Qualitative Reasoning Methodology for the Generation of Process Plant Operating Procedures

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The candidate confirms that the work submitted is his own and that appropriate credit has been given where reference has been made to the work of others.

"This is not the end.

It is not even the beginning of the end.

But it is, perhaps, the end of the beginning."

Winston Churchill

To my husband,

whose love, patience and dedication

made the unbearable bearable,

and without whom all my efforts

would have been meaningless.

Abstract

The analysis of operating procedures in the early stages of design can lead to safer and higher performance plants. Qualitative reasoning techniques hold considerable promise in supporting generations of operating procedures, since they are able to describe possible trajectories of a system based on non-quantitative information and provide explanation about process behaviour in a way which gives insight into the underlying physical processes. Despite this potential, existing techniques still present limitations related to the tendency for generating non-real behaviour patterns and the inability to describe distributed parameter systems.

This study presents a qualitative reasoning methodology, weighted digraph (WDG) approach, for describing the dynamics of complex chemical processes, and in particular of distributed parameter systems, with a considerable reduction in the generation of spurious solutions. It is based on a generalisation of the signed digraph approach and retains its main advantages, such as the ability to easily represent intuitive and causal knowledge and a graph structure which makes apparent the flow of information between variables. In addition, it incorporates several new features, making use of *functional weighting*, *differential nodes* and *temporal edges*, which enable the procedure to qualitatively describe complex patterns of behaviour.

The effectiveness of the approach is demonstrated by considering the qualitative modelling and simulation of the dynamic behaviour of several chemical processes: heat-exchanger, CSTR with and without temperature control and distillation column.

The proposed weighted digraph approach is used to support generation of start-up procedures with reference to two case studies: a network of heat-exchangers and an integrated system composed of a CSTR and a feed/effluent heat-exchanger. It is shown that the digraph based strategy has the ability to generate feasible operating procedures in the presence of operational constraints and identify the need for modifications of the process topology in order to allow the start-up of the system.

Results also indicate that work is still needed in order to further improve the methodology and create an interactive computer based interface to help with reasoning about complex patterns of behaviour.

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Nomenclature

A	heat transfer area, m^2.
A_{cs}	cross-sectional area, m^2.
B	bottom product flow rate, mol/s.
C_a	concentration of component A, kgmol/m^3.
C_{ao}	inlet concentration of component A, kgmol/m^3.
C_p	heat capacity, kJ/kg K.
D	top product (distillate) flow rate, mol/s.
E	activation energy, kJ/kgmol.
F	flow rate, m3/h.
F_f	feed flow rate, mol/s.
F_j	jacket flow rate, m3/h.
F_{jo}	jacket inlet flow rate, m3/h.
F_L	liquid feed flow rate, mol/s.
F_o	inlet flow rate, m3/h.
F_V	vapour feed flow rate, mol/s.
H	enthalpy, kJ/kgmol.
K_{fl}	gain of the proportional controller, m^3/h K.
K_i	equilibrium constant for component i.
k_o	pre-exponential factor, h^{-1}.

l	length, m.
L	liquid flow rate, mol/s.
L_T	level, m.
M	liquid hold-up, m.
O_i^j	partial derivative of x_i (or δx_i) with respect to x_j.
Q	heat transfer rate; kJ/h, J/s.
R	perfect-gas constant, kJ/kgmol K.
r_a	reaction rate, kgmol/m^3 h.
R_f	reflux flow rate, mol/s.
S	constant.
s_{ij}	sign of the edge from x_i to x_j.
T	temperature, K.
t	time; h, s.
T_f	feed temperature, K.
T_j	jacket temperature, K.
T_{jo}	jacket inlet temperature, K.
T_o	inlet temperature, K.
U	overall heat transfer coefficient, kJ/h K m^2.
V	vapour flow rate, mol/s.
V_j	volume of coolant in the jacket, m^3.
V_R	volume of reactor, m^3.
w	*weight*.
$x_{1,2 \text{ or } 3}$	mole fraction of components 1, 2 or 3 in the liquid phase.
x_i	algebraic variable.
$y_{1,2 \text{ or } 3}$	mole fraction of components 1, 2 or 3 in the vapour phase.
$z_{1,2 \text{ or } 3}$	mole fraction of components 1, 2 or 3 in the feed.

Greek Letters

Δ	change of the value or state of a variable.
δ	qualitative change of a variable with time.
δx_i	differential variable.
λ	heat of reaction, kJ/kgmol *A*.
θ	qualitative state of a variable.
ρ	density, kg/m^3.
ψ_i	represents a generic variable x_i or δx_i.

Superscripts and Subscripts

BT	bottom.
C	cold fluid.
cd	condensate.
CD	condenser.
Ci	inlet of cold fluid.
Co	outlet of cold fluid.
cw	cooling water.
D	distillation column.
E	heat-exchanger.
f	feed.
FT	feed tray.
H	hot fluid.
Hi	inlet of hot fluid.
Ho	outlet of hot fluid.
hw	hot water.
j	jacket (for CSTR).
L	liquid.

N	number of nodes or variables.
R	reactor.
RB	reboiler.
RS	rectifying section.
sp	set point.
ss	steady-state.
SS	stripping section.
T	tank.
t	time.
V	vapour.
vp	steam.

Chapter 1

Introduction

1.1 Process Engineering Environment

Process design is the art of building process systems to convert raw materials into desired products, while meeting prescribed specifications and satisfying economic and environmental constraints.

The design of large chemical plants call for a merging of a range of engineering disciplines, each of which produces a complex network of information that is used by each of the engineering groups in a different way and passed on to the others. The dramatic improvement in computer technology during the last decades has stimulated the generation of ever larger amounts of information that have to be interpreted and interchanged between the design teams. The variety of type of information that includes complex data structures and relationships associated with multiple solutions and conflicting goals, adds to the difficulty in finding solutions, transferring information and communicating decisions. Recently, efforts have been made to create a support system that can facilitate the manipulation of information and group decision-making. The motivation is to build an integrated concurrent process engineering environment to replace the conventional sequential approach.

It is intended to encompass all aspects of conceptual and detailed design, as well as construction, and the emphasis is on the consideration of all phases of the product life-cycle, from initial concept to disposal, including operating procedures at the design stage.

The traditional sequential design approach often requires many iterations that involve successive revisions of previous design parameters and/or structures in order to meet all design criteria (Lin, 1991). Design problems are divided into unconnected pieces and the design process evolves in a sequential procedure. In the philosophy of the concurrent process engineering design the multitude of multidisciplinary design activities are carried out integrated and in parallel to meet the multiple criteria involved in process design (Jiang, 1994). Data and information are easily shared by the design team and lateral communication and cooperative tasks are encouraged. The goal is to increase design productivity and quality and reduce the product development cycle. The general philosophy has been discussed by McGreavy (1985) and Lu *et al.* (1994), and demonstrated by Wang *et al.* (1994) through an application to the design of a fluid catalytic cracking unit. The authors emphasise the need to approach design through an interactive environment which includes a set of numerical and non-numerical tools for graphical representation, scientific visualisation and database management, and also provides platforms for knowledge representation and building expert systems. The mixing of various techniques offers potential for flexible integration of the multitude of engineering design tasks resulting in better quality designs. In such context, the tools from artificial intelligence tailored to model knowledge and deal with non-numerical information, such as artificial neural networks and qualitative reasoning, have a very important role to play. Artificial neural networks can handle non-linear problems (McGreavy *et al.*, 1994; Hashimoto *et al.*, 1994), qualitative information and non-continuous data, and simulate the inverse of a process (Hunt and Sbarbaro, 1991; Guimarães, 1992). They adopt an empirical learning procedure that leads to a non-linear functional

approximation of the process (Stephanopoulos and Han, 1994). Another advantage of artificial neural networks is the speed of computation which is a crucial issue in the integrated concurrent process engineering environment (Guimarães and McGreavy, 1995). This technique has been intensively applied to process identification, fault diagnosis, data rectification, among others. A reference book on this subject has been published by Rumelhart and McClelland (1986).

Qualitative reasoning is a technique created to cope with qualitative knowledge, i.e. knowledge that is difficult to quantify and represent by conventional mathematical methods. Although it has been experimented in several areas, such as control of non-linear processes and dynamic analysis of processes, its main application is still related to fault diagnosis. There have been some attempts in applying it to the synthesis of operating procedures and explanation of quantitative solutions generated by numerical simulators. However, the limitations of the existing techniques, mainly related to their inability to describe dynamic behaviour of complex chemical processes, such as distributed parameter systems, and the generation of large amounts of ambiguous solutions, have hindered its widespread use.

1.2 Process Plant Operations

The design and operation of process plants are increasingly constrained by safety and economic factors which have to be satisfied during the process life-cycle. This associated with frequent changing economic policies has stimulated the design of highly integrated processes, which are intrinsically more dynamic than previous conceptions and subject to frequent changes between steady-states. This leads to the need to consider operating procedures at the design stage in order to achieve a safer and higher performance plant.

Process plant operations is a wide definition that encompasses commissioning, start-up, normal operation, process change-over, emergency fall-back and shut-down, each of which is characterised by different goals. The synthesis of operating procedures is directed to find a sequence of actions to be performed by plant operators or computers in order to lead the process system from an initial state to the goal state.

Although computational power has been largely explored in design and control, the use of computers to support comprehensive methodologies for generating operating procedures has not yet been explored to the same extent. This is due to the fact that planning, scheduling and implementation of chemical plant operations are heavily based on non-quantitative information from heuristic knowledge of human operators and experience of the designer, which are very difficult to translate into computer programs and cannot be handled by conventional mathematical procedures. This is even more critical in the early design stages when detailed numerical information about model parameters is not yet generally available or accurate enough to allow numerical simulations.

The operational objectives of chemical processes may be *local*, involving a particular processing unit, or *global*, involving several processing units. *Global* objectives include the consideration of production requirements, optimum economic operation, safety in the presence of faults or other disturbances and flexibility for start-up, shut-down or change-over (Stephanopoulos, 1987). All these have to comply with heavy environmental regulations and technological constraints. The multi-objective character of plant operations associated with the lack of appropriate supporting procedures makes it a task essentially dependent on empirical methods and personal skills. Therefore it is by no means a trivial and well defined problem, which has tended to become even more complex due to the increasing restriction on the availability of raw materials and energy. These have stimulated the

formulation of highly integrated topologies with better energy management and the consideration of more recycles to reduce waste of raw materials. As a consequence, the units have become tightly coupled, allowing for strong interactions and acute operational problems (Stephanopoulos, 1983).

There is a growing interest in the process industries for systematic procedures for the analysis of process dynamics in order to achieve a high quality design and efficient plant. However, despite this trend, the conceptual design of the process flowsheet and its optimisation aiming at maximum plant efficiency and minimum capital costs is still mainly based on information of steady-state operation.

Steady-state analysis may miss important transient responses of the system which can affect the controllability and operability of the process or even have disastrous consequences during start-up and shut-down, since these are usually the most hazardous plant operations. Although they are not usually frequent operations in continuous plants, they involve drastic and complex process variations that can potentially exceed equipment design conditions and so threaten the integrity of the plant. Moreover, control is manual rather than automatic and trip systems are often disarmed or bypassed and this makes them intrinsically more dangerous operations.

Dynamic simulation at the design stage is important when comparing different design alternatives for control and operational policies, as well as in identifying potential operating problems. Poor characterisation of the dynamic behaviour of a process during the design stage and inadequate assessment of process performance during start-up and shut-down can give rise to difficulties during plant operation. This can considerably increase project costs, since it may be necessary to make modifications to prevent unstable or unsafe operations, or even to allow the start-up. As plant efficiency is a direct consequence of smooth plant operations, including a successful start-up with minimum loss of raw materials, energy consumption and production of off-specification products (Scott and Crawley, 1992), process

dynamics analysis is essential for achieving good quality designs. Up to now the main focus of dynamic simulation has been on the use of conventional quantitative approaches.

It is now commonly the case that the analysis of plant operations comes during the construction of the piping and instrumentation diagram (P&ID) along with analysis of control strategies. The procedure has tended to be based on experience and heuristics and involves: (1) assumption of a policy for plant operation, (2) design of additional facilities for the start-up, and (3) evaluation of the performance of the process for the proposed operating procedure. If a policy does not satisfy the *global* operational objectives, another is proposed and the procedure repeated. In extreme cases, it may be necessary to return to the conceptual stage, which considerably increases the costs. Clearly, the earlier plant operations is considered the less the risk in needing changes or the possibility of process failures. Also, aspects related to safe start-up and shut-down, special requirements for equipment size and conditions during these and other critical operations, intermediate storage and the need for auxiliary equipment need to be considered during the conceptual design of the process flowsheet.

Clearly, the synthesis of operating procedures during the design of a plant involves a very complex decision-making process. It is an expensive, time-consuming and error-prone task both with respect to the process engineering specifications of procedures and automation which needs to consider sequence control codes from the process specifications. The interface between these two groups is a further potential source of error, since the boundary is ill-defined and involves people from different backgrounds who make assumptions without adequate understanding of all aspects of the problem (Crooks *et al.*, 1994). This reinforces the need for the integrated concurrent process engineering environment.

An integrated concurrent process engineering environment makes possible analyses of the life-cycle performance of the plant at the design stage. This approach avoids the decomposition of the highly integrated process structures often present in new projects and allows the exploration of operational features of the process, such as stability, flexibility and operability together with the overall structure of the flowsheet while evaluating design alternatives. Interactions between process synthesis, analysis and evaluation not only lead to a more effective way to reach the optimal solution but also allow the assessment and elimination of potentially hazardous situations at the design stage, including those related to start-up and shut-down procedures. In particular it allows the analysis of plant operating procedures at the very early conceptual stage of the process flow diagram (PFD), which can lead to the identification of potential bottlenecks of critical, unreliable and/or ineffective operations when modifications are not so costly.

During the design stage, data are generated and analysed and decisions are made based on interpretations using this information. Flexibility, operability, controllability and safety are largely assessed by subjective interpretation of such data. Generation of feasible operating procedures, which include potentially dangerous situations during start-up and shut-down, are also based on process analysis and identification of dynamic trajectories and patterns of process behaviour. The interpretation of raw data aiming at the assessment of qualitative information can be a very complex task, relying heavily on interpretation skills and visualisation capabilities of graphical outputs. A tool capable of formalising the deep knowledge involved in reasoning about process behaviour would therefore be expected to play a very important role in a design or operational environment.

Early stages in process design are characterised by lack of precise data and information about the system. This makes process analysis based on interpretation of data even more difficult. Qualitative reasoning techniques from artificial intelligence

have shown that little numerical information about system parameters may be required to describe feasible trajectories of the system. This indicates that these techniques hold considerable promise in supporting generation of operating procedures at early stages in process design.

1.3 Qualitative Reasoning as a Basis for Synthesis of Plant Design and Operating Procedures

Engineering is based on the knowledge of the laws of nature, such as mass and energy conservation, which constitute the framework of mathematical models. Developing a model that reflects the essential features of a system is a complex task, and an important problem is to determine the numerical values of the parameters. If these constants are not well defined the set of equations loses its practical sense in terms of the usefulness of the solutions.

Most of the techniques employed for the analysis of engineering tasks are of classical quantitative nature: analytical or statistical. However, these precise formal tools have a limited range of application and do not contribute as much as expected to a better modelling and understanding of highly cognitive tasks, such as fault diagnosis and generation of operating procedures, which are among the most complex and ill-defined of engineering problems.

A process engineer is required to solve problems reliably and in doing so often has to be innovative and imaginative. This particularly applies for ill-defined situations where information is incomplete, imprecise and sometimes inconsistent. A specific solution is built by using an intricate combination of strategies, general knowledge and information and experience. In many cases it is necessary to make a decision without any numerical calculation. During the reasoning process, equations may be used to perform logical deductions rather than calculations. Even when

calculations are used, the interpretation of results, in order to make a decision, is an intuitive process and the reasoning is based on qualitative information extracted from the numerical results (Muratet and Bourseau, 1993). For instance, it is possible to describe the operation of a system by a sequence of events caused by prior actions (cause and effect relations), without the need for complex mathematical models or numerical parameters. This makes use of intuitive knowledge about the causality of the processes to reason about system behaviour, without causality being explicitly expressed in any mathematical model. Another example is the design of large physical systems based not only on accumulated knowledge from previous experience but also on creativity. This is often used to choose between design alternatives or in selecting new technologies, when the designer has no direct previous experience in relation to the system being designed.

The ability to reason with incomplete knowledge and be creative seems to be related to the qualitative understanding of how physical systems work. This is based not only on the knowledge acquired from the set of conservation laws, equilibrium relations and other mathematical models used to describe the system, but also from intuitive knowledge associated to causality, continuity, feedback and so on which represent general principles. Qualitative reasoning is typically used to analyse how effects are propagated, make assessments of relative strength of influences and order-of-magnitude of effects, and neglect those which do not contribute to the understanding of the problem.

The classical approach used in qualitative analysis is based on the analytical solutions of the problem (Dohnal, 1991a). For example, a system described by the differential equation (1.1) has the general solution described by Eq. (1.2) and the auxiliary equation (1.3).

$$\frac{d^2y}{dx^2} + ay = 0 \tag{1.1}$$

$$y = e^{\alpha x}.(C_1.\sin\beta x + C_2.\cos\beta x) \tag{1.2}$$

$$m^2 + a = 0 \quad \Rightarrow \quad m = \sqrt{-a} \tag{1.3}$$

$$\text{if} \begin{cases} a \langle 0 \quad \Rightarrow \quad m = \sqrt{a} \quad \Rightarrow \quad \alpha = \sqrt{a},\ \beta = 0 \\ a \rangle 0 \quad \Rightarrow \quad m = \pm a i \quad \Rightarrow \quad \alpha = 0,\ \beta = a \end{cases}$$

where a, C_1, C_2 , α and β are constants;

x and y are the independent and dependent variables, respectively.

An examination of the solution shows that, depending on the sign of the parameter '***a***', the system may describe different trajectories, as illustrated in Fig. 1.1. This shows that the system has distinctly different qualitative behaviour depending on whether the constant '***a***' is positive or negative. Thus the different patterns of behaviour are divided into two different classes or domains, although the specific responses will depend on the precise numerical value of '***a***' and the initial conditions.

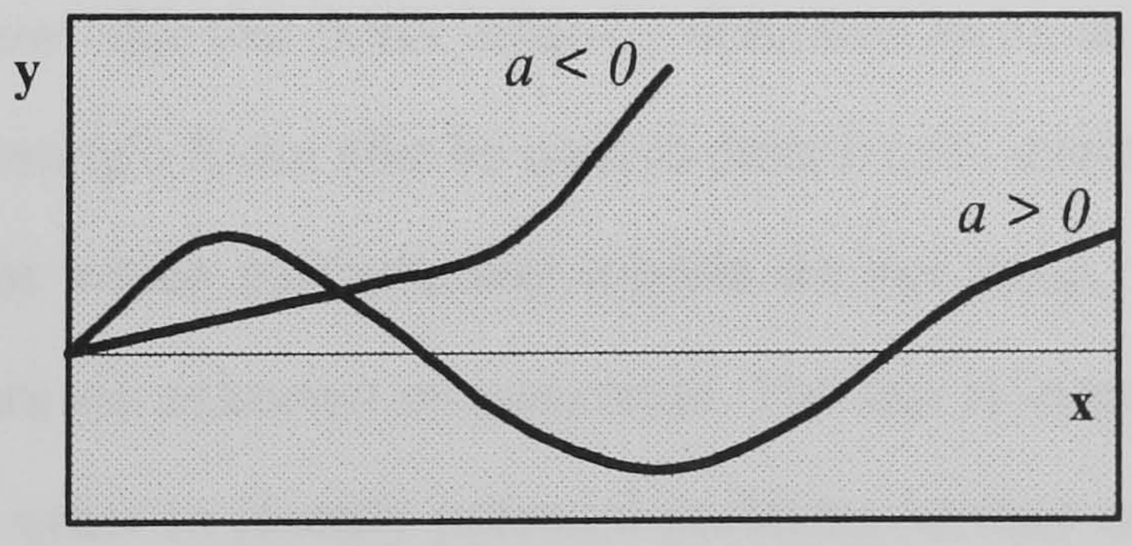

Figure 1.1 Qualitative trajectories of a system described by Eq. (1.1).

Most practical problems in chemical engineering cannot be solved analytically and classified in this straightforward way. Nevertheless, equivalent general criteria apply, i.e. certain parameters will define domains with similar types of functional behaviour exhibiting distinctive features. However, a more flexible tool is needed to

enable similar qualitative analyses to be applied in more complex problem domains where the solutions, or indeed the mathematical models, may be unknown. Such concepts appear to have common links with those being developed in artificial intelligence.

The translation into computer procedures of the reasoning process that people intuitively apply to solve problems is seen as a major challenge of artificial intelligence (Muratet and Bourseau, 1993). In meeting this objective, there is a need to understand how qualitative information can be managed as part of reasoning processes.

Qualitative reasoning techniques from artificial intelligence provide a framework for representing qualitative information and reasoning about aspects of the physical world. The goal is to capture the way people reason about a problem and formalise both the intuitive and other knowledge underlying quantitative calculations. The methodology involves analysis, modelling, qualitative simulation, causal reasoning and qualitative symbolic algebra. It is well suited for modelling intuitive and engineering knowledge that is not naturally described by mathematical equations, e.g. "reactor is operating". It can also be used to describe the qualitative behaviour of physical systems for which mathematical models do not exist, or if they do the numerical parameters are unknown or inaccurate. The aim is to generate all possible system solutions in order to identify potential problems so they can be addressed at an early stage during design. For example, it is important to examine start-up procedures at early stages in process design so that any changes which need to be made can be carried out with minimum cost.

The main motivation of applying qualitative reasoning to chemical engineering problems is related to the need to create a framework to help to understand how information flows through the system in order to be able to reason about process behaviour and explain why it takes a particular form. As pointed out by Grantham

and Ungar (1990) "one cannot ask a numerical simulator why does the temperature of the reactor rise in this simulation". Qualitative reasoning provides a bridge which enables numerical simulation results to be explained. Moreover, it can be used to help carrying out highly cognitive tasks, such as process analysis and data interpretation, supporting design decisions, fault diagnosis and generation of operating procedures. Therefore, qualitative reasoning can be expected to play a very important role in an integrated concurrent process engineering environment.

Figure 1.2 illustrates the relationship between qualitative reasoning and quantitative simulation. It also shows how visualisation, coupled with qualitative reasoning, supports process analysis, design decisions and synthesis of operating procedures.

Qualitative reasoning has been extensively applied to fault diagnosis (Umeda *et al.*, 1980; Oyeleye and Kramer, 1988; Savkovic-Stevanovic, 1995). A smaller number of studies concerned with the description of process dynamics have been reported (Kuipers, 1984,1986; Dalle Molle *et al.*, 1988) but they are mainly directed to lumped parameter systems (described by ordinary differential equations) and cannot be easily used to analyse distributed parameter systems (described by partial differential equations). This is a significant limitation since these systems are frequently found in chemical processes in the form of equipment such as distillation columns or tubular reactors.

Investigations on the use of qualitative reasoning to support generation of operating procedures have already been reported, as for example Fusillo and Powers (1987, 1988) and Hangos *et al.* (1991). However, these approaches present limitations related to either the inability to deal with complex chemical processes or the tendency to generate large numbers of ambiguous and spurious (non-real) solutions which make the problem intractable.

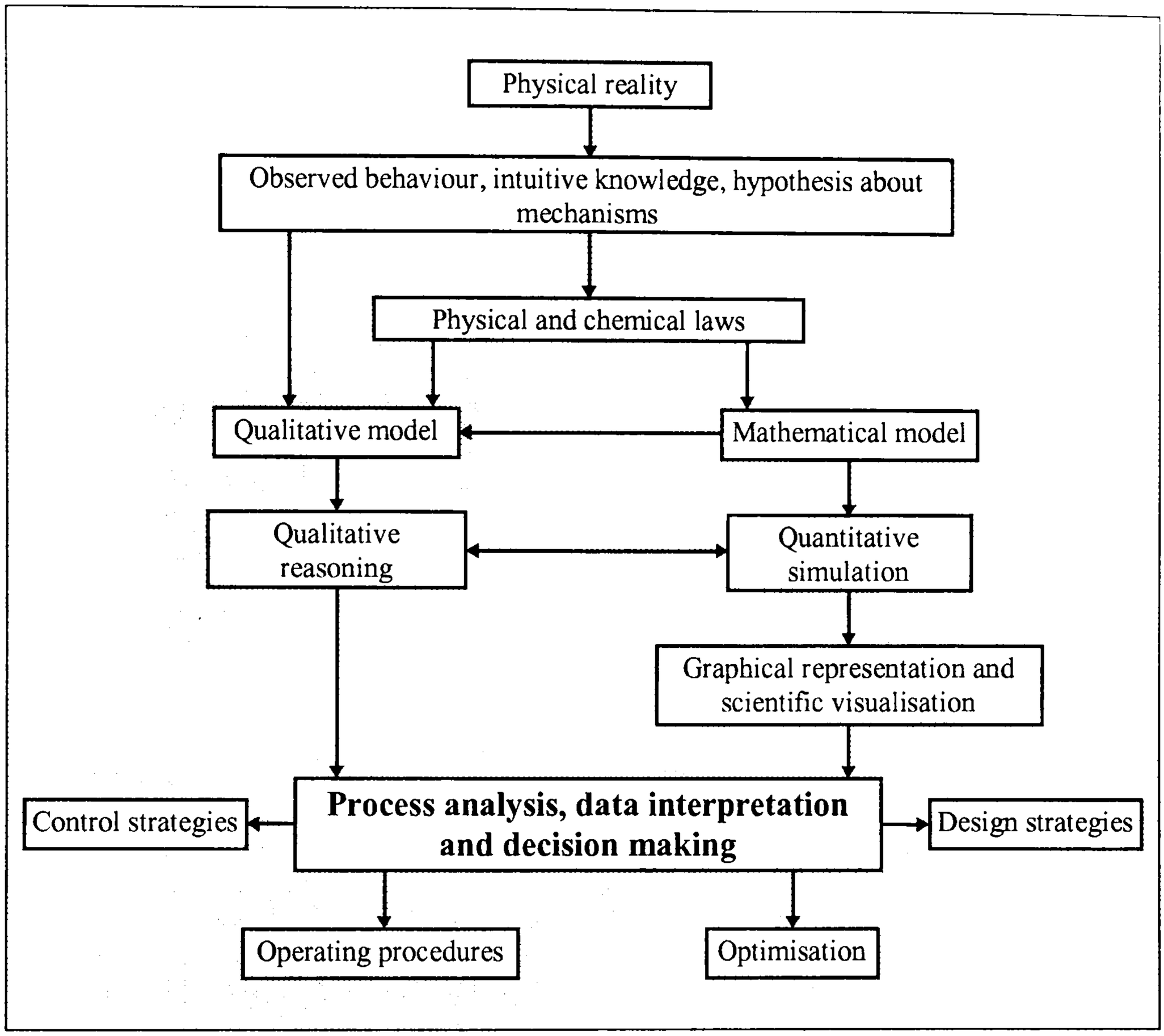

Figure 1.2 Qualitative reasoning in the design environment.

Clearly, there is considerable scope for developing a qualitative reasoning methodology capable of describing transient responses of complex chemical processes, especially for distributed parameter systems. The methodology must be versatile enough to be used for the generation of operating procedures at early stages in process design, when data and information about the system are minimal and mainly qualitative. Such a methodology would form part of a framework to support process analysis and data interpretation, with the aim of improving the conceptual design stage of a process. It should also be capable of explaining operating behaviour and supporting assessment of control strategies. The ability to describe complex

dynamic behaviour is clearly essential when evaluating operating procedures for start-up and shut-down at the early stages of design.

1.4 Research Objectives

Qualitative reasoning techniques from artificial intelligence are used to generate descriptions about possible dynamic trajectories of a system and provide a means of explaining behaviour in a way which gives insight into the underlying physical processes. They can be a very powerful tool in a design environment which, along with good visualisation, can assist in reasoning about the behavioural characteristics of the process. Despite this potential, there are still problems arising from the tendency to generate non-real behaviour patterns and inability to describe distributed parameter systems.

The overall objective of this research is to develop a qualitative reasoning methodology for describing complex patterns of process behaviour, which is sufficiently robust to be used in the early stages of process design to appraise possible operating procedures. In particular, it addresses the problems of distributed parameter systems and generation of ambiguous solutions during qualitative simulation.

To do this it is necessary to extend the set of qualitative states {+,0,-} used by existing approaches and mix qualitative and intuitive knowledge with crude quantitative information of the relative strength of influences, based on observed behaviour or order-of-magnitude analysis of a mathematical model.

The approach is based on using weighted digraphs (WDG) and, in many respects, is analogous to model reduction since it extracts the essential characteristics of system responses and complex systems are built from a minimal set of elemental structures .

To provide a reference base, systems with known solutions are considered in the first instance to build up a set of basic models which can be used to describe more complex systems. This is extended to the development of algorithms for the generation of start-up procedures based on process flow diagrams (PFD). Qualitative models are used not only for finding feasible sequences of operations, but also for describing the dynamic responses of the system.

1.5 Thesis Organisation

This thesis comprises seven chapters and one appendix. Following this introduction which covers aspects related to the need for a qualitative reasoning tool to support synthesis of operating procedures at early stages in process design, Chapter 2 presents a review of the existing qualitative reasoning techniques. In particular the signed digraph approach is discussed in terms of the main characteristics which make it a suitable candidate for devising a new procedure for qualitative reasoning. The limitations are also discussed in terms of coping with process dynamics and the generation of ambiguous solutions. The discussion is illustrated by a case study.

Chapter 3 looks at a detailed description of the proposed weighted digraph (WDG) methodology for qualitative reasoning with nonsteady-state processes. The modelling and simulation algorithm are illustrated and discussed with reference to a simple case study which uses this novel procedure to overcome the main limitations of the conventional signed digraph approach.

Chapter 4 is devoted to the application of weighted digraphs for describing the dynamic behaviour of several chemical processes. It covers the systematic modelling of chemical processes at different levels of complexity, including distillation columns and the multiple thermal steady-states of continuous stirred-tank reactors (CSTR). The aim is to illustrate the effectiveness, functionality and flexibility of the approach

in dealing with process dynamics and the ability to reject spurious solutions, and investigate how these characteristics are related to model elements. Emphasis is given to the way in which particular behaviour patterns arise and information flows through the model, in order to demonstrate how a graphical approach can assist the designer in understanding process behaviour.

In Chapter 5 a review of methodologies for synthesising feasible operating procedures is presented. An algorithm for generation of start-up procedures based on the proposed weighted digraph methodology is described. Aspects related to the qualitative representation of process flow diagrams, specification of qualitative operational constraints and scheduling are emphasised. The procedure involves two steps: (1) sequencing of valve operations and (2) description of dynamic trajectories. In the first step a start-up procedure is generated by using weighted digraph models in steady-state. It looks at the propagation of cause and effects between process variables. The qualitative models are then used in dynamic mode to predict the trajectories of the system using the proposed sequence of operations. The aim is to test the feasibility of the start-up procedure and identify potential bottlenecks related to the process topology. This can be used to determine whether auxiliary equipment may have to be added or process modifications may be required to make it possible to start-up the process.

Chapter 6 evaluates the performance and limitations of the algorithm proposed in the previous chapter. The suitability of the method in predicting the scheduling of valve operations in the presence of qualitative constraints is illustrated and discussed, based on a network of heat exchangers as a case study. The importance and effectiveness of the qualitative reasoning methodology in predicting dynamic trajectories and the need for auxiliary equipment is analysed, based on an integrated process flow diagram composed of a CSTR and a feed/effluent heat exchanger.

Chapter 7 draws together the main topics studied by presenting a summary of the work together with concluding remarks. Suggestions for the direction of future work are also made. The appendix provides supplementary material on numerical data for generating quantitative predictions that serve to assess the effectiveness of the weighted digraph methodology.

Chapter 2

Qualitative Reasoning

2.1 Introduction

The first use of artificial intelligence in process engineering was in the early 1980s with the development of rule-based expert systems. However, the limitations of these kind of tools based solely on associative ("shallow") knowledge were soon revealed. This motivated the development of methodologies for modelling knowledge based on physical and chemical laws. The goal was to capture both intuitive and engineering knowledge from basic principles underlying the process behaviour without having to resort to the simulation of quantitative models. Since only general patterns of behaviour were used, this became known as qualitative reasoning. This provides a framework for intuitive reasoning about process problems in much the same way that individuals do in their everyday life. In particular, the aim is to provide explanations of behaviour and the general nature of the way changes occur when a system is perturbed, making use only of structural information and incomplete descriptions.

In this chapter, a critical review of qualitative reasoning techniques in terms of the advantages and drawbacks from the viewpoint of their application to describe the

dynamics of chemical processes and the ability to assist in generation of operating procedures, is presented. The discussion is intended to set the context of the problem by identifying the limitations of earlier work. A qualitative reasoning technique is chosen to be used as a platform for the formulation of a new approach, and is critically evaluated so as to highlight the problems to be overcome so that the new approach can be successfully used.

2.2 Main Characteristics of Qualitative Reasoning and Applications in Process Engineering

Simulation of the behaviour of chemical plants is normally based on algebraic and differential equations representing the basic laws of physics and chemistry. By solving the equations and analysing the essential features of the solution, the patterns of expected behaviour can be examined. However, many aspects of reasoning about the behaviour of chemical plants during the preliminary design of a new project, or in planning start-up procedures, do not require this level of detail. Much information is often available from experience and numerical simulation comes later to fix design values and eliminate unsatisfactory design alternatives. Qualitative analysis is then used to interpret results, being valuable in supporting decisions about design and operating strategies. It is also used by operators to identify the effects of changes in the process or the origin of faults.

Qualitative reasoning techniques are invaluable in providing a basis for identifying the essential features of problems that cannot be effectively handled by conventional mathematical approaches. Some of these problems and the value of adopting qualitative reasoning are:

- Numerical algorithms require a complete set of equations together with associated parameters to obtain a solution. Qualitative reasoning is

possible with incomplete models and data or with models derived from common-sense and experience (de Kleer, 1990), examples of which are commonly found in biochemical engineering (Dohnal, 1987, 1989, 1991a, b). In the early stages of design it is important to know if the proposed process flow diagram will create operating difficulties. The qualitative description of process trajectories gives valuable insight into system behaviour, so preliminary design decisions can be made. The use of qualitative reasoning to support synthesis of operating procedures has already been investigated (Fusillo and Powers, 1988; Csaki *et al.*, 1991; Hangos *et al.*, 1991);

- Some engineering tasks need a description of all possible operational trajectories. For example, a fault-tree must capture all failure modes. Numerical simulators predict a specific trajectory for the prescribed set of parameters and initial conditions. Simulation of all combinations of parameters and input data may be computationally expensive or even non-feasible. Moreover, it is always difficult to guarantee that all combinations have been identified. Qualitative reasoning has the ability to identify all possible trajectories of a system (de Kleer, 1990);

- In some situations a rapid and rough estimation of behaviour, rather than a very precise prediction based on many unsupported assumptions is acceptable (Forbus, 1990). For instance, in considering several design alternatives, it is only necessary to know roughly how changes in one process variable will affect others and the consequential effect on design of equipment. Qualitative reasoning techniques are well suited for rapid and rough estimation of behaviour and so can be used for screening design alternatives;

- There is the need for a tool to support reasoning with highly cognitive tasks, such as process analysis, data interpretation, fault diagnosis, generation of operating procedures and explanation of process behaviour in relation to decision making. The identification of the relationship and information flow between variables is necessary in decision-making which relies on reasoning about the effects of changes. For instance, it is very important to understand how and why counter-intuitive responses can arise, so that decisions related to equipment design and control strategies can be effectively made. A systematic method to support explanation of process behaviour and patterns of response, coupled with graphical representation and scientific visualisation, is very important in interactive design, so that the skills of the designer can be better exploited;

- An integrated concurrent engineering environment and the global control of complex chemical plants need information to be easily shared. Qualitative reasoning can be used to pass on (share) expert knowledge;

Against these must be set some important limitations:

- Most techniques do not use dynamic information to constrain the solution space or to eliminate spurious solutions, as for example signed digraphs (Iri *et al.*, 1979) and qualitative process theory (Forbus, 1984);

- Quantitative knowledge about strength and order of magnitude of influences tend to be ignored. In those cases where this information is used it is limited by the use of the triple qualitative state descriptor {+,0,-}, which is not adequate for describing complex systems (Mavrovouniotis and Stephanopoulos, 1987, 1988);

- Qualitative reasoning techniques can generate non-real solutions in addition to real ones. There is no systematic procedure to identify and

eliminate the wrong solutions. This is the case of the qualitative simulation method (QSIM) proposed by Kuipers (1986).

