singular.

The authors go on to describe a number of potential models which may have been used to account for the cost of maintaining inventory. In the end, they used a "storage capacity capital cost" model, which determines the inventory costs from the maximum inventory level over a cycle.

The design algorithm for continuous multiproduct processes proposed by Cantrell and Luyben is shown in figure B.1.

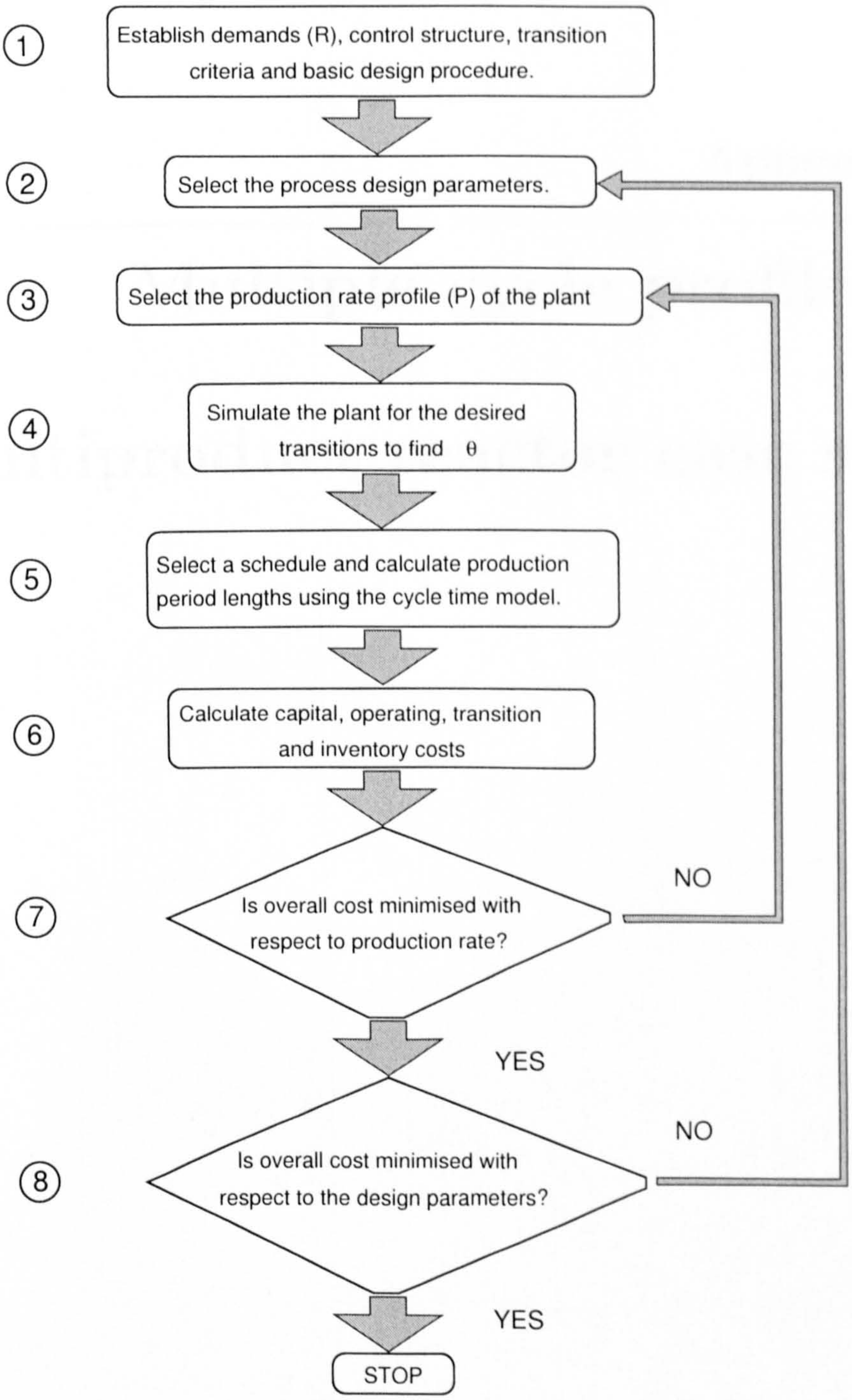


Figure B.1: Procedure for continuous multiproduct process design proposed by *Cantrell and Luyben*

Appendix C

Multiple cycle profiles for multiproduct reactor case study

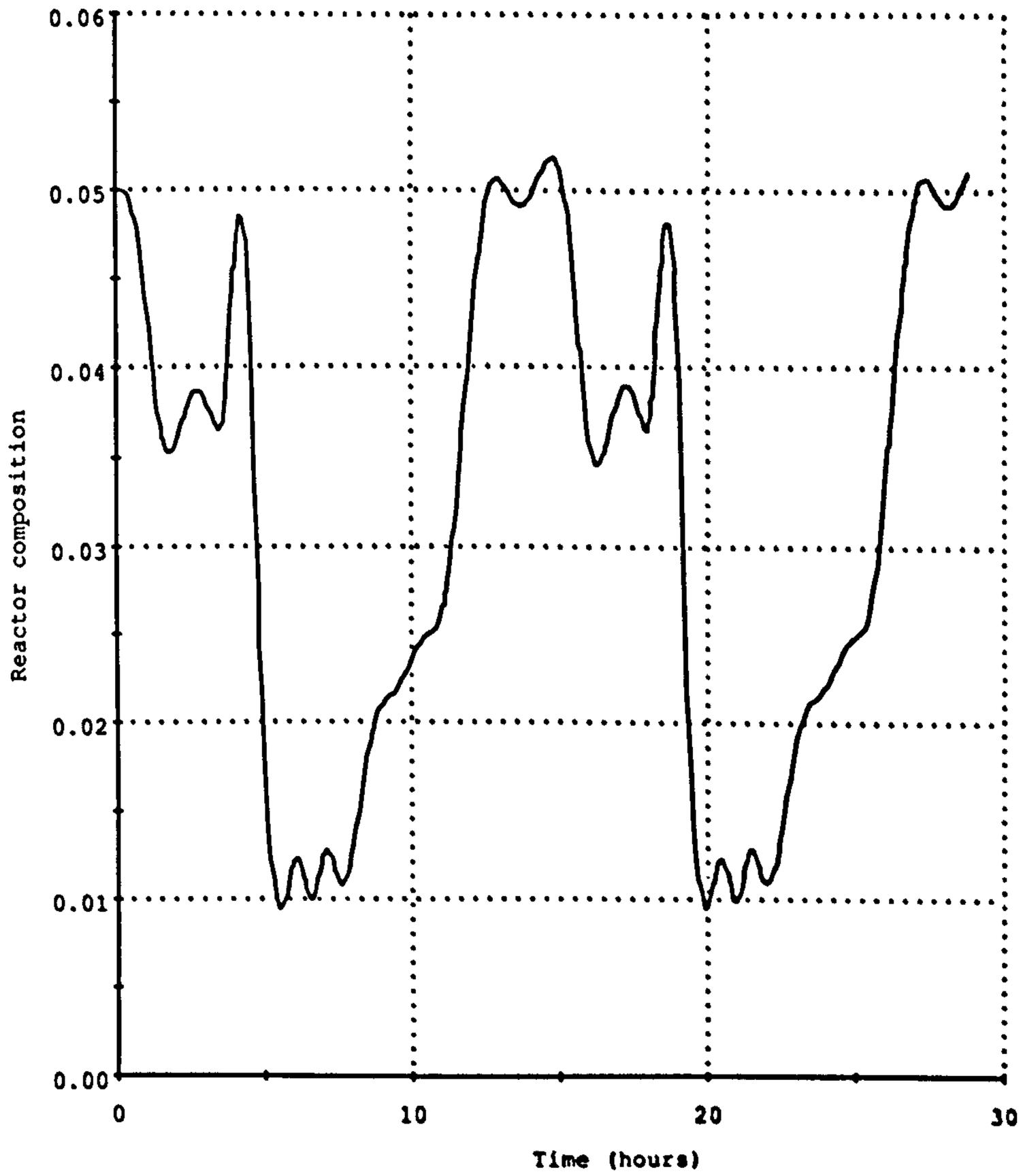
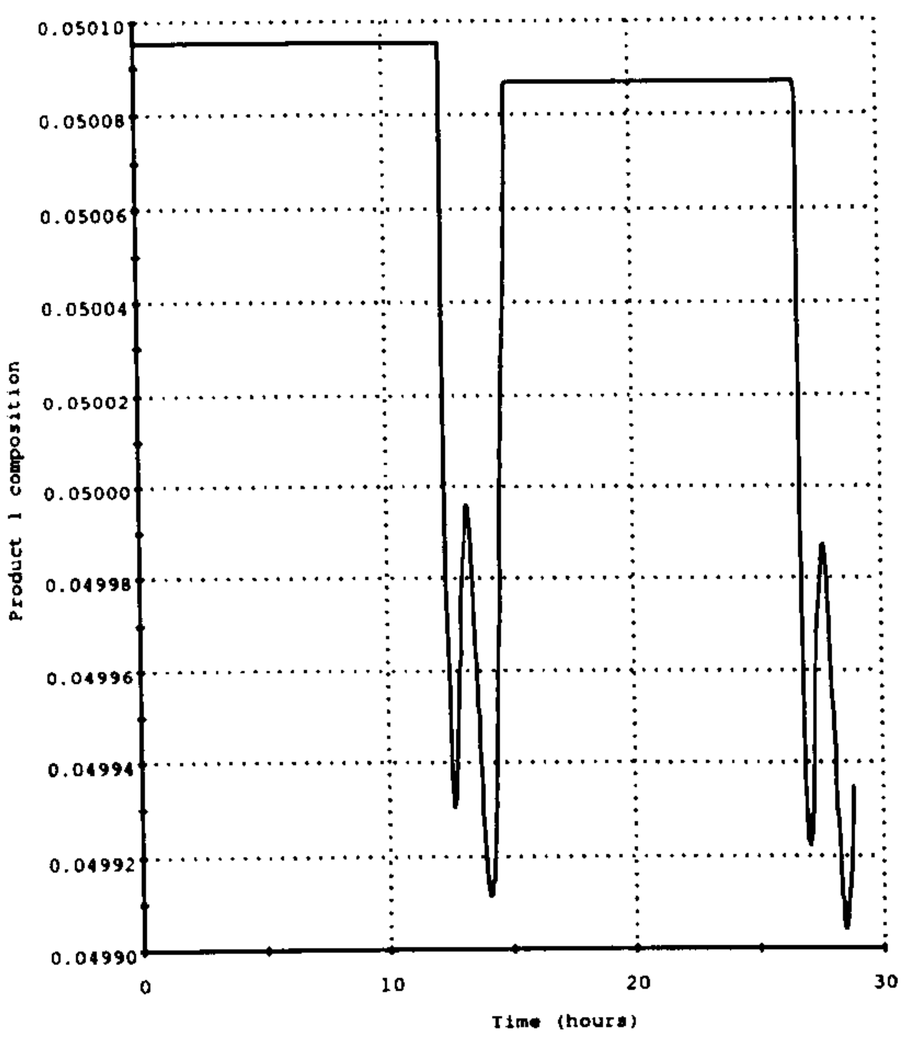
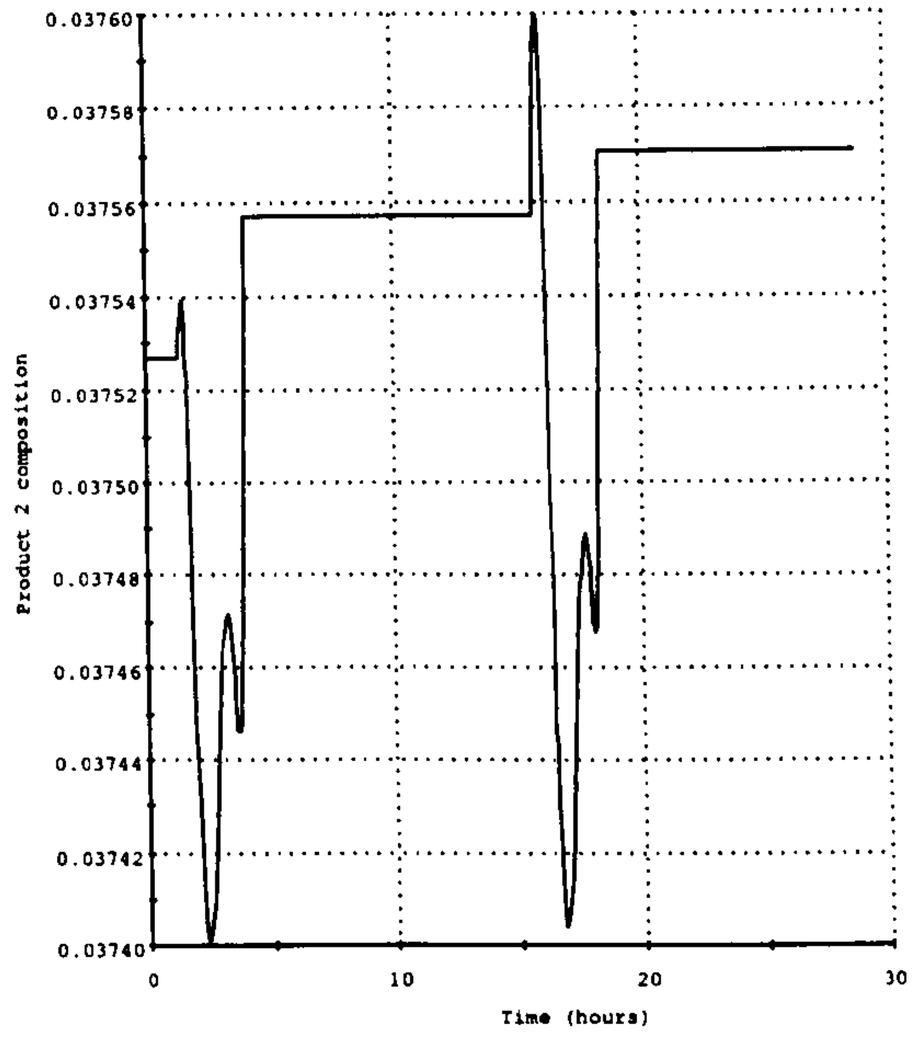


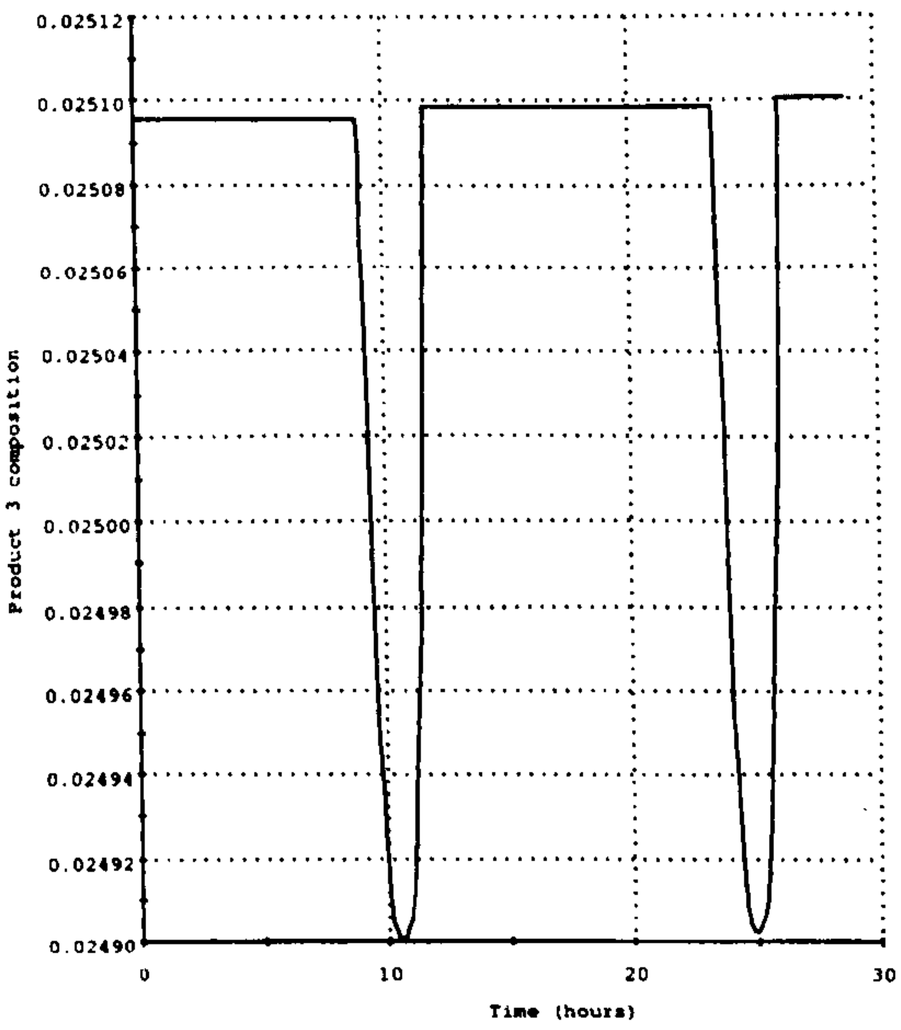
Figure C.1: Reactor composition optimised for 2 cycles minimising cost