These limitations have prevented the use of qualitative reasoning for describing complex systems found in the process industry, e.g. distributed parameter systems. However, there are some areas where there has been some success, such as fault diagnosis and process control. This is mainly because faults and control strategies can be analysed using steady state or quasi-steady state models. On the other hand, operational supervision, dynamic simulation and synthesis of operating procedures need non-steady state models, which are not very effectively handled by most qualitative reasoning techniques.

During the operation, the control and monitoring of process variables and their fluctuations within allowable limits are crucial to maintain product quality and safety. Diagnosis of process malfunctions is a very difficult task which has depended mostly on human judgement. However, even experienced operators may have difficulties in handling unanticipated events and low-probability failures (Kramer, 1987). Because of these difficulties, several methods of automated fault diagnosis based on qualitative reasoning have been proposed. Diagnosis by cause-and-effect analysis using patterns of process alarms is discussed by Iri *et al.* (1979), Tsuge *et al.* (1985a, b) and Shiozaki *et al.* (1985a, b). A related technique has been proposed for alarm analysis by Andow and Lees (1975). Umeda *et al.* (1980); Oyeleye and Kramer (1988) and Chang and Yu (1990) present improved qualitative methods for predicting propagation of disturbances. Tsuge *et al.* (1989) and Savkovic-Stevanovic (1992, 1995) present qualitative simulators for estimation of plant behaviour during abnormal situations. There are several other interesting approaches, such as those by Rich and Venkatasubramanian (1987), Mavrovouniotis and Stephanopoulos (1988), Grantham and Ungar (1990), Yu and Lee (1991) and Wang *et al.* (1995), among others, who also propose fault diagnosis methods. Up to

now fault diagnosis has been, by far, the main application of qualitative reasoning in process engineering.

Qualitative reasoning has been used in process control to describe open-loop responses of linear, non-linear and multivariable processes (Dalle Molle *et al.*, 1988), providing a framework for building qualitative versions of process models (Dalle Molle and Edgar, 1989) and verification of controller behaviour (Gazi *et al.*, 1994). Govind and Powers (1982) present a systematic procedure to support synthesis of control structures based on the cause-and-effect representation of the process and Féray-Beaumont *et al.* (1991) have applied qualitative transfer functions to represent the process model of a distillation column. Hangos (1991) discusses possible applications of qualitative techniques in control engineering, including evaluation of characteristic properties, such as structural stability, observability and controllability, as well as generation of operating procedures using a qualitative reasoning approach proposed by Németh *et al.* (1992).

Although the design of chemical plants makes use of non-quantitative procedures, qualitative reasoning has not been used to support it to the same extent as for fault diagnosis and process control. Grantham (1990) presents a prototype first-principles based system which aides in the conceptual development of batch processing systems. It suggests which phenomena would help to achieve the design goals, determines the processing conditions required to activate them and performs a qualitative analysis of the resulting behaviour, pointing out positive and negative aspects.

2.3 Qualitative Reasoning Techniques

Qualitative reasoning techniques are concerned with modelling the various forms of knowledge and establishing the basis for reasoning about the physical world in a way

that mimics human reasoning. They usually consist of a modelling methodology for the representation of knowledge about the process and an inference strategy usually referred to as qualitative simulation. As the field is very young, different approaches coexist with still unsolved theoretical, methodological and application problems (Hangos, 1991).

The literature on the subject is extensive. The book by Weld and de Kleer (1990) contains the most important foundation articles on the subject and presents a detailed historical discussion from the point of view of artificial intelligence. Hurme (1992) presents a very extensive review of the different approaches, forms of knowledge representation and applications to process engineering.

In order to be able to have a consistent basis for describing the various techniques, it is useful to adopt the following definitions:

- **Spurious solutions** - Solutions that cannot be exhibited by the physical system. This type of solution is generated when competing qualitative influences arise during the simulation and the algorithm cannot determine which influence prevails, e.g. when one influence tends to increase the variable while another tends to decrease it;

- **Ambiguous solutions** - A set of solutions which does not give a definitive answer;

- **State** - Indicative value of variables or derivatives;

- **Qualitative state descriptor** - Set of states that the variables and derivatives can assume during qualitative simulation;

- **Causality** - The definition of causality is a topic that has provoked a lot of controversy and discussion. The "mythical causality" proposed by de Kleer and Brown (1984) uses a finer time granularity to order events that

theoretically occur simultaneously in "reality", i.e. if "A causes B" than A occurs before B in the "mythical" time. Iwasaki and Simon (1986) propose the "causal ordering" method to determine the direction of influences from mathematical models. However, the method is of limited application, since it is not always possible to know the mathematical models which describe the system. The most accepted definition in engineering relates causality with the cause-and-effect relationships between two variables as in the expression: "A causes B". It is accepted that causality is the basic component of human reasoning. Therefore, methods based on causality can easily represent intuitive knowledge and are well suited to explain process behaviour and flow of information between process variables. In non-causal models, the explanation of process behaviour is very difficult since the causal links between variables are not explicitly represented. Because causality is not explicitly represented in mathematical models, the most effective way to determine causality is to analyse all mechanisms influencing the variables and make assumptions about the controlling mechanisms (Iwasaki and Simon, 1986), or by experiments. Skorstad (1992) presents a causal theory for thermodynamic properties. The author discusses that although the ideal gas law PV=RT defines a functional relationship between pressure (P) and temperature (T), the causal dependency is not clearly defined. Experiments have shown that a pressure drop caused by an expansion on a throttle without an accompanying work or heat flow has no effect on the temperature (Skorstad, 1992), but a heat flow will affect T and consequently P, although the volume is constant. Causality is usually represented by causal graphs in form of digraphs, which are discussed in section 2.5.

Table 2.1 summarises the most noted qualitative reasoning techniques and their main area of application. The techniques use different modelling languages and

concepts to describe physical systems. The choice of the most adequate approach to handle a particular problem depends on the objectives of the model and the desired type of response.

Table 2.1 Qualitative reasoning techniques.

Main qualitative reasoning techniques	Reference	Main area of application
Confluences	de Kleer and Brown (1984)	Description of quasi-steady state systems for validation of sensor data and fault diagnosis
Qualitative simulation (QSIM)	Kuipers (1984, 1986)	Modelling and simulation of the dynamic behaviour of chemical processes described by ODE[1] and by incomplete or uncertain knowledge about quantitative parameters.
Qualitative process theory (QPT)	Forbus (1984)	Description of steady-state chemical processes for explanation of system behaviour and fault diagnosis.
Signed digraphs (SDG)	Iri *et al.* (1979)	Modelling of steady-state chemical processes for fault diagnosis

[1] Ordinary differential equations

Confluences (de Kleer and Brown, 1984) and qualitative simulation - QSIM (Kuipers, 1984, 1986) translate the system of ordinary differential equations (ODE) and algebraic relations into qualitative differential equations (QDE). These qualitative equations preserve the structural form of the quantitative models and replace numerical values of variables and parameters by qualitative values. Both methods assume that the system being analysed is described by continuously differentiable functions of time. This greatly limits the use of non-quantitative, intuitive information. Therefore they use only a small part of available knowledge and are of limited application.

Confluences assumes that variables and time derivatives are described by a restricted set of possible qualitative states {+,0,-}, and uses sign algebra. The major disadvantage of sign algebra is that addition and subtraction operations involving variables with opposite signs are undetermined, i.e. the result can be any one of the three qualitative values {+,0,-}. This gives rise to a tree of possible behaviour, containing non-real (spurious) solutions. In some cases the number of ambiguous solutions can be very large, imposing severe restrictions on the use of the method. Confluences is usually classified as a device-centred approach, since it assumes that the behaviour of complete systems can be determined from the behaviour of individual process units and their interconnectivity. There is no attempt to provide insight into how to develop the models for the individual units (Grantham and Ungar, 1990). The simulation is based on the concept of "mythical causality" that assumes infinitesimal changes in the neighbourhood of an equilibrium point. This means it cannot be used to describe process dynamics for critical operations. As the method does not explicitly represent causality, it also cannot be used to explain process behaviour. It has mainly been used in validation of sensor data and fault diagnosis.

Qualitative simulation (QSIM) is usually classified as a constrained-based approach, since it is heavily based on constraints more than any other approach. It uses a much more flexible qualitative state descriptor than confluences. Variables and derivatives can assume intermediate qualitative values inside the initial allowed set. For example, initially a variable can assume the qualitative values {0,+∞}, but during the qualitative simulation new landmarks can be established and the variable can reach intermediate values, such as l1 and l2 ordered as follows: {0,l1,l2,+∞}. However, the method assumes a very complex methodology for qualitative operations with the states of the variables, which contributes to the generation of trees of possible solutions. These include real and spurious solutions and inconsistent branches. The explosive number of ambiguous solutions is among

the main problems of this approach. Many efforts have been made (Kuipers, 1987; Kuipers and Chiu, 1987; Kuipers and Berleant, 1988; Lee and Kuipers, 1988; Dalle Molle, 1989; Kuipers *et al.*, 1991) to combine qualitative with quantitative information and include information about higher-order derivatives, aiming at improving the qualitative description and eliminate spurious solutions. These works have improved the performance of the method but also made it far more complex than the original approach. QSIM is mainly a simulation tool and requires very detailed descriptions of systems, even for simple cases such as tanks. However, it does not provide any support for building the qualitative models in order to minimise the number of ambiguous solutions. For the simulation of a shell-tube heat exchanger in steady-state, Vianna (1992) obtained a tree of solutions with nine branches (including non-feasible trajectories). Probably this result can be improved by a better modelling. Attempts have been made to develop a model-builder (Crawford *et al.*, 1990; Richards *et al.*, 1992; Farquhar, 1994). QSIM cannot be used to explain how process behaviour is generated, since it does not explicitly represent causality and the simulation is very complex. QSIM has been used for process control and prediction of dynamic behaviour of lumped parameter systems (Dalle Molle *et al.*, 1988; Dalle Molle and Edgar, 1989, 1990; Kuipers, 1989), and process monitoring (Dvorak and Kuipers, 1989).

QSIM is not suited to describe distributed parameter systems. Kuipers (1992) says that the extension of QSIM to cope with this class of problem faces two major problems:

- **Boundary conditions** - QSIM works with landmarks and the clear definition of limit values for variables and derivatives. This implies that for a distributed parameter system the explicit representation of boundary conditions is needed. However QSIM cannot be easily adapted to deal with boundary conditions (Kuipers, 1992);

- **QSIM can only reason along one path each time** - Because of the simulation technique, a particular path, e.g. space, has to be simulated independently of others, e.g. time (Kuipers, 1992). However, a distributed parameter system has to be simulated by considering changes in time and space simultaneously.

Qualitative process theory (QPT) is significantly different from previously mentioned approaches. It is based on the representation of the knowledge of the basic physical and chemical phenomena underlying the process behaviour, such as mass and heat transfer, phase equilibrium and chemical reaction (Grantham, 1990; Grantham and Ungar, 1990). Because of this, it is usually referred to as a process-based approach. Models are created using a description of substances, objects and the basic process. Influences impose changes on system parameters while relations constrain the propagation of the influences between process variables. Causality is used to impose order on the events and can also be interpreted backwards to discover the cause of a specific event or to realise which variables must change in order to produce a desired effect. The method includes a graphical representation of the relations between process variables. It has been used to explain how results from numerical simulation are generated (Forbus and Falkenhainer, 1990, 1992), in diagnosis of faults (Grantham and Ungar, 1990) and for building intelligent tutoring systems for undergraduate students (Forbus and Whalley, 1994). QPT presents the following limitations: (1) the algebra involved in determining changes is not trivial; (2) inadequacy of the triple qualitative descriptor {+,0,-}; (3) generation of spurious solutions; (4) the consideration of intuitive knowledge (not based on the laws of physics and chemistry) is not easy, although possible; and (5) it only represents steady-state processes. Extension to deal with dynamics and distributed parameter systems is very difficult, if indeed possible.

The signed digraph (SDG) approach (Iri *et al.*, 1979) is based on graph theory from mathematics. It has been used in engineering for a long time (Hurme, 1992). A signed digraph model is a graphical structure which represents the physical and chemical processes underlying the system behaviour. It is composed of nodes, edges and signs. Nodes represent process variables while edges represent the local influence between variables. The influences can be either positive or negative. Variables are described by the limited triple set of possible qualitative states: {+,0,-}. The construction of signed digraphs is heavily based on the concept of causality, i.e. on the determination of variables that cause changes and those that change. The method allows the representation of causality in feedback loops, which is a great advantage over other methods. Models are easy to construct from intuitive and engineering knowledge and the graph structure makes it well suited to explain process behaviour. A signed digraph can be constructed from observed plant operation data, experience or using the structural information from mathematical models. The latter represents a more robust approach. Iri *et al.* (1979) use the signed digraph approach for qualitative modelling of steady-state chemical processes and apply it to fault diagnosis.

The qualitative simulation of signed digraphs is done by introducing a disturbance to the set of input variables, where the initial state of all variables is [0], and propagating the disturbance through nodes and edges (Hurme, 1992). The simulation does not involve sign algebra but the logic "or" approach is used, i.e. the stronger influence prevails.

The conventional signed digraph (SDG) approach presents several characteristics that make it a very attractive tool for reasoning with chemical processes in a design or operational environment. The characteristics include:

- **Easy to construct from intuition or engineering knowledge -** The signed digraph technique is based on the direct representation of the

cause and effects relationships between process variables in a digraph structure. It requires little compilation of knowledge and unlike other representation it does not require the development of qualitative equations or the use of complex qualitative algebra;

- **Visualisation facilities** - The model assumes a graphical approach which is very well suited to the visualisation of flow of information;

- **It may be used to describe complex process topologies** - Coupled systems or processes with recycles generate cycles or loops in the digraph structure. These can be easily handled by signed digraph models, since nodes can receive multiple branches and information can flow in any direction. Moreover these structures do not impose problems during the reasoning stage;

- **Reasoning with SDG is intuitive** - Individuals usually do not use equations or complex mathematical relations to mentally describe a problem. They reason about the cause and effects of the relevant influences and use a very simple qualitative algebra and an order-of-magnitude-like approach to eliminate weaker influences in order to simplify the model structure. As the signed digraph is a causal relationship based approach, it is able to capture the intuitive way people reason about problems;

- **Computationally undemanding** - The inference algorithm for describing process behaviour is based on search methods, and the logic "or" for solving multiple influences, which are not computationally demanding if compared with methods that use complex algebra. It can be programmed in any language and therefore does not require the use of LISP language or LISP machines, as do most other qualitative reasoning techniques.

The above advantages and the effectiveness in dealing with fault diagnosis have stimulated many researches with signed digraphs aiming at overcoming some of its main limitations: (1) generation of spurious solutions, (2) inability to describe dynamic behaviour and (3) inadequacy of the triple qualitative descriptor {+,0,-}. However, the existing approaches have been restricted to fault diagnosis, and no attention has been directed at other areas of process engineering, such as generation of start-up procedures or supervision of plant operations.

Umeda *et al.* (1980) have extended the conventional SDG approach to allow the description of the dynamic behaviour of systems described by ordinary differential equations by assuming a multi-stage approach. Shiozaki *et al.* (1985a, b) and Tsuge *et al.* (1985a, b) extend the possible qualitative values of the variables to a five-range pattern (+,+?,0,-?,-) to deal with states that are outside the range of normal changes but still within threshold limits. Tsuge *et al.* (1985a) also use delays to help in ordering the causes of failures and a multi-stage approach to enhance knowledge representation and allow diagnosis of impulse type failures. Oyeleye and Kramer (1988) develop an extended signed digraph (ESDG) and a method to convert the ESDG to a set of confluences (relations, in de Kleer and Brown's approach), which are used to eliminate spurious interpretations produced by non-causal confluences. The ESDG includes non-physical feedforward edges that represent inverse and compensatory responses due to negative feedback. Dynamic effects were ignored and only qualitative steady-state equations are used. No numerical data is required. Hashimoto *et al.* (1991) propose a three-layer approach in order to capture subsequent transitions that variables can undergo due to actions of operators or controllers. Mohindra and Clark (1993) attach the logical "and" to the edges, extending the applicability of the conventional SDG, which is limited to the "or" logic approach. Wilcox and Himmelblau (1994a, b) propose the possible cause and effect graph (PCEG), which limits the statements that can be used to describe the root

cause of a fault based on material and energy balances. This reduces the size of the search space and consequently reduces the number of spurious solutions.

Although the above works have greatly enhanced the functionality and applicability of the conventional SDG approach, it is still limited to fault diagnosis and cannot be applied to systems described by partial differential equations.

There are many other qualitative reasoning methods, such as formal order-of-magnitude (Mavrovouniotis and Stephanopoulos, 1987, 1988), which uses the rough magnitude of parameters and effects to improve the qualitative description of chemical processes. According to the latter authors, qualitative reasoning pays too much attention to values of single parameters and neglects the importance of the relations between them. In engineering problems, besides information about signs of quantities, there is also information about relative order-of-magnitude and rough numerical values describing intervals of changes. These approaches are based on previous ideas about using order-of-magnitude in artificial intelligence (Raiman 1986, 1991). Other researches mix the idea of order of magnitude with other qualitative reasoning methods (Hurme and Järveläinen, 1991; Dague, 1993 and Yip, 1993). The main problem with such approaches is related to the complexity of the algebra used to reason with order-of-magnitude entities. Much work has also been done in terms of fuzzy-logic based approaches (Shen and Leitch, 1992; Huang and Fan, 1993; and Wang *et al.*, 1995).

From the analysis of the existing techniques it is clear that qualitative reasoning techniques need to be extended to handle the dynamic description of distributed parameter systems so that they can be used to support tasks involving critical transients, such as generation of operating procedures.

2.4 Generation of Operating Procedures Using Qualitative Reasoning

The synthesis of operating procedures makes use of non-quantitative causal knowledge which reflects an understanding of how behaviour arises. This insight provides a basis for making decisions about optimal operating strategies. The concept of causality is fundamental in explaining the results of simulation whether qualitative or quantitative. Causal models encode more knowledge than non-causal approaches because they attempt to rationalise knowledge and extend the formal algebraic relationships. The usual approach is to use a graphical representation to visualise the links between variables and indicate how information flows through the system. As a consequence it is a natural choice for generating operating procedures.

Therefore, since confluences and QSIM are based on non-causal models, they are not generally capable of supporting synthesis of operating procedures. Moreover, the models required by such approaches are often very complicated, even for simple physical systems, and consequently the number of equations necessary to describe a complete plant become overwhelming and the resulting set of constraints intractable (Feráy-Beaumont *et al.*, 1991). Such complexity associated with the algebra used in the qualitative calculus results in an explosion of ambiguous solutions. This means that it is impossible to extend these methods to deal with distributed parameter systems.

The application of qualitative process theory (QPT), which is based on causal models, for the description of process dynamics and representation of intuitive knowledge is not straightforward. It requires a clear definition of substances and equipment involved in each process, which becomes very complex when modelling a complete plant.

The main characteristics of the signed digraph approach in terms of ease to construct models, visualisation facilities and ability to represent complex process topologies, causal relationships and intuitive knowledge, make it well suited to the new approach. The goal is to overcome the limitations of existing qualitative reasoning techniques, by avoiding generation of spurious solutions and enabling distributed parameter systems to be handled.

The next section contains a detailed description of the signed digraph approach and a discussion about how the limitations are related to basic structural elements. This is directed to showing that the method can be modified to effectively handle process dynamics, and so be used in a framework for the generation of operating procedures.

2.5 Signed Digraphs

The signed digraph (SDG) methodology used by Iri *et al.* (1979) is based on the graph theory and was originally developed for diagnosis of system failures in chemical processes. It was motivated by the difficulties in finding the first cause of a failure during plant operation because of the large number of state variables which need to be considered.

2.5.1 Graphs and Digraphs

Equations are a very effective means of representing the structural relationships of the variables of a system. However, they are mainly computational tools and give no insight into the way in which information flows through the system or solutions arise. Visual images are a much more appealing mode of conveying this type of detail.

Graph theory is a branch of mathematics which explicitly is concerned with representing this kind of structure. It provides a key to successful problem solving and to gain insight into the general solution of a problem.

Mathematically a graph is an abstraction of the structural relationship between discrete objects, represented by a set of *nodes* , N=$\{n_1, n_2,...,n_n\}$. The relationship between any two objects n_i is represented by an unordered pair of nodes called an *edge*, $e_k\{n_i,n_j\}$. Nodes are denoted by letters and edges are line segments drawn between the nodes, as shown in Fig. 2.1a. The choice of the discrete objects and the relationship between them depends on the application (Mah, 1990).

Directed graphs, or digraphs for short, are used to represent asymmetric relationships where the direction of flow is important for the problem formulation, e.g. the direction of flow in a pipeline or in a process flowsheet. In a digraph, each edge e_k is mapped onto an ordered pair of nodes $\{n_i,n_j\}$. It is drawn as a line segment with an arrow directed from n_i to n_j. The edge e_k is *incident to* the node n_j and *emergent from* the node n_i. Figure 2.1b illustrates a digraph.

It is useful to summarise the terminology used in the graph-based methods:

- *adjacent nodes* - two nodes n_i and n_j linked by an *edge* e_k;
- *incident to* - e_k is *incident to* the node n_i, if n_i is the terminal node of the edge e_k;
- *emergent from* - e_k is *emergent from* the node n_i, if n_i is the initial node of the edge e_k;
- *ascendant node* - node from where the *edge* e_k is *emergent from*;
- *descendant node* - node to where the *edge* e_k is *incident to*;
- *parallel edges* - edges sharing the same pair of end nodes;

- *self-loop* - an edge with the same two end nodes;

- *path* - sequence of distinct and consecutive edges linking any two nodes but not intersecting a node more than once. For instance, in Fig. 2.1b the sequence {a,b},{b,d},{d,e} is a path between 'a' and 'e' (terminal nodes);

- *loop* - two different paths leading from the same initial node to the same terminal node, but the initial node is different from the terminal one. The sequences {b,c},{c,d} and {b,d} in Fig. 2.1b enclose a loop.

- *cycle* - a path with the two terminal nodes being the same. In Fig. 2.1b the sequence {b,d},{d,e},{e,b} is a cycle.

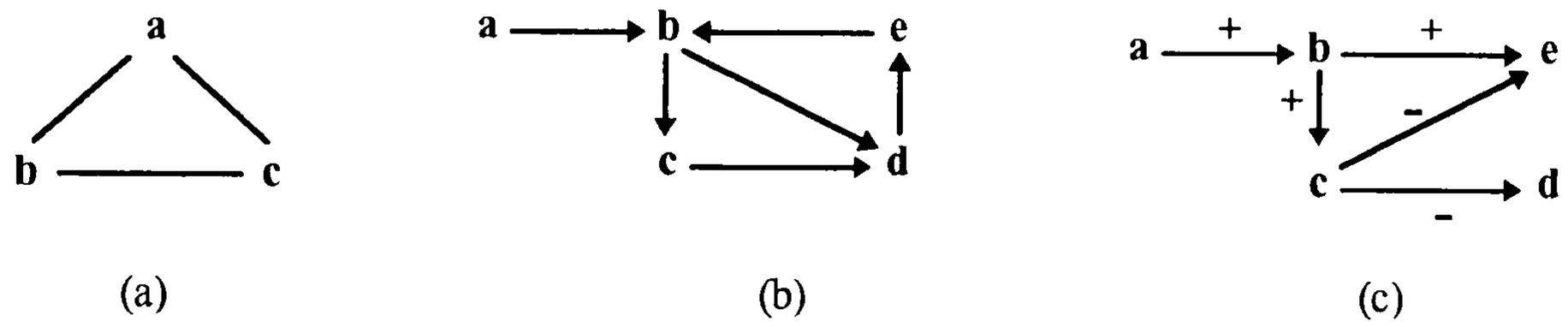

Figure 2.1 (a) Graph; (b) digraph; and (c) signed digraph.

2.5.2 Signed Digraph Structure

Iri *et al.* (1979, 1980) generalised the digraph approach by representing the positive and negative influences between process variables as a signed digraph (SDG). The proposed structure is a graph: **G=(N,E)** where **N** is a set of nodes $\{n_1, n_2,...,n_n\}$ and **E** is a set of edges $\{e_1, e_2,...,e_m\}$. Each edge is identified by an ordered triple (n_i, n_j, s_k) where the nodes n_i and n_j define the direction of the influences by mapping the edges to their initial and terminal nodes, respectively. The component s_k represents the sign {+,-} of the influences between the variables.

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Positive influences are characterised by variables changing in the same direction, while negative influences represent variables changing in opposite directions. Figure 2.1c illustrates a signed digraph.

Signed digraphs are qualitatively derived from operating data and/or the characteristic equations of the process and used as the model for representing the influences on the elements of the system.

Generally, systems can be described by algebraic equations and a set of ordinary differential equations which can be written as follows:

$$\frac{dx_i}{dt} = f_i(x_1, x_2, \ldots, x_n) \tag{2.1}$$

If $\partial f_i / \partial x_j \neq 0$, there is an edge from x_j to x_i with the same sign of the derivative ($\partial f_i / \partial x_j$). No self-loop is defined, even if $\partial f_i / \partial x_i \neq 0$. Iri *et al.* (1979, 1980) use a buffer tank to illustrate their methodology applied to fault diagnosis. For fault diagnosis purposes the variables can assume three state values: high (+), normal (0) and low (-).

The state (θ) of each node is calculated by multiplying the state of the *ascendant node* by the sign of the *incident edge* as shown in Eq. (2.2). If more than one edge *is incident to* a node and they are of opposite signs, Iri *et al.* (1979, 1980) assume that the influences will compensate each other for purposes of fault diagnosis.

$$\theta n_i = \theta n_j . s_{ji} \tag{2.2}$$

where θn_i is the state of the node n_i;

s_{ji} is the sign of the edge from node n_j to node n_i;

$i = \{1, \ldots, n_n\}$ and $j = \{1, \ldots, n_{ic}\}$;

n_n is the number of nodes of the model; and

n_{ic} is the number of edges *incident to* n_i.

The basic principle of the inference algorithm is to trace the causes of a failure back along the directed graph by calculating the state of the nodes which compose the digraph structure.

Although signed digraphs are very effective for fault diagnosis they do have limitations, as discussed below in relation to process dynamics.

2.5.3 Limitations of Signed Digraphs in Describing Process Dynamics

Consider the gravity-flow tank shown in Fig. 2.2a subject to step disturbances in the feed flow rate, F_i. The outlet flow rate and level are represented by F_o and L_T, respectively. The tank is described by Eqs. (2.3) and (2.4) and the causal relationships (2.5) to (2.7) derived from these equations. Figure 2.2b shows the SDG model generated for the system, based on the causal relationships.

$$\frac{dL_T}{dt} = S_{T1}.(F_i - F_o) \tag{2.3}$$

$$F_o = S_{T2}.L_T \tag{2.4}$$

$$F_i \xrightarrow{+} L_T \tag{2.5}$$

$$L_T \xrightarrow{+} F_o \tag{2.6}$$

$$F_o \xrightarrow{-} L_T \tag{2.7}$$

where t = time; S_{T1} and S_{T2} = constants.

Qualitative simulation of signed digraphs is done by introducing a disturbance to the set of input variables, where the initial state of all variables is [0]. The disturbances are propagated from node-to-node. To describe the dynamics, it is assumed that the variables can take one of three state values: increase (+), constant (0) and decrease (-). Simulation does not involve algebra of signs: rather a logic "or" is used which implies that the stronger influence prevails. However, when two influences of opposite signs are *incident to* a node, the result is ambiguous because any of the three solutions {+,0,-} is possible.

Table. 2.2 and Fig. 2.3 show the result of reasoning with the SDG model (Fig. 2.2b) when the tank is subject to a positive step disturbance in the feed flow rate (F_i). It can be seen that the method correctly captures the first response of the system, i.e. the level and outlet flow rate increase with the increase in the feed flow rate. However, at subsequent time steps, the prediction is uncertain because the model is unable to distinguish the strength of the influences from conflicting actions: (1) positive influence from F_i and (2) negative compensatory influence from F_o. As the signed digraph does not contain enough information to constrain the solution space, the method fails to distinguish the final response of the system and cannot predict unambiguously the behaviour of the system over the next period. Therefore, all possible solutions need to be generated, including spurious ones. Figure 2.4 shows an expected (quantitative) dynamic response for the system.

The unsatisfactory prediction of the qualitative dynamic behaviour of the tank can be attributed to the following reasons:

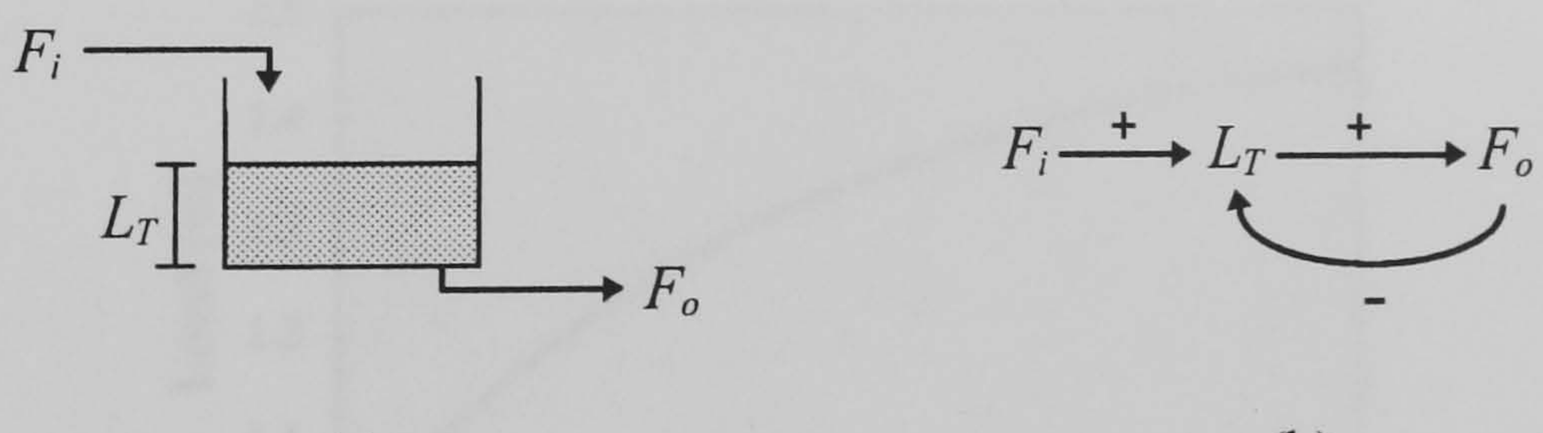
- **Inadequacy of the triple qualitative state descriptor** - The value space of the variables is restricted to the set {-,0,+} which is not sufficiently discriminatory to assess the relative strength of the influences;

- **Lack of dynamic information** - The model does not take account of information on the system dynamics, i.e. how variables change with time;

- **Inadequate reasoning algorithm** - The reasoning methodology proposed by Iri *et al.* (1979, 1980) for fault-diagnosis is inadequate for description of system dynamics. It searches for the origin of failures and is not intended to trace system behaviour through time.

In summary, the signed digraph approach is well suited to the description of the initial response of the variables of a system, but is not intended to describe the magnitude of the influences and delays, because it does not include information about changes in the state of the variables with time and space and is restricted by the state descriptor {+,0,-}.

It is clear that there is considerable scope to the development of a methodology based on the conventional signed digraph to deal with distributed parameter systems and support several process engineering tasks, such as process design and generation of operating procedures.



(a) (b)

Figure 2.2 (a) Gravity-flow tank and (b) signed digraph model.

Table 2.2 Qualitative dynamic behaviour for the gravity-flow tank subject to a positive step disturbance in the feed flow rate.

Time	F_i	L_T	F_o
0	0	0	0
1	+1	0	0
2	+1	+1	0
3	+1	+1	+1
4	+1	?	?

N.B. ? = indeterminate behaviour

(a) (b)

Figure 2.3 Qualitative dynamic behaviour for the gravity-flow tank: (a) inlet flow rate and (b) level.

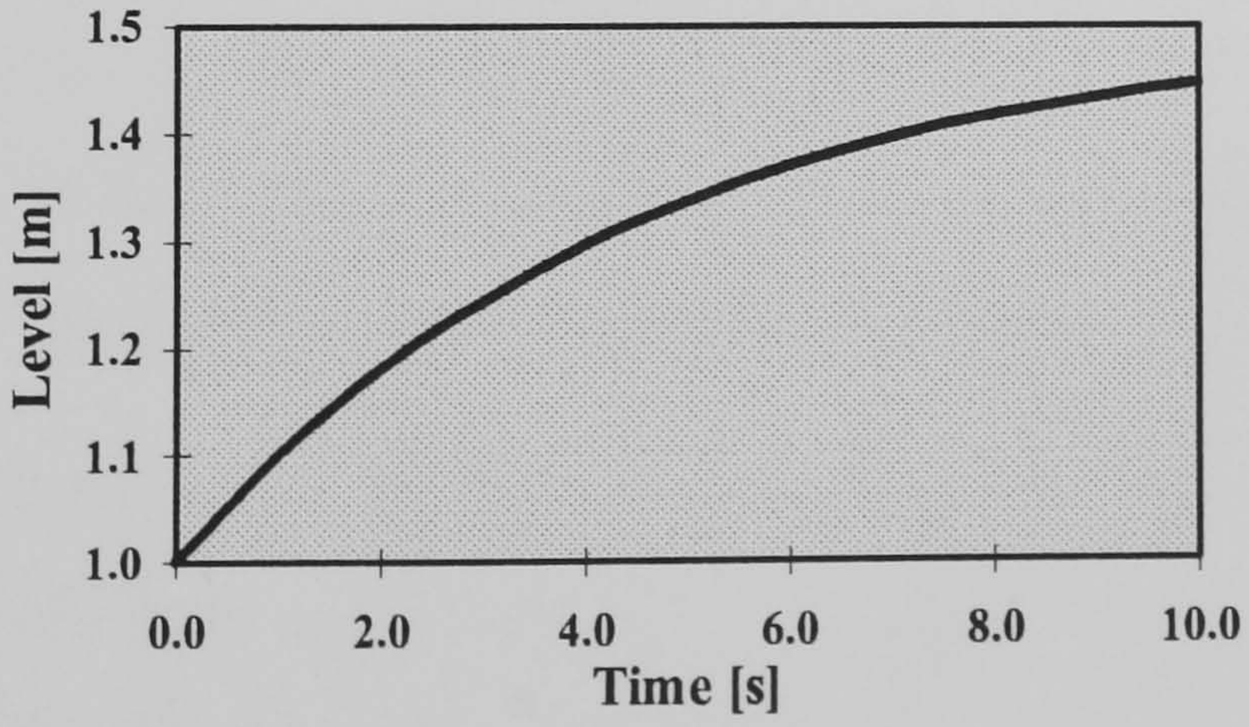


Figure 2.4 Expected dynamic behaviour of the level of the gravity-flow tank.

2.6 Concluding Remarks

In this chapter an overview on qualitative reasoning has been presented and the existing techniques discussed in terms of their effectiveness in dealing with process engineering problems.

Qualitative reasoning has the ability to deal with incomplete knowledge and support cognitive tasks. Some techniques can express causality embedded in intuitive and engineering knowledge and have great potential for use in explaining process behaviour. Signed digraphs have several advantages, mainly in terms of visualisation capabilities, ease to construct models and ability to deal with coupled systems and recycles. They can therefore be used to explain process behaviour and support non-quantitative tasks, such as process analysis and data interpretation, as well as to generate optimal design and operating strategies.

Existing qualitative reasoning techniques, including signed digraphs, do have several limitations, mainly related to the inability to cope with process dynamics in terms of distributed parameter systems and generation of spurious solutions. Therefore, there is a need for a flexible tool capable of dealing with several classes of chemical engineering problems and robust enough to describe complex system trajectories.

Chapter 3

Weighted Digraphs for Qualitative Description of Process Dynamics

3.1 Introduction

The simulation of process dynamics has been recognised as a task of major importance during the analysis of the life-cycle performance of a plant, and is essential in ensuring high standards of design. Despite this, the use of dynamic simulators in process design has been very limited, even for supporting the synthesis of procedures such as operating and control. The reasons are mainly related to the difficulties involved in generating dynamic models, determining precise numerical values of parameters and interpreting the large amounts of data and information generated by numerical simulators. Until now, these highly cognitive tasks have been carried out mainly based on the experience and skills of talented designers.

The current trend towards the creation of an integrated concurrent engineering environment has stimulated the development of "intelligent" support systems to help with model building and reasoning with information generated by the simulators. The aim is to provide a platform to make explicit the relations between process variables and the way in which responses develop in order to help to explain and

interpret solutions and so be valuable in supporting design decisions. Such a framework can be used to reason about the effects of changes in process design and operating conditions and to trace back to problem sources so that modifications can be targeted more specifically on desired goals.

Qualitative reasoning derives from artificial intelligence and is well suited to be used in conjunction with numerical simulation to provide an "intelligent" support system. Qualitative representation of process dynamics is essential in assessing flow of information through a process because it reveals the nature of the interaction between process variables which is fundamental in understanding the solutions.

At present, existing qualitative reasoning techniques are not able to effectively describe the general dynamic behaviour of process systems, since the models are very difficult to generate and the simulation tends to produce large numbers of spurious (non-real) solutions. Moreover, they are unable to cope with distributed parameter systems. Consequently, this has hindered their widespread use in design and synthesis of operating procedures. This calls for a more robust approach.

In this chapter a qualitative reasoning approach, referred to as weighted digraphs (WDG), is presented. The methodology is based on a generalisation of the signed digraph (SDG) approach by Iri *et al.* (1979). It is directed to retaining the main characteristics of the conventional approach, such as the ease of model construction from intuitive or engineering knowledge and visualisation facilities, but introduces several new features to overcome the main limitations due to poor representation of knowledge and lack of dynamic information.

The potentialities and functionalities of the approach are discussed in terms of the flexibility and ability to describe complex process dynamics and distributed parameter systems. The procedure is illustrated by reference to a simple case study based on the gravity-flow tank.

3.2 Difficulties Involved in Describing Dynamic Behaviour

The description of process behaviour depends on an understanding of the underlying basic physical and chemical phenomena, which identify the principal variables and their relationships since these determine the system dynamic trajectories. It is a highly demanding task based on extensive process analysis and requiring considerable expertise.

The dynamics can usually involve complex patterns of behaviour, such as inverse responses, unstable transitions and bifurcations, arising from a combination of factors, such as non-linearities, delays, feedback and compensatory response. For example, inverse response may arise from differences in the rate of change of interacting variables. Luyben (1990) discusses the very interesting example of the inverse response of the bottom composition and base level of a distillation column. It is known that an increase in vapour boil-up must carry more of the least volatile component up the column and therefore decrease the mole fraction of the light component at the bottom, x_1^B. However, the tray hydraulics can alter this pattern of behaviour during the transient period. The increase in the vapour flow rate through a tray may cause two opposite effects: (1) decrease in the liquid flow rates due to back up of more liquid in the downcomers to overcome the increase in the pressure drop through the trays, or (2) increase in the liquid flow rates due to the reduction in the density of the liquid and vapour froth on the active part of the tray which increases the height of liquid flowing over the weir. Establishing which effect will dominate requires knowledge of the tray design and operating policy. In valve trays, the latter effect prevails, since the pressure drop changes little with vapour flow rate. In this case, the increase in vapour boil-up provokes a transient increase in the liquid flow rate down the column which carries more light component to the base of the column and increases x_1^B. However, as these effects are associated with the tray hydraulics

and so are fast, the liquid flow rate returns to normal quickly and x_1^B decreases due to the increase in the vapour boil-up. In fact, the inverse response is a consequence of the competition between the strength of the influences, which change with time depending on the speed of the rates of change. Some influences are inevitably faster than others and so reach their peak when other variables are still not responding. Later responses reach a peak towards the end of the transient period. Therefore, in dealing with the dynamics the characterisation of the speed of the various responses and their relative strength through time is important.

Unusual patterns of behaviour, such as the above, are difficult to capture using qualitative reasoning techniques. To handle these complexities of the process dynamics, any new approach has to possess the ability to:

- Deal with fast and slow dynamics;
- Capture delays generated by the process topology;
- Represent compensatory responses and feedback loops;
- Characterise the relative strength of the influences and their pattern of change with time;
- Represent first and higher-order derivatives.

The weighted digraph (WDG) approach has been developed on the basis of creating a qualitative reasoning procedure which incorporates all the above characteristics and consequently is able to represent complex patterns of dynamic behaviour, as those arising in distributed parameter systems.

3.3 Qualitative Modelling

The formulation of a qualitative model involves several steps, as shown in Fig. 3.1, and places emphasis on subjective judgements. The starting point is the choice of a modelling technique capable of capturing the important features of the process. As argued by Shen and Leitch (1992), the critical task is to decide on the form of abstract representation of system variables and the relationships between them. The definition of the objectives of the model, including the domain to be described and the restrictions, also plays a very important role at this stage, since different objectives may lead to different models. A qualitative model intended for support of operating decisions has different characteristics from that supporting decisions in an engineering design environment (Vianna and McGreavy, 1995a).

After the definition stage, the formulation of the model evolves as an interactive process, as illustrated in Fig. 3.1. Specifically it includes: (1) knowledge acquisition, (2) knowledge representation and (3) inferring with respect to process behaviour. These stages involve several activities:

- Hypothesising about the dominant mechanisms which underlie the system behaviour, e.g. heat transfer, mass transfer, reaction and mixing. The description of a system in terms of these mechanisms is fundamental to causal analysis (Iwasaki and Simon, 1986);

- Definition of system boundaries - The boundaries of the solution space to be described must be explicitly stated since WDG models are developed for specific regions of the solution space. If boundaries are to be crossed, another model will normally need to be developed. Shen and Leitch (1992) propose dividing the model space into sub-spaces with different model structures being used in accordance with the model objectives;

- Simplifying assumptions - Assumptions have to be made to make the model manageable yet able to capture the essential behaviour of the process;

- Selection of variables and identification of their functionality - Variables can be classified as one of three types: *input*, *structural* and *output*. The state of the *input* variables is determined externally. They are responsible for carrying information from the "outside world" to the system. An initial value for each of them must be known. *Structural* variables compose the main body of the model and are responsible for propagating disturbances from the input to the output variables. *Output* variables characterise the response of the system to input disturbances. The type and importance of the variables depend on the objectives of the model. Important variables for a model to be used in an engineering environment may not be valuable for a model dedicated to operational supervision, so a *structural* variable in one model may be an *output* variable in another. The variables can be intensive, extensive, equipment related and first and higher-order derivatives;

- Description of the relationships between process variables is necessary in order to characterise the dominant mechanisms. These relations are determined based on experience, intuition, physical and chemical laws as well as mathematical models;

Following the above activities, a model structure is proposed and associated to an inference procedure to provide an overall response. Verification of the predictive performance of the model is carried out by comparing predicted and observed responses. This is used to suggest any necessary changes to the model or system boundaries.

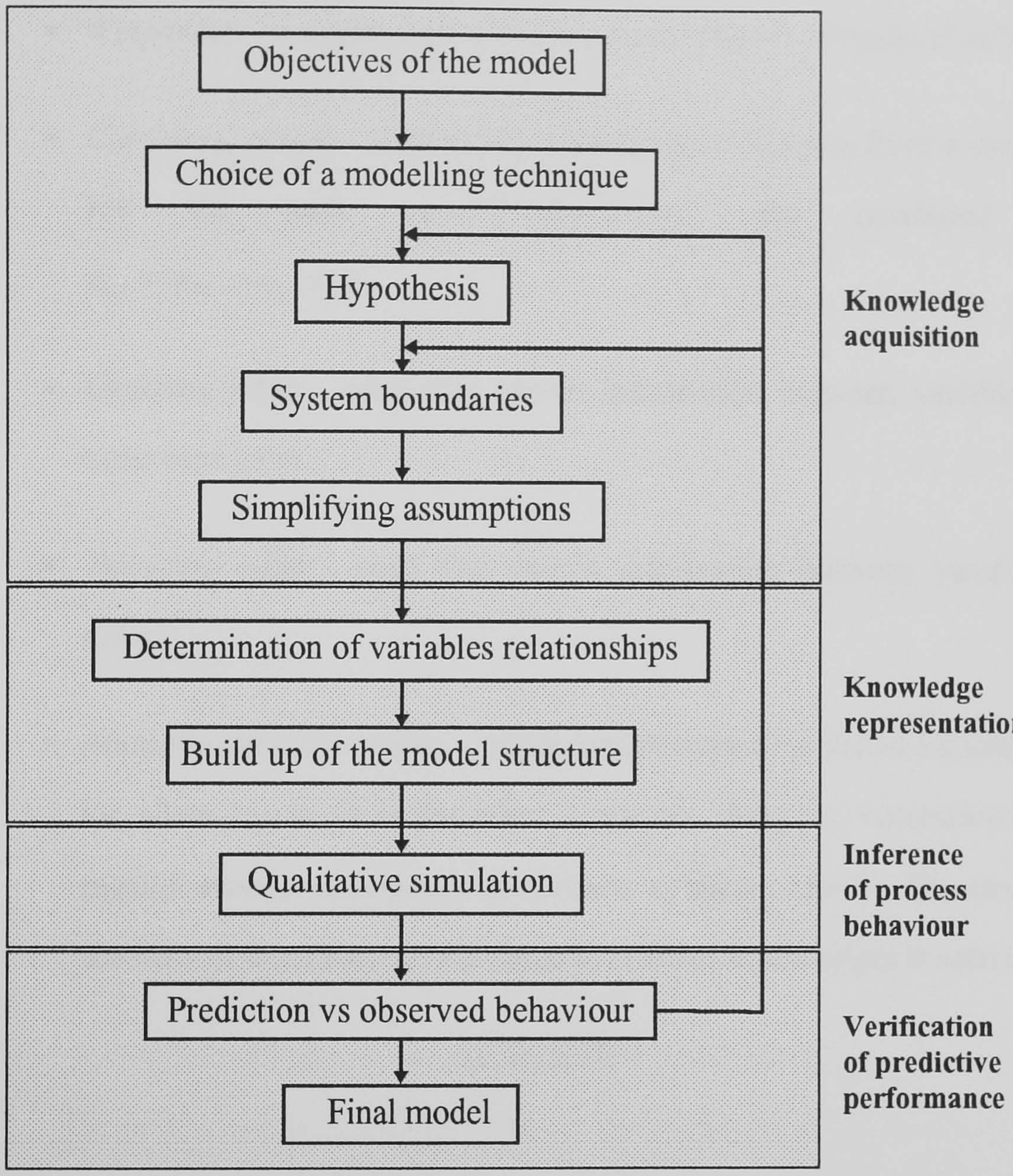


Figure 3.1 Steps involved in formulating a qualitative model.

3.4 Weighted Digraph Models

Weighted digraphs (WDG) are an extension of the signed digraph (SDG) approach aiming at enhancing the qualitative knowledge representation to cope with process dynamics. In addition to the definitions used for signed digraphs in Chapter 2, the following are added:

- *State value (or state)* - qualitative value of a node at time *t*;

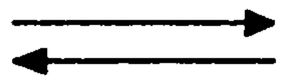
- *Weight (w)* - concept used to represent the relative strength of influences;

- *Combined weight* - obtained by multiplying all *weights* from a simple path. For the path: $a \xrightarrow{+w} b \xrightarrow{-w} c$, the *combined weight* $w_{a,c} = w_{a,b} \cdot w_{b,c} < 0$;

- *Ordinary edge* - edge that carries information between variables at the same time level;

- *Temporal edge* - edge that carries information between variables over time;

- *Mutually exclusive edges* - edges which cannot be active simultaneously, i.e. when one is active the other is inactive. They are represented by two parallel arrows in opposite directions, as shown below. The direction of the flow of disturbances will determine which of the edges is active;



- *Edge listing* - résumé of the information on edge connections, type of influences and *weights* of edges.

A weighted digraph **D** is represented by a graph structure: **D** = (**P**,**R**), where each component is defined as follows:

- **P** is a set of nodes $\{p_1, p_2, \ldots, p_n\}$ that represents process variables. Each node is identified by a pair (θ, α) where θ describes the state value of the node and α identifies its type: *algebraic* ($\alpha = 0$) or *differential* ($\alpha = 1$). *Algebraic nodes* (x_i) represent the instantaneous value of process variables, while *differential nodes* (δx_i) represent the rate of change of the variables with time. The inclusion of nodes dedicated to representing the

trends of first-order time derivatives not only introduces into the basic model knowledge about system dynamics but also constrains the solution space, and assists in eliminating spurious solutions. In a conventional signed digraph model, if the state of the node p_i is positive, three different qualitative dynamic behaviour patterns are possible: (1) p_i will increase continuously, (2) p_i may increase and stabilise, or (3) p_i may reach a maximum and start to decrease, although still be positive. However, if it is known that the first-order derivative will be positive during the subsequent time steps, the system will follow trajectories (1) or (2). In the following sections, it will be seen that information about second-order time derivatives is implicitly represented in weighted digraph models. This constrains the solution space even more and can successfully describe the shape of a variety of functions. Information of this type leads to a more robust model;

- **R** is a set of edges $\{r_1, r_2,...,r_m\}$ which represents causal relationships between process variables. Each edge is identified by a quadruple set $(p_i, p_j, \beta_k, w_{ij})$, where p_i and p_j define the direction of the influence by mapping the edges to their initial and terminal nodes, respectively. Component β_k assumes a value from the set {0,1}, which defines the type of the edge: *ordinary* ($\beta_k = 0$) and *temporal* ($\beta_k = 1$). *Ordinary edges* propagate information through the graph structure by considering the state of the *ascendant nodes* at time t. They are represented as full line arrows. *Temporal edges* propagate information through time by considering the state of the *ascendant nodes* at time t-Δt. They are represented as dashed arrows and used to describe: (1) delays in the response of the system due to the process topology, (2) variables with different speed of response (slow and fast dynamics), (3) compensatory responses due to negative feedback, and (4) feedback control loops. Their main advantage is in

allowing the representation of the complex patterns of behaviour which commonly arise from variables with different speeds of response, and in breaking loops and cycles. The latter reduces the complexity of the graph structure and consequently eliminates dubious solutions and avoids problems of convergence during simulation. The component w_{ij} represents a *weight* which characterises the strength of the relationship between variables p_i and p_j. This means that if $w_{ij} > w_{kj}$ then p_i exerts a stronger influence over p_j than p_k . Although *weights* may assume any value in the real interval [-1,+1], they are usually attributed values of +1 or -1 for positive and negative influences, respectively. In specific situations such as loops and cycles where potentially conflicting influences (those with the same absolute value but opposite signs) arise, *weights* with values different from +1 or -1 are attributed to the influences, according to their strength, in order to avoid conflict. This greatly reduces the generation of ambiguous solutions. The methodology for calculating the value of the *weights* will be described in section 3.5.3;

Figure 3.2 summarises the main concepts of the basic weighted digraph structure.

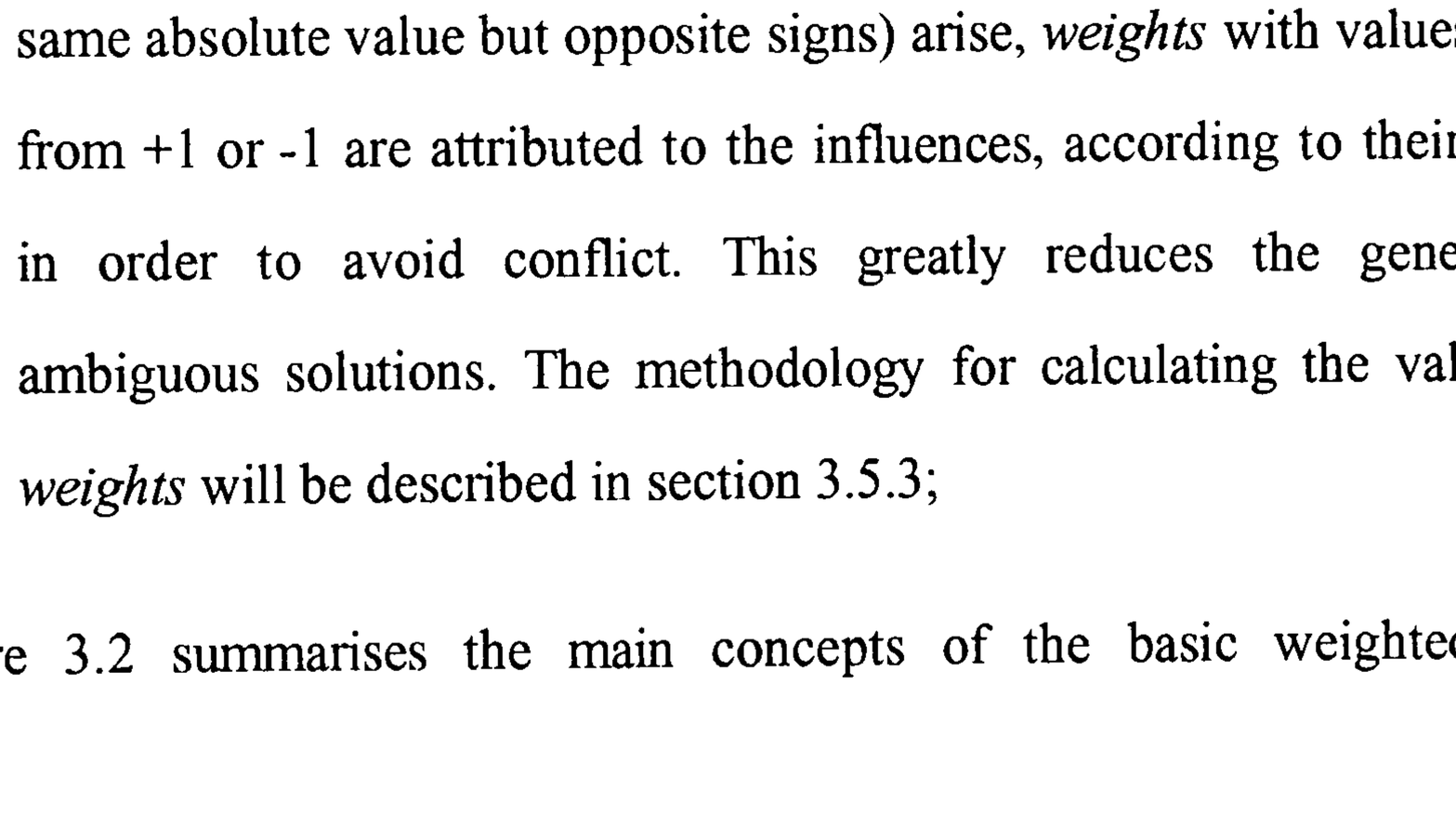


Figure 3.2 Main concepts of the basic weighted digraph structure.

3.5 Methodology for the Formulation of Weighted Digraph Models

3.5.1 Knowledge Acquisition

For the construction of weighted digraph models, any of the following sources of knowledge may be used:

- Experience expressed as heuristic rules;
- Observed behaviour from pilot and real plants;
- Physical and chemical laws, such as conservation of mass, energy and momentum;
- Conventional mathematical models, e.g. sets of differential and algebraic equations.

Although it is possible to build a weighted digraph model from heuristic rules and observed behaviour only, the resulting model will not be flexible and robust enough to describe complex patterns of behaviour. Therefore the use of fundamental physical and chemical laws in the general time-dependent form is essential for building powerful models. If mathematical models derived from the basic laws are also available, they constitute the best source of knowledge in building weighted digraphs, since they encompass a large amount of knowledge from experts. However, there is no need to know the precise numerical values of parameters. Their order-of-magnitude and rough numerical information about the range of the variables is useful, but are not essential in the model building.

Some researchers argue that qualitative methodology should not be based on mathematical models, because it is redundant. However, the motivation of

developing weighted digraph models is not to create a tool to replace mathematical models or numerical simulation, but to find a procedure which explicitly encodes causal and engineering knowledge embedded in mathematical models and experience-based procedures for process design and operation. In particular, the objective is to build graphic-based models suited to supporting explanations of process behaviour, so that they can be used to help the engineer to analyse and reason about results from quantitative simulators and the consequences of changes in design or process conditions. In such cases, mathematical models are often known, although accurate values of parameters and initial conditions may not be available, as for example in the early stages of process design. The procedure can also be used to help operators to reason about the possible effects of changes in operating conditions or controller set-points.

The weighted digraph methodology has many similarities to model-reduction, where complex systems are built from a minimal set of elemental structures. Thus a weighted digraph model for a distillation column can be based on qualitative models of a few flash stages and heat-exchangers. This has considerable attractions when considering generation of operating procedures.

3.5.2 Determination of Causal Relationships

Expressing causality is important in reducing spurious solutions and explaining systems trajectories. It aims at making explicit the directional (asymmetric) relationships between process variables and is useful in developing visual representations of information which assist in understanding process behaviour.

Translating heuristic rules and observed behaviour into causal relationships is an intuitive activity. For example, it is known that fouling (F) reduces heat transfer (Q)

in a heat exchanger, and this can be conveniently expressed in digraph terms as: $F \xrightarrow{-w} Q$.

However, the translation of physical and chemical laws into causal relationships is significantly more complex, since there is no explicit representation of causality. To provide this capability it is necessary to postulate on the existence of a set of algebraic and first-order ordinary differential equations which are capable of describing the system behaviour. However, to gain insight into the patterns of behaviour, accurate values of parameters are not needed. Restricting the approach to first-order systems is not a problem because higher-order equations can be cast as a set of first-order differential equations.

Algebraic equations must be available in the explicit form shown by Eq. (3.1), and first-order differential equations must be represented by Eqs. (3.2) and (3.3). Differential equations are discretized by finite-differences as shown in Eqs. (3.4) and (3.5).

$$x_i = g_i(x_1, x_2, \ldots, x_N) \tag{3.1}$$

$$\delta x_i = f_i(x_1, x_2, \ldots, x_N) \tag{3.2}$$

$$\text{where} \quad \delta x_i = \frac{dx_i}{dt} \tag{3.3}$$

$$\delta x_i^{(t)} = \frac{x_i^{(t)} - x_i^{(t-\Delta t)}}{\Delta t} \tag{3.4}$$

$$\text{or} \qquad x_i^{(t)} = \delta x_i^{(t)} . \Delta t + x_i^{(t-\Delta t)} \tag{3.5}$$

The vector (x_1, x_2,..., x_N) represents not only the dependent (*structural*) variables but also the *input* variables which must be independent or a function of time. Neither f_i or g_i should be explicit functions of time.

The conserved quantities (left-hand side of the differential equations), δx_i, are represented as *differential nodes* in the digraph structure, while the other variables, x_i, are represented as *algebraic nodes*.

The causal relationship between process variables is determined by *structural causality*, which is defined as follows:

- Expressing important structural knowledge implicit in the algebraic and differential equations into causal relationships makes use of partial derivatives (O_i^j) of the variables represented on the left-hand side (lhs) of Eqs. (3.1) and (3.2) with respect to each variable from the right-hand side (rhs), while keeping all other variables constant, as shown in Eq. (3.6). This means that if $O_i^j \neq 0$, then the variables x_j, from the rhs of Eqs. (3.1) or (3.2) are responsible for changes in the variables ψ_i , from the lhs. Therefore, an edge from x_j to ψ_i is defined and identified by a *weight* (w_{ji}) with the same sign of O_i^j. In the digraph, this means that the state values of the nodes representing ψ_i are determined by the state value of the nodes that represent x_j;

$$O_i^j = \left(\frac{\partial \psi_i}{\partial x_j} \right)_{x_{k \neq j}} \tag{3.6}$$

where $\psi_i = \delta x_i$ or x_i (lhs of Eqs. (3.1) and (3.2))

x_j = variables on the rhs of Eqs. (3.1) and (3.2)

$i, j, k = \{1,....,N\}$

N = number of nodes (variables)

- The sign of O_i^j is important in distinguishing positive and negative influences, while its order-of-magnitude is used to rule out negligible influences in order to simplify the model;

- If $\partial(\delta x_i)/\partial x_i \neq 0$, the variable x_i is self-regulating, since the time derivative is dependent on x_i. In such cases it is possible to define a special structure: *self-regulating group*.

In the WDG methodology, some system features are represented by specific structures as follows:

- ***Self-regulating group*** - If $\partial(\delta x_i)/\partial x_i \neq 0$, the *self-regulating group* is composed of a *self-regulating variable* x_i, and the time derivative, δx_i. The causal relationships of the *self-regulating group* are defined as a positive *ordinary* influence from $\delta x_i^{(t)}$ to $x_i^{(t)}$ based on Eq. (3.5) and a past time *(temporal)* influence from $x_i^{(t-\Delta t)}$ to $\delta x_i^{(t)}$ based on Eq. (3.2). The latter may or may not stabilise the system, depending on the sign of the *weight* attributed to the *temporal edge* and the sign of $x_i^{(t-\Delta t)}$. Figure 3.3a shows a stabilising *self-regulating group*, where the *temporal edge* has a negative *weight*, and Fig. 3.3b shows an unstable *self-regulating group*;

- **Fast and slow dynamics** - Chemical processes are usually described by a mixture of fast and slow dynamics. For example, in a distillation column while the hydraulic dynamic response occurs very rapidly (a few seconds per tray), the composition and temperature dynamic responses are much slower, minutes or even hours (Luyben, 1990). In the WDG approach, fast influences are propagated by *ordinary* and slow by *temporal edges*. The main advantage in scheduling influences in accordance to the speed of their dynamic responses is to break loops, since the influences act at different times. This greatly reduces the generation of ambiguous solutions. Another potential application of scheduling influences is in describing inverse responses, as shown in Fig. 3.4. Variables with fast dynamics determine the response during the initial time steps, and only afterwards does the influence of the slow dynamics prevail;

- **Compensatory responses and feedback loops-** *Temporal edges* are also used to represent compensatory responses and feedback control loops. For example, the gravity-flow tank can be described by Eqs. (3.7) and (3.8). If the inlet flow rate (F_i) increases, the level (L_T) tends to increase according to Eq. (3.7) and the outlet flow rate (F_o) consequently increases as shown by Eq. (3.8). However, the outlet flow rate exhibits a compensatory response as a result of negative feedback on the level, because of Eq. (3.7). Therefore, a negative weighted *temporal edge* from F_o to L_T has to be used in the digraph. Figure 3.3c shows such a generic *compensatory structure*.

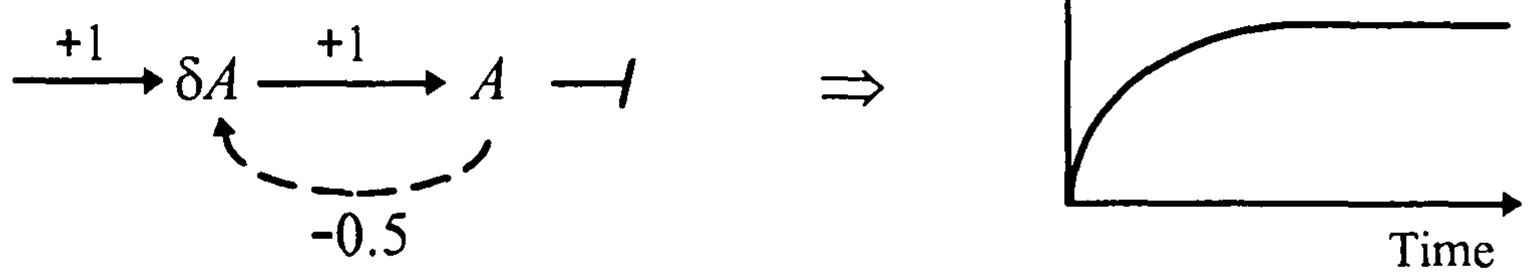
$$\frac{dL_T}{dt} = S_{T1}.(F_i - F_o) \tag{3.7}$$

$$F_o = S_{T2}.L_T \tag{3.8}$$

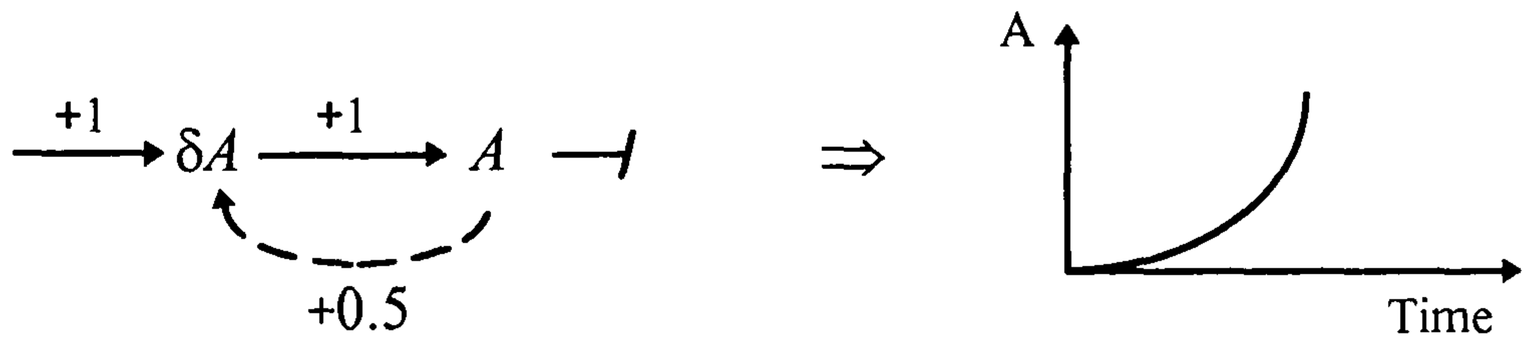
- **Multiple influences** - Some influences may change sign depending on the values of reference variables and/or with time, i.e. the sign of O_i^j may vary depending on the situation. In this case, a structure containing *multiple edges* and the logic "*or*" is introduced in the digraph structure to represent each influence, as shown in Fig. 3.3d. This case is well illustrated by the continuous-stirred tank reactor (CSTR) where the behaviour may change, depending on the range of variation of the inlet temperature and concentration. Thus the reactor may reach a new steady-state, suffer a run-away of temperature or result in a blow-out. This requires *multiple edges* to describe the different types of behaviour, in accordance with the qualitative ranges of the inlet temperature and concentration.

Weighted digraph structure	**Function represented** (for a positive step disturbance)

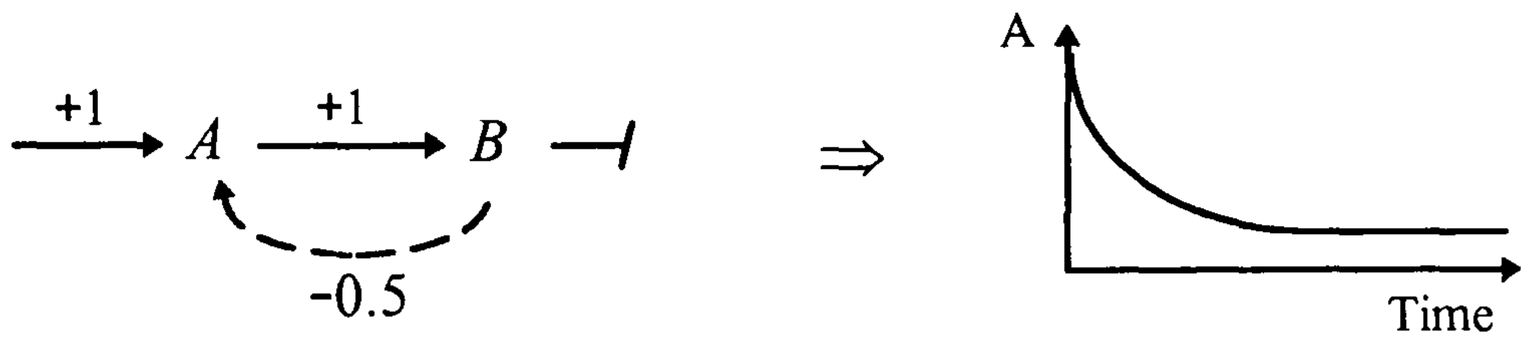
(a) stable *self-regulating group:*



(b) unstable *self-regulating group:*



(c) *compensatory structure:*



(d) *multiple edges:*

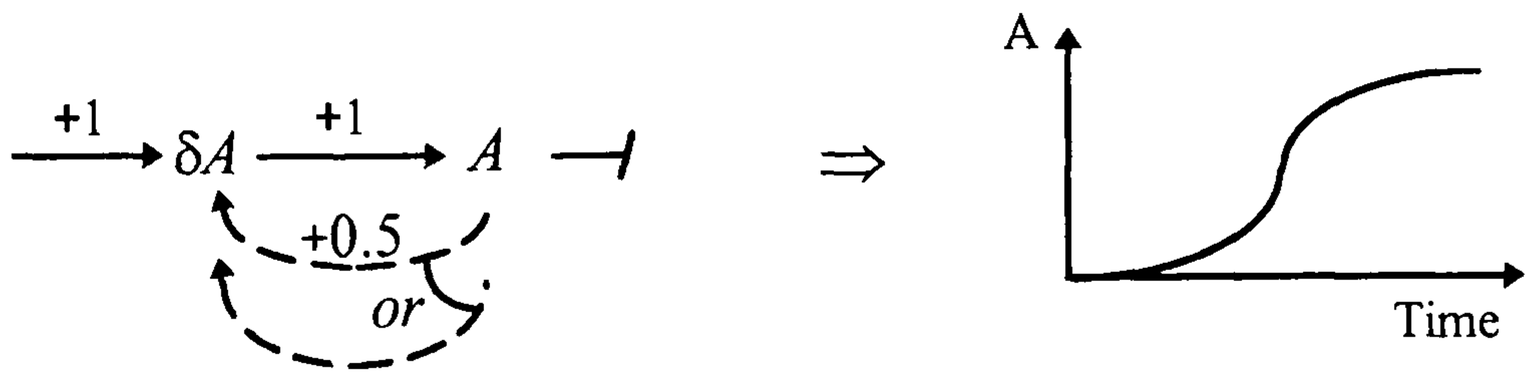


Figure 3.3 Special WDG structures and the functions they represent for a positive step disturbance: (a) stable *self-regulating group*, (b) unstable *self-regulating group*, (c) *compensatory structure* and (d) *multiple edges*.