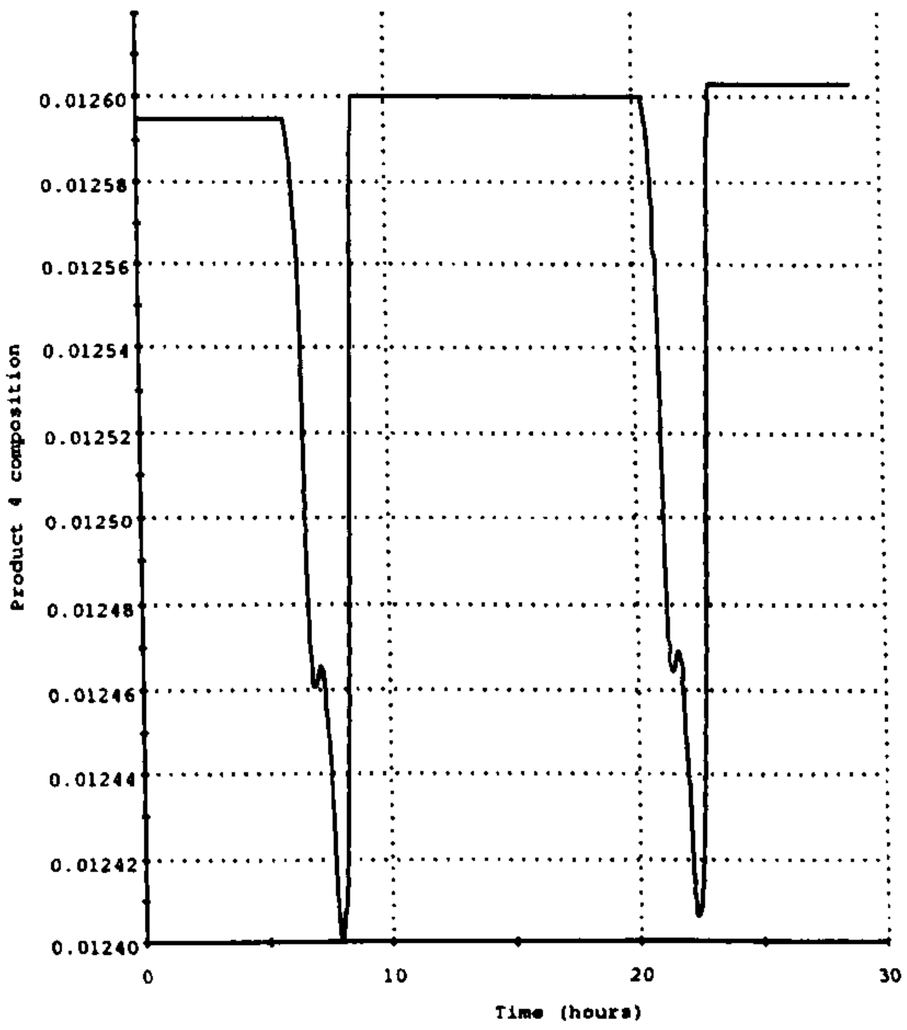
(a) Storage tank 1 composition



(b) Storage tank 2 composition



(c) Storage tank 3 composition



(d) Storage tank 4 composition

Figure C.2: Storage compositions optimised for 2 cycles minimising cost

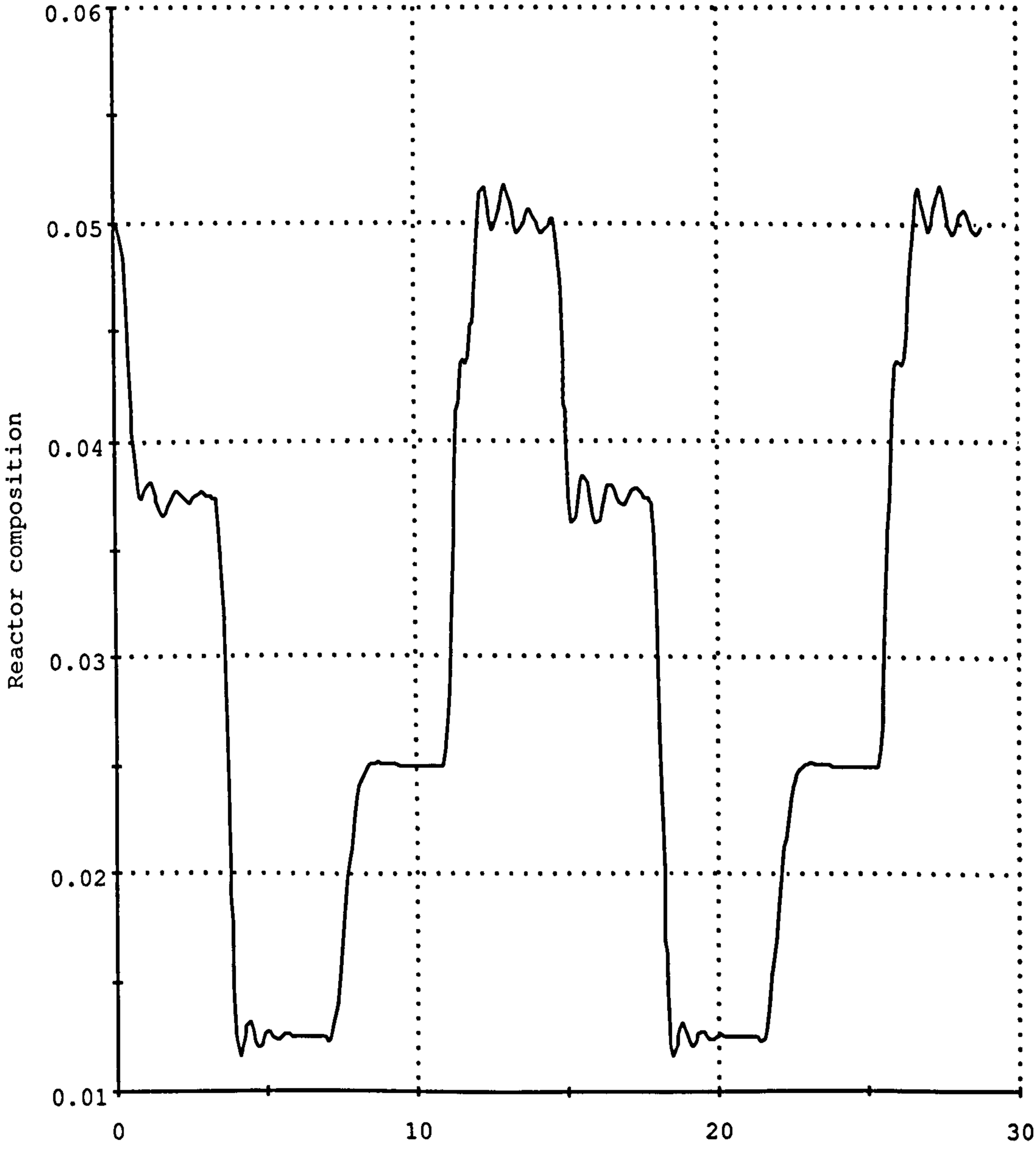
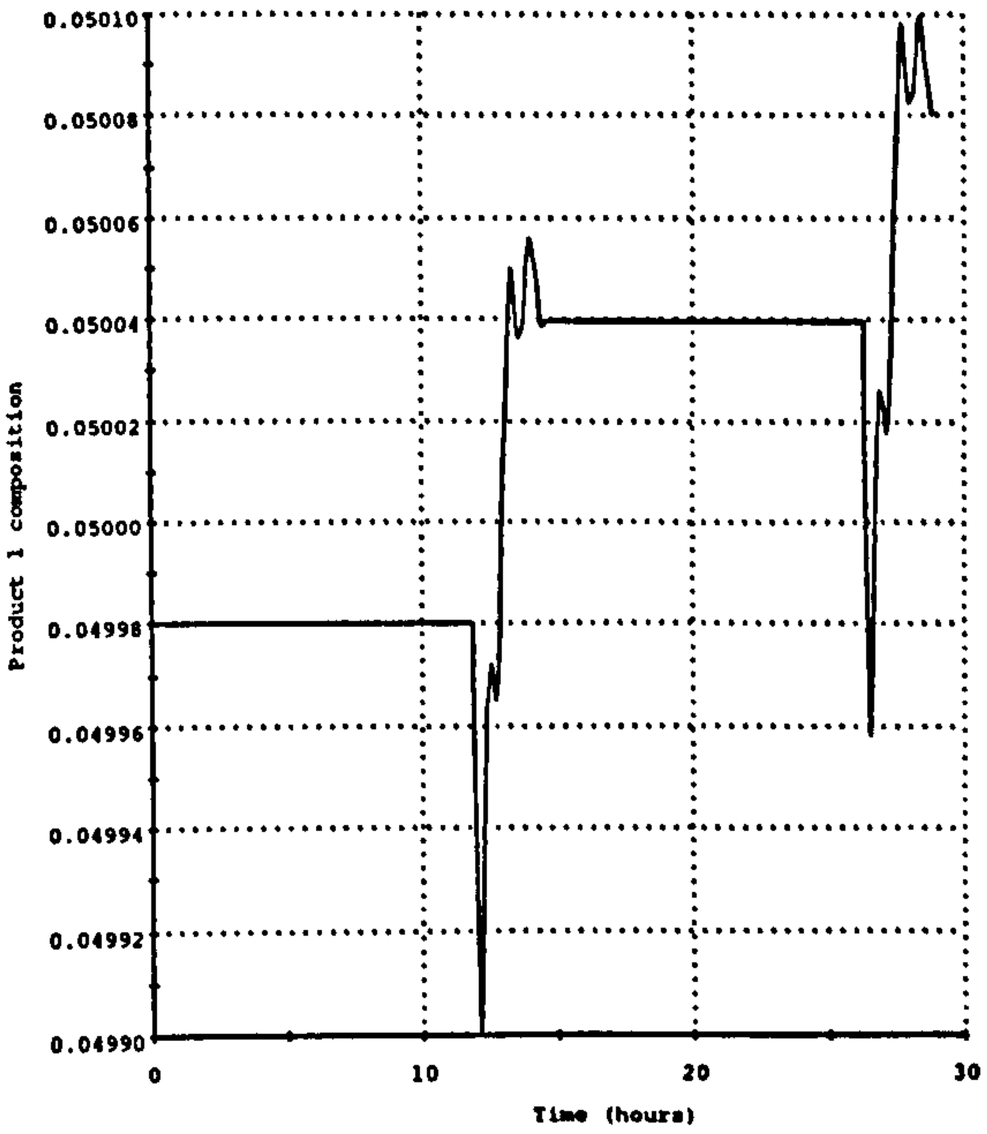
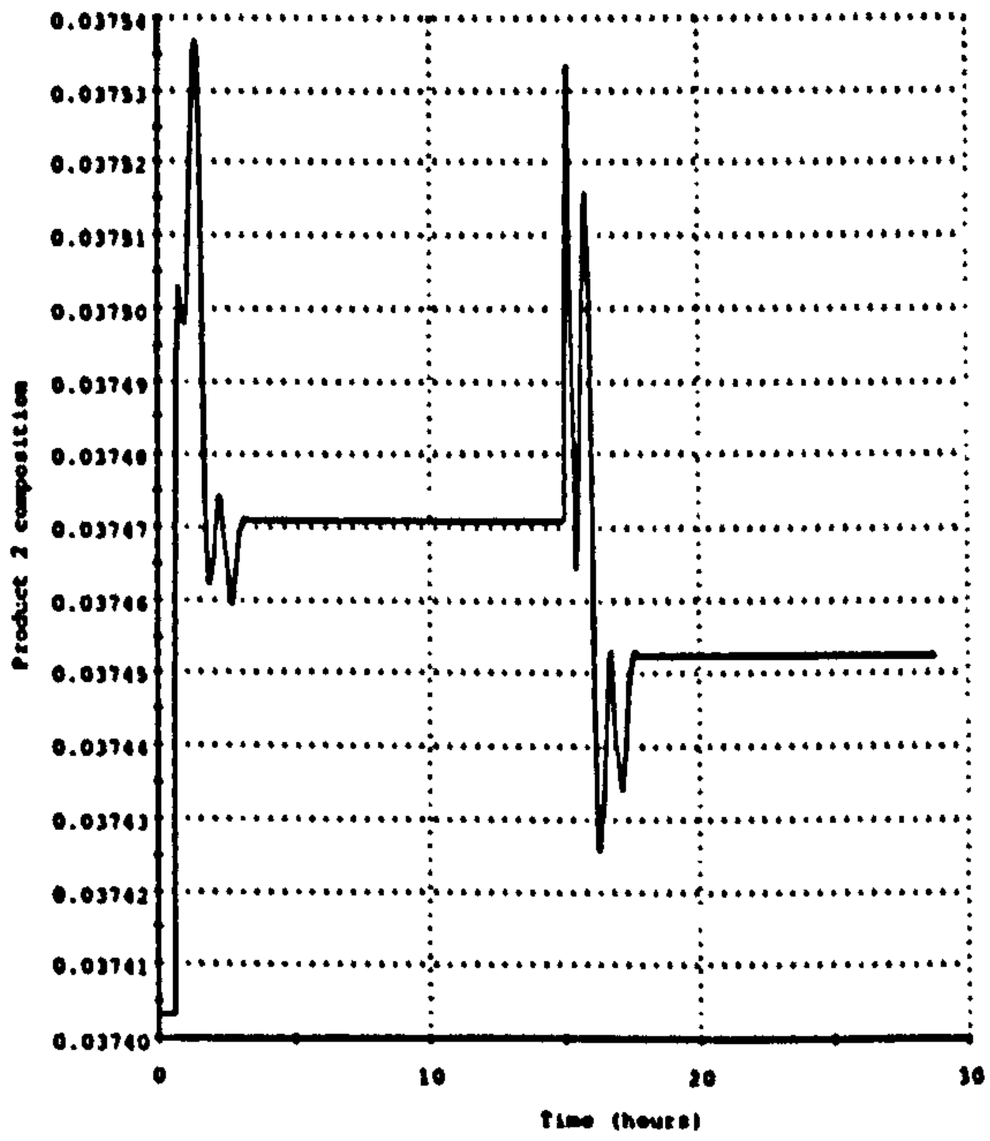


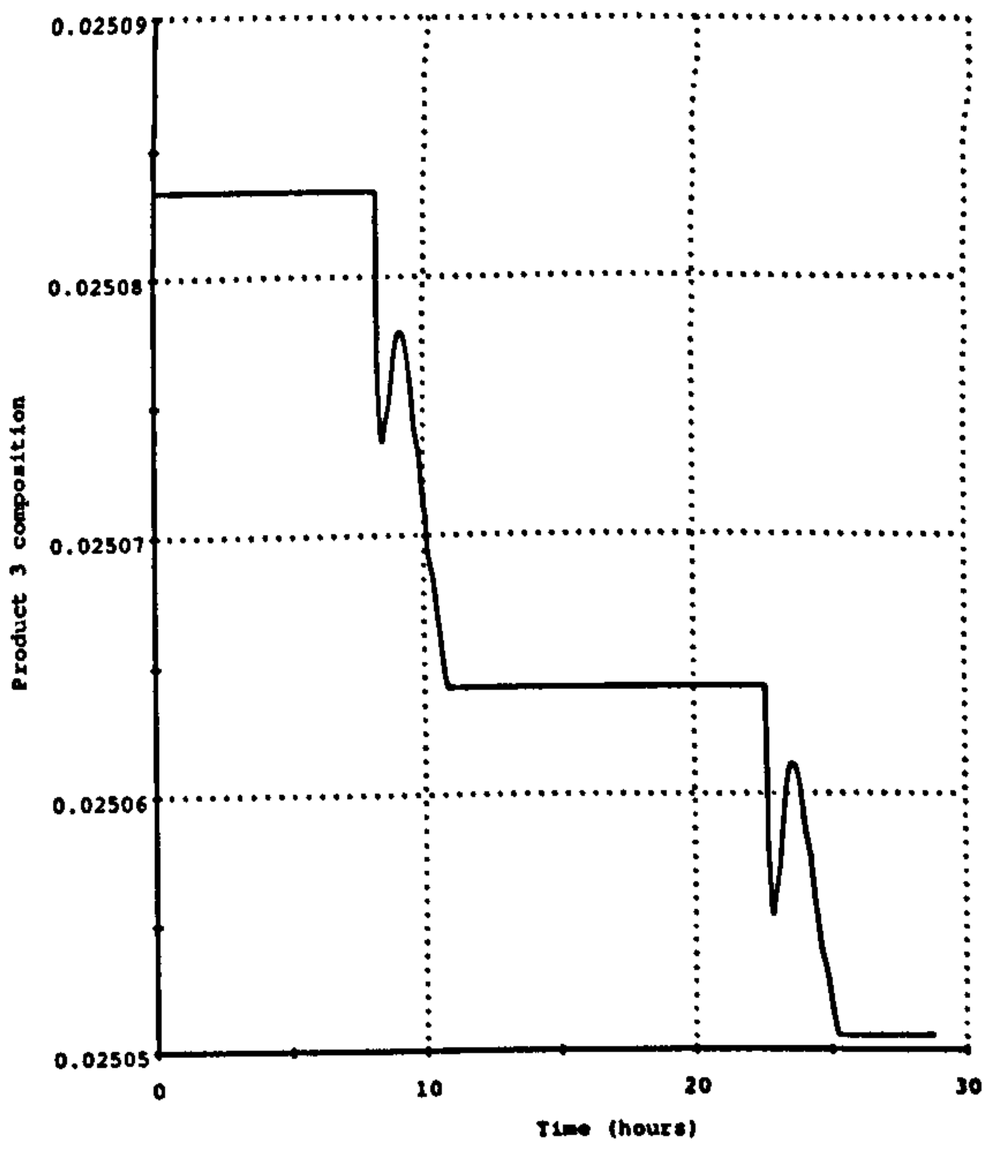
Figure C.3: Reactor composition optimised for 2 cycles minimising ISE



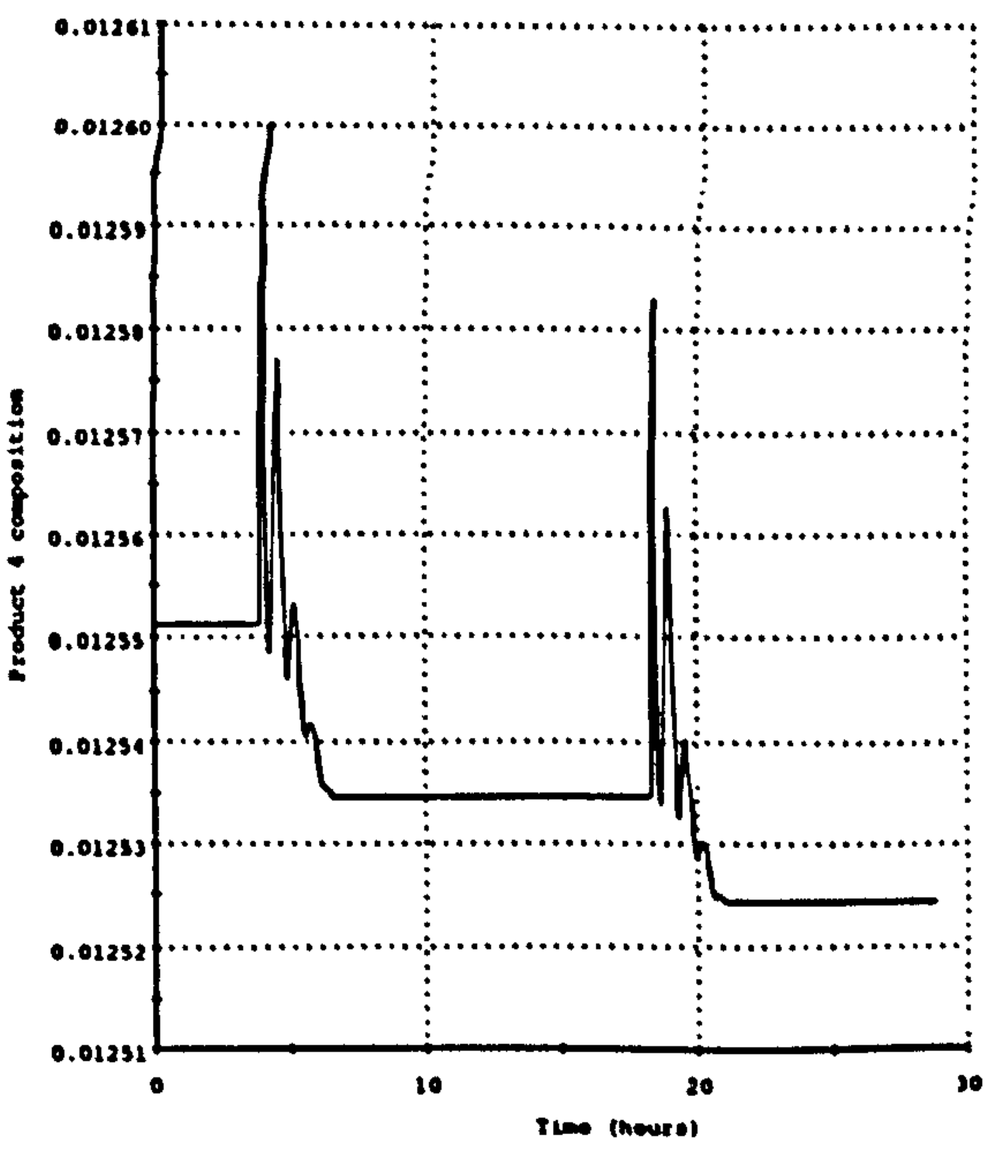
(a) Storage tank 1 composition



(b) Storage tank 2 composition



(c) Storage tank 3 composition



(d) Storage tank 4 composition

Figure C.4: Storage compositions optimised for 2 cycles minimising ISE

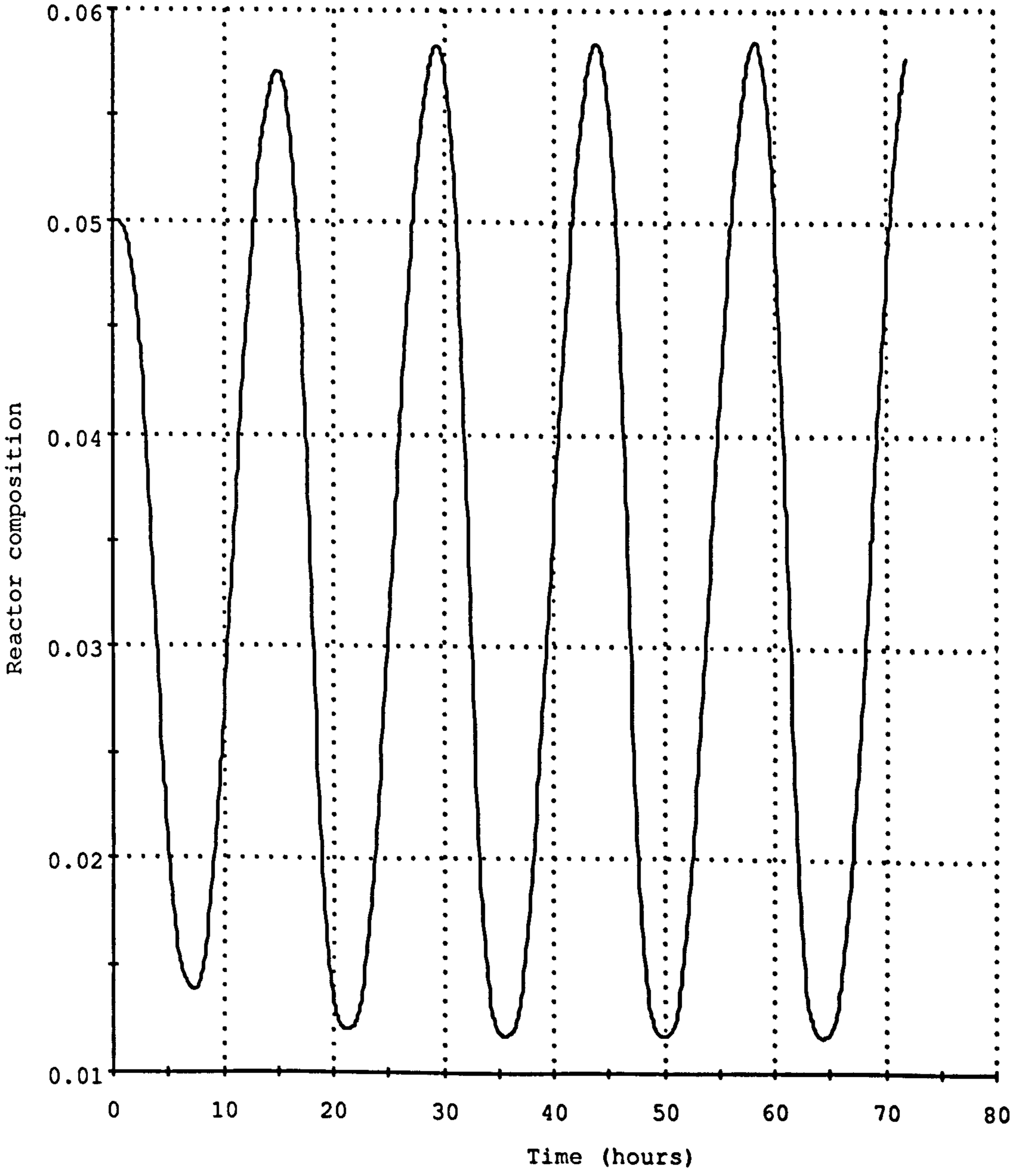
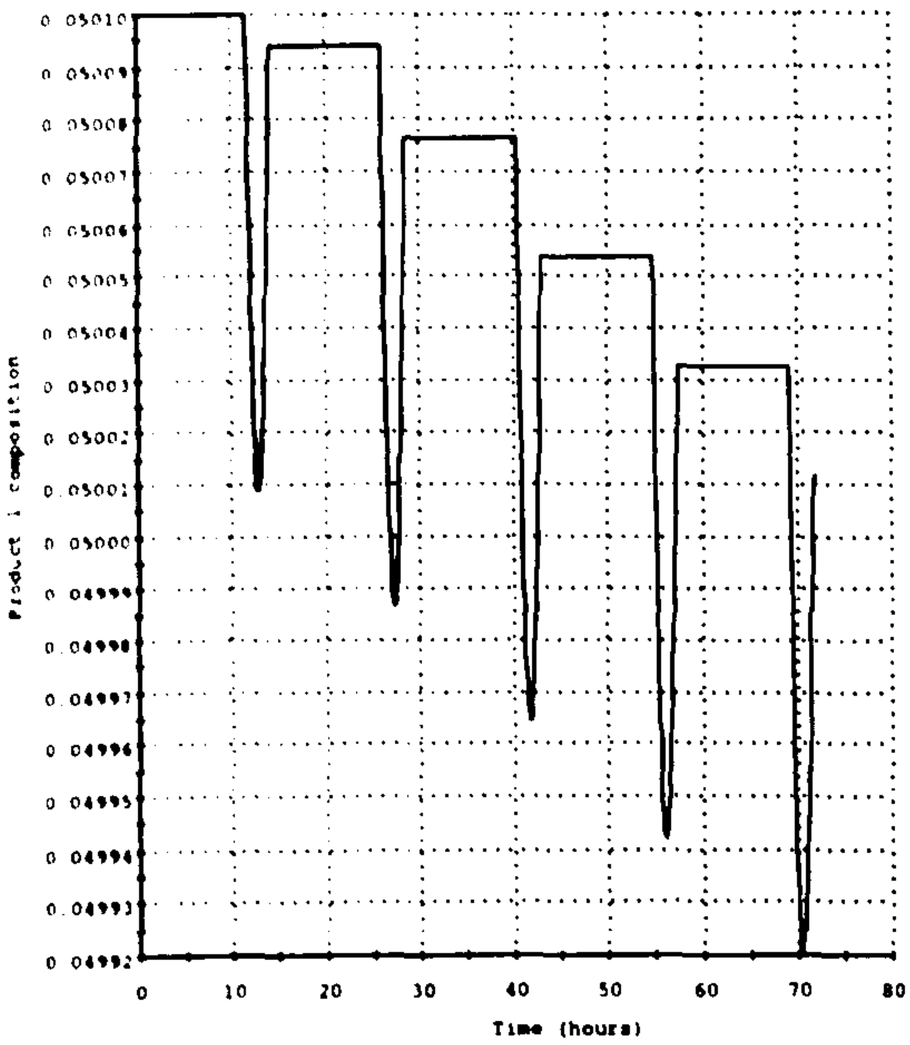
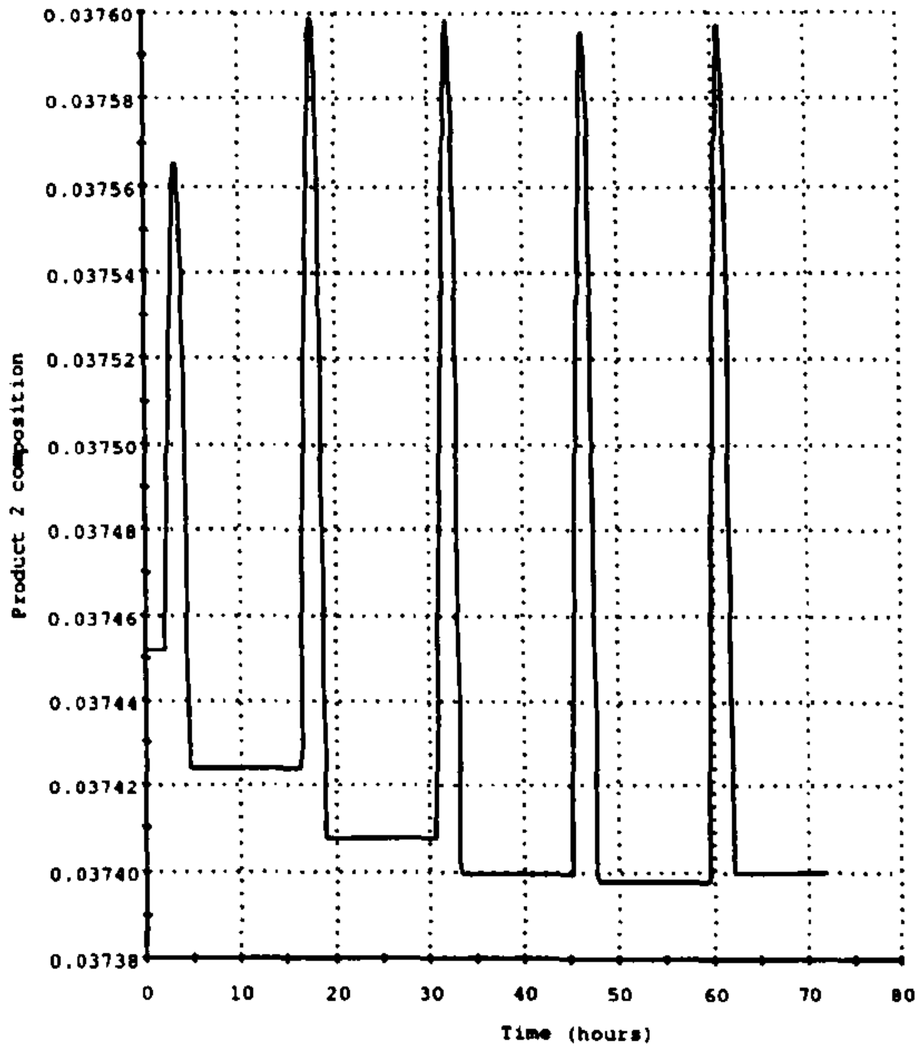