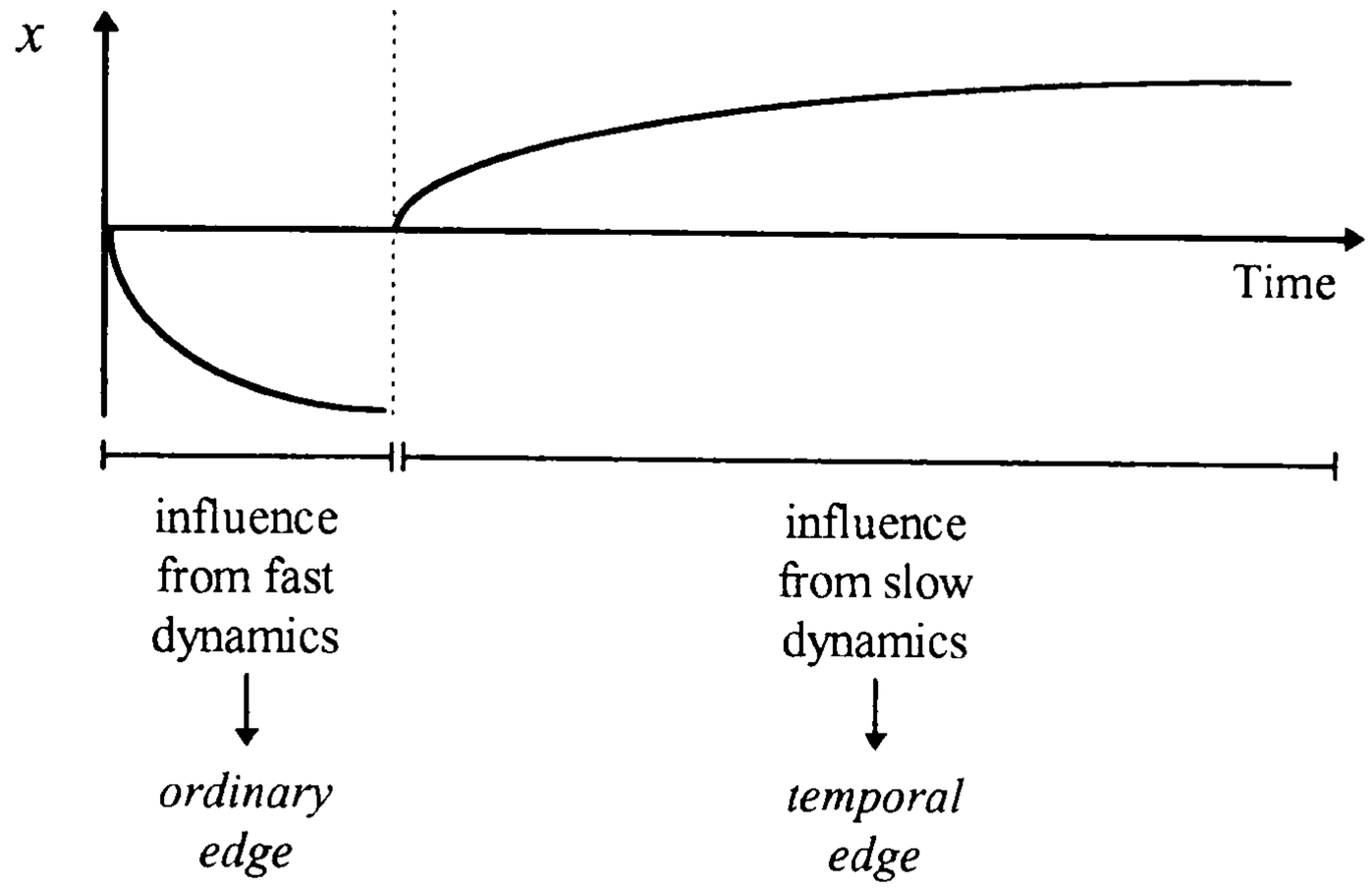


Figure 3.4 Scheduling of fast and slow responses.

In cases where the direction of the influences cannot be unambiguously determined by the algebraic and differential equations or heuristic rules, they can be inferred from the direction of flow of mass and energy or from experience.

3.5.3 Determination and Functionality of *Weights*

The weighting concept is used to reflect the relative sensitivity of the influences of the process variables and can be continuously updated based on experience or changes in process behaviour. It extends the SDG approach by allowing the edges to be mapped into the interval [-1,+1] instead of the double set {+,-} proposed originally. This has the advantage of improving the knowledge content of the qualitative model and formalising knowledge about the relative strength of the influences affecting the analysed process without requiring detailed calculations. This, coupled with other features, brings several advantages to the WDG approach, including the reduction in the number of ambiguous solutions and the ability to model the shape of monotonic and non-monotonic functions, which is not possible with existing qualitative reasoning techniques.

Generally, *weights* are attributed values of -1 and +1 to represent negative and positive influences, respectively. However, in situations involving conflicting influences, i.e. influences with the same strength but opposite signs, the *weights* receive values in the interval [-1,+1] in accordance with their relative strength. If the relative strength of the influences varies with certain system conditions, the values of the *weights* must change, according to the situation, and a structure with *multiple edges* of different *weights* used. These situations are identified by experience or by examining the signs and/or order-of-magnitude of the derivatives O_i^j together with changes in the order-of-magnitude of certain variables and signs of parameters. For instance, the *weights* of the *multiple edges* describing the possible qualitative behaviour of a CSTR are determined by analysing the possible influences that changes in temperature and concentration may have on the energy balance equation and the signs of the derivatives O_i^j obtained from this equation. This case will be discussed in detail in Chapter 4.

From the above it is seen that *weights* are determined by:

- Experience;
- Observed behaviour of an existing plant;
- Analysing the signs and/or order-of-magnitude of the derivatives O_i^j in relation to ranges of variation of specific variables and signs of parameters.

Functional weighting is an effective means of avoiding ambiguous solutions, since it can be used to break loops and reshape cyclic influences embedded in the digraph structure. Some usual situations of potential generation of ambiguous behaviour which can benefit from the use of *weights* are described below, although other situations also arise during model formulation:

- **Conflicting influences in loops** - Figure 3.5a illustrates a situation where a disturbance in a variable *A* would potentially generate a conflict

represented by two possible different types of behaviour associated with the same variable C due to the loop containing conflicting paths. The conflict can be solved by attributing a smaller *combined weight* to the path of the relatively weaker influence. In Fig. 3.5a, it is assumed that the path of constant B has stronger influence on C than the one where B changes with disturbances in A. Therefore, the ambiguous behaviour of C is avoided;

- ***Self-regulating groups*, feedback control loops and compensatory responses (cycles) -** These also can potentially generate ambiguous solutions. The use of different values for the *weights* of the active (*ordinary edge*) and reactive (*temporal edge*) influences solves the conflict. Figure 3.5b illustrates a *self-regulating group* for a first-order response, where the *weight* of the reactive influence is smaller than that for the active one;

- **Complex model structures -** Models generated for distributed parameter systems, mainly those which involve countercurrent flows, such as distillation columns, are characterised by complex structures which frequently give rise to conflicts and problems of convergence during the inference procedure used in describing process behaviour. In such cases, the process structure has to be analysed in order to identify situations which can result in different values for the *weights* and so reduce model complexity. For example, in a distributed parameter system the distance from the source of the disturbance plays a very important role in the strength of the influences and consequently on the shape of the responses. In such cases, the values of the *weights* need to vary according to the distance from the source of the disturbance, as will be illustrated in Chapter 4.

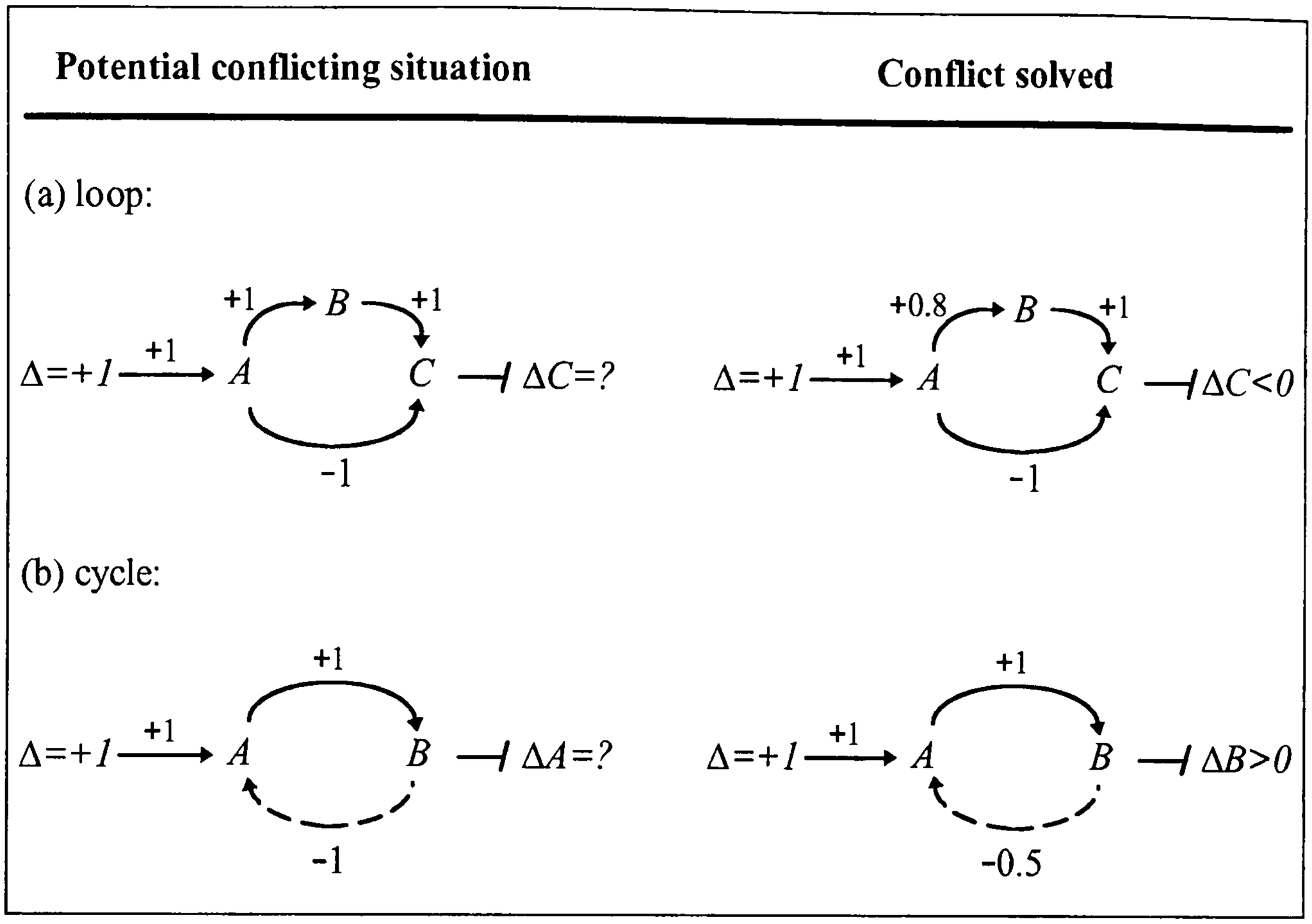


Figure 3.5 Conflicting situations and the use of *weights*: (a) loop and (b) cycle.

Functional weighting plays a major role in the description of the shape of system trajectories. Most qualitative models cannot distinguish between different shapes of monotonic functions. For example, exponential, linear and logarithmic functions are all simply described as *monotonic increasing functions* and represented by a line with positive slope. In the WDG procedure the proper use of *weights* coupled with *compensatory* or *self-regulating* structures enables different shapes of several monotonic functions to be described. Moreover, the use of *weights* having adjustable values depending on the qualitative state of a reference variable can effectively describe non-monotonic functions. A first-order system subject to a positive step disturbance in the input variable v (Fig. 3.6a) is used to illustrate the application of functional weighting to describe the shape of the trajectory. The response u shown in Fig. 3.6b is a first-order exponential rise to the new steady-state. This kind of response is characterised by a time constant, i.e. the time to reach 63.2 percent

of its new steady-state value (Luyben, 1990). It can be seen from Fig. 3.6 that the output variable *u* changes more slowly than the input variable *v* and the derivative is positive and decreases with time. This shape is simulated by the use of a *self-regulating structure* where the *weight* of the reactive influence (*reactive weight*) is smaller than the *weight* of the active one (*active weight*). The value of the *reactive weight* determines the speed of the response and can be seen as a time constant. The greater the *reactive weight* the faster the response. Figure 3.3 shows this and other WDG structures and the shapes of functions they represent, for a positive step disturbance in the input variables. These curves are generated by applying the inference algorithm described in section 3.6.

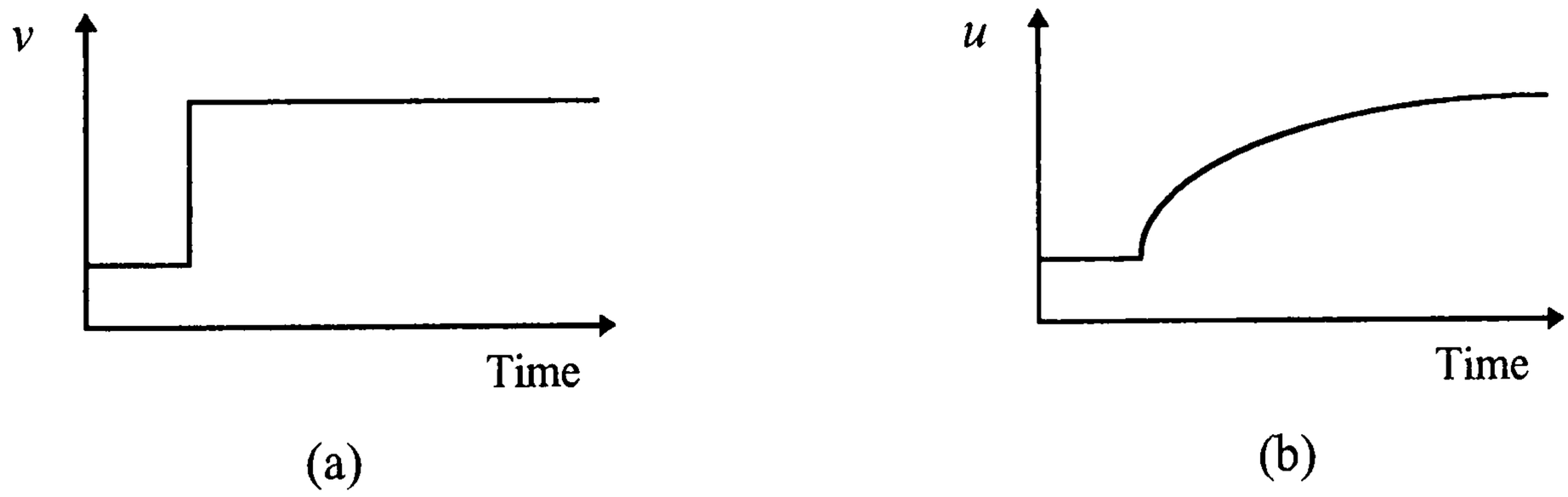


Figure 3.6 Qualitative first-order response: (a) step disturbance in the input variable *v* and (b) response of variable *u*.

3.5.4 The Build up of the Model Structure

The appropriate level of detail of a qualitative representation depends on the objectives of the model, the region of the solution space to be described and the accuracy required.

Shen and Leitch (1992) define *model resolution* as the number of variables incorporated within a particular model. The concept is extended by also considering

the number of paths between process variables, since there may be several possible paths between a limited number of variables.

Chemical processes are usually described by a large number of variables and functional relationships. The direct translation of this into a qualitative model results in a highly connected structure, which is so complex that the visualisation of flow of information and understanding of process behaviour is virtually impossible. Moreover, the possibility of generating ambiguous solutions is increased significantly. Therefore, simplifications involving the weakening of functional dependencies and reduction in the number of variables are necessary to reduce the complexity of the model. However, these simplifications must not compromise the description of important features which characterise the process behaviour.

Less detailed (lower resolution) models are obtained by neglecting influences (paths) or aggregating variables. A common simplification is to ignore some fast dynamics by assuming that some variables respond instantaneously to process disturbances. This leads to the elimination of some *differential nodes* and their related edges.

The simplification of model structure by eliminating redundant influences, cycles or loops must involve a careful analysis, since apparent duplicated relationships may carry different information which may be crucial in characterising the dynamics and describing complex patterns of process behaviour. For example, the relation : $a \xrightarrow{-w} c$ is included in the graph: $a \xrightarrow{+w} b \xrightarrow{-w} c$, since the negative path between a and c is also represented in the latter structure ($w_{a,c} = w_{a,b}.w_{b,c}$). However, if the influence between a and c exists even when b is constant, the first graph is essential in describing this condition and should not be eliminated.

Clearly, variables which do not improve the understanding of the problem and add complexity to the graph structure should be avoided. For instance, if the coolant

temperature (T_C) of a heat exchanger increases, the heat removed (Q) tends to decrease and consequently the hot fluid temperature (T_H) tends to increase. This information can be represented by the digraph: $T_C \xrightarrow{-w} Q \xrightarrow{-w} T_H$, but depending on the problem, the variable Q can be omitted which simplifies the digraph structure without altering the information content: $T_C \xrightarrow{+w} T_H$.

Once the appropriate resolution of the model has been defined, the causal relationships are put together to build up the weighted digraph structure. Some structures usually appear in WDG models and are characterised by special names, as shown in Fig. 3.3.

3.5.5 Representation of Distributed Parameter Systems

The weighted digraph (WDG) model of a distributed parameter system (DPS) is a three-dimensional structure where time and spatial dependencies are taken into account by assuming a multi-layer approach (Vianna and McGreavy, 1995a, b), as depicted in Fig. 3.7.

Space (position coordinate) is divided by cells usually linked by ordinary edges. Cells and linking edges present the following characteristics:

- Edges that link different cells are *incident to* and *emergent from* nodes that represent the variables on the left-hand side of Eqs. (3.1) and (3.2);

- Cells are macroscopic entities dedicated to the representation of sections of the system characterised by similar qualitative behaviour. For instance, a distillation column is divided into five cells which represent the reflux drum, rectifying section, feed tray, stripping section and bottom, since each of these sections has a typical behaviour. A counter-current heat exchanger is represented by one only cell, since all the solution space is

characterised by the same monotonic functions and there is no point in dividing the equipment into cells with identical qualitative behaviour. Each cell contains a digraph structure which characterises the behaviour of the particular section of the system. If the sections are described by different relations, the cells must contain related qualitative models. However, this is not usually the case and the cells contain mainly the same digraph structure which may differ only in the value of certain *weights*.

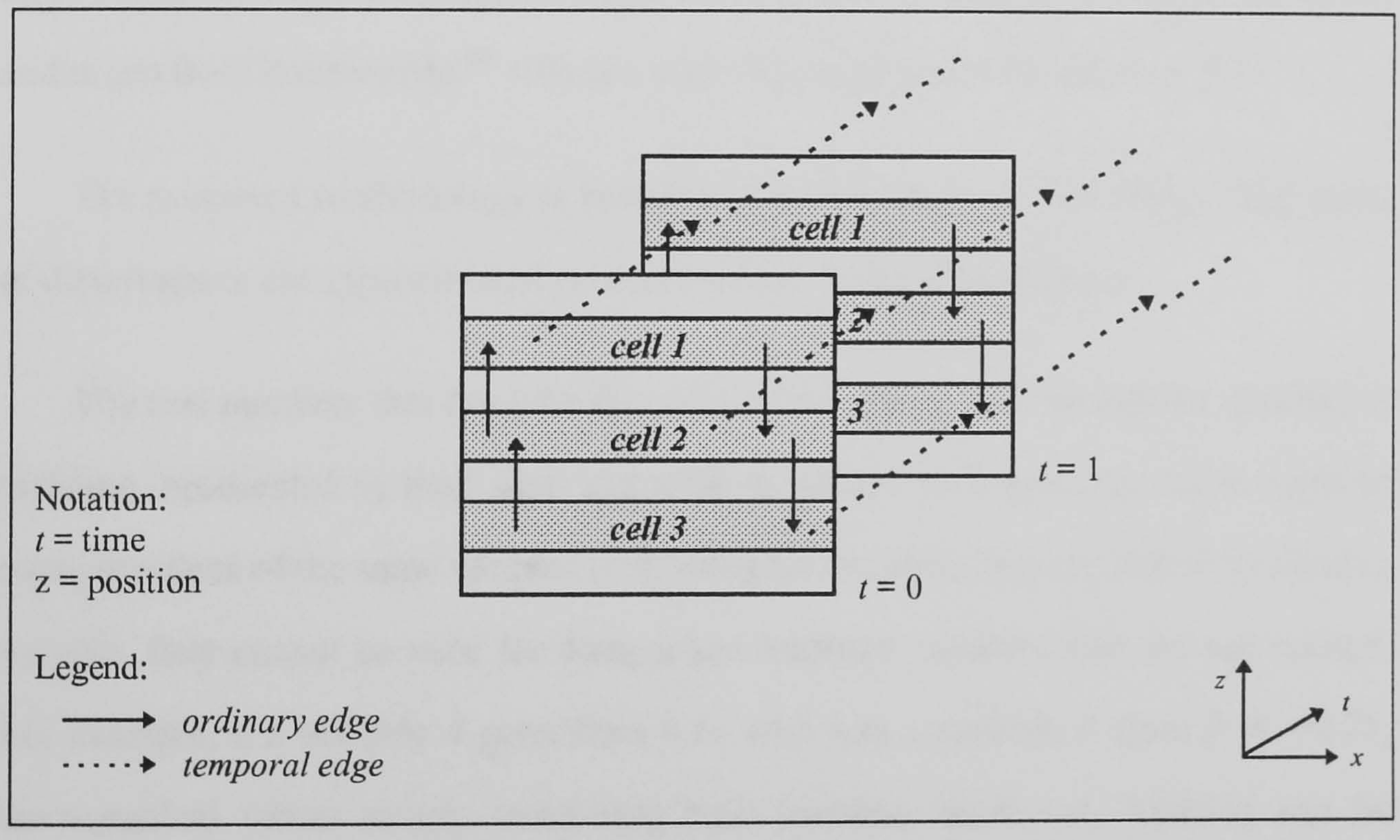


Figure 3.7 Schematic representation of distributed parameter systems.

The basic digraph structure composed of nodes and *ordinary edges* is repeated at each time level. Consecutive time levels are linked by *temporal edges*. As the basic model is repeated at each time level, a bidimensional representation where *ordinary* and *temporal edges* are drawn with different line styles and in the same plane is commonly used to flatten the 3D structure. *Temporal edges* are represented as dashed arrows while *ordinary edges* are shown as full arrows.

The notion of time step is introduced to impose movement through consecutive time levels with the aim of mimicking an explicit numerical integration algorithm based on Euler's method. The actual numerical value is not important and usually considered as 1.

3.5.6 Characterisation of the State of Process Variables

States can be defined as snapshots of the variables in time. The propagation of disturbances through the graph structure takes place by changing the state (θ) of the nodes (p_i) from inactive ($\theta p_i^{(0)} = 0$) to a real ($\Re$) numerical value ($\theta p_i^{(t)} \in \Re$).

The proposed methodology is based on step disturbances. Therefore other types of disturbances are approximated as a succession of step disturbances.

The real numbers that describe the state of the nodes have mainly the qualitative meaning represented by their signs and relative values. Although they can be used to compare states of the same variable or of different variables that are linked by relative *weights*, they cannot be used for comparison between variables that are not related. For example, if a variable *A* goes from 0 to +0.5 and a variable *B* from 0 to +0.75, the numerical values simply mean that both variables increased. Nothing can be concluded about their relative rate of increase, i.e. *B* is not necessarily greater than *A* in the final state. The initial state (0) means that both variables started from their usual normal states before the disturbance. Each non-related variable changes in accordance with its own scale. If a comparison between variables is desired, they have to be put on a single scale by referring to a common base, or *weights* may be applied to ensure normalisation of the influences. In this study only *weights* are used.

In general, positive or negatives numbers for *algebraic nodes* mean that the new state of the variable is greater or smaller, respectively, than the previous state. Zero means that the state of the variable is unchanged. For *differential nodes* and

variations in the state of nodes, $\Delta(\theta p_i^{(t)})$, positive, zero or negative numbers imply increase, constant or decrease, respectively.

The state of a node i at time t ($\theta p_i^{(t)}$) is calculated by adding to its past state value the value of the disturbance (Δ), as shown in Eq. (3.9). An exception is made for *self-regulating groups*, as described later in this section.

$$\theta p_i^{(t)} = \Delta(\theta p_i^{(t)}) + \theta p_i^{(t-1)} \tag{3.9}$$

The absolute value of the change in the state of a node i at time t ($|\Delta(\theta p_i^{(t)})|$) is determined by Eqs. (3.10) and (3.11) and means that the stronger influence will always prevail. Influences with the same but opposite strengths will result in a null influence. The sign of $\Delta\theta$ is equal to the sign of the prevalent influence.

$$\left|\Delta(\theta p_i^{(t)})\right| = max\,(\ldots max\,(max\,(A_1, A_2), A_3), \ldots, A_j) \tag{3.10}$$

$$A_j = \left|w_{ji}.\Delta(\theta p_j^{(\tau)})\right| \tag{3.11}$$

where $i = (1,2,\ldots, N)$,

$j = (1,2,\ldots, NI)$,

N = number of nodes,

NI = number of *ascendant nodes* of p_i,

θ = state of the node,

$\Delta\theta$ = variation in the state of the node,

τ = time t if the edge from p_j to p_i is an *ordinary edge* or past time (t-Δt) if it is a *temporal edge*.

Since the *weights* w_{ij} and the state θ are real numerical values, the mathematical operations involving them obey the rules of conventional algebra.

If the node p_i is part of a *self-regulating group*, $(\delta x_i, x_i)$, changes in the state $(\Delta\theta)$ of nodes δx_i and x_i are determined by Eqs. (3.10) and (3.11). The state of δx_i is calculated by Eq. (3.9), while the state of x_i depends on the type of the *ascendant node* which influence is prevalent. If this comes from a node different from δx_i, the state of x_i is also calculated by Eq. (3.9), but if it comes from δx_i, the state θ and not the variation of it $(\Delta\theta)$ is used to determine the state of x_i, as shown in Eq. (3.12), which is the qualitative equivalent to Eq. (3.5):

$$\theta x_i^{(t)} = \theta\delta x_i^{(t)} + \theta x_i^{(t-1)} \tag{3.12}$$

Using a qualitative analogy with Eq. (3.5), the following algebra may be derived:

$$\theta\delta x_i^{(t)} = \theta x_i^{(t)} + \theta x_i^{(t-1)} \tag{3.13}$$

$$\Delta(\theta\delta x_i^{(t)}) = \theta\delta x_i^{(t)} + \theta\delta x_i^{(t-1)} \tag{3.14}$$

Analysing Eqs. (3.13) and (3.14) the following equivalence is obtained:

$$\Delta(\theta\delta x_i) \Leftrightarrow d^2 x_i \approx \Delta(\Delta x_i) \tag{3.15}$$

$$\theta\delta x_i \Leftrightarrow dx_i \approx \Delta x_i \tag{3.16}$$

From Eqs. (3.15) and (3.16) it can be concluded that second-order derivatives $(d^2 x_i)$ are implicitly represented in the WDG structure. This is a significant improvement over the conventional SDG approach, since the constraint of the solution space by using information about higher-order derivatives is essential in reducing the number of ambiguous behaviour and eliminating spurious solutions. For example, if $\theta x_i > 0$ and $\theta\delta x_i > 0$, there are three possible behaviour for x_i : (1) increase at a non-constant positive rate; (2) increase at a constant rate; and

(3) increase at a non-constant negative rate. However, if it is known that $\Delta(\theta\delta x_i) < 0$, behaviour (1) and (2) can be eliminated and only behaviour (3) is possible, i.e. the system tends to stabilise at a new steady-state.

Equations (3.9) to (3.14) are the qualitative equations which mathematically describe a weighted digraph model.

3.6 Inference of Qualitative Behaviour from Weighted Digraphs

Once the model structure has been defined a problem-solving technique has to be applied not only to infer behaviour from the information embedded in the digraph structure, but also to help with the visualisation of the flow of information through the graph structure and explanation of how and why solutions are generated.

This section is dedicated to the description of a procedure for inference of dynamic behaviour from WDG structures. No attempt is made to establish a visualisation procedure or an explanatory methodology.

The inference procedure uses a node-to-node propagation algorithm based on a modification of the depth-first search algorithm (Tarjan, 1972).

3.6.1 Procedure for Propagation of Disturbances

A list (*edge-listing*) containing nodes and edges connections, types of nodes and edges, and values of *weights* is used. Nodes and edges receive identification numbers. The propagation procedure works forwards or backwards.

The control strategy starts by identifying active *input nodes*, i.e. nodes with non-zero states. It propagates the step changes in the states of the *input nodes* throughout

the model structure by calculating changes in the states of the *descendant nodes*, based on Eqs. (3.10) and (3.11). If a node has more than one *emergent edge*, the algorithm follows the edge which leads to the *descendant node* with the lower identification number, unless this has already been evaluated or receives other edges from nodes not yet evaluated. Evaluated nodes have their states determined by Eqs. (3.9) or (3.12). Non-evaluated nodes are put on a stack for later assessment. The state of the nodes and any changes are calculated as described in section 3.5.6. *Mutually exclusive edges* are activated in accordance with the direction of propagation of the disturbance. *Temporal edges* are only taken into account in relation to the nodes they are *incident to*, since by definition they carry information from a past time. The procedure is terminated when all nodes have been evaluated.

The propagation procedure can be explained by reference to Fig. 3.8. Suppose that nodes 1,2 and 3 are *input nodes* so their states are known. Node 1 is activated. The control strategy proceeds to calculate the state of the *descendant node* 4. This node receives two more edges from nodes 6 and 8 which are not yet evaluated. The edges linking nodes 4 and 6 are *mutually exclusive* and, as the flow is from 4 to 6, the edge from 6 to 4 is deactivated. The edge from 8 is *temporal* so carries information from the previous value of 8, which is already known (the states of all internal nodes are initialised as zero). Therefore node 4 can be evaluated and the control strategy can proceed to one of the two *descendant nodes* 5 or 6. The one with lower identification number (node 5) is chosen and evaluated, while node 6 is put on the stack to be evaluated later on. Node 9 is the direct *descendant* of node 5, but cannot be evaluated yet because the present state of 8 is still unknown, so 9 is also put on the stack. As there is not any other *descendant node* from 5, the control strategy looks for the first node on the stack, which is node 6. It evaluates the state of this node and subsequently those of nodes 7 and 8, since node 3 is an *input node* and consequently already evaluated. By definition, the *temporal edge* that is

emergent from node 8 is not considered at this stage. Finally, node 9 can be evaluated and the search is terminated.

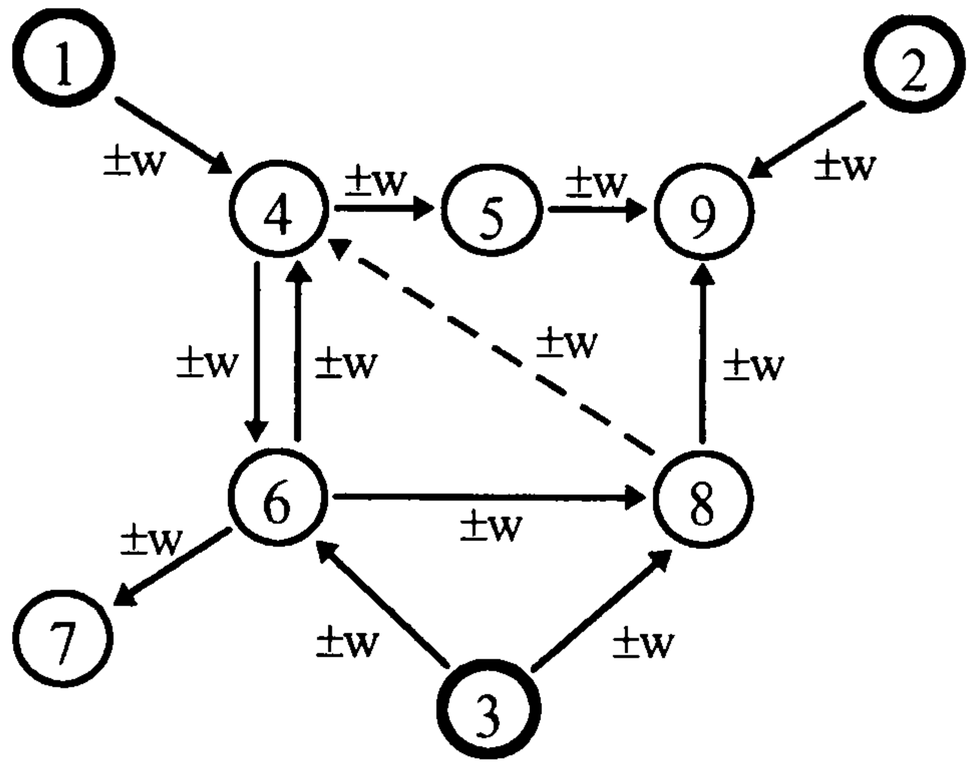


Figure 3.8 Digraph for illustrating the procedure of propagation of disturbances.

3.6.2 Inference Algorithm

The inference algorithm uses a qualitative analogy of the explicit numerical integration approach based on Euler's method for solving ordinary differential equations, which is qualitatively expressed in Eqs. (3.9) and (3.12). The algorithm steps out in time and does not involve iteration. A unitary time step is assumed. The procedure involves the calculation of the state of the nodes which comprise the WDG model at each time t, based on the previous state at (t-1) and step disturbances introduced by the *input nodes*, which are propagated node-to-node through the graph structure by the previously described propagation procedure.

Convergence is reached when the relative variation in the states (θ) of the nodes between any two consecutive time steps is smaller than a given tolerance (*tol*):

$$\varepsilon_i = \left| \frac{\theta_i^{(t)} - \theta_i^{(t-\Delta t)}}{\theta_i^{(t)}} \right| \leq tol \qquad (3.17)$$

The algorithm is described as follows and schematically represented in Fig. 3.9:

1. Initialisation of time (0) and designation of all variables as being inactive: $\theta p_i^{(0)} = 0$;

2. Reading of the *edge listing* describing the model structure;

3. Reading of the changes in the states of the *input nodes*;

4. Creation of the *identification numbers list* for the nodes and edges;

5. Step in time: $t=t+1$;

6. Calculation of the state of the nodes at time t by using the propagation procedure (section 3.6.1) and Eqs. (3.9) to (3.14). If a node cannot be evaluated it is put on the stack;

7. If there is another node on the stack, return to stage 6;

8. Checking for convergence using Eq. (3.17);

9. If convergence is achieved, the program stops; if not the control returns to stage 5.

3.7 Gravity-Flow Tank - Case Study

To illustrate the approach, a gravity-flow tank subject to a step disturbance in the inlet flow rate is used. As already shown in Chapter 2, the conventional SDG approach is unable to describe dynamic responses of chemical processes. This case study provides an example of the additional capabilities of the weighted digraph approach.