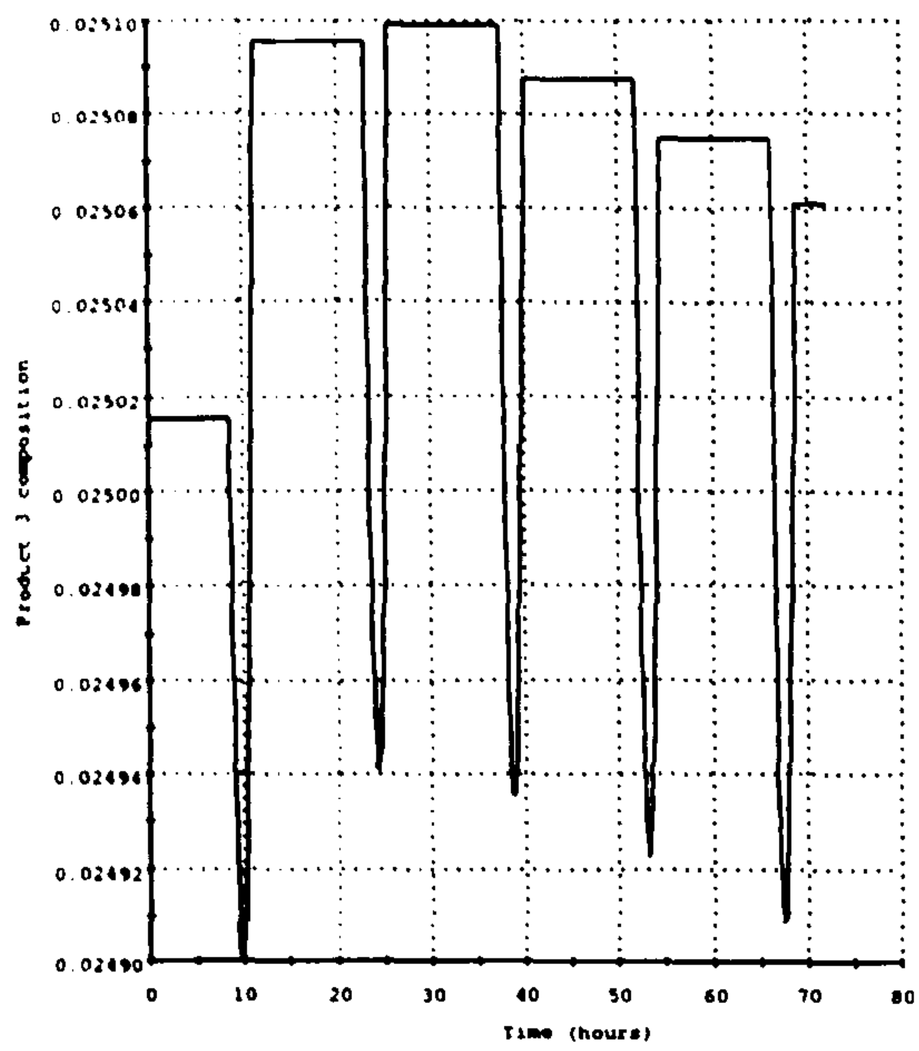
Figure C.5: Reactor composition optimised for 5 cycles minimising cost



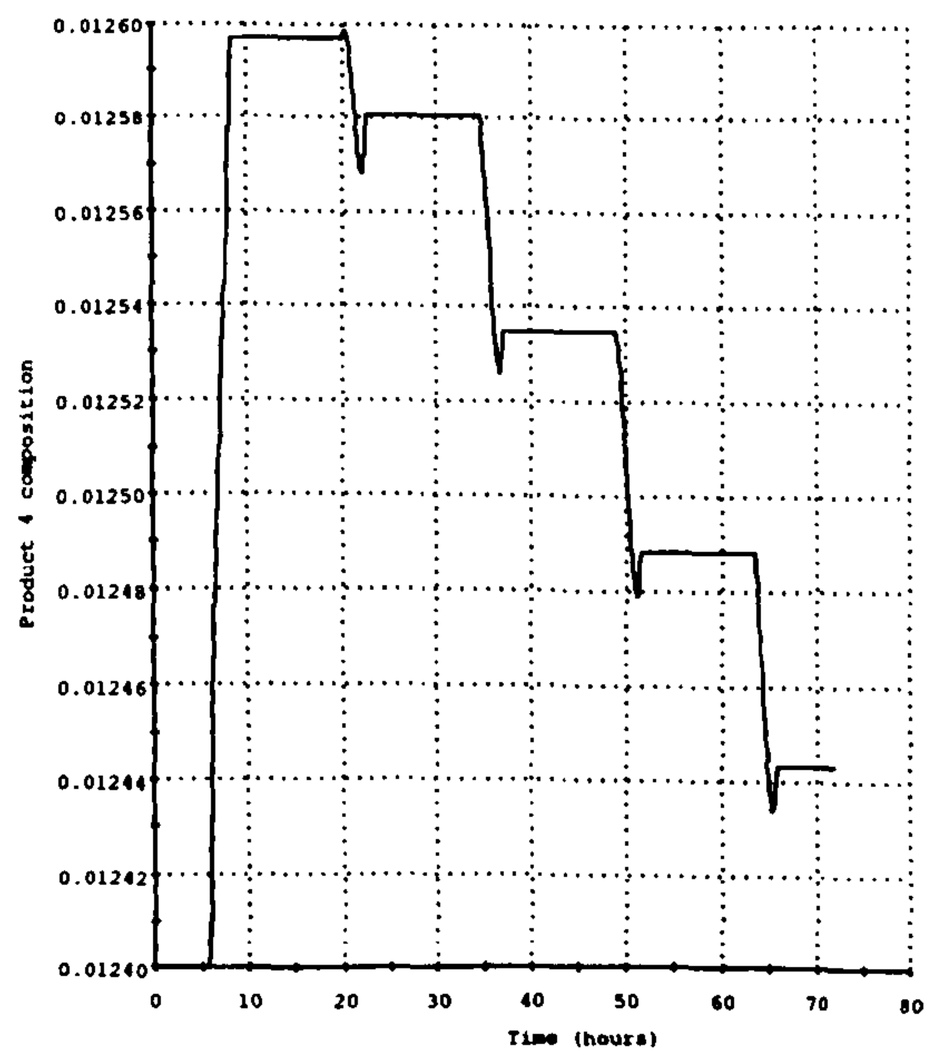
(a) Storage tank 1 composition



(b) Storage tank 2 composition



(c) Storage tank 3 composition



(d) Storage tank 4 composition

Figure C.6: Storage compositions optimised for 5 cycles minimising cost

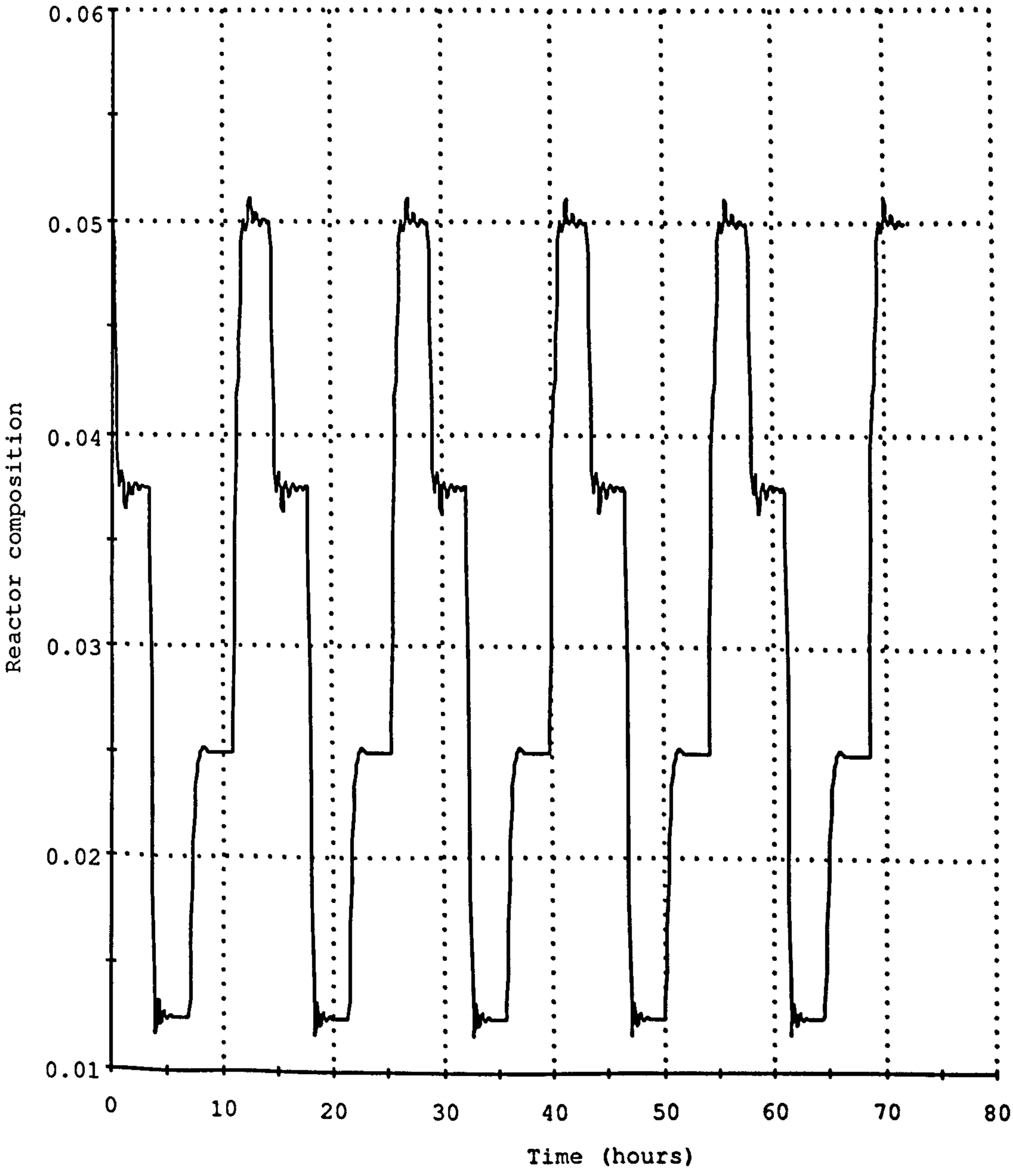
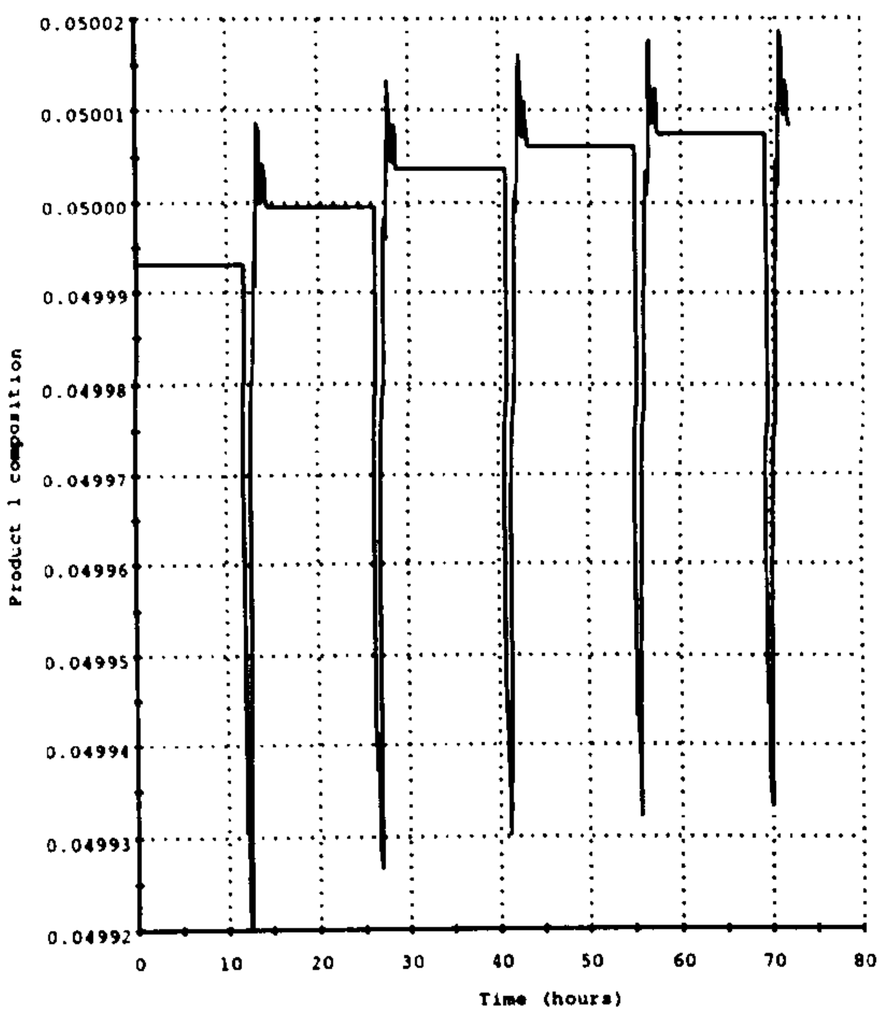
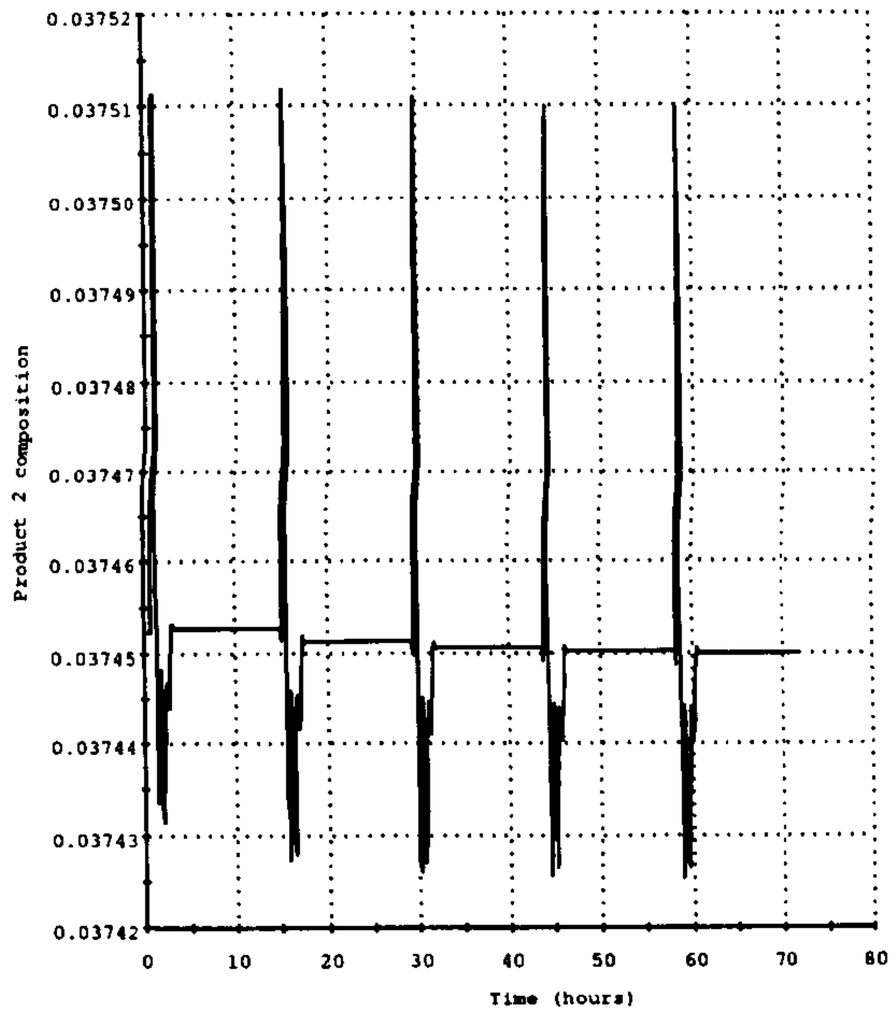


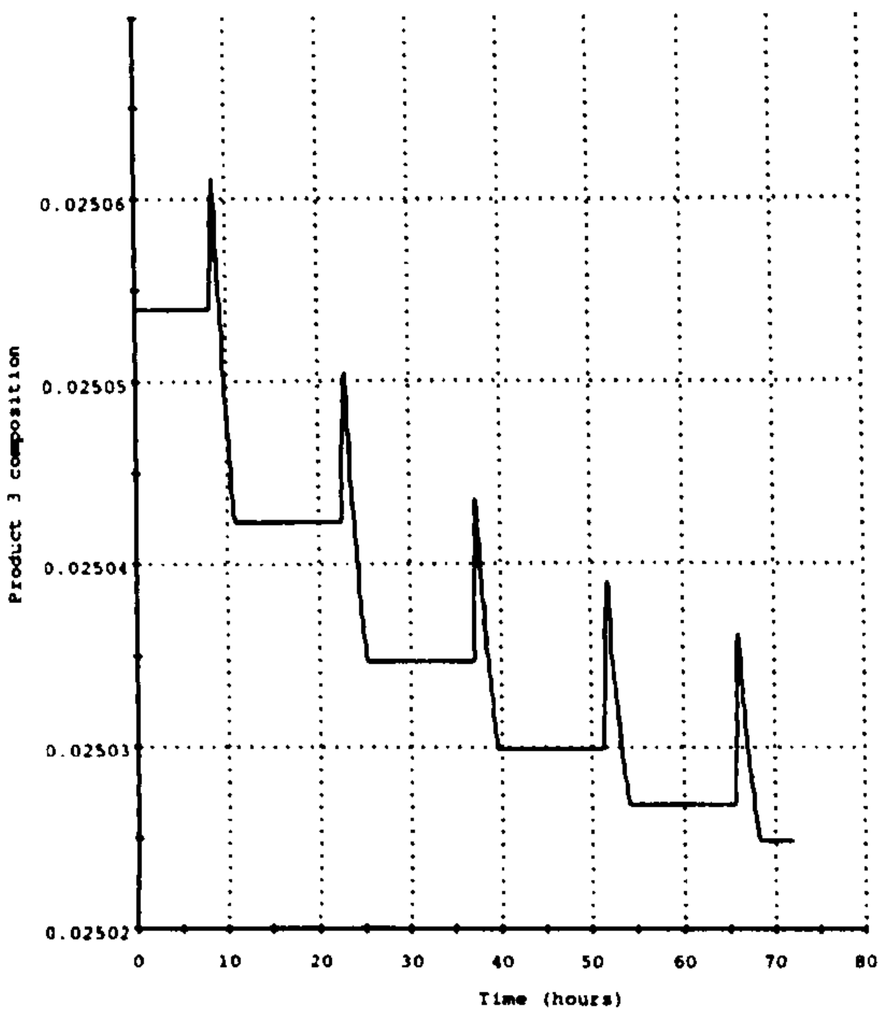
Figure C.7: Reactor composition optimised for 5 cycles minimising ISE



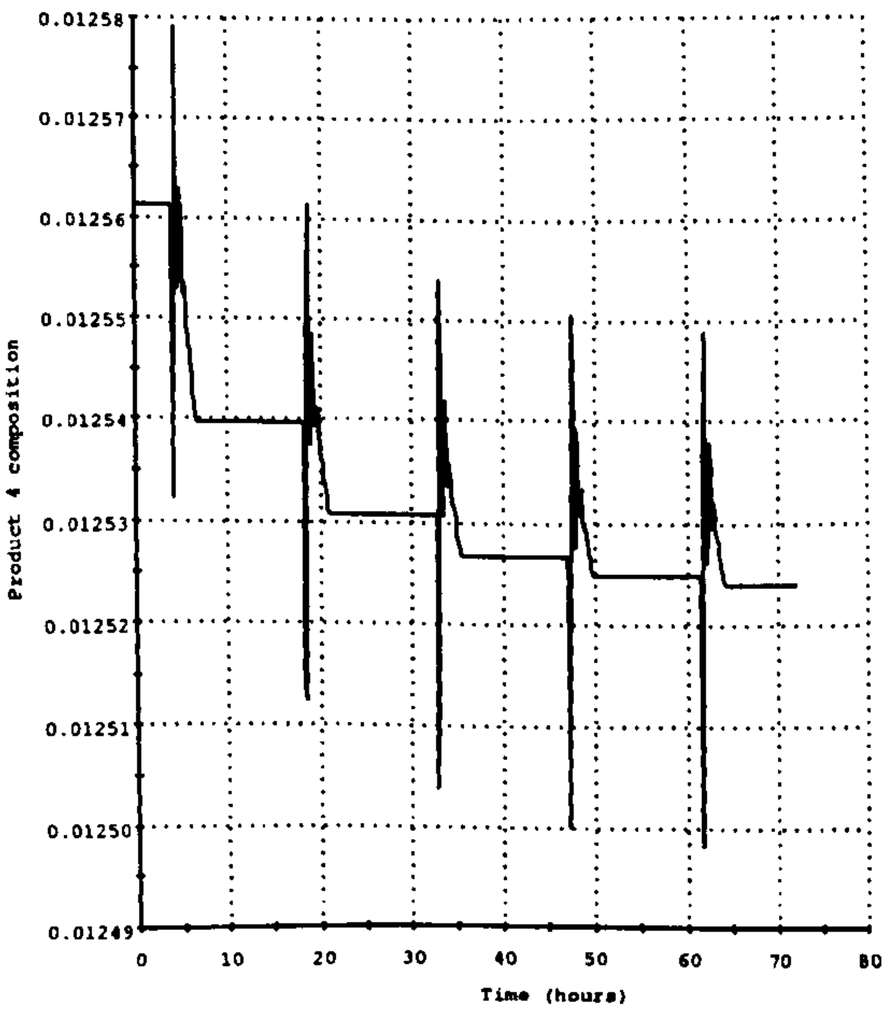
(a) Storage tank 1 composition



(b) Storage tank 2 composition



(c) Storage tank 3 composition



(d) Storage tank 4 composition

Figure C.8: Storage compositions optimised for 5 cycles minimising ISE

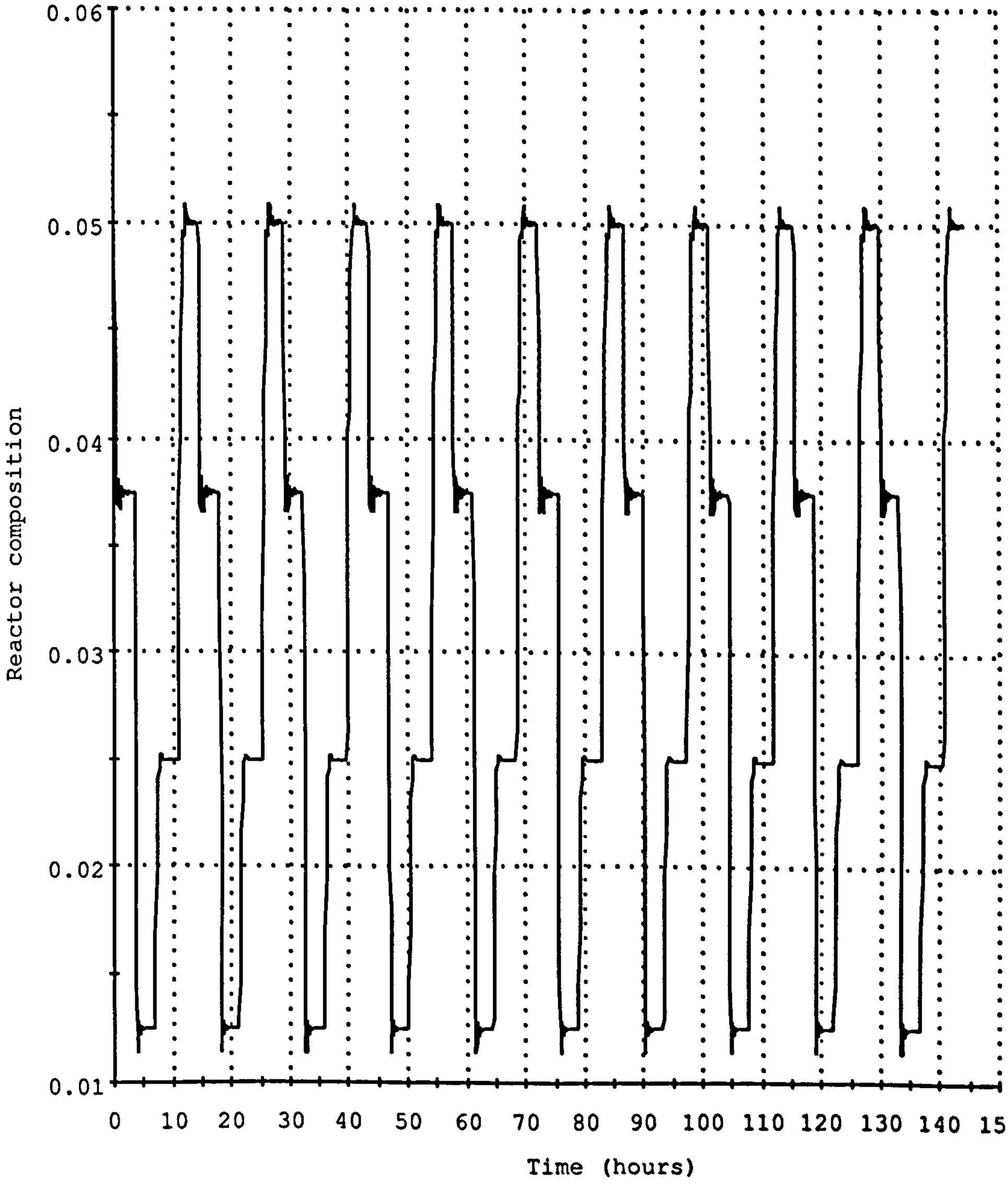
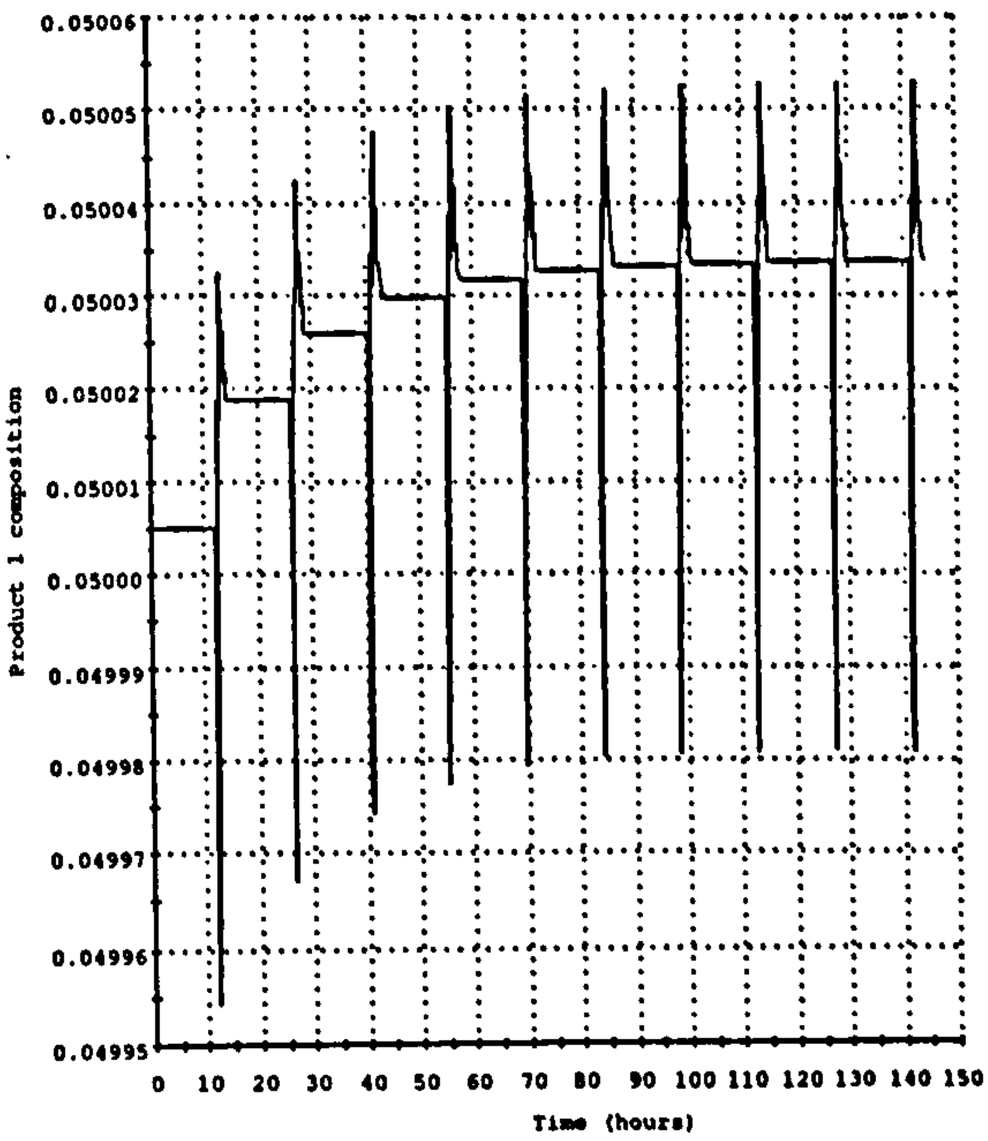
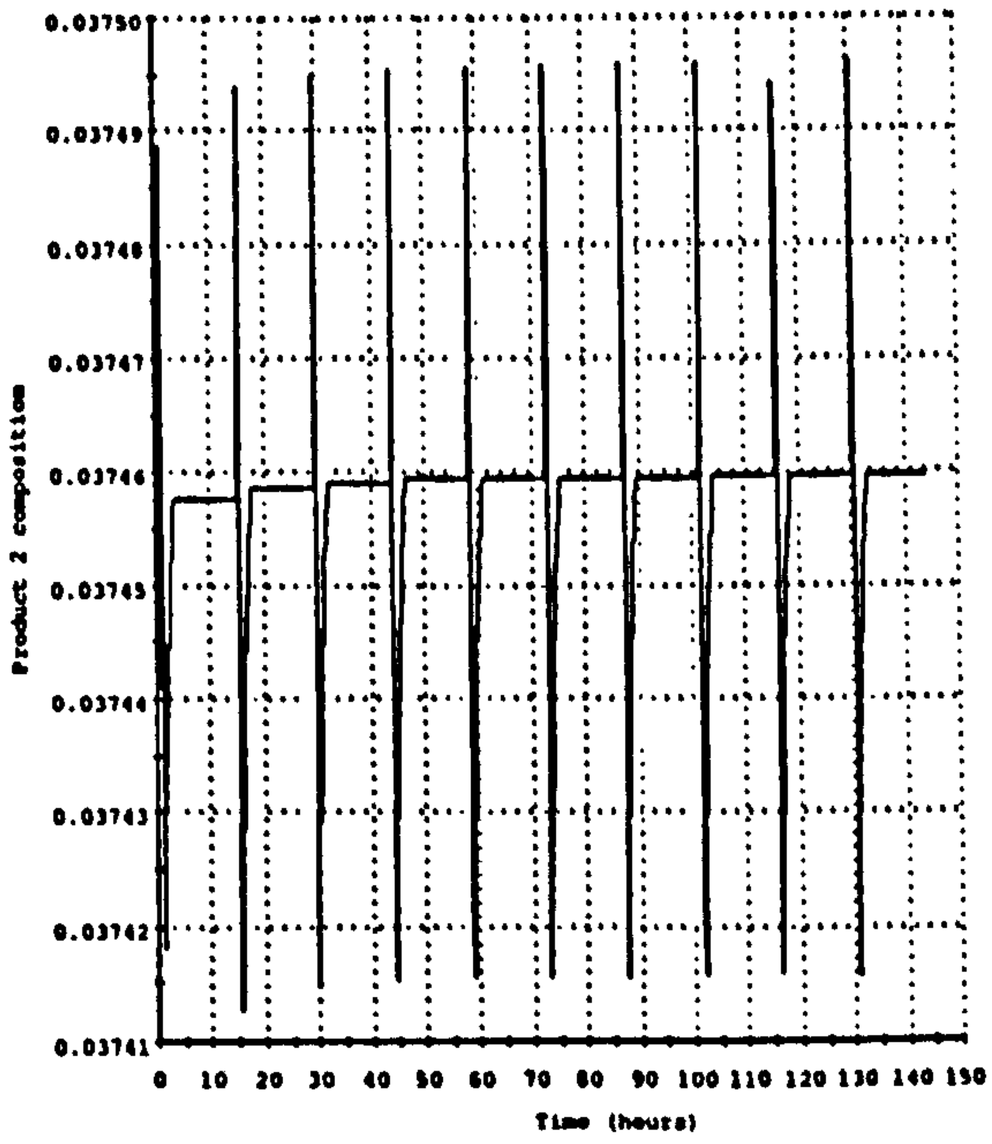