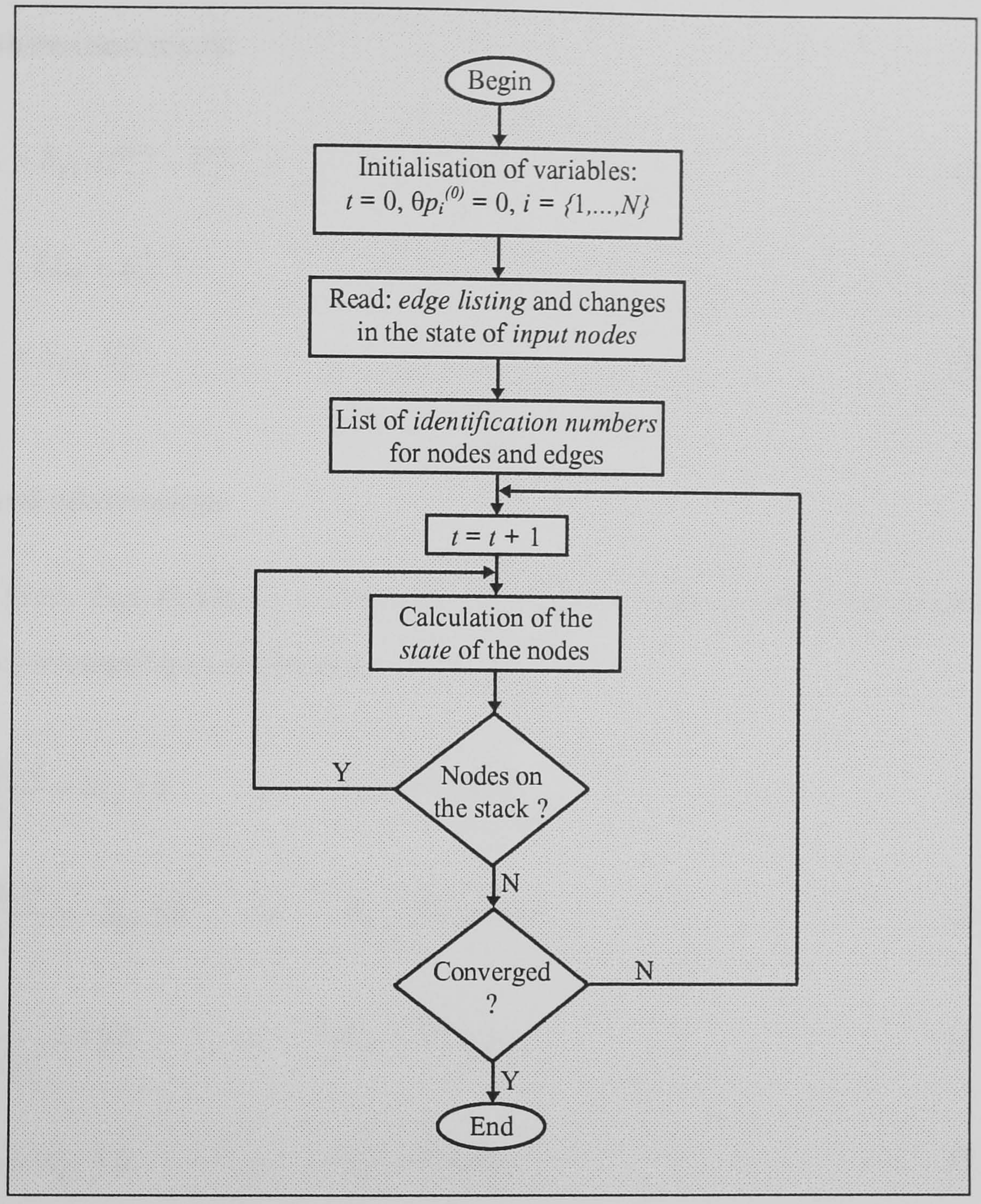


Figure 3.9 Inference algorithm.

Knowledge acquisition

Consider the tank shown in Fig. 3.10a and described by Eqs. (3.18) to (3.20), where F_i, F_o and L_T are the volumetric inlet flow rate, volumetric outlet flow rate and level, respectively. Assume that θF_i is the qualitative state of the inlet flow rate, θF_o is the qualitative state of the outlet flow rate, θL_T is the qualitative state of the level in the tank and $\theta \delta L_T$ is the state of the rate of change of the level with time.

Mathematical model

$$\delta L_T = S_{T1} \cdot (F_i^{(t)} - F_o^{(t-1)}) \tag{3.18}$$

$$L_T^{(t)} = \delta L_T + L_T^{(t-1)} \tag{3.19}$$

$$F_o^{(t)} = S_{T2} \cdot L_T^{(t)} \tag{3.20}$$

Causal relationships

From Eqs. (3.18) to (3.20), the following derivatives and consequently the causal relationships are obtained:

$$\frac{\partial(\delta L_T)}{\partial F_i} = S_{T1} \rangle 0 \qquad \Rightarrow \qquad F_i \xrightarrow{+w} \delta L_T \tag{3.21}$$

$$\frac{\partial(\delta L_T)}{\partial F_o} = -S_{T1} \langle 0 \qquad \Rightarrow \qquad F_o \xrightarrow{-w} \delta L_T \tag{3.22}$$

$$\frac{\partial L_T}{\partial(\delta L_T)} = 1 \rangle 0 \qquad \Rightarrow \qquad \delta L_T \xrightarrow{+w} L_T \tag{3.23}$$

$$\frac{\partial F_o}{\partial L_T} = S_{T2} \rangle 0 \qquad \Rightarrow \qquad L_T \xrightarrow{+w} F_o \tag{3.24}$$

Build up of the WDG model

Causal relationships are put together to build up the model structure. Simplifications are made whenever possible or necessary. If the above causal relationships are just assembled, the structure described in Fig. 3.10b is obtained. It can be seen that the edge (F_o, δL_T) is a potential source of conflict, since it is a *compensatory response* and therefore determines a loop, which is on the same time level of the edge (F_i, δL_T), and where both paths have *combined weights* with

opposite signs. This loop can be broken by attributing a *temporal edge* to (F_o,δL_T). Therefore Fig. 3.10c is the best model structure for the system.

Determination of *weights*

As the influence (F_o,δL_T) is a *compensatory response*, and it is known that the dynamics of the system is fast, the *weight* related to this influence can receive a relatively high value in absolute terms within the interval [-1,+1]. An arbitrary value of -0.75 is chosen, and therefore this edge can be described by the set: (F_o,δL_T,1,-0.75), which defines its direction, type and *weight*. All other edges receive *weights* of +1 or -1. The resulting weighted digraph model is shown in Fig. 3.10d.

Inference of Dynamic Behaviour

The inference algorithm is applied to the structure described in Fig. 3.10d. Table 3.1 is generated for a tolerance of 0.04. Figure 3.11 shows the qualitative dynamic trajectory of the level of the tank by using data from Table 3.1. Figure 3.12 shows the result from a numerical simulation based on the following parameters: $S_{T1} = 1.0\ m^{-2}$, $S_{T2} = 2.0\ m^2/h$, time step = 0.1, disturbance:$\Delta F_i = 1.0\ m^3$, and initial values: $L_T^{(0)} = 1.0$ m, $F_i^{(0)} = 2.0\ m^3/h$, $F_o^{(0)} = 2.0\ m^3/h$.

No attempt has been made to normalise the qualitative values of the variables so the numerical values generated by the qualitative simulation cannot be used to compare variables quantitatively. For example, the outlet flow rate in the new steady-state is not greater than the inlet flow rate. The qualitative value of +1.25 means that the value of F_o in the new steady-state is greater than its own value in the initial steady-state.

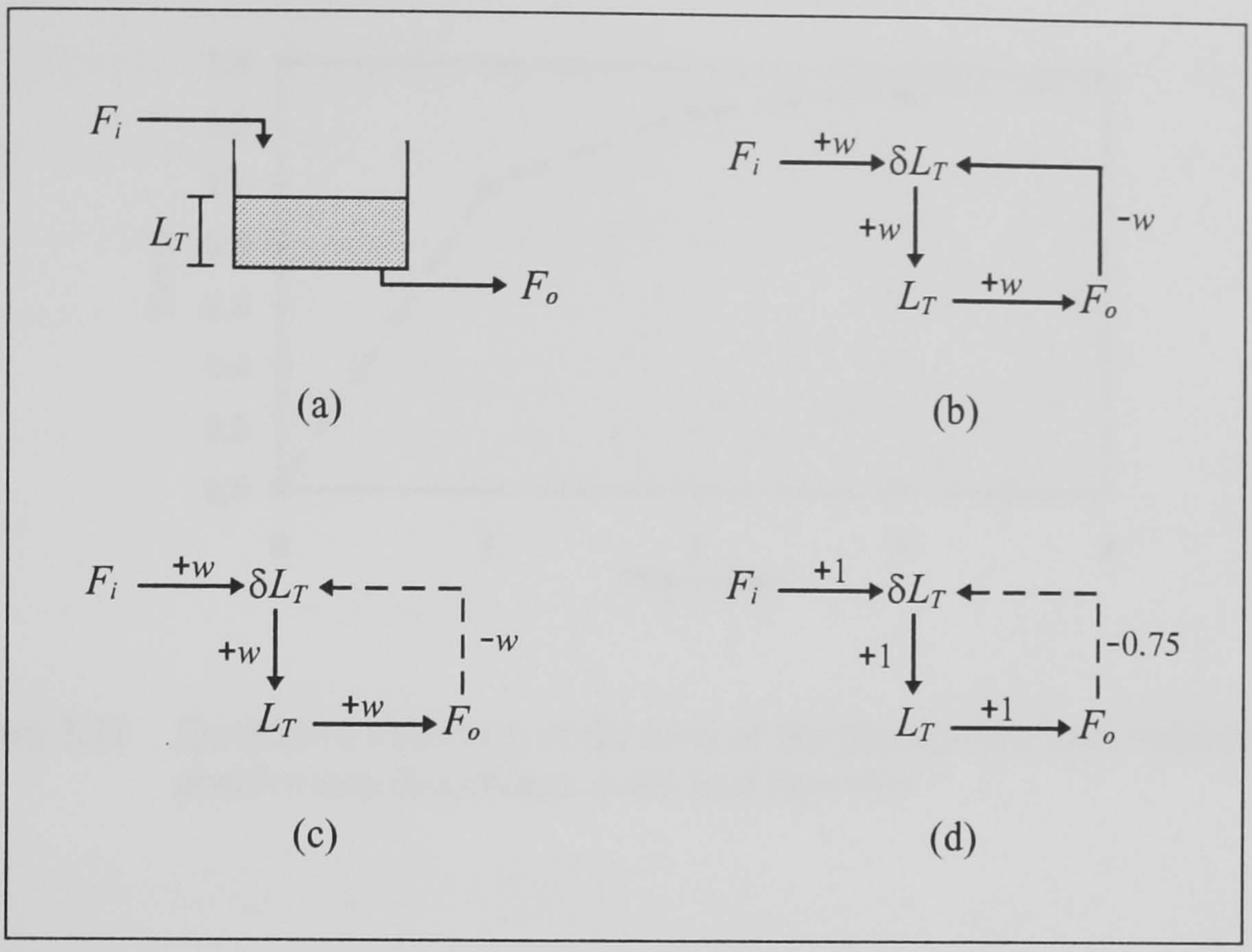


Figure 3.10 Gravity-flow tank: (a) schematic representation, (b) basic digraph structure, (c) use of *temporal edge*, and (d) weighted digraph.

Table 3.1 Qualitative results for the simulation of the gravity-flow tank subject to a positive step disturbance in the inlet flow rate.

	$t = 0$	$t = 1$	$t = 2$	$t = 3$	$t = 4$
$\Delta(\theta F_i)$	0.0	+1.0	0.0	0.0	0.0
θF_i	0.0	+1.0	+1.0	+1.0	+1.0
$\Delta(\theta \delta L)$	0.0	+1.0	-0.75	-0.188	-0.016
$\theta \delta L$	0.0	+1.0	+0.25	+0.063	+0.047
θL	0.0	+1.0	+1.25	+1.313	+1.360
$\Delta(\theta F_o)$	0.0	+1.0	+0.25	+0.063	+0.047
θF_o	0.0	+1.0	+1.25	+1.313	+1.360

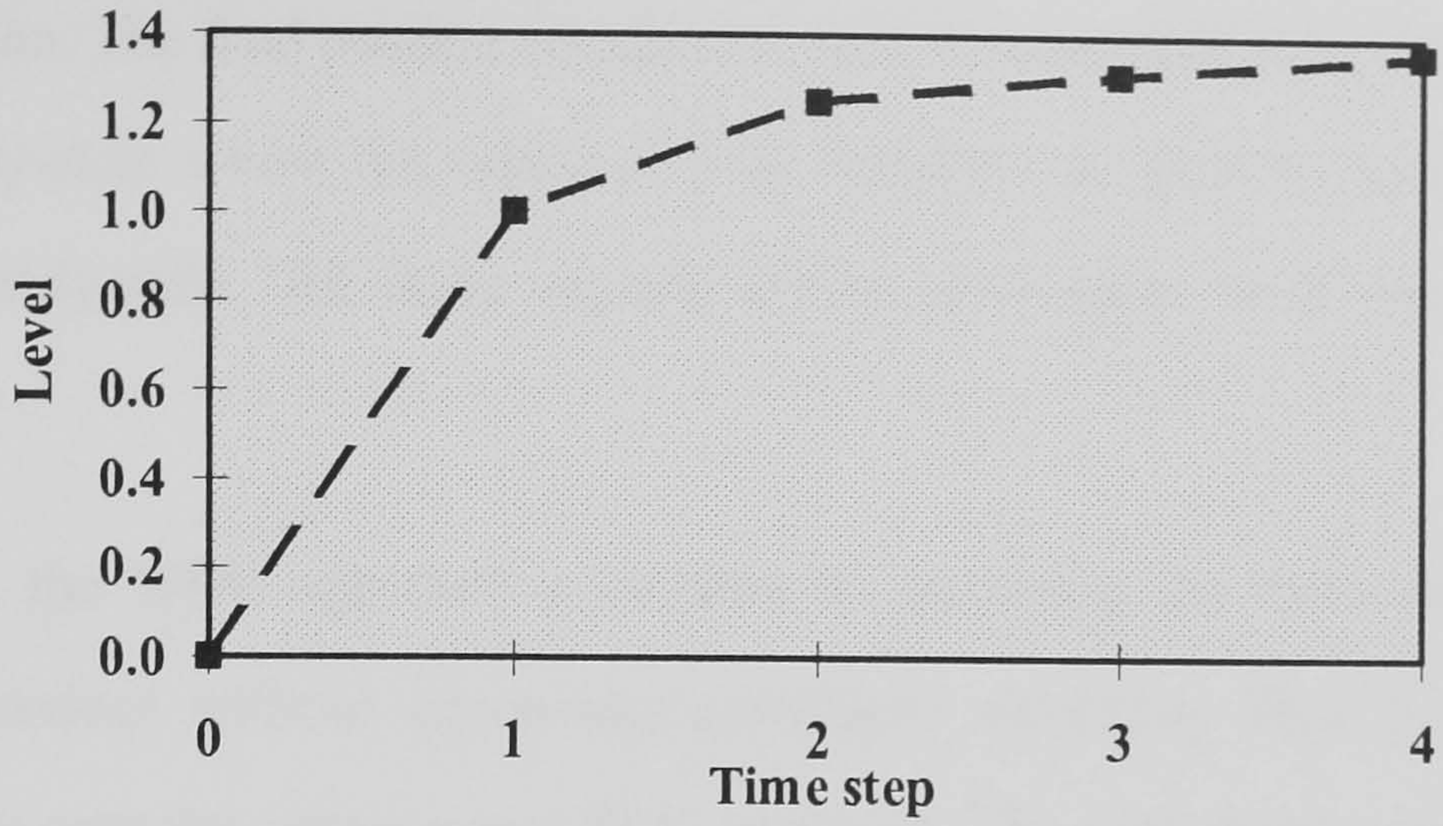


Figure 3.11 Qualitative trajectory of the level of the gravity-flow tank subject to a positive step disturbance in the feed flow rate.

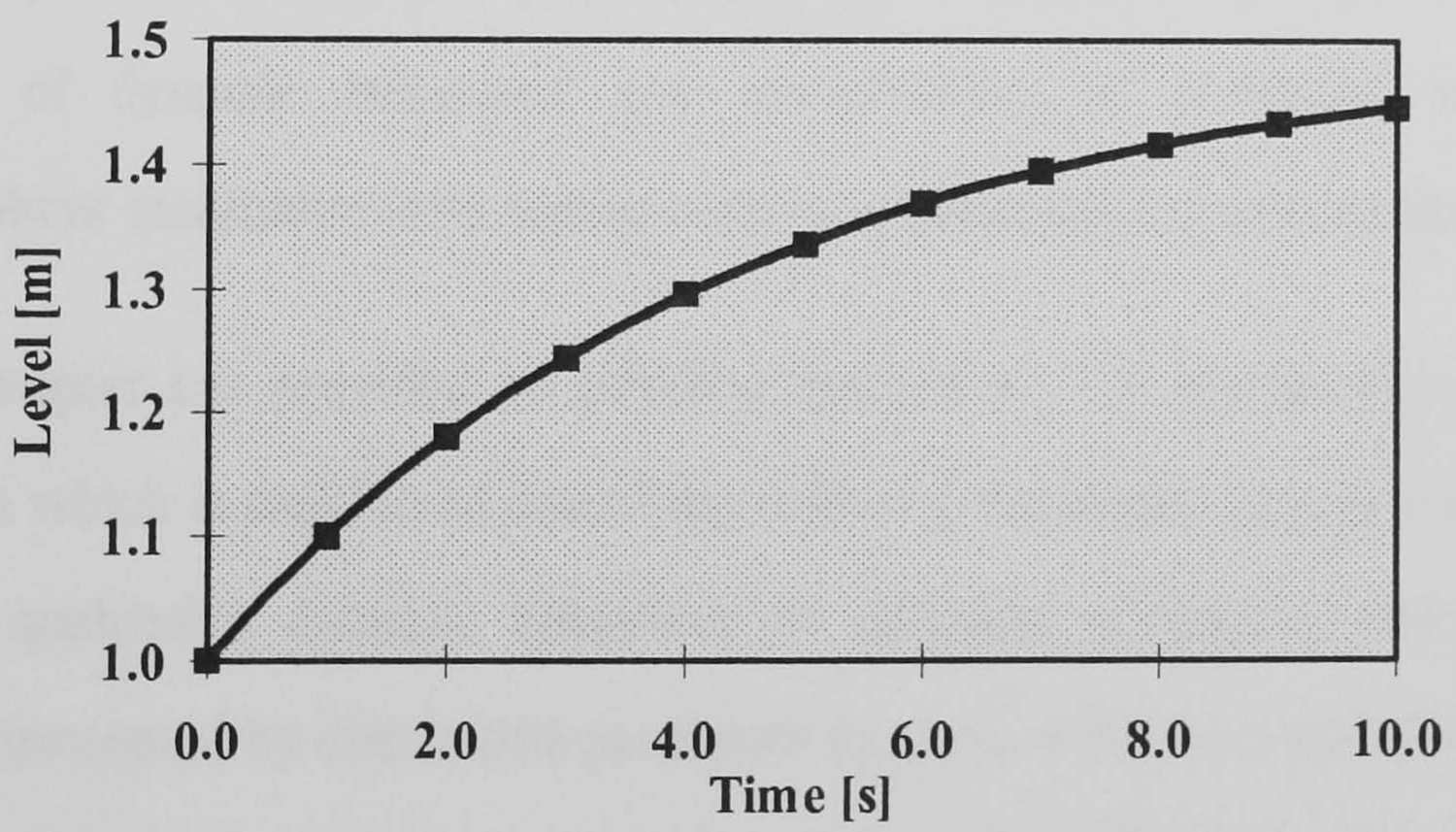


Figure 3.12 Dynamic trajectory of the level of the gravity-flow tank - numerical simulation.

The results presented in Table 3.1 and Figs. 3.11 and 3.12 show that the WDG procedure has the ability to capture the most important dynamic features of the system. The positive values of the first-order derivative ($\theta\delta L_T$) indicate that the level tends to increase, while the negative values of the second-order derivative ($\Delta(\theta\delta L_T)$) show that the rate of increase of the level tends to slow down with time, which leads

to stabilisation. The final positive values of L_T and F_o mean that the system will reach a new steady-state where the values of both variables are greater than those in the previous steady-state. All these aspects are in accordance with the real system behaviour.

Clearly, the WDG approach is capable of describing the dynamic patterns of process behaviour without generating ambiguous solutions. This is a significant improvement over the conventional SDG approach. The next chapter is dedicated to the analysis of more complex systems to provide evidence of the enhanced capability.

3.8 Concluding Remarks

Successful process design involves quantitative numerical simulation, including evaluation of dynamic behaviour and interpretation of graphical images, and reasoning about qualitative information used in synthesising an optimal strategy.

This chapter has provided an effective framework for representing qualitative information which is capable of describing different functional shapes and reasoning about the qualitative dynamic behaviour of chemical processes, including those elements represented by distributed parameter systems, with a considerable reduction in the generation of ambiguous solutions. These capabilities are provided by the combination of several features which include the explicit consideration of *differential variables*, the use of *temporal* and *multiple edges*, functional weighting, a multi-layer approach and a comprehensive qualitative state descriptor.

The weighted digraph approach successfully solves the problems not handled by conventional methods and extends the application of qualitative reasoning to a wide variety of chemical engineering problems. This represents a considerable help not only in applying qualitative reasoning, but also in gaining more insight into process design and operation. To be able to qualitatively describe dynamic trajectories of

chemical process and formalise the way in which information flow between process variables is a considerable advantage when carrying out process analysis and data interpretation, especially in early stages in process design where the majority of data and information are qualitative. This enables decisions to be taken about improvements in process performance, control and operating strategies based on the understanding of the interaction between process variables. The procedure also has the potential for being used in synthesis of operating procedures.

Chapter 4

Weighted Digraph Models for Chemical Processes

4.1 Introduction

A chemical plant is composed of a large number of units, such as heat-exchangers, tanks, reactors, pumps, distillation columns, etc., which are connected to each other and through which there is a flow of materials and energy. Most chemical engineering tasks, e.g. the design of control systems and generation of operating procedures, require a description (model) of each unit in order to make possible the analysis of process behaviour for different inputs or operating conditions.

In this chapter the weighted digraph (WDG) methodology is used to qualitatively describe the dynamic behaviour of several chemical processes. The systematic procedure of modelling systems of different levels of complexity, such as a heat-exchanger, reactor and distillation column, is presented and discussed. The aim is not only to generate qualitative models for process units which are going to be used in the following chapters to support the synthesis of operating procedures, but also to demonstrate the effectiveness of the weighted digraph approach in dealing with process dynamics of complex systems as well as revealing the flow of

information between process variables to allow the understanding of how these responses arise.

4.2 Process Modelling Strategy

The approach to be used is based on the decomposition of the chemical process into several process units (pieces of equipment), which may be further decomposed into sub-units. For example, a distillation column may be decomposed into flashes and heaters (condenser and reboiler). The description of units and sub-units is based on the fundamental laws (mass and energy balances, equilibrium relations, etc.) which translate information about the basic mechanisms (heat and mass transfer, reaction ,etc.) into the underlying process behaviour. The description of the whole plant is based on the integration of the several qualitative models describing the behaviour of each unit and sub-unit. Figure 4.1 depicts the process decomposition methodology.

The modelling of the process units, heat-exchanger, reactor and distillation column, is based on the WDG methodology previously described in Chapter 3 and that can be summarised in the following steps:

- Knowledge acquisition;
- Determination of causal relationships;
- Build up of model structure;
- Comparison of predicted with expected behaviour.

The phase of knowledge acquisition is based on mathematical models, whenever they are available and simple enough to reveal *structural causality*, or on experience, intuition or graphical representations of responses.

Weighted digraphs are developed for limited regions of the solution space and therefore simplifying assumptions and system boundaries need to be well defined.

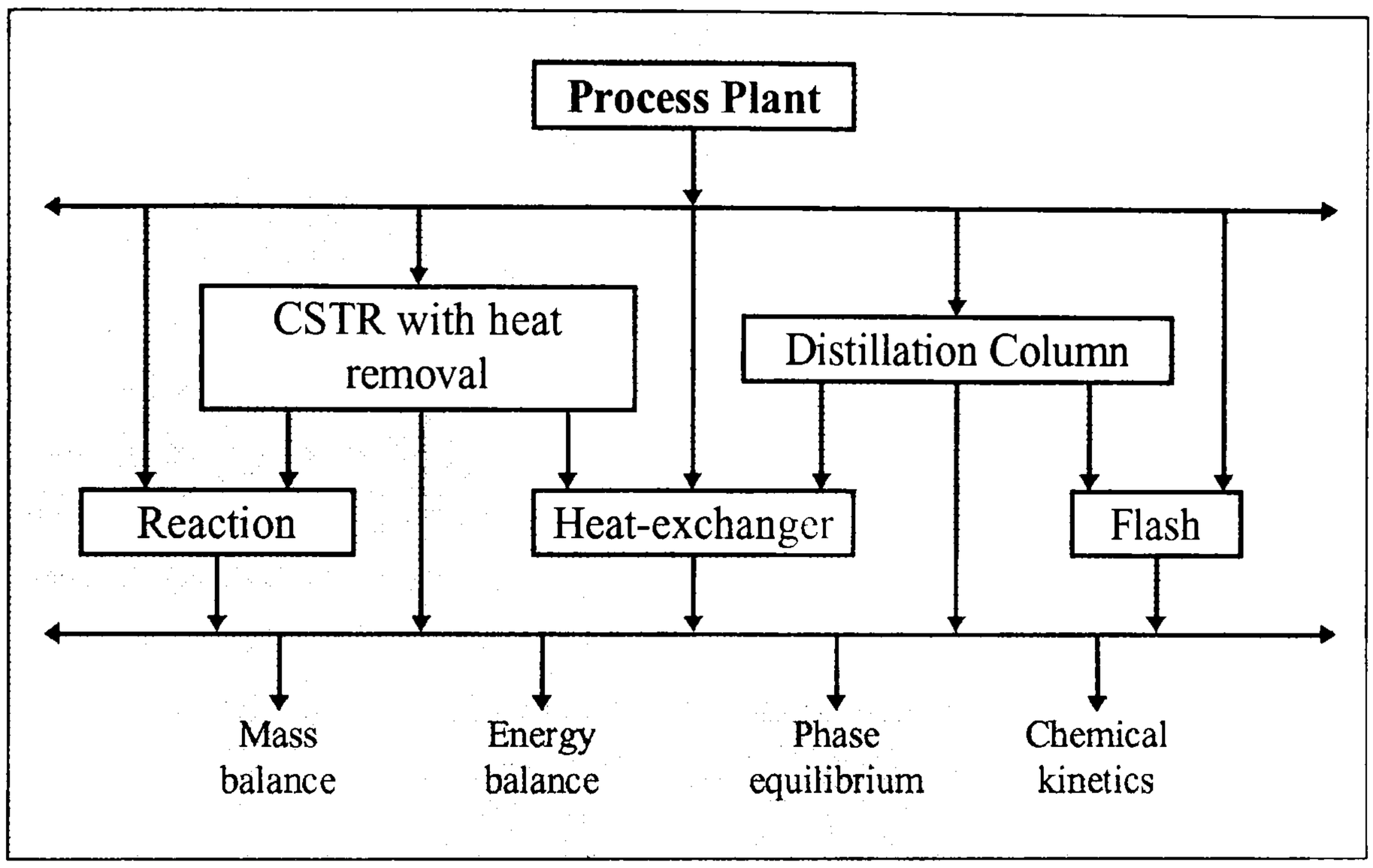


Figure 4.1 Decomposition of a process flow diagram.

4.3 Comparison of Qualitative and Quantitative Simulations

A WDG model is considered to adequately describe a chemical process when the results from a simulation matches that of the target plant or a quantitative (numerical) simulation. In this study, the latter is used because this enables detailed knowledge of the way in which responses develop to be obtained.

Qualitative trajectories are determined by applying the algorithm for inference of behaviour, described in Chapter 3, to the WDG models obtained in the following sections. Quantitative results are derived from solving the system of algebraic and differential equations that describes each process.

Numerical values generated by qualitative simulation cannot be analysed quantitatively, since they only reveal the general qualitative states of the variables. Scales of qualitative representations usually involve positive, zero and negative values, to represent states above, on and below desired or normal conditions, respectively. Therefore, numerical values of qualitative and quantitative simulations cannot be compared. Comparisons can only be made in terms of the general shape of dynamic trajectories. This means that only the most important features of the system behaviour can be taken into account, as for example turning points, tendency to approach the steady-state, oscillations and large deviations from normal operating conditions.

In the following sections WDG models for heat-exchangers, CSTRs and distillation columns are developed.

4.4 WDG Model for Shell-Tube Heat-Exchangers

Consider the shell-tube heat-exchanger shown in Fig. 4.2a. The hot fluid enters at temperature T_{Hi} and flow rate F_{Hi}, and leaves at temperature T_{Ho} and flow rate F_{Ho}. Similarly, the cold fluid enters at temperature T_{Ci} and flow rate F_{Ci} and leaves at temperature T_{Co} and flow rate F_{Co}.

4.4.1 Simplifying Assumptions and System Boundaries

The WDG model is based on the following considerations and assumptions:

- Pressure drop is considered negligible;
- Transient reversal of heat flow is ignored;

- In condensers and reboilers the following considerations are made:

  : Only latent heat is considered for the fluid which suffers phase change;

  : There is always enough hot fluid available to absorb changes on the cold side. If the equipment is the overhead condenser of a distillation column, it is assumed that there is always sufficient cooling water available to condense all vapour;

  : Condensation and vaporisation temperatures are assumed constant, even for mixtures;

- The overall heat transfer coefficient (U) is assumed to be constant;

- Uniform temperature across the radius of the pipe;

- The following boundary conditions are assumed:

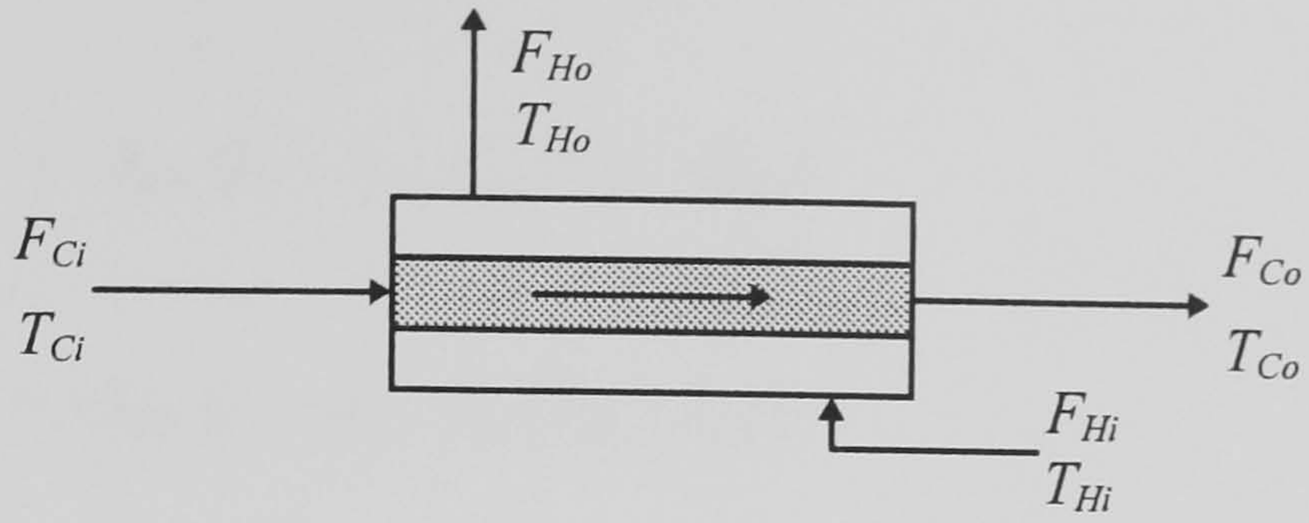
  : $T_{Hi} > T_{Ci}$;

  : $T_{Ho} > T_{Co}$;

  : $T, F > 0$;

- Dynamic trajectories of temperature as a function of position, for heat-exchangers subject to a single step disturbance in any input variable, are monotonic. This means that the qualitative state of temperature can be considered as independent of position and so these equipment can be described by one single qualitative space cell, as defined in Chapter 3. The implication is that only time derivatives are included in weighted digraph models and a unit length is used.

(a)



(b)

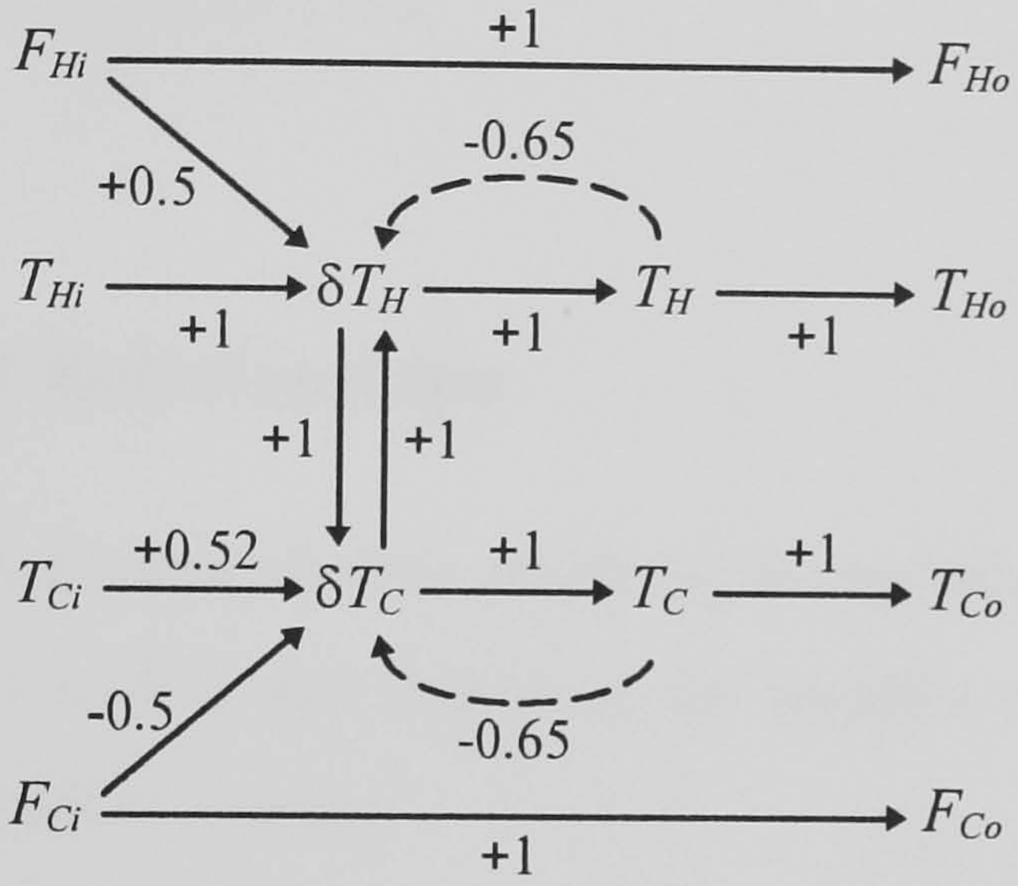


Figure 4.2 Shell-tube heat-exchanger: (a) schematic representation and (b) weighted digraph model.

4.4.2 Mathematical Model

The following mathematical relations for describing the behaviour of heat-exchangers are based on models described by Stephanopoulos (1984). An arithmetic mean is assumed for the temperature differences used for calculating the heat transfer rate (Q_T). Constant terms, such as heat transfer area (A), overall heat transfer coefficient (U), etc., are grouped into the positive constants S_{E1} to S_{E5}, described in Appendix A.

$$F_{Hi} = F_{Ho} \tag{4.1}$$

$$F_{Ci} = F_{Co} \tag{4.2}$$

$$\delta T_{Ho} = \frac{dT_{Ho}}{dt} = -S_{E1}\ Q_T + S_{E2}\ F_{Hi}\left(T_{Hi} - T_{Ho}\right) \tag{4.3}$$

$$\delta T_{Co} = \frac{dT_{Co}}{dt} = +S_{E3}\ Q_T + S_{E4}\ F_{Ci}\left(T_{Ci} - T_{Co}\right) \tag{4.4}$$

$$Q_T = S_{E5}\ \frac{\left(T_{Hi} - T_{Co}\right) + \left(T_{Ho} - T_{Ci}\right)}{2} \tag{4.5}$$

4.4.3 Causal Relationships

Causal relationships are determined by calculating the derivatives of the left-hand side terms of Eqs. (4.1) to (4.5) with respect to the variables on the right-hand side, keeping all other variables constant:

$$\frac{\partial\left(\delta T_{Ho}\right)}{\partial F_{Hi}} = S_{E2}\left(T_{Hi} - T_{Ho}\right) \rangle\ 0 \quad \Rightarrow \quad F_{Hi} \xrightarrow{+w} \delta T_{Ho} \tag{4.6}$$

$$\frac{\partial\left(\delta T_{Ho}\right)}{\partial T_{Hi}} = S_{E2}\ F_{Hi} \rangle\ 0 \quad \Rightarrow \quad T_{Hi} \xrightarrow{+w'} \delta T_{Ho} \tag{4.7}$$

$$\frac{\partial\left(\delta T_{Ho}\right)}{\partial T_{Ho}} = -S_{E2}\ F_{Hi} \langle\ 0 \quad \Rightarrow \quad T_{Ho} \xrightarrow{-w'} \delta T_{Ho} \tag{4.8}$$

$$\frac{\partial\left(\delta T_{Ho}\right)}{\partial Q_T} = -S_{E1} \langle\ 0 \quad \Rightarrow \quad Q_T \xrightarrow{-w} \delta T_{Ho} \tag{4.9}$$

$$\frac{\partial\left(\delta T_{Co}\right)}{\partial F_{Ci}} = S_{E4}\left(T_{Ci} - T_{Co}\right) \langle\ 0 \quad \Rightarrow \quad F_{Ci} \xrightarrow{-w} \delta T_{Co} \tag{4.10}$$

$$\frac{\partial\left(\delta T_{Co}\right)}{\partial T_{Ci}} = S_{E4}\ F_{Ci} \rangle\ 0 \quad \Rightarrow \quad T_{Ci} \xrightarrow{+w} \delta T_{Co} \tag{4.11}$$

$$\frac{\partial(\delta T_{Co})}{\partial T_{Co}} = -S_{E4}\, F_{Ci} \langle 0 \quad \Rightarrow \quad T_{Co} \xrightarrow{-w} \delta T_{Co} \tag{4.12}$$

$$\frac{\partial(\delta T_{Co})}{\partial Q_T} = S_{E3} \rangle 0 \quad \Rightarrow \quad Q_T \xrightarrow{+w} \delta T_{Co} \tag{4.13}$$

$$\frac{\partial(\delta Q_T)}{\partial T_{Hi}} = \frac{S_{E5}}{2} \rangle 0 \quad \Rightarrow \quad T_{Hi} \xrightarrow{+w} Q_T \tag{4.14}$$

$$\frac{\partial(\delta Q_T)}{\partial T_{Ci}} = -\frac{S_{E5}}{2} \langle 0 \quad \Rightarrow \quad T_{Ci} \xrightarrow{-w} Q_T \tag{4.15}$$

4.4.4 Weighted Digraph Model

The weighted digraph (WDG) model for the heat-exchanger shown in Fig. 4.2b is built up based on Eqs. (4.6) to (4.15) subject to the following considerations:

- The model is built up by decomposing the heater into two processes: heating and cooling, coupled by the flow of energy through the differential variables (δT_C and δT_H);

- Variable Q_T is eliminated by combining Eqs. (4.9), (4.11) and (4.15), and Eqs. (4.7), (4.13) and (4.14) assuming that the direct positive influence: $T_i \xrightarrow{+w} \delta T \xrightarrow{+w} T_o$, prevails over the negative influence through the path which includes Q_T : $T_i \xrightarrow{+w} Q_T \xrightarrow{-w} T_o$;

- Relations (δT_{Ho}, T_{Ho}) and (δT_{Co}, T_{Co}) are *self-regulating*, and therefore edges representing relations (4.8) and (4.12) are *temporal*. To characterise these *self-regulating groups*, while still maintaining a clear definition of *input*, *structural* and *output* variables, dummy variables (δT_H, T_H) and (δT_C, T_C) are created, as shown in Fig. 4.2b. In fact these dummy variables

represent the behaviour inside the qualitative cell (*structural variables*), and T_{Ho} and T_{Co} the outputs of the cell. The reactive (*temporal*) influences are known to be very fast in a heat-exchanger, and therefore a high absolute *weight* within the interval [-1, 1] can be assumed for relations (4.8) and (4.12), e.g. -0.65;

- From observed behaviour of typical systems it can be assumed that influences from flow rates on δT are weaker those from the inlet temperature. Therefore the *weight* for (F, δT) will be smaller than for (T, δT);

- The relative *weights* for relations (4.6), (4.7), (4.10) and (4.11) depend on the hot and cold fluids, since the rate of change of temperature is a function of fluid properties, such as specific heat and density. In the model of Fig. 4.2b it is assumed that the hot fluid is more sensitive to changes in temperature than is the cold fluid;

4.4.5 Qualitative Behaviour for the Start-up of the Heat-Exchanger

Figures 4.3 and 4.4 compare results of qualitative and quantitative simulations for the start-up of a heat-exchanger. Figure 4.3 shows the behaviour of flow rate through time and Fig. 4.4 shows the resulting temperature profiles.

The qualitative trajectories are derived from the model shown in Fig. 4.2b. The adjustment of hot and cold flow rates is made by giving positive qualitative step disturbances of +0.65 to both flow rates. The temperatures are also changed by +0.65, because at start-up the heater is empty and at ambient temperature.

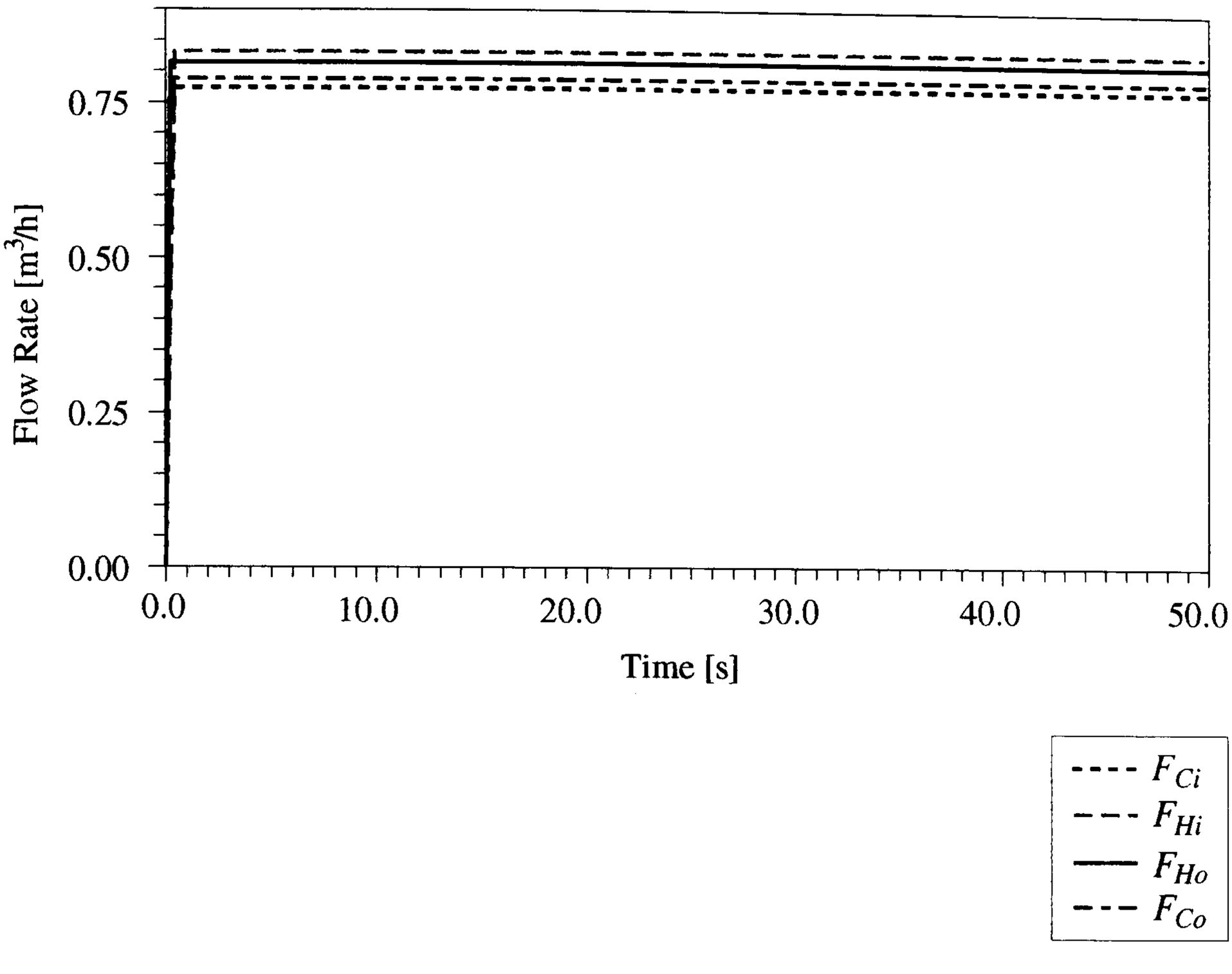
The quantitative (numerical) simulation is based on Eqs. (4.1) to (4.5), using parameters listed in Tables A.1 and A.2 in Appendix A.

Qualitative and quantitative results are compared in terms of the shape of the trajectories, as discussed in section 4.3. It can be seen from Figs. 4.3 and 4.4 that the qualitative model captures the general process behaviour, including the approach to the steady-state for both flow rates and temperature, without generating spurious solutions.

The graphical representation of the WDG model shown in Fig. 4.2b reveals how variables interact to generate the simulation results. For example, the positive step disturbance in the hot flow rate (F_{Hi}) increases the time derivative δT_H, representing a temporary accumulation of energy. This energy will flow to the cold side and increase the time derivative δT_C. The change in the state of the differential nodes causes an increase in the outlet temperature of both fluids. This tends to reduce the energy accumulated in the differential nodes by means of the negative *temporal edges*. As the absolute value of the negative *weights* from T to δT are smaller than the positive *weights* from δT to T the system will not return to the initial steady-state but settle at a higher temperature, what is in accordance with the behaviour of the reference model.

The WDG model for heat-exchangers is effective in describing dynamic behaviour and in explaining how and why solutions are generated. It provides an effective visualisation of the flow of information between the variables of the system during the changes caused by disturbances in *input* variables.

(a)



(b)

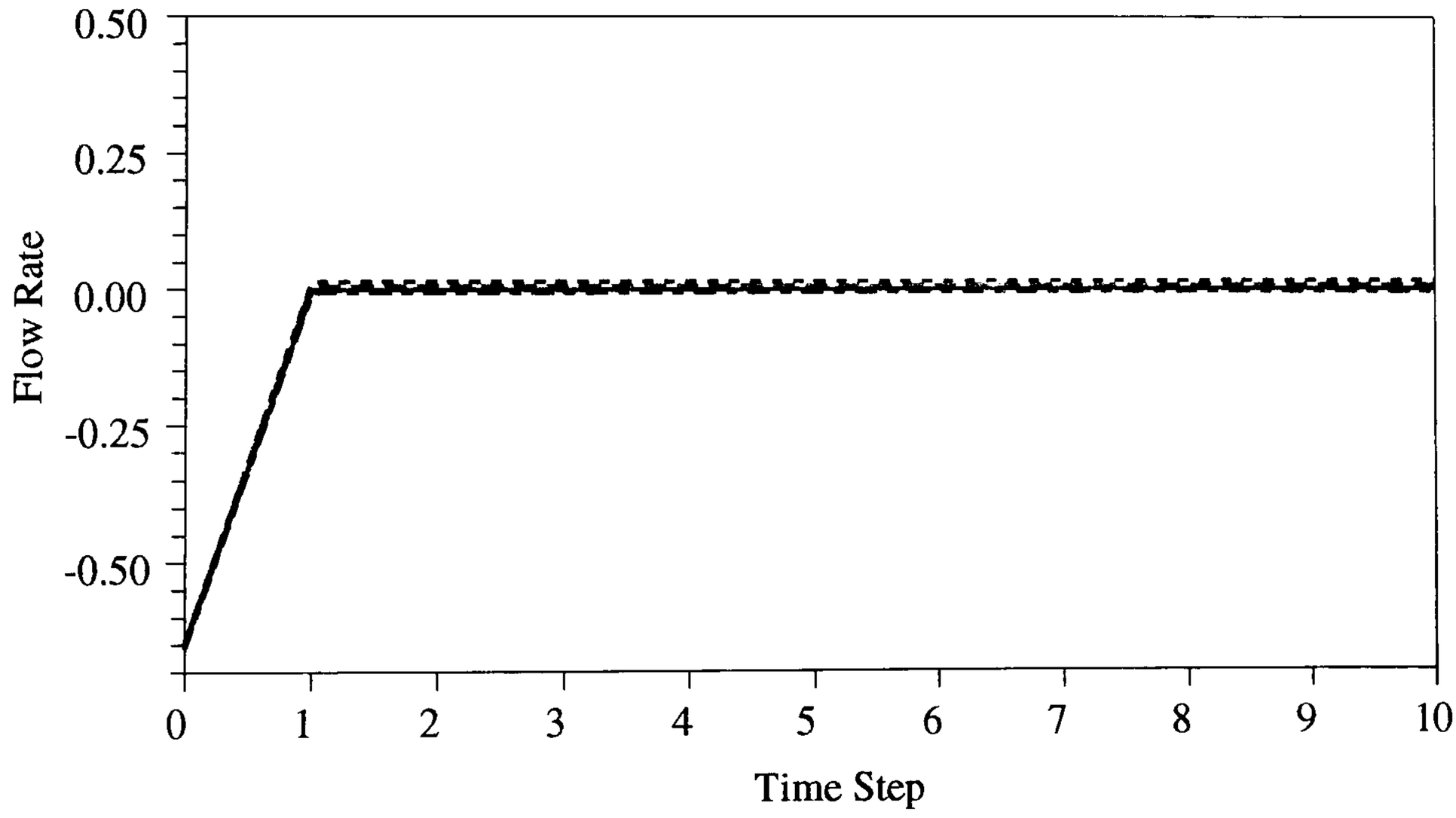
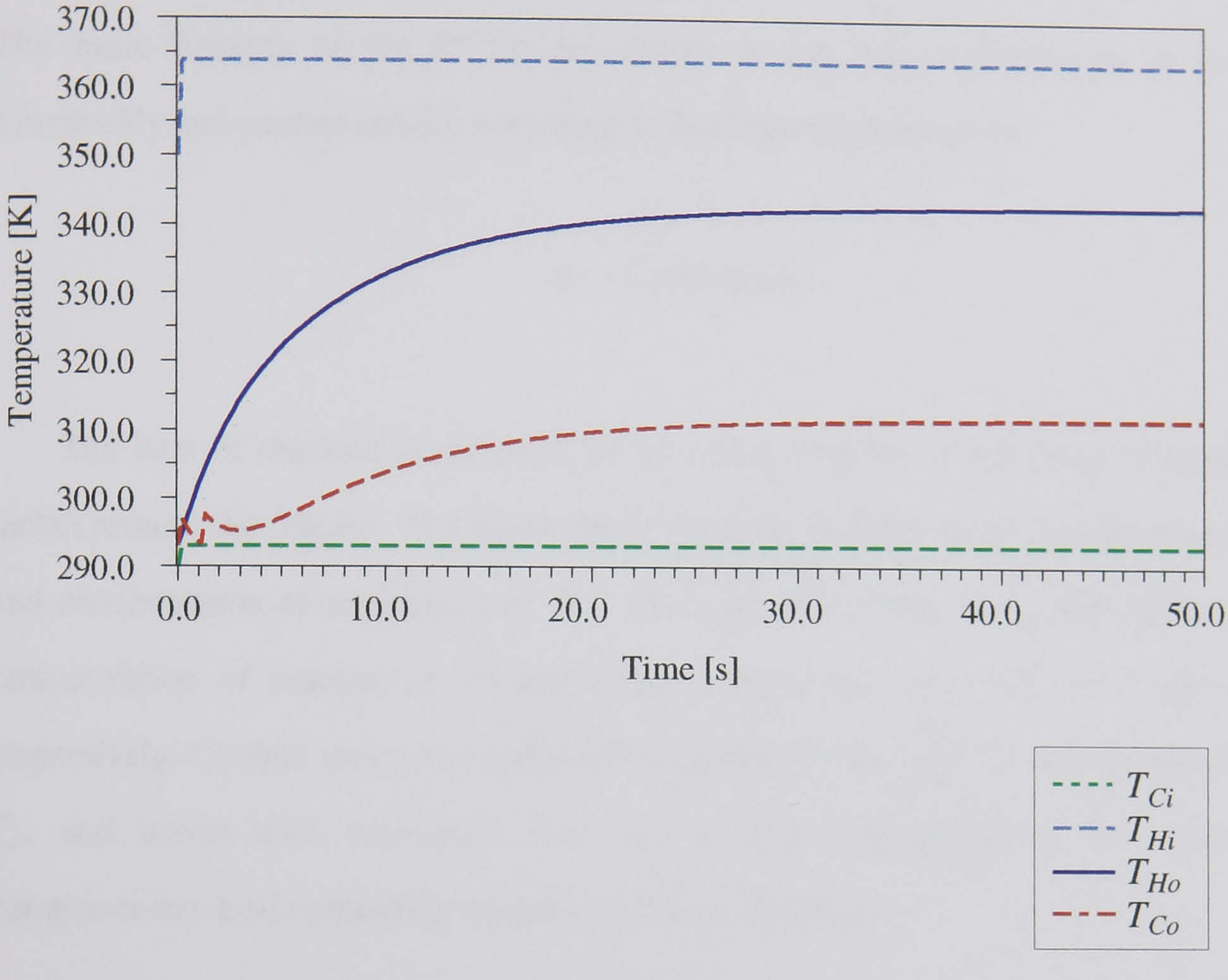


Figure 4.3 Start-up of the heat-exchanger - flow rate profiles: (a) numerical simulation; (b) qualitative simulation.

(a)



(b)

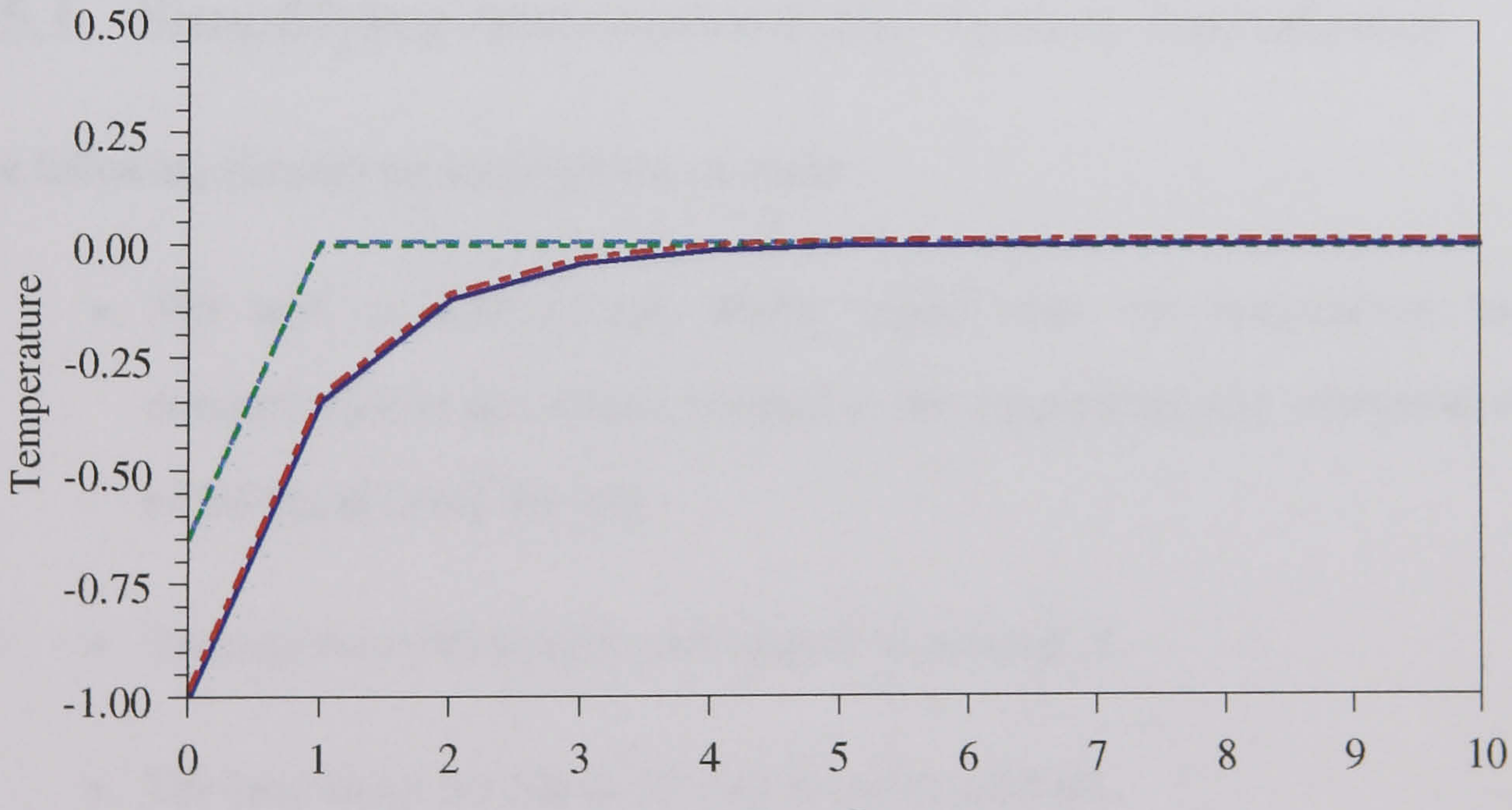


Figure 4.4 Start-up of the heat-exchanger - temperature profiles:
(a) numerical simulation; (b) qualitative simulation.

4.5 Continuous Stirred-Tank Reactor (CSTR)

The basic features of the CSTR are shown in Fig. 4.5a. Component *A* reacts irreversibly and exothermically according to the following expression:

$$A \longrightarrow Products$$

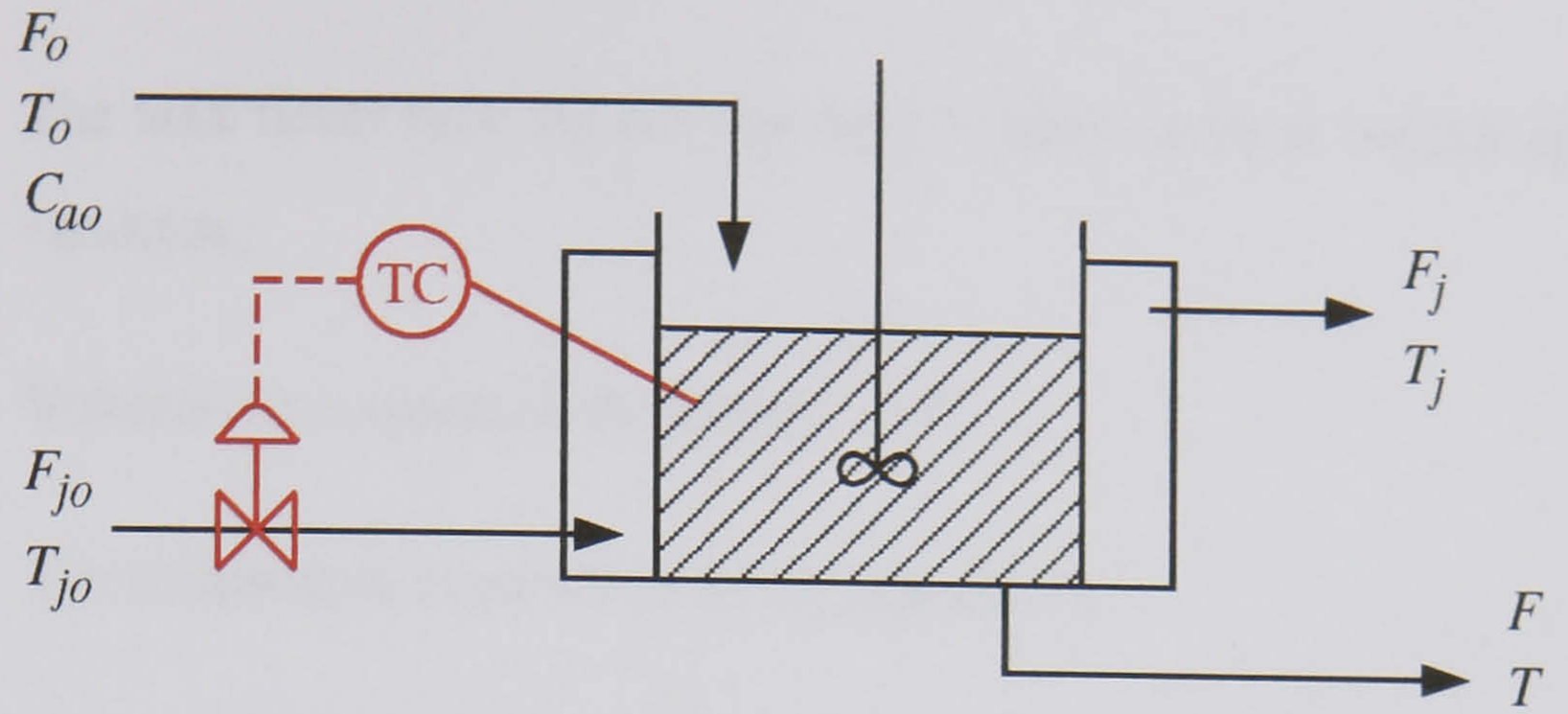
The heat of reaction is removed by a coolant medium which flows through a jacket around the reactor. The liquid enters the tank at flow rate F_o, temperature T_o and concentration of component *A*, C_{ao}. The volumetric flow rate, temperature and concentration of reactant *A* on the stream leaving the tank, are F, T and C_a, respectively. Coolant enters the jacket with volumetric flow rate F_{jo} and temperature T_{jo}, and leaves with volumetric flow rate F_j and temperature T_j. The reactor comprises two basic processes: reaction and heat-exchange.

4.5.1 Simplifying Assumptions and System Boundaries

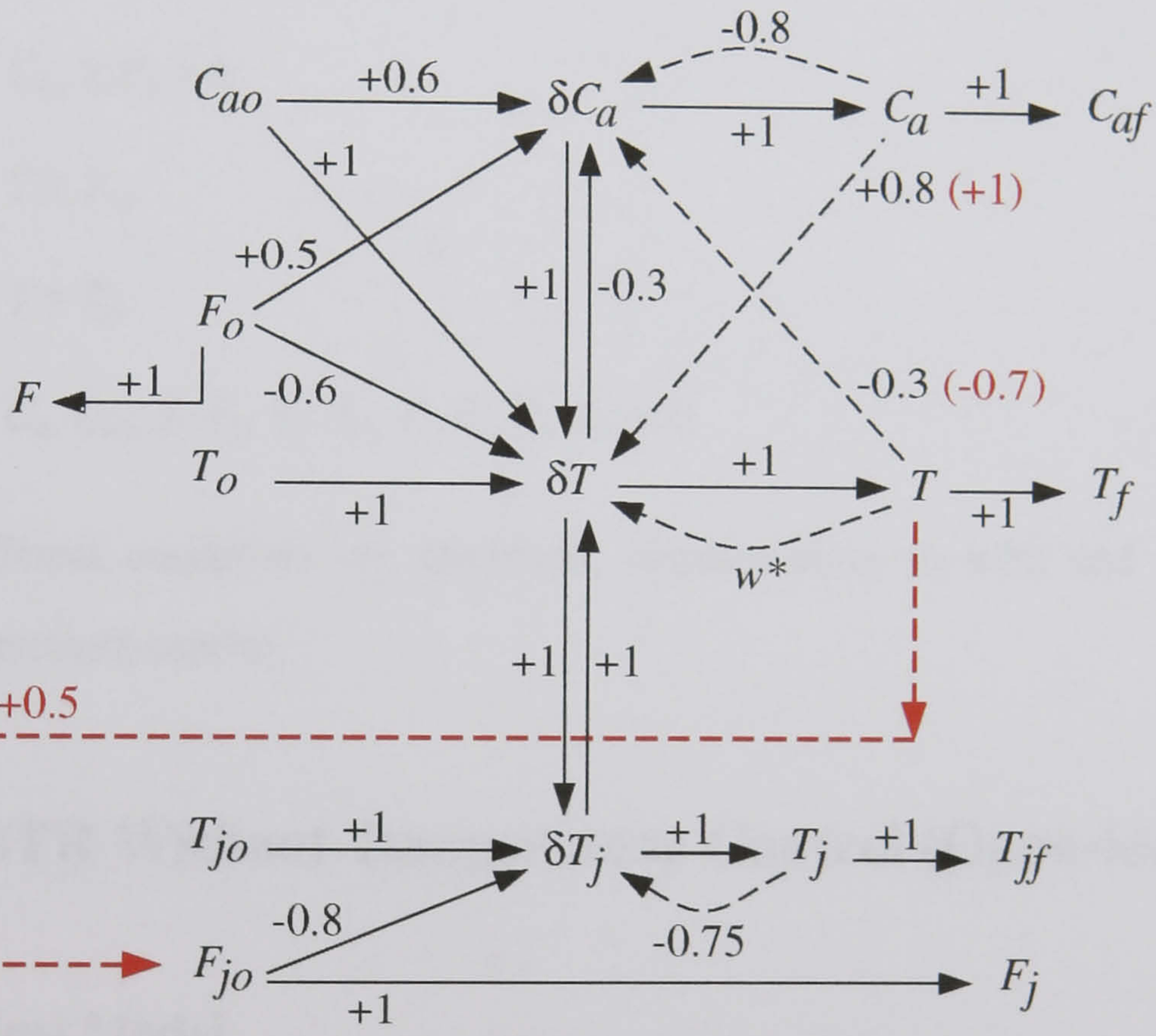
The following simplifying assumptions are made:

- The tank is well stirred, which implies that the temperature and concentration of the effluent is equal to the temperature and concentration of the liquid inside the tank;
- The reaction is first-order with respect to reactant *A*;
- The heat losses are negligible and densities constant;
- *Thermal capacitance* of the equipment is not considered;
- The overall heat transfer coefficient (U) is constant;
- Heat exchange does not vary with liquid hold-up;

(a)



(b)



N.B. (1) *Multiple edges*:

$$w^* = \begin{cases} -0.75, & \text{if } T < T1 \\ +0.8, & \text{if } T1 \le T \le T2 \\ -0.75, & \text{if } T > T2 \end{cases}$$

(2) *T1* and *T2* are arbitrary qualitative limits.

(3) Information in red refer to the CSTR with temperature control.

Figure 4.5 CSTR with cooling system: (a) schematic representation; (b) weighted digraph model.

- The tank never runs dry and the level is taken to be at normal operating condition;
- Volume (V) is constant, therefore $F_o = F$;
- The temperature everywhere in the jacket is T_j;
- Volume of coolant in the jacket (V_j) is constant. Therefore $F_{jo} = F_j$;
- The boundary conditions are:

  : $C_{ao} \geq C_a \geq 0$;

  : $T \geq T_o$;

  : $T \geq T_j$;

  : $C_a, C_{ao}, T, T_o, T_j, T_{jo}, F, F_o, F_j, F_{jo} > 0$.

Two different conditions are examined, corresponding to with and without reaction temperature control.