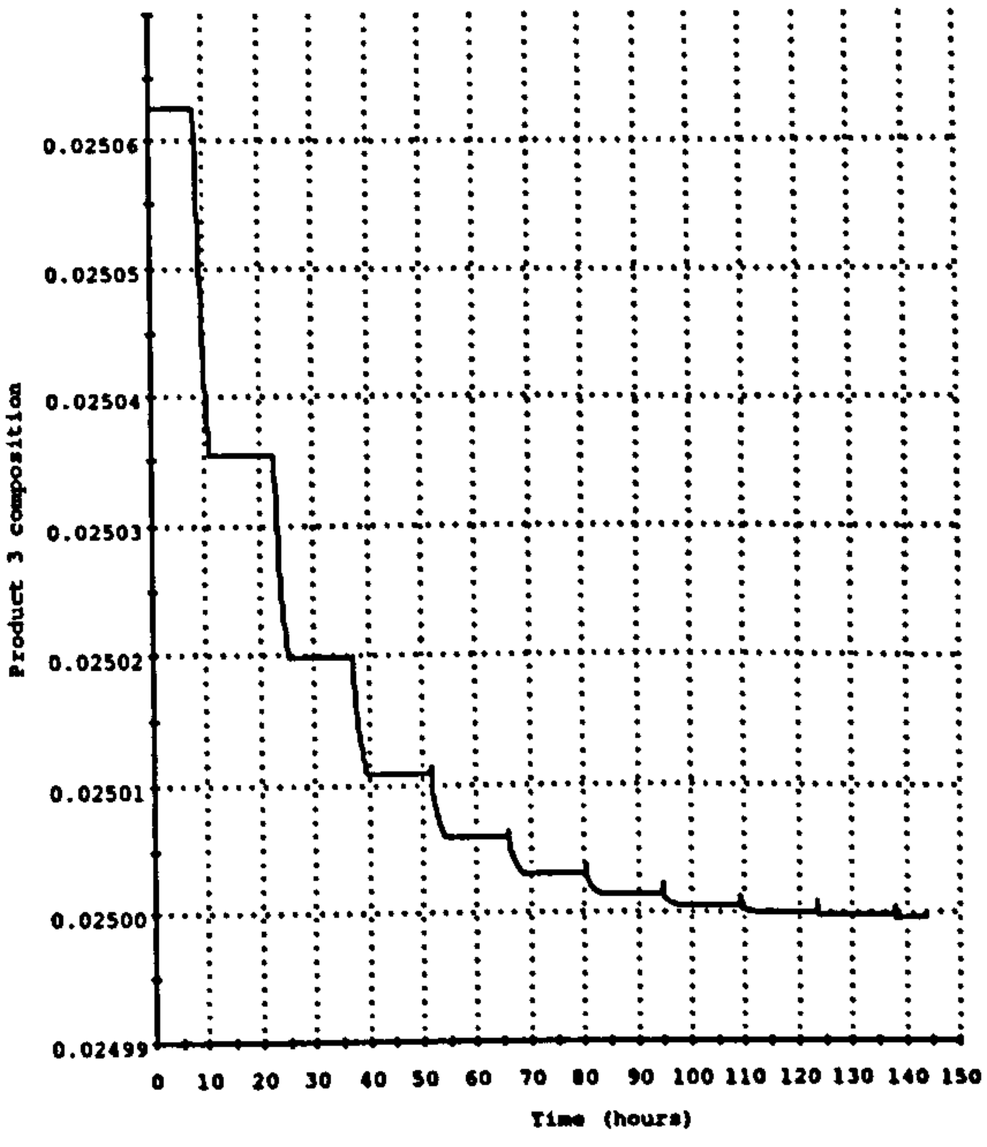
Figure C.9: Reactor composition optimised for 10 cycles minimising ISE



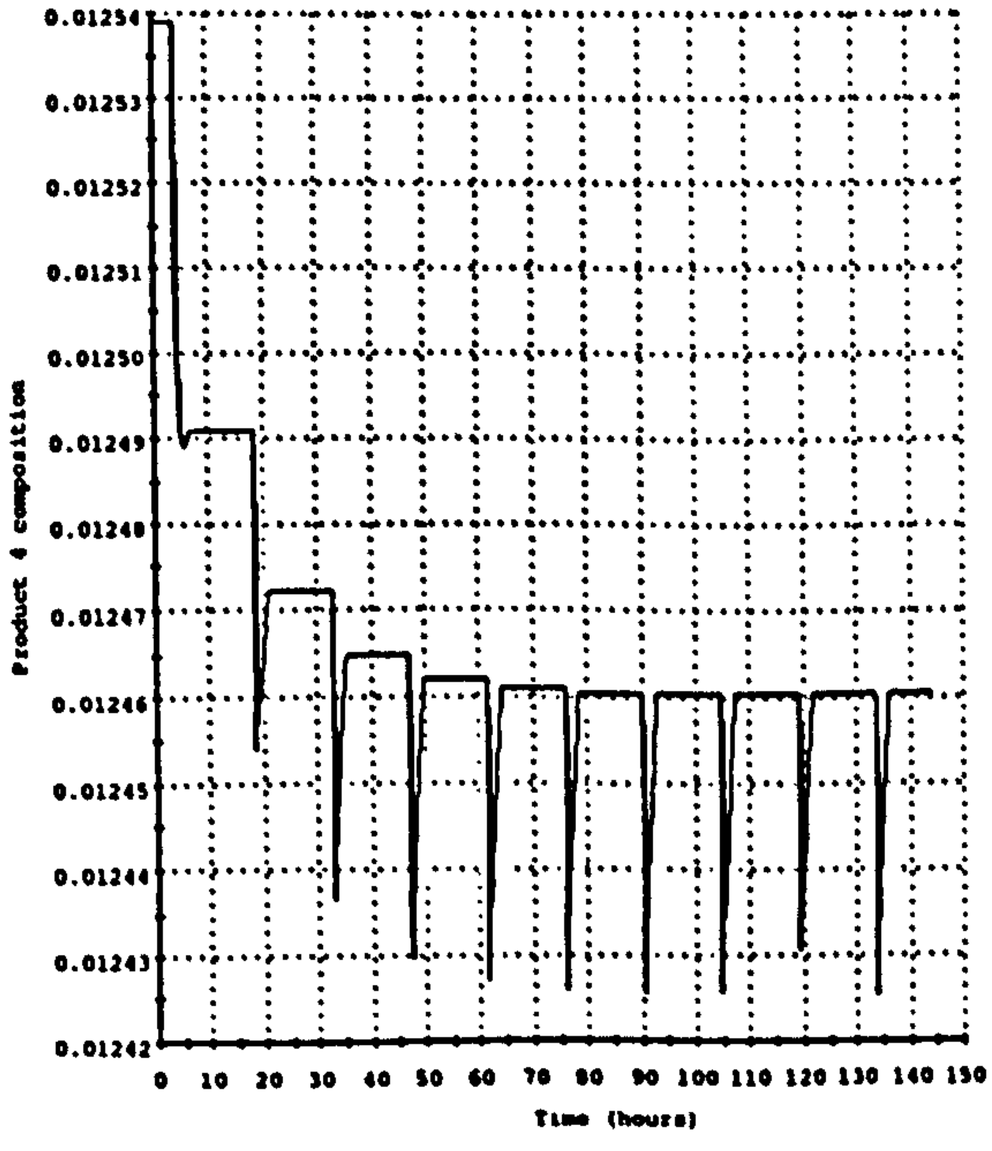
(a) Storage tank 1 composition



(b) Storage tank 2 composition



(c) Storage tank 3 composition



(d) Storage tank 4 composition

Figure C.10: Storage compositions optimised for 10 cycles minimising ISE

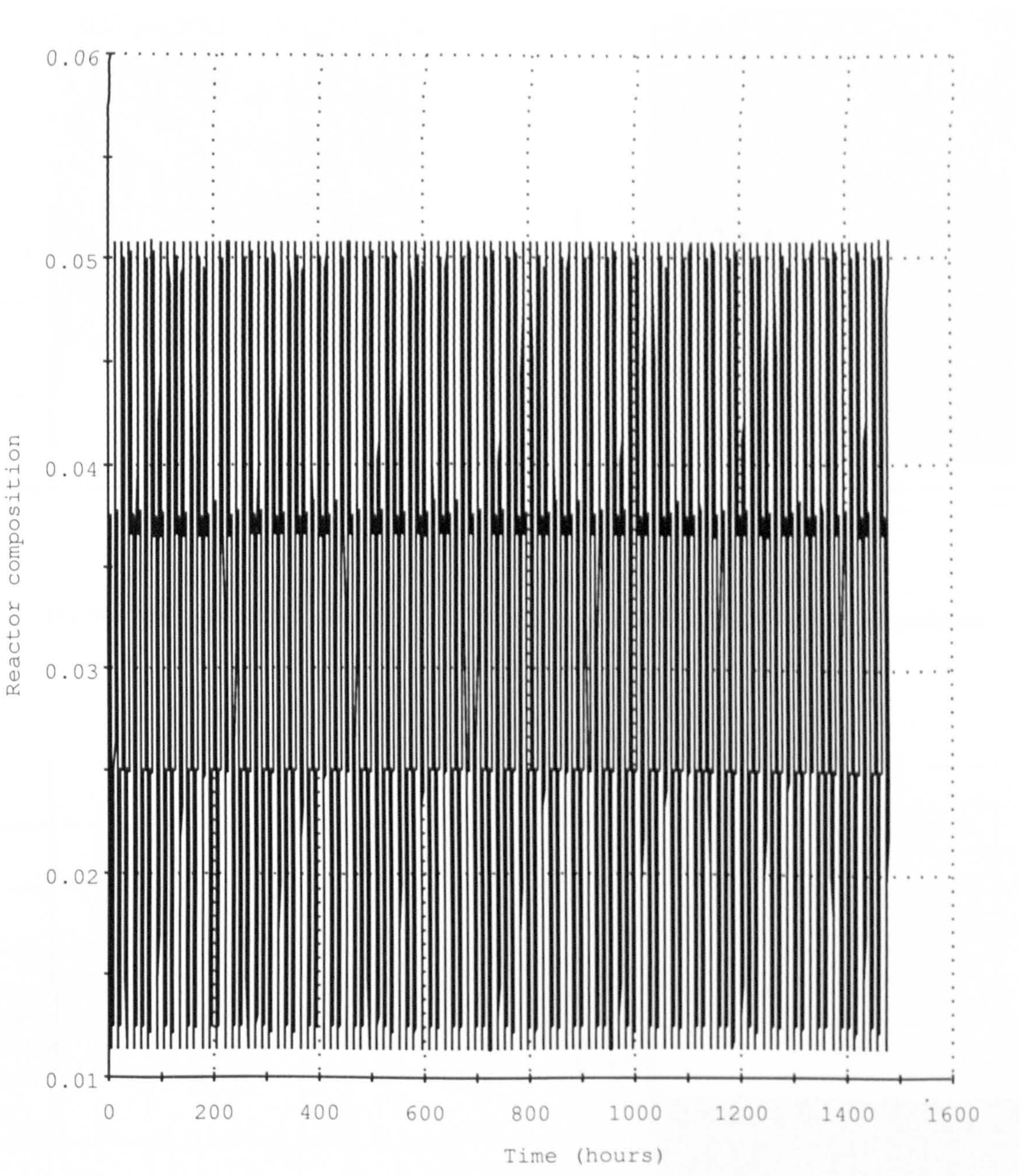
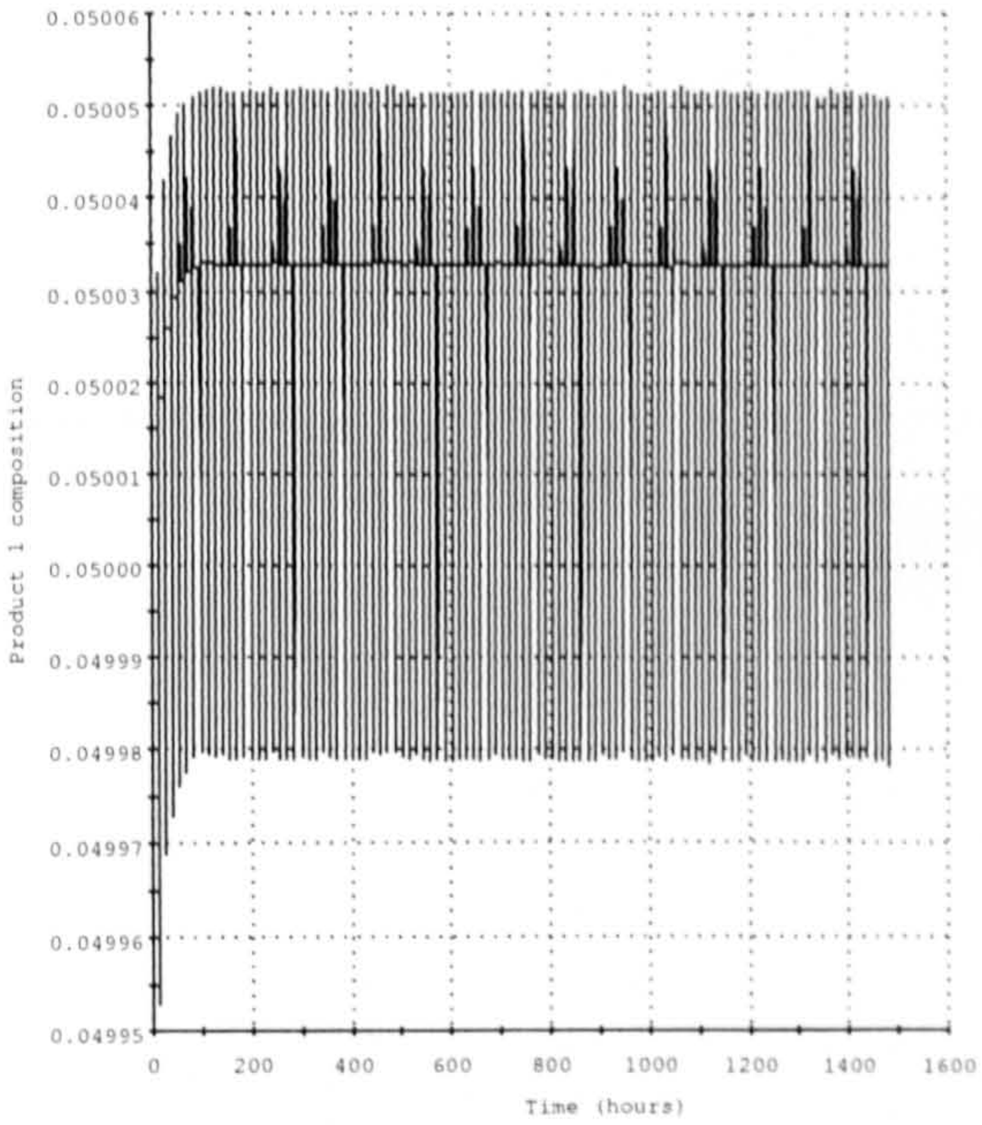
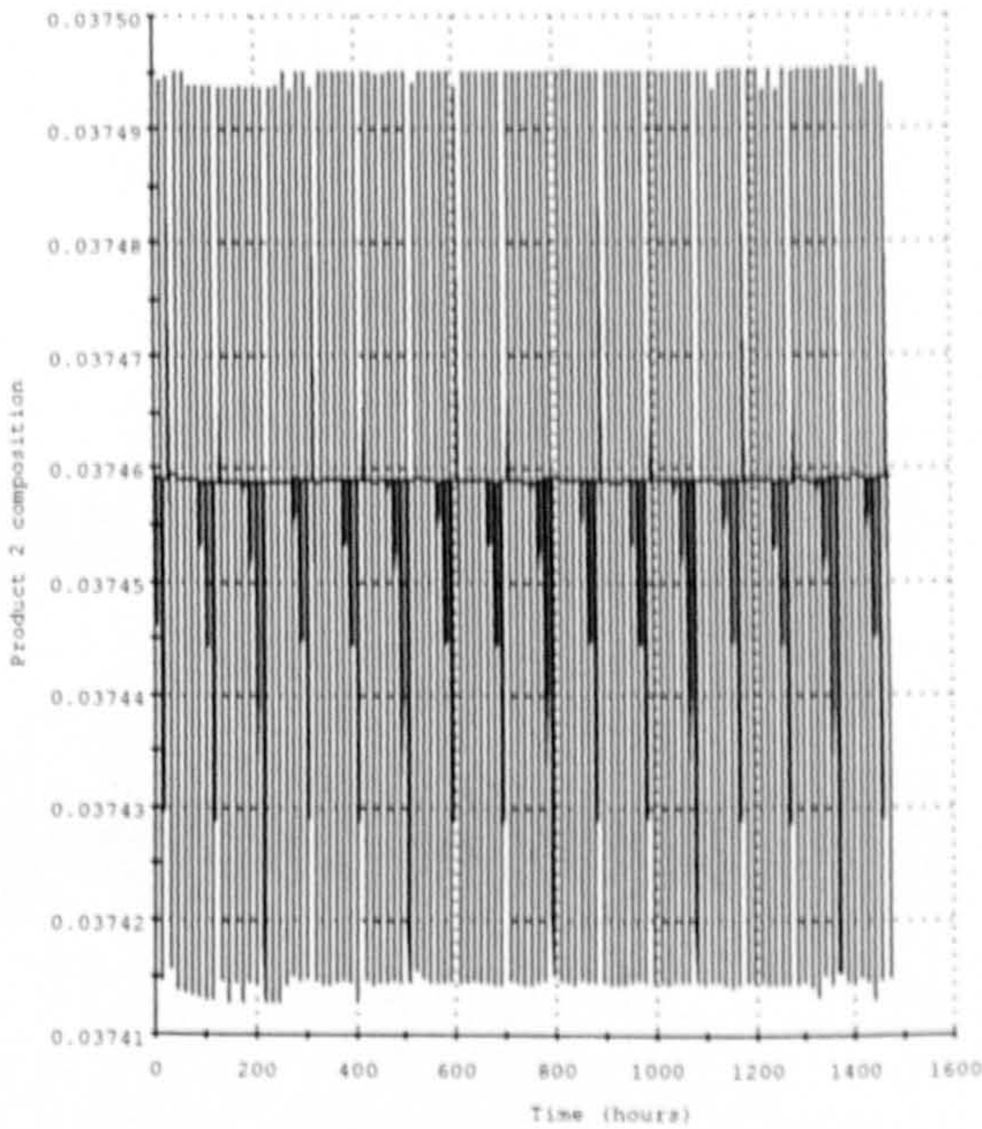


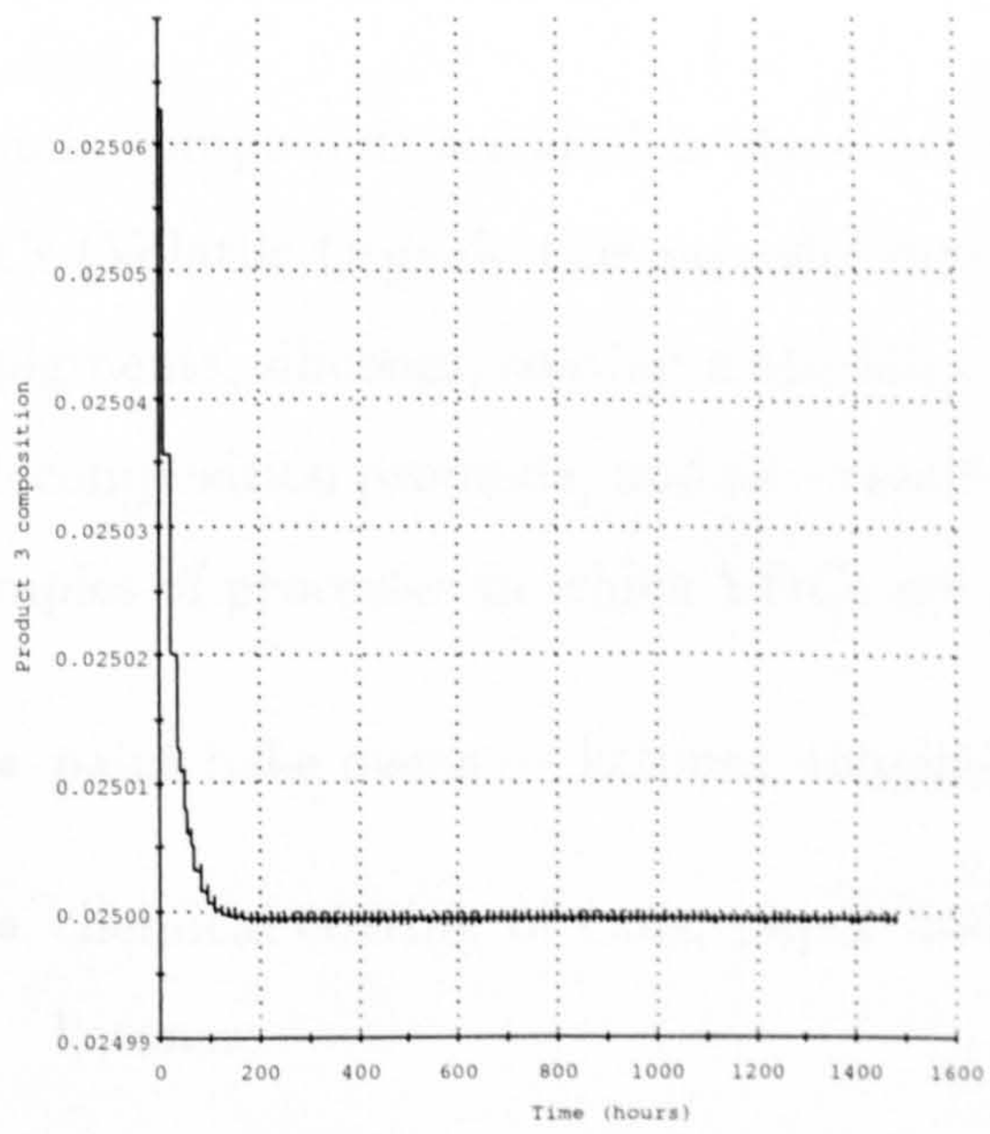
Figure C.11: Reactor composition simulated for 100 cycles using results from the 10 cycle case



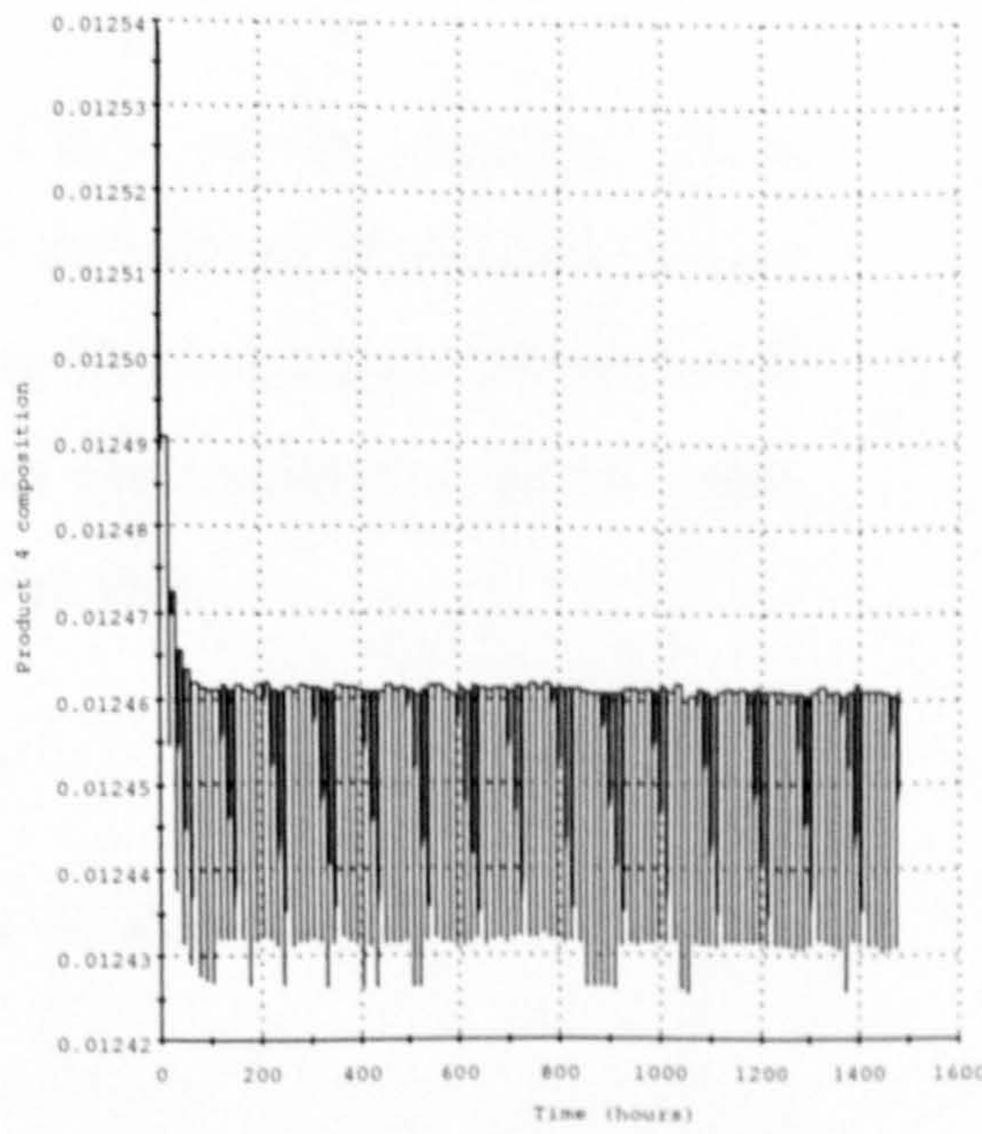
(a) Storage tank 1 composition



(b) Storage tank 2 composition



(c) Storage tank 3 composition



(d) Storage tank 4 composition

Figure C.12: Storage compositions simulated for 100 cycles using results from the 10 cycle case

Appendix D

Catalytic abatement of hydrocarbons

D.1. Introduction

Organic compounds are used in the manufacture of most consumer goods. These VOCs (Volatile Organic Compounds) may be used as solvents serving as carriers for pigments, silicones, coating materials etc., or are found as unreacted feedstock or decomposition products, and as a result appear in plant effluent streams. Some examples of processes in which VOCs are emitted include:

- paint bake ovens — ketones, toluene, xylene;
- chemical coating of cans, paper and fabrics — alcohols, phenols, toluene, ketones;
- metal coating operations — ketones, toluene, xylene;
- wire enameling operations — phenols, xylenols and methylated benzene;

- chemical plants — formaldehyde, phthalic anhydride, acrylonitrile;
- printing — toluene, n-hexane, ethyl acetate, propanol;
- textile plants — alcohols, phenol, toluene;
- electronic plants — cellulose acetate butyrate and alkanes.

The generalised reaction for the catalytic oxidation of VOCs is

$$HC + O_2 \rightarrow CO_2 + H_2O \qquad \Delta H \ -ve \qquad \text{(D.1)}$$

The heat generated by this exothermic reaction may be used to preheat the incoming pollutant-laden gas. It also effects a steep conversion versus temperature curve; the region in which conversion increases rapidly with increasing temperature is referred to as the "light-off" temperature. As can been seen from Table D.1 and recent modeling work by Sawyer and Abraham (1995), the light-off temperature varies inversely with hydrocarbon reactivity; reactivity for oxidation in turn decreases in the order

$$alcohols > ethers > aldehydes > alkenes > aromatics > ketones > esters > alkanes$$

Reactor design for oxidation of dilute hydrocarbon effluents is reasonably straightforward, as the reaction is usually mass-transfer controlled. Thus the reaction rate is simply

$$r = k_m(C_{HC})_b \tag{D.2}$$

where k_m is the mass-transfer coefficient which may be calculated from correlations for monolithic catalysts (Hegedus, 1973). Diffusivities of hydrocarbons in air or N_2, used in these correlations, can be calculated from the Chapman-Enskog formula (Smith, 1981).

Further information on catalytic oxidation of VOCs can be found in reviews by Jennings et al. (1985) and Chu and Windawi (1996), and in a guide book edited by Bertelsen (1992). The technical content for this proposal was taken from "Fundamentals of Industrial Catalytic Processes" (Farrauto and Bartholomew, 1997).

D.2. Motivation

A 90% reduction in the emissions of 189 toxic chemicals is called for by the 1990 Clean Air Act, of which 70% are VOCs. As the VOC concentration in plant effluent streams is usually well below the minimum flammability limit, sizeable amounts of additional fuel are required to reach temperatures sufficient for the non-catalytic thermal incineration of the VOCs ($500 - 800°C$). The use of catalysts for the oxidation of VOCs (typically alumina-supported Pt and Pd, coated on metal or ceramic monoliths) offers a considerable utility advantage by lowering the operating temperature to $150 - 400°C$ (see Table D.1). In addition to energy savings, the lower costs of construction and reactor materials are an economic benefit. Hence catalytic removal is often the choice for abatement (Spivey, 1987; Noordally et al., 1993; Freidel et al., 1993).

Molecule	*Catalytic, Pt/Al_2O_3 catalyst (°C)*	*Thermal (°C)*
Benzene	200	560
Toluene	200	480
Formaldehyde	150	430
Ketone	250	515
Propane	410	500

Table D.1: Temperatures for initiation of oxidation

Zeneca is known to use catalytic oxidation units for VOC abatement. After a discussion with a manager, he described them as "huge and troublesome to control". A "responsive" version would ideally remedy these problems by being smaller and more versatile. Incidentally, Rowe (1997) used distributed manufacturing as a motivation for responsiveness. A more controllable and economically attractive design creates the possibility of replacing a single massive monolith reactor into which all effluent lines must flow, with several smaller units each attached to a single line, fulfilling Rowe's vision.

D.3. The design problem

Recall, a responsive plant is defined as being capable of responding *instantly* to frequent and major changes in customer demand for product quantity and/or quality. In this case, the problem is somewhat reversed. There is no "customer" except the atmosphere, which has no demands apart from imposing product composition constraints (Clean Air Act, 1990). Instant response to the supplier is now needed: it is envisaged that as soon as some critical concentration of VOC is exceeded in the buffer before the catalytic oxidation unit, the reactor must be turned on (i.e. the temperature needs to be raised to the light-off temperature of the predominant species about to enter). As soon as the VOC concentration in this buffer falls below an acceptable background level, the reactor is turned off.

Feasible and plausible process and control structures that can "automatically" satisfy the supplier must be developed using an integrated design methodology. The optimal cyclic operation of such a facility must also be determined. Some challenges of this design problem are highlighted below:

- to determine how small the preceding buffer can be,
- to select the optimal temperature profile for operation,
- *to find the* level that the reactor concentration must fall to at shutdown, in order to compensate for *the time delay incurred* later during startup as the temperature is raised from ambient to "light-off", *whilst satisfying the* outlet composition constraint.