4.5.2 CSTR Without Temperature Control (Open-loop)

Mathematical Model

Consider the algebraic and ordinary differential equations described in Eqs. (4.16) to (4.24), based on the model given by Luyben (1990).

$$F_o = F \tag{4.16}$$

$$\delta C_a = \frac{dC_a}{dt} = \frac{F_o}{V_R}\left(C_{ao} - C_a\right) + r_a \tag{4.17}$$

$$\delta T = \frac{dT}{dt} = \frac{F_o}{V_R}\left(T_o - T\right) + S_{R1} r_a - S_{R2}\left(T - T_j\right) \tag{4.18}$$

$$\delta T_j = \frac{dT_j}{dt} = \frac{F_{jo}}{V_j}\left(T_{jo} - T_j\right) + S_{R3}\left(T - T_j\right) \tag{4.19}$$

$$r_a = -k_o\, e^{(-S_{R4}/T)}\, C_a \tag{4.20}$$

$$S_{R1} = \frac{\lambda}{\rho\, C_p} \langle\ 0 \qquad \text{(exothermic reaction)} \tag{4.21}$$

$$S_{R2} = \frac{U\,A}{\rho\ C_p\, V_R} \rangle\ 0 \tag{4.22}$$

$$S_{R3} = \frac{U\,A}{\rho_j\, C_{p_j}\, V_j} \rangle\ 0 \tag{4.23}$$

$$S_{R4} = \frac{E}{R} \rangle\ 0 \tag{4.24}$$

Causal Relationships

Based on the above, the following causal relationships are obtained:

$$\frac{\partial\left(\delta C_a\right)}{\partial F_o} = \frac{C_{ao} - C_a}{V_R} \rangle\, 0 \quad \Rightarrow \quad F_o \xrightarrow{+w} \delta C_a \tag{4.25}$$

$$\frac{\partial\left(\delta C_a\right)}{\partial C_{ao}} = \frac{F_o}{V_R} \rangle\, 0 \quad \Rightarrow \quad C_{ao} \xrightarrow{+w} \delta C_a \tag{4.26}$$

$$\frac{\partial\left(\delta C_a\right)}{\partial C_a} = -\frac{F_o}{V_R} - k_o\, e^{(-S_{R4}/T)} \langle\, 0 \quad \Rightarrow \quad C_a \xrightarrow{-w} \delta C_a \tag{4.27}$$

$$\frac{\partial(\delta C_a)}{\partial T} = -k_o \frac{S_{R4}}{T^2} C_a e^{(-S_{R4}/T)} \langle 0 \quad \Rightarrow \quad T \xrightarrow{-w} \delta C_a \tag{4.28}$$

$$\frac{\partial^2(\delta C_a)}{\partial T^2} = k_o C_a e^{(-S_{R4}/T)} \frac{S_{R4}}{T^2}\left[\frac{-S_{R4}+2T}{T^2}\right] \langle 0 \quad \Rightarrow \quad \delta T \xrightarrow{-w} \Delta(\delta C_a) \tag{4.29}$$

$$\frac{\partial(\delta T)}{\partial F_o} = \frac{T_o - T}{V_R} \langle 0 \quad \Rightarrow \quad F_o \xrightarrow{-w} \delta T \tag{4.30}$$

$$\frac{\partial(\delta T)}{\partial T_o} = \frac{F_o}{V_R} \rangle 0 \quad \Rightarrow \quad T_o \xrightarrow{+w} \delta T \tag{4.31}$$

$$\frac{\partial(\delta T)}{\partial C_a} = -k_o S_{R1} e^{(-S_{R4}/T)} \rangle 0 \quad \Rightarrow \quad C_a \xrightarrow{+w} \delta T \tag{4.32}$$

$$\frac{\partial(\delta T)}{\partial T_j} = S_{R2} \rangle 0 \quad \Rightarrow \quad T_j \xrightarrow{+w} \delta T \tag{4.33}$$

$$\frac{\partial(\delta T)}{\partial T} = -\frac{F_o}{V_R} - S_{R2} - k_o S_{R1} e^{(-S_{R4}/T)} C_a \frac{S_{R4}}{T^2} \quad \begin{matrix}\rangle \\ \langle\end{matrix} \quad 0 \quad \Rightarrow \quad T \xrightarrow{\pm w} \delta T \tag{4.34}$$

$$if \quad \begin{cases} low\ or\ medium\ T & \Rightarrow & T \xrightarrow{-w} \delta T \\ high\ T & \Rightarrow & T \xrightarrow{+w} \delta T \\ very\ high\ T \quad \Rightarrow \quad C_a \cong 0 & \Rightarrow & T \xrightarrow{-w} \delta T \end{cases}$$

$$\frac{\partial^2(\delta T)}{\partial T^2} = S_{R1} k_o C_a e^{(-S_{R4}/T)} \frac{S_{R4}}{T^2}\left[\frac{-S_{R4}+2T}{T^2}\right] \rangle 0 \quad \Rightarrow \quad \delta T \xrightarrow{+w} \Delta(\delta T) \tag{4.35}$$

$$\frac{\partial(\delta T_j)}{\partial F_{jo}} = \frac{T_{jo} - T_j}{V_j} \langle 0 \quad \Rightarrow \quad F_{jo} \xrightarrow{-w} \delta T_j \tag{4.36}$$

$$\frac{\partial(\delta T_j)}{\partial T_{jo}} = \frac{F_{jo}}{V_j} \rangle 0 \quad \Rightarrow \quad T_{jo} \xrightarrow{+w} \delta T_j \tag{4.37}$$

$$\frac{\partial(\delta T_j)}{\partial T_j} = \frac{-F_{jo}}{V_j} - S_{R3} \langle 0 \quad \Rightarrow \quad T_j \xrightarrow{-w} \delta T_j \tag{4.38}$$

$$\frac{\partial(\delta T_j)}{\partial T} = S_{R3} \rangle 0 \quad \Rightarrow \quad T \xrightarrow{+w} \delta T_j \tag{4.39}$$

Weighted Digraph Model without Temperature Control

If relations (4.25) to (4.39) are used with *weights* of ±1 attributed to all influences, ambiguities arise during simulation with consequential spurious behaviour. Some structural adjustments are therefore needed which are achieved with *weights* different from ±1. Using the above relations and making the adjustments described below the model shown in Fig. 4.5b (excluding red lines and symbols) is obtained.

- Influences of T on C_a, and T_j on T, and vice-versa, are exerted through the differential nodes δT, δC_a and δT_j. An exception is when the second-order derivative is different from zero. For example, in relation (4.29), the second-order derivative of δC_a with respect to T is also a function of T, and therefore a direct influence of T on δC_a is necessary. In the case of relation (4.35), although the left-hand side only involves T and δT, the right-hand side involves not only T but also C_a, and therefore the influence has to be exerted along a path which involves both C_a and T, i.e. $T \rightarrow \delta C_a \rightarrow C_a \rightarrow \delta T$. Influences generated by second-order derivatives are represented by *temporal edges* since they enclose cycles involving δC_a and δT and therefore are potential sources of conflict;

- Influence (4.34) has to be represented by *multiple edges* since the sign of the *weight* changes with the range of T. This is due to the dependency of the third term of this relation on T. This term represents the heat generated

by the reaction and so is positive. The sign of the *weight* will depend on the dominant term, i.e. heat generated or removed. For low or medium temperatures the third term is negligible or equivalent to the negative terms, respectively. For very high temperatures it tends to zero with C_a. In such cases the negative terms will prevail, or at least neutralise the third, and therefore the *weight* is negative, causing the system to tend to a steady-state. For relatively high temperatures, the third term (heat generated) always outweighs the other two (heat removed), so the *weight* is positive and tends to lead to runaway of temperature;

- Influences (4.27), (4.34) and (4.38) are compensatory influences of the *self-regulating groups*: (δC_a, C_a), (δT, T) and (δT_j, T_j), and therefore are represented by *temporal edges*. In such cases dummy variables, C_{af}, T_f and T_{jf}, are created to differentiate between *structural* and *output* variables;

- By analysing Eqs. (4.16) to (4.24) the following conclusions about the relative strength of the influences, and consequently on the values of the *weights*, can be drawn:

  a) δT depends linearly on F_o and exponentially on T, and therefore the influence from T_o is stronger than the influence from F_o;

  b) There is a mutual influence between C_a and T, and therefore δT is also strongly affected by C_{ao};

  c) δT_j is more strongly affected by T_{jo} than by F_{jo}.

- Because the CSTR is well-mixed, and the dynamics are fast, the *reactive weights* in the *self-regulating groups* can have high absolute values in the interval [-1, +1], e.g. ±0.75, ±0.80, etc.;

- By analysing the order-of-magnitude of relations (4.28) and (4.32) it can be concluded that the influence of T on C_a is relatively weaker than of C_a on T, for most of the time:

$$\textbf{OM}\ [(4.28)] \cong [10^{-4}, 10^{1}]$$

$$\textbf{OM}\ [(4.32)] \cong [10^{0}, 10^{4}]$$

  Therefore, *weights* in the direction $T \rightarrow C_a$ must be smaller than in the reverse direction;

- The following qualitative restriction has to be considered during the qualitative simulation:

  if $C_a \leq C_{a_{min}}$ $\Rightarrow$ all *weights* between T and C_a go to zero, because the reaction is quenched;

- In order to scale the influences of C_{ao}, T_o and T_{jo} on the system, a smaller *weight* is given to the relation (C_{ao}, δC_a) since the numerical values of C_a are usually one or two orders of magnitude smaller than those of T and T_j.

The above considerations provide a guide to assigning the values of the *weights* which should be different from ±1 by giving a more realistic value for the relative strength of influences and so avoiding ambiguous indications of trends. The exact values of the *weights* are determined empirically by comparing predicted and observed behaviour. In this study, the observed behaviour is provided by the quantitative (numerical) simulation. In the case of operating plants, the values would be obtained from observations. In other cases, it can be obtained from previous experience.

The model is built up in a modular fashion by separating the two systems: reaction and heat-exchange. This means that the WDG model for the cooling side of the heat-exchanger previously formulated can be used to describe the behaviour of

the jacket. It is coupled with the reactor model through the differential variables δT and δT_j representing the flow of energy, as can be seen from Fig. 4.5b. The reaction model represents any first-order exothermic reaction of the type $A \rightarrow Products$, occurring in a CSTR, since specific numerical values of parameters are not required in building the qualitative model.

The model of Fig. 4.5b is well adapted to deal with multiple disturbances, i.e. changes in more than one input variable at once. However, when dealing with one single disturbance at a time, it is not necessary to consider *weights* different from ± 1 for the *input edges*, since there are no competing influences from input variables.

Qualitative Behaviour without Temperature Control

Figures 4.6 to 4.9 compare results from qualitative and quantitative (numerical) simulations for positive step disturbances in the inlet flow rate, concentration, temperature and coolant temperature.

Qualitative trajectories are obtained by applying the algorithm for inference of behaviour to the model shown in Fig. 4.5b and giving a positive step disturbance of +1 in each of the input variables, in turn. The quantitative (numerical) simulation is based on Eqs. (4.16) to (4.24), for which parameters, steady-state values and measuring units are described in Tables A.3 and A.4 in Appendix A. Positive step disturbances of +10% are considered for each inlet variable, in turn.

It can be seen in Figs. 4.6 to 4.8 that the functions are monotonic for disturbances in concentration, flow rate and temperature, respectively. The qualitative model captures the shape of each trajectory and the approach to the new steady-state, as can be concluded by comparing Figs. 4.6a to 4.8a with Figs. 4.6b to 4.8b, respectively. For the step disturbance in the coolant temperature (Fig. 4.9) the shape is different from the previous ones since after the disturbance the

coolant is no longer able to remove the heat generated by the reaction and the reactor temperature runs away with a consequent sharp drop in concentration. Even in this extreme situation the qualitative model behaves quite well with the *multiple edges* capable of describing the sudden change in the system trajectories. When the system reaches the *T2* limit (Fig. 4.5b) the *weight* of the influence (T, δT) is switched from -0.75 to +0.8 and the behaviour changes from a smooth to a sharp increase in the temperature. As the temperature reaches a very high qualitative value the simulation is halted which is characteristic of runaway.

The effectiveness of the WDG model in describing the various types of qualitative trajectories of the CSTR is because of the use of first-order derivatives in the model structure, together with the *weights* representing the relative strength of the influences. These are essential for shaping the qualitative responses and describing the dynamics. In particular, the use of *temporal edges* is essential in scheduling influences, solving ambiguities and breaking loops and cycles which would otherwise give rise to conflicting tendencies.

The qualitative model for the CSTR is more complex than for the heat-exchanger but it is still possible to visualise the flow of information and understand how the responses are generated. For example, for a positive step disturbance in the inlet temperature (T_o), the time derivatives δT and δT_j increase, as can be seen from Fig. 4.5b. The increase in δT causes a decrease in δC_a since the rate of reaction (not represented in the model) increases. As a consequence of the changes in the time derivatives, temperatures T and T_j increase and the concentration (C_a) decreases. The drop in C_a tends to decrease δT which in turn tends to increase δC_a and, as a consequence, the changes in the time derivatives decrease with time so the system stabilises at a new steady-state.

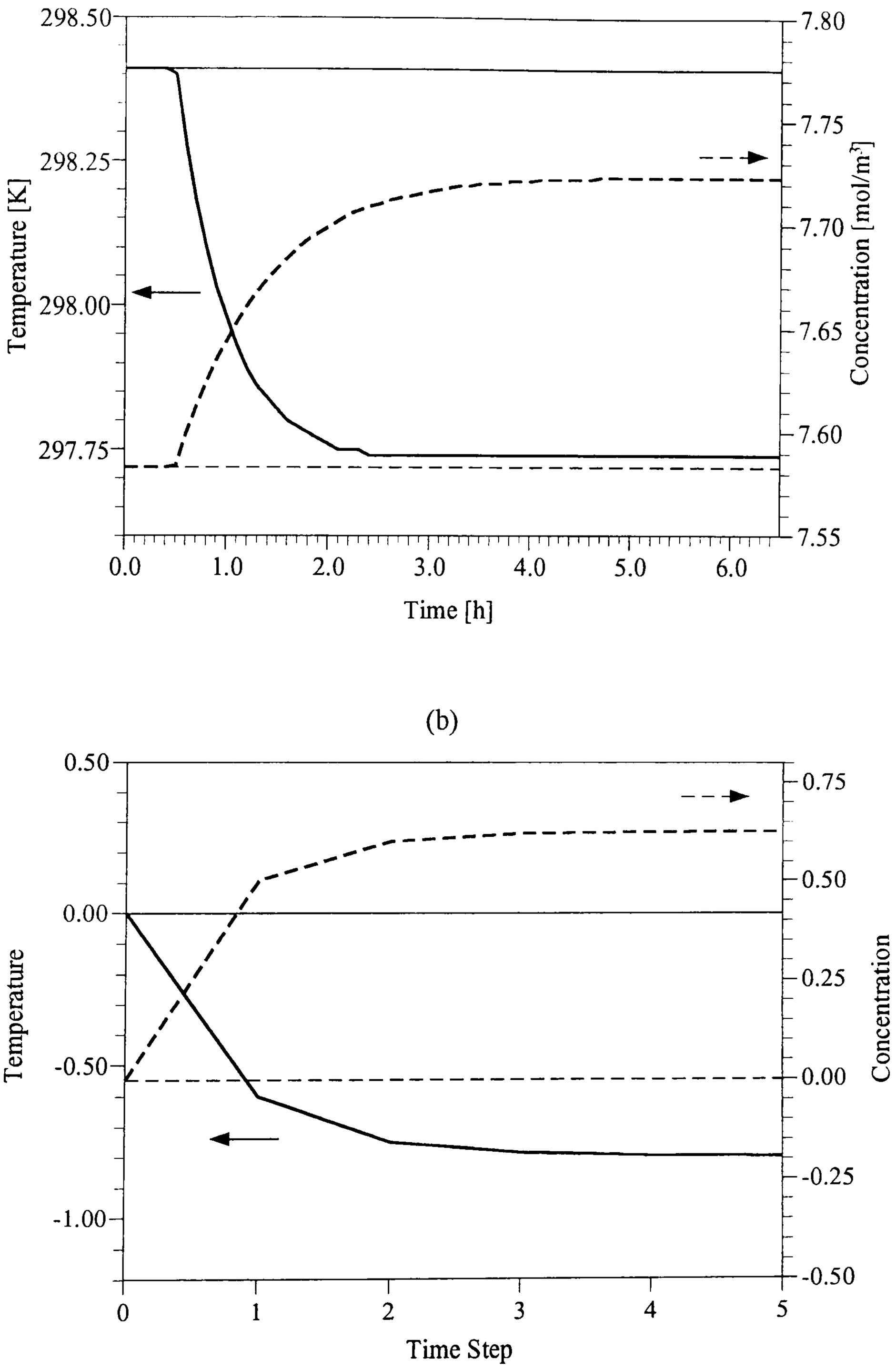


Figure 4.6 CSTR without temperature control - temperature and concentration profiles for a positive step disturbance in inlet flow rate: (a) numerical simulation; (b) qualitative simulation.

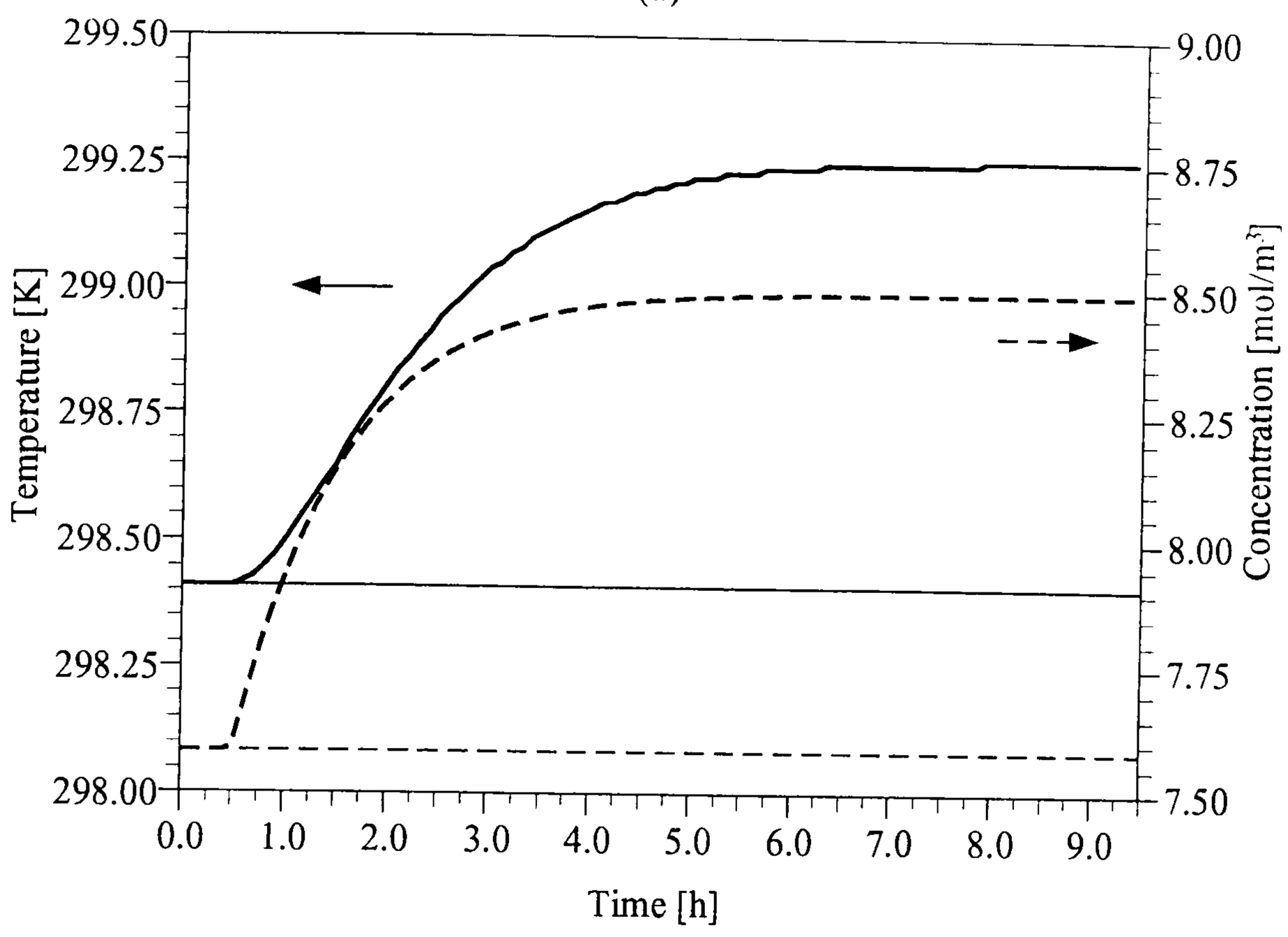


Figure 4.7 CSTR without temperature control - temperature and concentration profiles for a positive step disturbance in inlet concentration: (a) numerical simulation; (b) qualitative simulation.

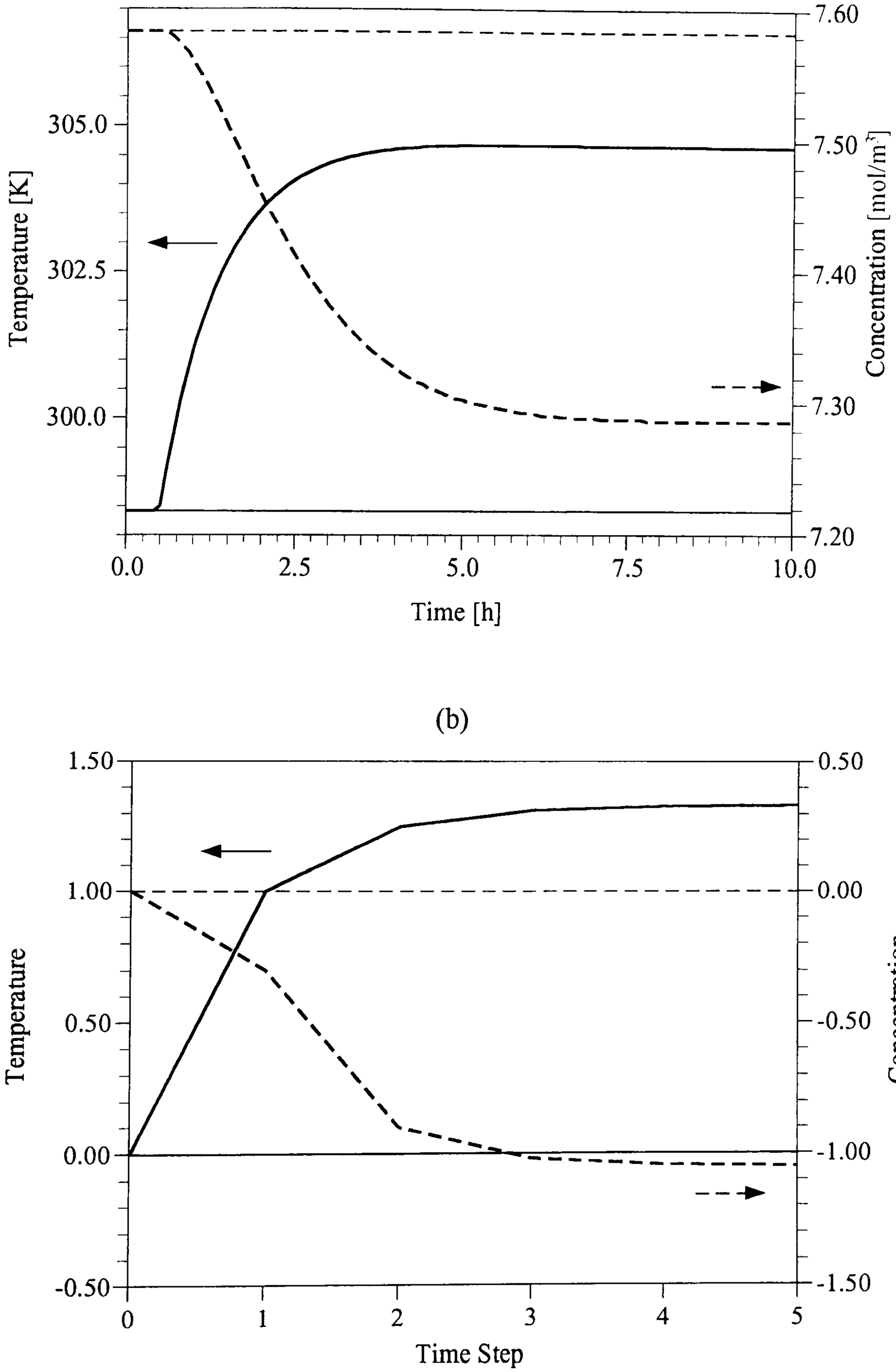


Figure 4.8 CSTR without temperature control - temperature and concentration profiles for a positive step disturbance in inlet temperature: (a) numerical simulation; (b) qualitative simulation.

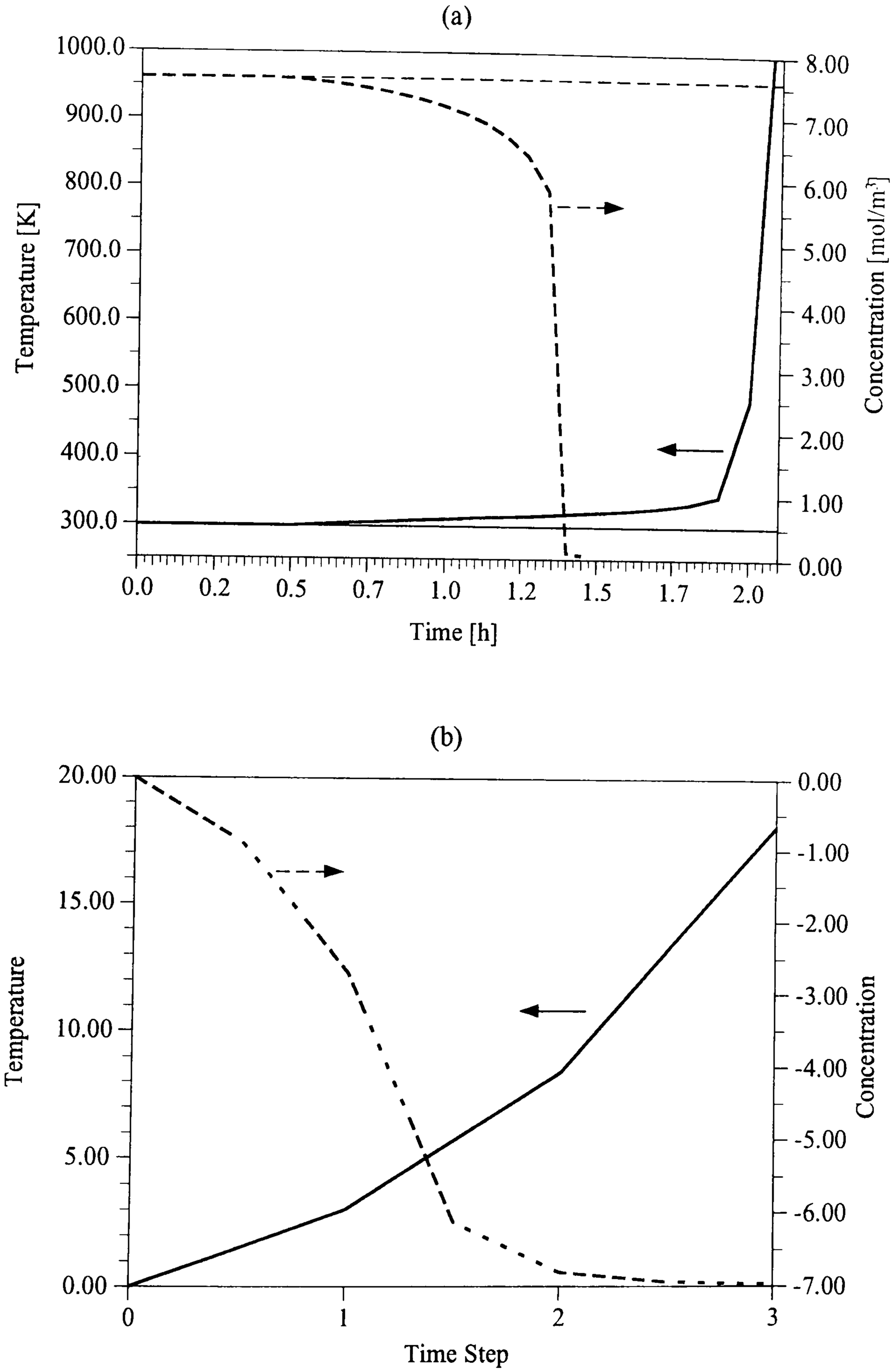


Figure 4.9 CSTR without temperature control - temperature and concentration profiles for a positive step disturbance in inlet coolant temperature: (a) numerical simulation; (b) qualitative simulation.

4.5.3 CSTR With Temperature Control (Closed-loop)

A CSTR with proportional control of the reaction temperature is shown in Fig. 4.5b, with the red lines and symbols relating specifically to this case.

The assumptions, system boundaries, mathematical relations (4.16) to (4.24), causal relationships (4.25) to (4.39) and the WDG model without temperature control remain valid for the present case. However, the effect of the controller has to be considered and the appropriate causal relationships included in the WDG model.

Model for the Controller

$$F_{jo} = F_{jo_{ss}} - \mathrm{K}_{ft}\left(T_{sp} - T\right) \tag{4.40}$$

Causal Relationship for the Controller

$$\frac{\partial F_{jo}}{\partial T} = \mathrm{K}_{ft} \rangle 0 \quad \Rightarrow \quad T \xrightarrow{+w} F_{jo} \tag{4.41}$$

Weighted Digraph Model with Temperature Control

The causal relationship for the controller is added to the basic WDG model for the reactor without temperature control, as shown in red in Fig. 4.5b. The use of the proportional controller alters the dynamic response of the system and some *weights* of the basic model have to be adjusted so the qualitative predictions match the observed system behaviour (numerical parameters in Appendix A.2). As can be seen from Fig. 4.5b, the *weight* for the influence (C_a, δT) is reset to +1.0 (from +0.8), and the *weight* for the influence (T, δC_a) changed from -0.3 to -0.7. This effectively means that the interactions between temperature and concentration are strengthened

by the presence of the controller. However, the relative strength between these influences is not altered. In the present case, the change in the *weights* can be explained by the higher temperature level at which the reactor operates after the introduction of the control which allows a higher conversion of component *A*. For the system without control the normal operating temperature is around 298.4 K, as can be seen in Figs. 4.6 to 4.9, while with control it is around 333.3 K, as shown in Fig. 4.10 to 4.12. The higher the temperature, the higher the reaction rate and consequently more strongly the concentration is affected by changes in temperature. Therefore, the *weights* representing interactions between T and C_a have higher absolute values for the system with temperature control.

Qualitative Behaviour with Temperature Control

Qualitative step disturbance of +1 and quantitative disturbance of +10% are given to one of the input variables, F_o, C_{ao} or T_o, for the qualitative and quantitative simulations, respectively. The dynamic behaviour with temperature control is very different from that of the same system without temperature control, as can be seen by comparing Figs. 4.6 to 4.8 with Figs. 4.10 to 4.12. With control, the responses are no longer monotonic and the system may even exhibit *inverse response*, as in the case of the temperature trajectory for the positive step disturbance in the inlet flow rate (Fig. 4.10). In this case the increase in the inlet flow rate causes a drop in the temperature, which causes the controller to respond by forcing an increase in the temperature while the concentration in the reactor is still high. The combination of high T and C_a results in a high heat generation rate, displacing the system to a region of slightly higher temperature. The system oscillates and reaches the steady-state, which is not the initial value because of the off-set introduced by the controller.

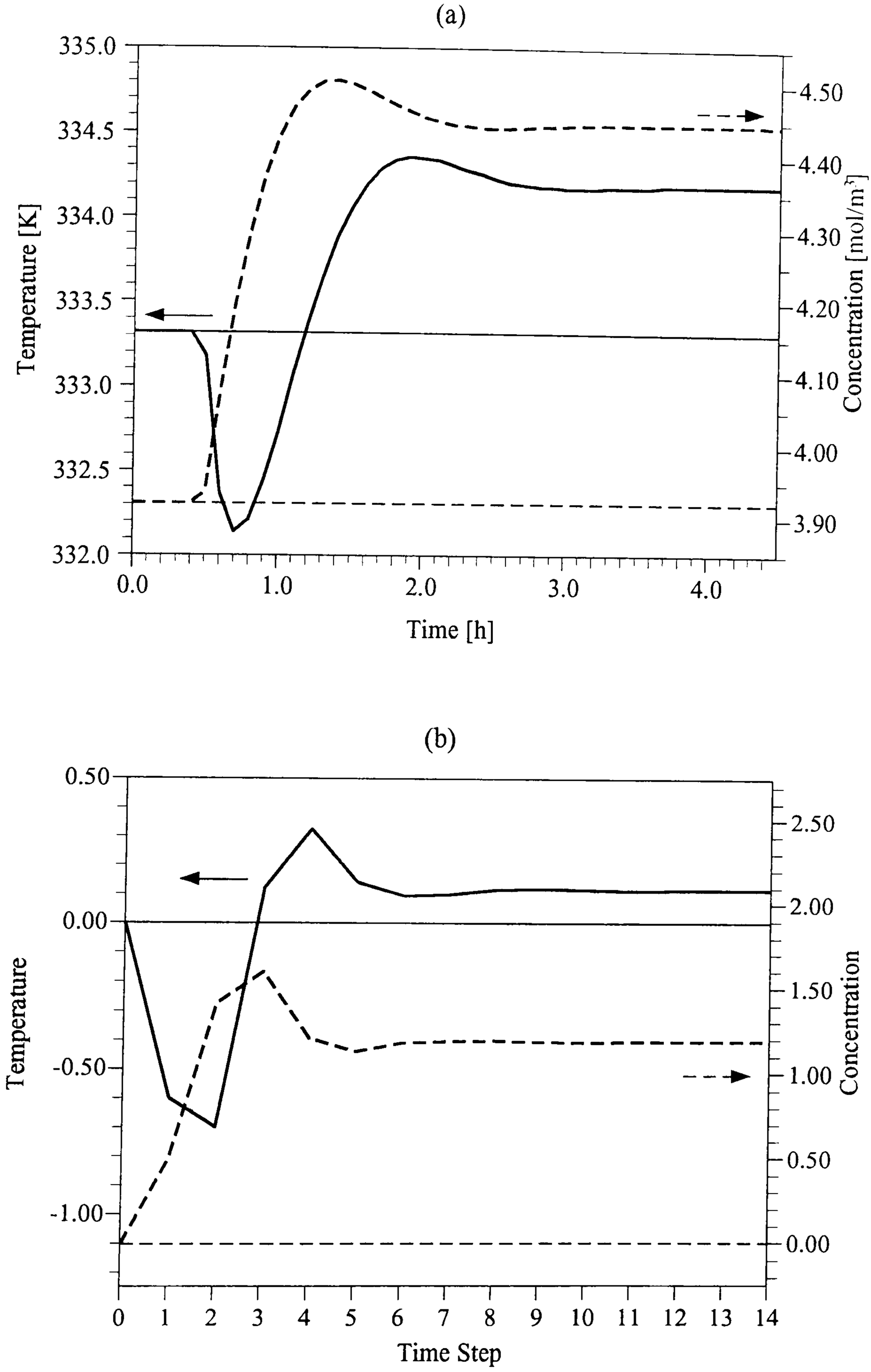


Figure 4.10 CSTR with temperature control - temperature and concentration profiles for a positive step disturbance in inlet flow rate: (a) numerical simulation; (b) qualitative simulation.

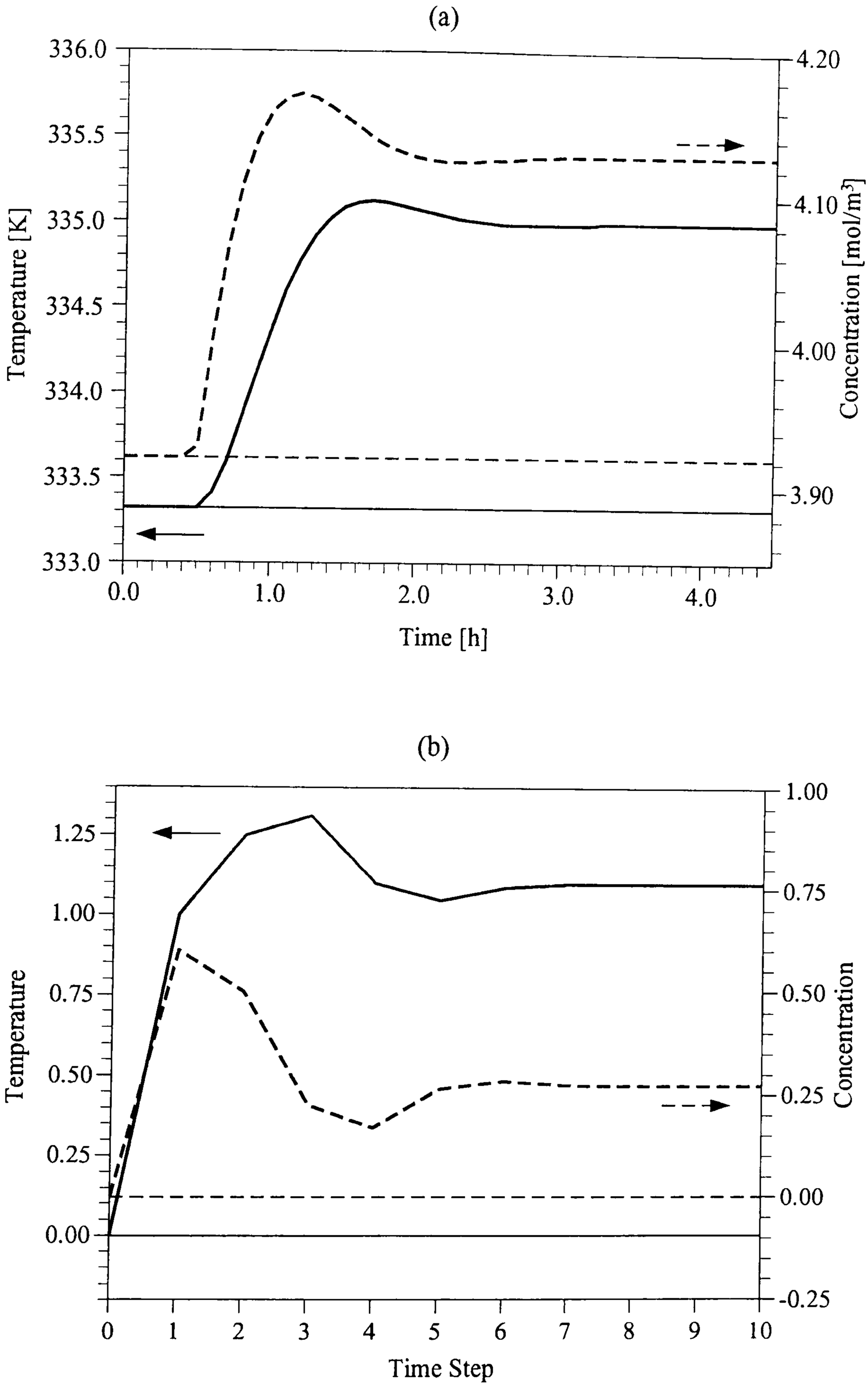


Figure 4.11 CSTR with temperature control - temperature and concentration profiles for a positive step disturbance in inlet concentration: (a) numerical simulation; (b) qualitative simulation.