D.4. Reactor design

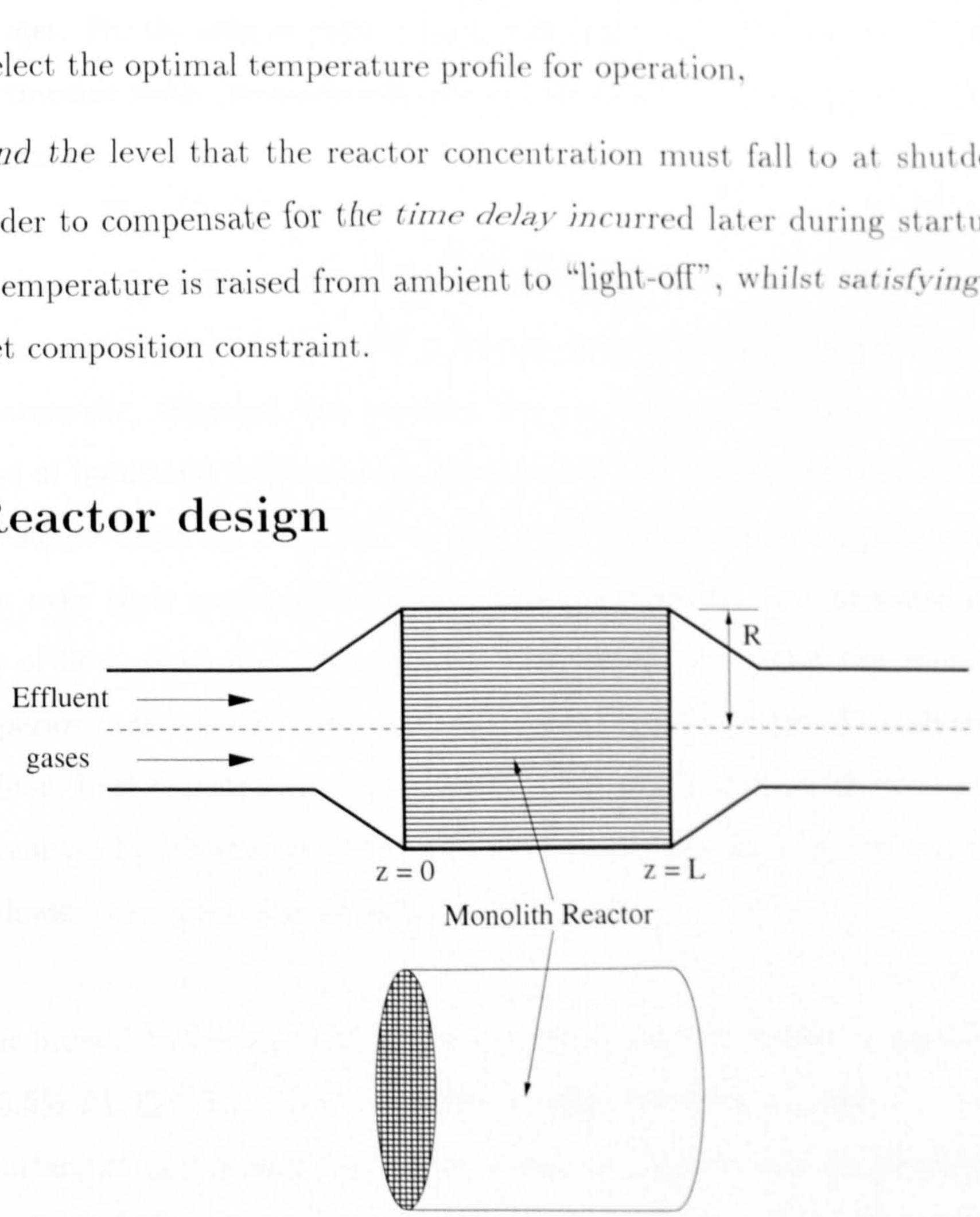


Figure D.1: Catalytic combustion system

Heterogeneous catalysis of VOCs is typically carried out in *monolith* reactors. Cybulski and Moulijn (1994) provide a general review covering various aspects of

these monoliths: structural characteristics/configurations, catalyst descriptions, gas phase reaction applications, and catagorisation of reactor models available from the literature. Monolithic catalysts are described as continuous unitary structures which contain many small, mostly parallel passages. A ceramic or metallic support is coated with a layer of material in which active ingredients are dispersed. The catalytically active material is present on or inside the walls of these passages. For the sake of simplicity, it is assumed that "monolithic" reactors have impermeable walls (reactors with permeable walls are categorised as "membrane" reactors).

Monolithic catalysts have recently found many applications in combustion, e.g. as afterburners of engine exhausts and in the combustion of fuels for gas turbines, boilers, heaters etc. However, the focus of this work will be on their role in the incineration of industrial off-gases containing unreacted VOCs. Investigations on monolith reactors have been carried out since the late 60s exploiting their many advantages over their conventional fixed-bed counterparts: low pressure drop, uniformity of flow, mass and heat transfer conditions throughout the monolith, and far superior heat transfer characteristics (when metal-supported catalysts are used). Indeed, heat transfer was improved to such an extent that the concentration of unreacted hydrocarbons in the effluent from a monolith reactor was up to five times lower than from packed beds.

Metallic monoliths are made of stainless steel (Cament) or steel alloys such as Kanthal (5.5% Al, 22% Cr, 0.5% Co) and Fecralloy (0.5-12% Al, 20% Cr, 0.1-3% Y). The surface of aluminium containing alloys is made Al-rich to improve the adherence of the alumina layer with which the support is covered. Monoliths made of foils of thickness 50-125μm are commercially available. The refractoriness of metallic supports is lower ($T_{max} \simeq 1670K$) than that of ceramics ($T_{max} \simeq 2470K$). In contrast, the thermal conductivity of metals is far higher than that of ceramics.

Due to the high voidage, the mass of metallic monoliths and their thermal capacity are lower than for ceramic monoliths. Therefore the ignition of a reaction mixture proceeds faster in metallic monoliths, increasing their potential for responsiveness.

D.4.1. Dynamic model of a monolith channel

Many of the models found in the literature are one-dimensional and only include steady state analysis. Obviously, due to the nature of this work, transient operation must be modelled. Although 1-D models are easy to implement and are computationally inexpensive, their disadvantages are many:

- it is impossible to calculate mass and heat transfer coefficients from simulation,
- it is often necessary to assume fully developed thermal flow (especially in the entrance region and during transient operation),
- the assumption of constant radial temperature, concentration and velocity profiles is dubious considering the heterogeneous reaction occurring at the wall.

A 3-D model would portray a non-circular cross-sectional passage most realistically but would be computationally prohibitive and probably not justified in terms of additional accuracy. A 2-D model representing monolith channels as cylinders of the same hydraulic diameter as the channels is deemed a suitable compromise. The model presented in this work is adapted from Hayes et al. (1992) and considers both heterogeneous and homogeneous reactions and the effects of radiation and conduction in the monolith support.

Component material balances:

$$\frac{\partial(\rho Y_i)}{\partial t} = - \rho V_z \frac{\partial Y_i}{\partial z} + \frac{\partial}{\partial z}\left(D\rho \frac{\partial Y_i}{\partial z}\right) + \frac{1}{r}\frac{\partial}{\partial r}\left(r D\rho \frac{\partial Y_i}{\partial r}\right) + \nu_i R_h \qquad i = 1 \ldots c \quad \forall\, z \in (0, L),\ r \in (0, R) \tag{D.3}$$

Boundary conditions:

$$Y_i = Y_{i,in} \qquad i = 1 \ldots c \quad @\ z = 0,\ \forall\, r \in (0, R) \tag{D.4}$$

$$\frac{\partial Y_i}{\partial z} = 0 \qquad i = 1 \ldots c \quad @\ z = L,\ \forall\, r \in (0, R) \tag{D.5}$$

$$\frac{\partial Y_i}{\partial r} = 0 \qquad i = 1 \ldots c \quad @\ r = 0,\ \forall\, z \in [0, L] \tag{D.6}$$

$$D\rho \frac{\partial Y_i}{\partial r} = \nu_i R_w \qquad i = 1 \ldots c \quad @\ r = R,\ \forall\, z \in [0, L] \tag{D.7}$$

Energy balance:

$$\rho C_{P_f} \frac{\partial T}{\partial t} = - \rho C_{P_f} V_z \frac{\partial T}{\partial z} + \frac{\partial}{\partial z}\left(k_f \frac{\partial T}{\partial z}\right) + \frac{1}{r}\frac{\partial}{\partial r}\left(r k_f \frac{\partial T}{\partial r}\right) - R_h \Delta H \qquad \forall\, z \in (0, L), r \in (0, R) \tag{D.8}$$

Boundary conditions:

$$T = T_{in} \qquad @\ z = 0,\ \forall\, r \in (0, R) \qquad \text{(D.9)}$$

$$\frac{\partial T}{\partial z} = 0 \qquad @\ z = L,\ \forall\, r \in (0, R) \qquad \text{(D.10)}$$

$$\frac{\partial T}{\partial r} = 0 \qquad @\ r = 0,\ \forall\, z \in [0, L] \qquad \text{(D.11)}$$

$$k_f \frac{\partial T}{\partial r} = -Q_{acc} + Q_{gen} + Q_{con} + Q_{rad} \qquad @\ r = R,\ \forall\, z \in [0, L] \qquad \text{(D.12)}$$

$$Q_{acc} = \delta \rho_w C_{P_w} \frac{\partial T}{\partial t} \qquad @\ r = R,\ \forall\, z \in [0, L] \qquad \text{(D.13)}$$

$$Q_{gen} = (-\Delta H) R_w \qquad @\ r = R,\ \forall\, z \in [0, L] \qquad \text{(D.14)}$$

$$Q_{con} = k_w \delta \frac{\partial^2 T}{\partial z^2} \qquad @\ r = R,\ \forall\, z \in [0, L] \qquad \text{(D.15)}$$

$$Q_{rad} = 0, \text{ neglected for now} \qquad @\ r = R,\ \forall\, z \in [0, L] \qquad \text{(D.16)}$$

Assuming ideal gas behaviour, the density for the fluid phase is given by:

$$\rho = \frac{P}{R_g T} \qquad \forall\, z \in [0, L], r \in [0, R] \qquad \text{(D.17)}$$

A fully developed laminar velocity profile is assumed to exist at the entrance and is maintained throughout the reactor, subject only to changes in density:

$$V_z = 2\overline{V}_{z,in} \left\{ 1 - \left[\frac{r}{R} \right]^2 \right\} \frac{T}{T_{in}} \qquad \forall\, z \in [0, L], r \in [0, R] \qquad \text{(D.18)}$$

Initially, only the reaction between propane (A) and oxygen (B) is considered, occurring homogeneously in the bulk fluid phase and heterogeneously at the wall, to produce carbon dioxide (C) and water vapour (D):

$$C_3H_8 + 5O_2 \Rightarrow 3CO_2 + 4H_2O \qquad \text{(D.19)}$$

Parameter	*Value*
A_w	$2.4 \times 10^5 ms^{-1}$
E_w	$89,791 Jmol^{-1}$
A_h	$2.00 \times 10^7 m^3 mol^{-1} s^{-1}$
E_h	$111,700 Jmol^{-1}$

Table D.2: Reaction rate parameters

The homogeneous reaction rate model is taken from Ahn (1983):

$$R_h = A_h exp\left\{-\frac{E_h}{R_g T}\right\} C_A C_B \qquad \forall\, z \in [0, L], r \in [0, R] \tag{D.20}$$

The heterogeneous reaction rate model is taken from Bennett et al. (1991):

$$R_w = A_w exp\left\{-\frac{E_w}{R_g T}\right\} C_A \qquad @\ r = R,\ \forall\, z \in [0, L] \tag{D.21}$$

The concentration for each component is simply the product of their respective molefraction and the density:

$$C_i = \rho Y_i \qquad i = 1 \ldots c \quad \forall\, z \in [0, L], r \in [0, R] \tag{D.22}$$

Initial conditions

$$Y_A = 0.0 \qquad \forall\, z \in (0, L), r \in (0, R) \tag{D.23}$$

$$Y_B = 21.0 \qquad \forall\, z \in (0, L), r \in (0, R) \tag{D.24}$$

$$Y_C = 0.0 \qquad \forall\, z \in (0, L), r \in (0, R) \tag{D.25}$$

$$Y_D = 0.0 \qquad \forall\, z \in (0, L), r \in (0, R) \tag{D.26}$$

$$T = 775K \qquad \forall\, z \in (0, L), r \in (0, R] \tag{D.27}$$

Parameter	*Value*
Diameter	1 mm
Wall thickness, δ	0.0735 mm
Density, ρ_w	$1710 kgm^3$
Heat capacity, C_{P_w}	$9.48 + 0.227TJkg^{-1}$
Thermal conductivity, k_w	$0.9558 - 2.09 \times 10^{-4}TWm^{-1}K^{-1}1$

Table D.3: Physical parameters of monolith model

D.4.2. Physical properties

The binary diffusion coefficient of propane in air was calculated using the Chapman-Enskog formula (Bird et al., 1960), which gives:

$$D = D_k \frac{T^{1.5}}{P(0.5 + 9T^{-0.5})} \tag{D.28}$$

The thermal conductivity of the gas was treated as a linear function of temperature, based on the values of air given in Incropera and De Witt (1985):

$$k_f = 0.0165 + 5 \times 10^{-5}T \tag{D.29}$$

The heat capacities of the fluid phase and the enthalpy change of reaction are defined in terms of 4th-order polynomials in temperature and standard enthalpies of formation, with values for the constants taken from Kyle (1984):

$$C_{P_i} = a_i + b_iT + c_iT^2 + d_iT^3 \qquad i = 1 \ldots c \tag{D.30}$$

Component	a	$b \times 10^2$	$c \times 10^5$	$d \times 10^9$	$\Delta h_f \times 10^3 Jmol^{-1}$
Propane (A)	-0.966	7.279	-3.755	7.580	-103.9
Oxygen (B)	6.085	0.3631	-0.1709	0.3133	0.0
Carbon dioxide (C)	5.316	1.4285	-0.8362	1.784	-393.8
Water vapour (D)	7.700	0.04594	0.2521	-0.8587	-242.0

Table D.4: Heat capacity and heat of formation data

$$C_{P_f} = \sum_{i=A}^{D} Y_i C_{P_i} \tag{D.31}$$

The enthalpy change of reaction is based on this heat capacity data and standard enthalpies of formation, also found in Kyle (1984):

$$\Delta H = \sum_{i=C}^{D} Y_i \left(\Delta h_{f,i} + \int_{T_{ref}}^{T} C_{P_i}(T) dt \right) - \sum_{i=A}^{B} Y_i \left(\Delta h_{f,i} + \int_{T_{ref}}^{T} C_{P_i}(T) dt \right) \tag{D.32}$$

D.5. Nomenclature

Symbols

A_h	Pre-exponential factor, homogeneous reaction	$m^3 mol^{-1} s^{-1}$
A_w	Pre-exponential factor, heterogeneous reaction	ms^{-1}
A, B, C, D	Propane, oxygen, carbon dioxide, water vapour	–
C_i	Concentration of component i	$molm^{-3}$
C_{P_f}	Mean specific heat capacity of the fluid phase	$Jmol^{-1}K^{-1}$
C_{P_i}	Specific heat capacity of pure component i	$Jmol^{-1}K^{-1}$
C_{P_w}	Specific heat capacity of the wall	$Jkg^{-1}K^{-1}$

D	Molecular diffusion coefficient	m^2s^{-1}
D_k	Diffusion equation constant (2.135×10^{-4})	$kgms^{-3}K^{-1.5}$
E_h	Activation energy for homogeneous reaction	$Jmol^{-1}$
E_w	Activation energy for heterogeneous reaction	$Jmol^{-1}$
$\Delta h_{f,i}$	Enthalpy of formation of component i at 298 K	$Jmol^{-1}$
ΔH	Enthalpy of reaction	$Jmol^{-1}$
k_f	Thermal conductivity of the gas	$Wm^{-1}K{-}1$
k_w	Thermal conductivity of the monolith support	$Wm^{-1}K{-}1$
L	Length of the monolith channel	mm
P	Pressure	Pa
r	radial coordinate	mm
R	Radius of the monolith channel	mm
R_g	Universal gas constant (8.314)	$Jmol^{-1}K^{-1}$
R_h	Rate of the homogeneous reaction	$molm^{-3}{}_s{}^{-1}$
R_w	Rate of the heterogeneous reaction	$molm^{-3}{}_s{}^{-1}$
t	Time	s
T	Temperature *of the gas*	K
T_{in}	Inlet temperature of the gas	K
V	Velocity	ms^{-1}
V_z	Axial velocity	ms^{-1}
$\overline{V}_{z,in}$	Average inlet velocity	ms^{-1}
Y_i	Gas phase mole fraction of component i	–
$Y_{i,in}$	Inlet gas phase mole fraction of component i	–
z	Axial coordinate	mm

Greek Symbols

δ	One-half the wall thickness between channels	mm
ν_i	Stoichiometric coefficient of component i	–

ρ	Gas density	$molm^{-3}$
ρ_w	Density of the support	kgm^{-3}

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