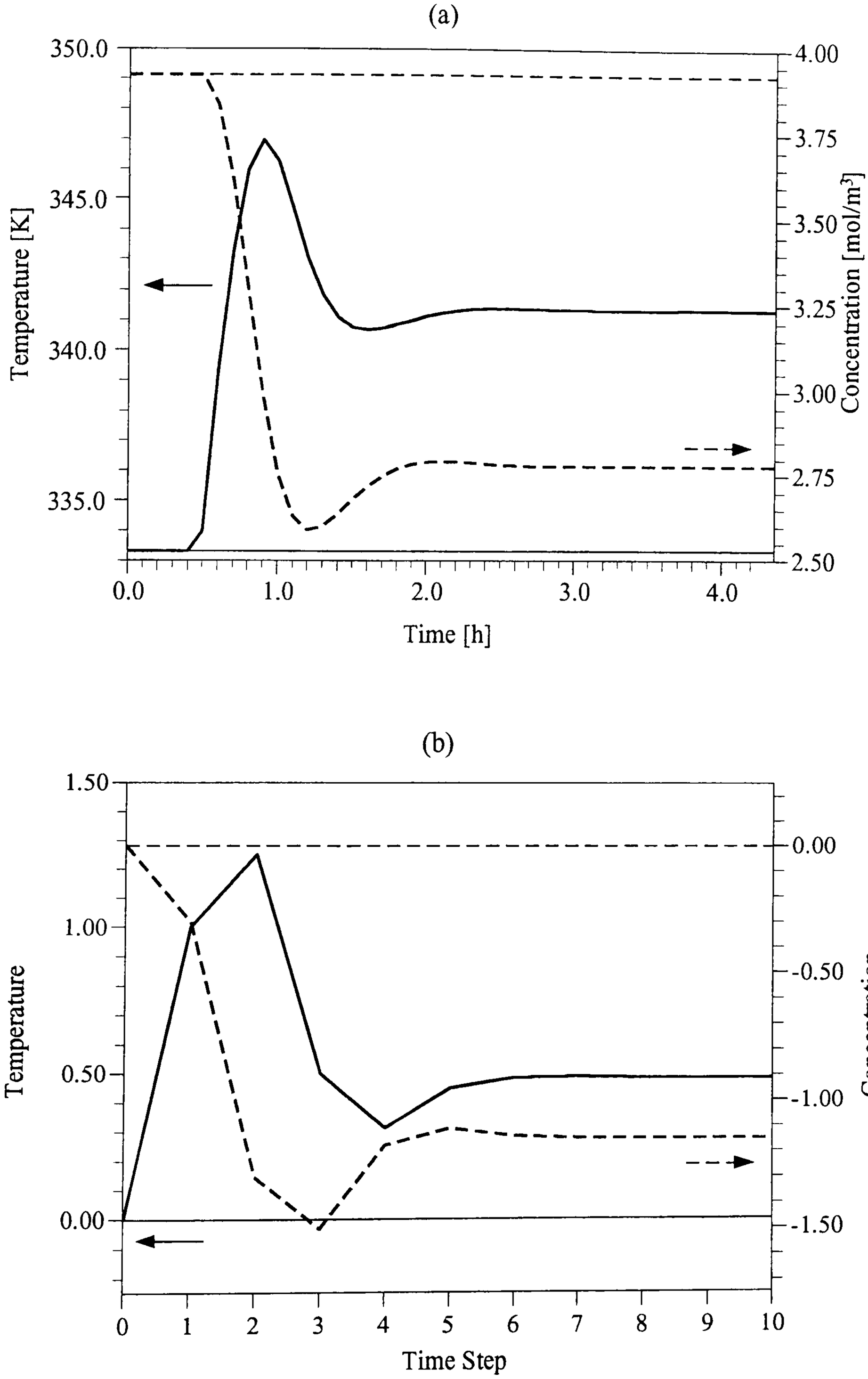


Figure 4.12 CSTR with temperature control - temperature and concentration profiles for a positive step disturbance in inlet temperature: (a) numerical simulation; (b) qualitative simulation.

As can be concluded by examining Figs. 4.10 to 4.12 the qualitative model described in Fig. 4.5b is able to capture the general behaviour of the system in all tests performed, including the complex dynamics of the *inverse response* for the disturbance in the inlet flow rate and the overshoot in all trajectories introduced by the action of the proportional controller. The qualitative model also captures the oscillatory (underdamped) behaviour of the system, although it shows to have some difficulty in distinguishing between small and large oscillations. The model is therefore able to capture the qualitative shape of the trajectories but not the magnitude of the changes.

The effectiveness of the WDG model in describing the complex patterns of behaviour presented by the CSTR with temperature control can be attributed to the association of *multiple edges*, *temporal edges* and *weights*, which allows the description of non-monotonic shapes and avoids ambiguous solutions. The use of *differential nodes* enables information about the rate of change of variables with time to be retained which is essential in describing the process dynamics. To illustrate how the WDG model deals with the several dynamic system features, it is instructive to look at the *inverse response* in temperature for the step disturbance in the feed flow rate in more detail. The following analysis is based on the model of Fig. 4.5b.

Step 1: The increase in F_o causes an increase in δC_a and a decrease in δT, and consequently C_a increases and T decreases;

Step 2: Following this, δT responds to several edges, but the influence from C_a prevails due to the combination of the high *weight* of the *temporal* influence with the state of C_a in the past time step. This combination makes the time derivative δT change from negative to positive and consequently the temperature rises;

Step 3: Because the qualitative value of C_a is high and T is relatively high as well, the qualitative limit, *T1*, is crossed and the *weight* from the *multiple edge* from T to δT becomes positive and this influence prevails. This results in a steep rise in δT. During this time step, C_a still increases slightly since δC_a is still positive;

Step 4: The high value of T causes a decrease in C_a. The influence from the controller prevails over δT reducing it and as a consequence the rate of increase of T is reduced relative to the previous step;

Step 5: From this step on, the influence from the controller always prevails over T, generating the characteristic underdamped oscillatory behaviour in all variables.

The above is a clear example of how the qualitative generation of complex patterns of process behaviour is associated with the effective use of *weights*, *temporal* and *multiple edges* and *differential nodes*.

4.6 Non-azeotropic Ternary Tray Distillation Column

Distillation columns are very difficult to model qualitatively because they are distributed parameter systems involving complex phase equilibria with counter-current flows. Féray-Beaumont *et al.* (1991) have adopted a qualitative version of the transfer function theory from control to model the dynamic behaviour of distillation columns around the steady-state without making any attempt to describe the physical phenomena inside the column.

In this study, the column is divided into five parts each of which has distinctive qualitative behaviour (cells). This is depicted in Fig. 4.13 and includes: (a) reflux

drum (*RD*); (b) rectifying section (*RS*); (c) feed tray (*FT*); (d) stripping section (*SS*); and (e) bottom (*BT*). The internal behaviour of each section is qualitatively described in terms of transient trajectories for vapour (V) and liquid (L) flow rates, temperature (T) and composition of the most and least volatile components, x_1 and x_3 respectively. It is assumed that top and bottom product specifications are given in terms of the most and least volatile components, respectively. Therefore, vapour and liquid compositions of the intermediate component, y_2 and x_2, are omitted from schematic representations and WDG models of the column. The overhead total condenser (*CD*) and the thermosyphon type reboiler (*RB*) are also considered as part of the qualitative model of the distillation column.

Figure 4.13 schematically shows the qualitative components and variables of the column. D and B are the top and bottom product flow rates, F_f the saturated liquid feed flow rate , before the pre-heater, at temperature T_f and compositions z_1 and z_3 for the most and least volatile components, respectively. Heat Q_{RB} flows from the reboiler to the column and Q_{CD} from the column to the condenser. The reboiler has steam supplied at a rate of F_{vp} which is condensed at a rate F_{cd}. Cooling water (F_{cw}) enters the condenser and leaves as hot water (F_{hw}). R_f is the reflux flow rate.

In terms of the underlying processes, the distillation column is made up of flash-based units (*RD*, *RS*, *FT*, *SS* and *BT*) and heat-exchange-based units (*CD* and *RB*). Therefore, WDG models for heat-exchangers are coupled with the WDG model of the main body of the distillation column. It is assumed that before entering the column the feed undergoes an isothermic flash, with adiabatic flashes of the liquid and the vapour flows in each section of the column.

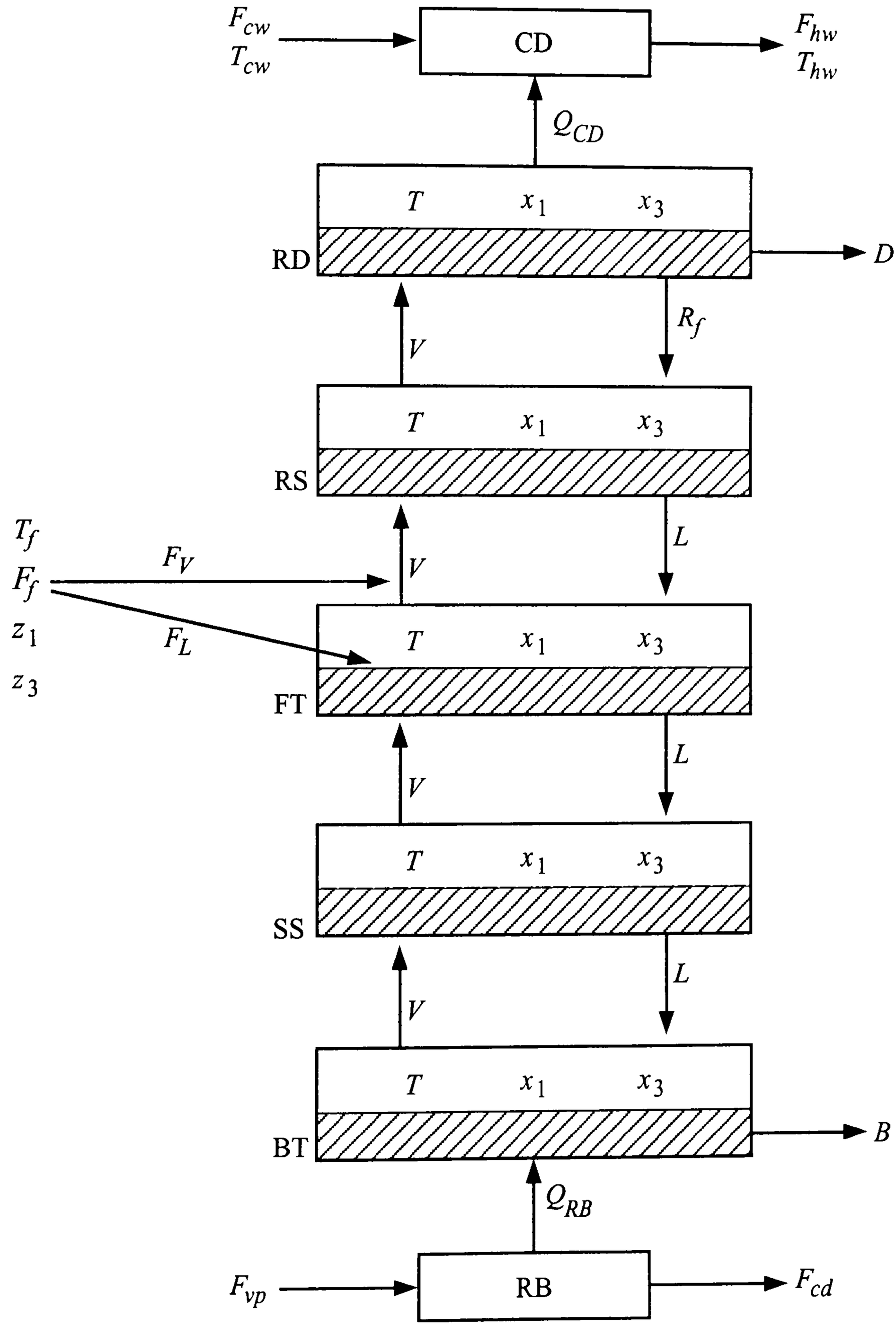


Figure 4.13 Schematic representation of a ternary distillation column.

4.6.1 Simplifying Assumptions and System Boundaries

The WDG model for the distillation column is based on the following considerations and assumptions:

- The system separates a non-azeotropic ternary mixture;
- Pressure is assumed constant throughout the column;
- Vapour hold-up is negligible;
- The liquid on each tray is well mixed;
- It is assumed that the mechanical details do not affect the qualitative behaviour of the distillation column;
- A reference state is assumed so that vapour phase enthalpy (H_V) is always positive while liquid phase enthalpy (H_L) is always negative:

$$H_V \rangle 0\,; \quad H_L \langle 0;$$

- It is also assumed that enthalpy is only a function of temperature and both liquid and vapour phase enthalpies vary in the same way with respect to temperature, i.e.

$$H_V = H_V(T)\,; \quad H_L = H_L(T)$$

$$\frac{dH_V}{dT} \rangle 0 \quad ; \quad \frac{dH_L}{dT} \rangle 0;$$

- Trays never run dry and the column is taken to be with normal liquid level on all trays, in the bottoms and reflux drum;
- Vapour and liquid phases are in equilibrium;

- The system boundaries and constraints are:

$$x_1,\ x_3 = [0,1]$$

$$F,\ T,\ V,\ L,\ R_f,\ Q_{RB},\ Q_{CD} \geq 0$$

$$\sum x_i,\ \sum y_i = 1;$$

- For separation of a non-azeotropic mixture it can be assumed that the liquid compositions of the most and the least volatile components (x_1 and x_3, respectively) always vary in opposite directions, i.e. when x_1 increases, x_3 decreases and vice-versa. Figure 4.14 shows the temperature profiles of a distillation column separating n-hexane (1); n-heptane (2); and n-octane (3) as a function of composition. It can be seen that x_1 (full blue line) always decreases with increase in temperature, while x_3 (full red line) always increases. Because the reboiler and condenser impose a temperature profile on the column, so that the temperature decreases from bottom to top, the following qualitative behaviour is always true for non-azeotropic ternary systems:

$$T \xrightarrow{+w} x_3 \xrightarrow{-w} x_1$$

It should be noted that qualitative relations similar to the above can also be defined for ternary azeotropic distillation columns. Figure 4.15 illustrates five operating profiles for a distillation column separating the azeotropic mixture acetone (1); ethyl-acetate (2); and ethanol (3). The triangular graph shows the planar projection of the vapour-liquid equilibrium surface for the system and the shading represents the equilibrium temperature. This graphical representation is discussed in detail in the works by Maciel (1989) and Shak (1994). The binary azeotrope composition is: $x_2 = 0.5374$ and $x_3 = 0.4626$, which is located on the $x_1 = 0.000$ side of the

triangle. Studies show (Doherty and Perkins, 1978a, b, 1979; Laroche *et al.*, 1992; Shak, 1994) that this kind of binary azeotrope defines a valley running from the azeotropic point to the vertex where $x_1 = 1.000$, dividing the surface into two regions having different qualitative behaviour. On the left side of the valley (Fig. 4.15) the temperature increases from $x_1 = 1.000$ to $x_2 = 1.000$, while on the right side of the valley the temperature reaches a maximum at $x_3 = 1.000$. This results in different distillation profiles, depending on which side of the valley the feed is located. In Fig. 4.15 it can be seen that for feeds 1, 3 and 5 the distillation profile is located on the left side of the valley, while for feeds 2 and 4 it is on the right side. Figure 4.16 shows the temperature versus composition profiles for feeds 4 and 5 located on opposite sides of the valley. It can be seen that for feed 4 the qualitative behaviour is the same as that obtained for the non-azeotropic mixture, i.e.

$$T \xrightarrow{+w} x_3 \xrightarrow{-w} x_1$$

However, for feed 5 the intermediate (2) and least volatile (3) components exchange behaviour patterns, as can be seen by comparing Figs. 4.16a and 4.16b, and the qualitative behaviour is:

$$T \xrightarrow{+w} x_2 \xrightarrow{-w} x_1$$

Therefore, if it is known on which side of the valley the feed is located, it is possible to predict the qualitative relationship between T, x_1 and x_2 or x_3. For other types of azeotropic systems, similar conclusions can be drawn if the vapour-liquid equilibrium surface and the equilibrium temperature profile are known.

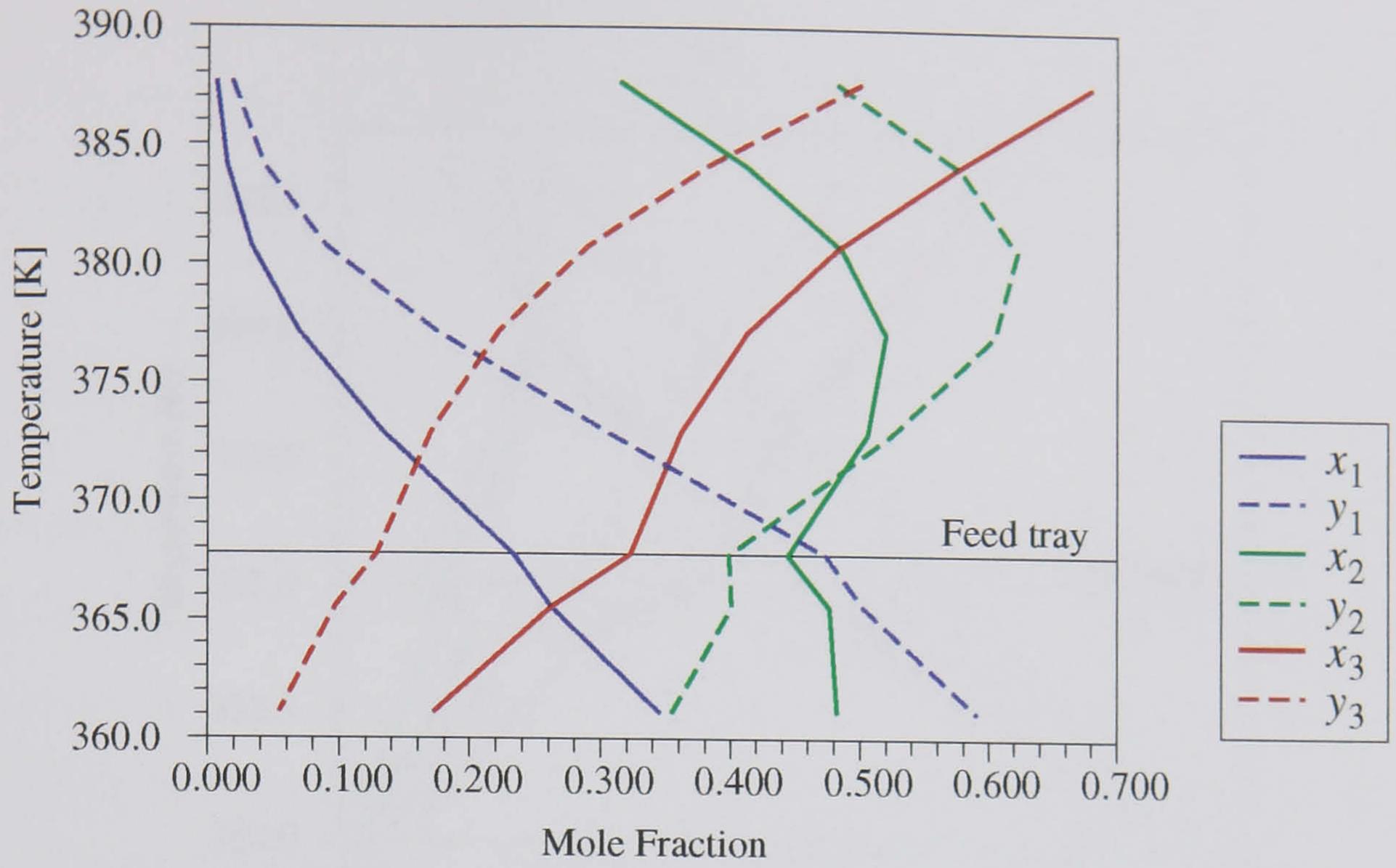
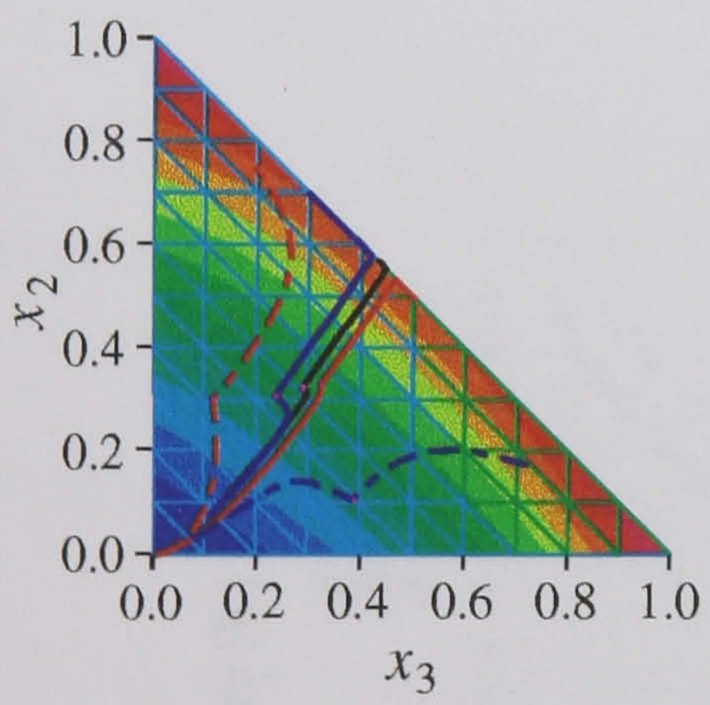


Figure 4.14 Temperature vs. composition profiles for a distillation column separating the non-azeotropic system n-hexane(1); n-heptane(2); n-octane(3).



VLE surface

Above diagonal plane ($x = y$)

Below diagonal plane ($x = y$)

Operating trajectories

Feed 1 (0.60/0.20/0.20)

Feed 2 (0.58/0.20/0.22)

Feed 3 (0.65/0.20/0.15)

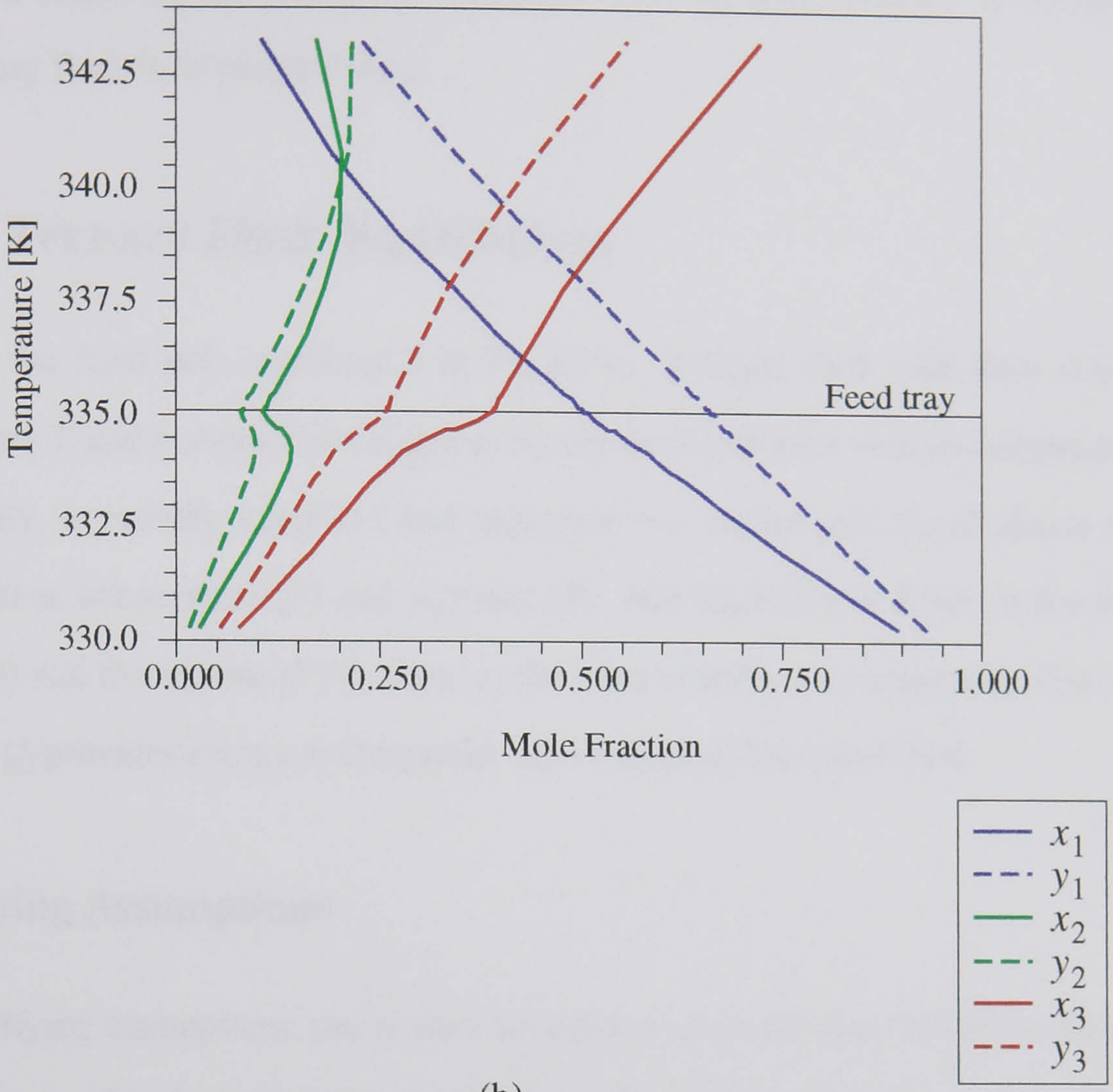
Feed 4 (0.50/0.10/0.40)

Feed 5 (0.50/0.40/0.10)

N.B. Compositions in mole fraction

Figure 4.15 Equilibrium surface and operating trajectories for the azeotropic system acetone(1); ethyl-acetate(2); ethanol(3).

(a)



(b)

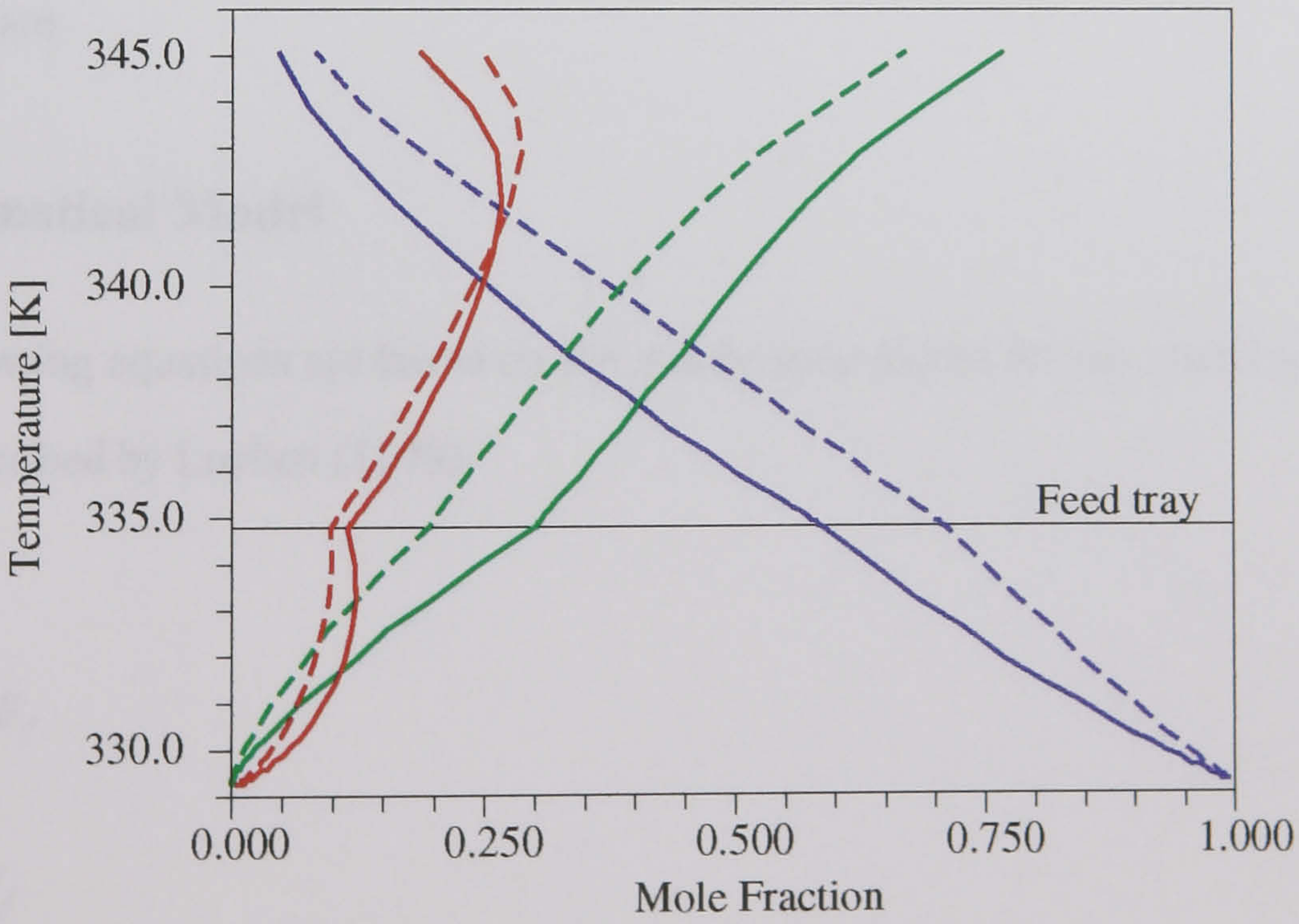


Figure 4.16 Temperature vs. composition profiles for a distillation column separating the azeotropic system acetone(1); ethyl-acetate(2); ethanol(3): (a) feed 4 (0.5/0.1/0.4); (b) feed 5 (0.5/0.4/0.1).

As the model for the distillation column is based on flash units the WDG model for a ternary flash is formulated next.

4.6.2 Ternary Flash Equilibrium

Consider the flash unit represented in Fig 4.17a. A liquid feed with flow rate F_f, temperature T_f and compositions z_1 and z_3 for the most and least volatile components, respectively, is partially vaporised and separated into vapour and liquid phases in a flash drum at temperature (T) and pressure (P). The liquid (L) is richer in the least volatile (3) and the vapour (V) is richer in the most volatile (1) component. The heat flow rate Q_f provides energy to the partial vaporisation of the liquid feed.

Simplifying Assumptions

The simplifying assumptions and system boundaries given for the distillation column also apply to the flash, together with the consideration that the system is at steady-state.

Mathematical Model

The following equations are based on the steady-state model for the multicomponent flash described by Luyben (1990).

$$V + L = F_f \tag{4.42}$$

$$\Psi = V/F_f \tag{4.43}$$

$$L/F_f = 1 - \Psi \tag{4.44}$$

$$\Psi\ y_3 + (1-\Psi)\, x_3 = z_3 \tag{4.45}$$

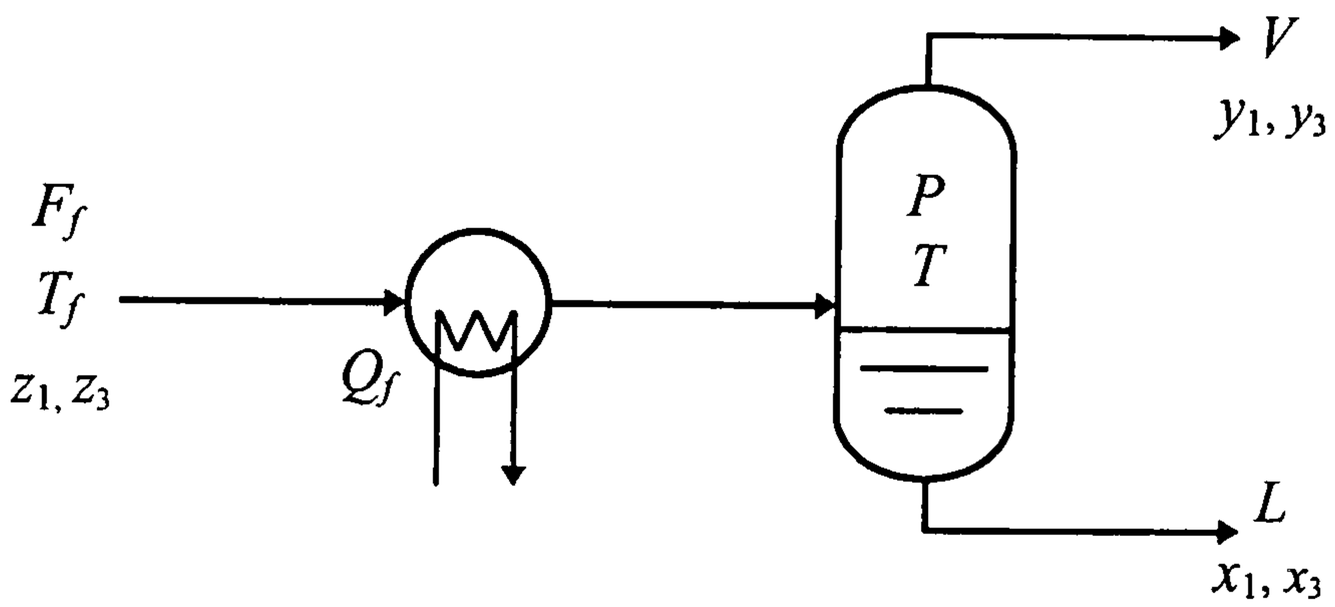
$$H_{in} = H_{out} \tag{4.46}$$

$$H_{in} = H_f\ F_f + Q_f \tag{4.47}$$

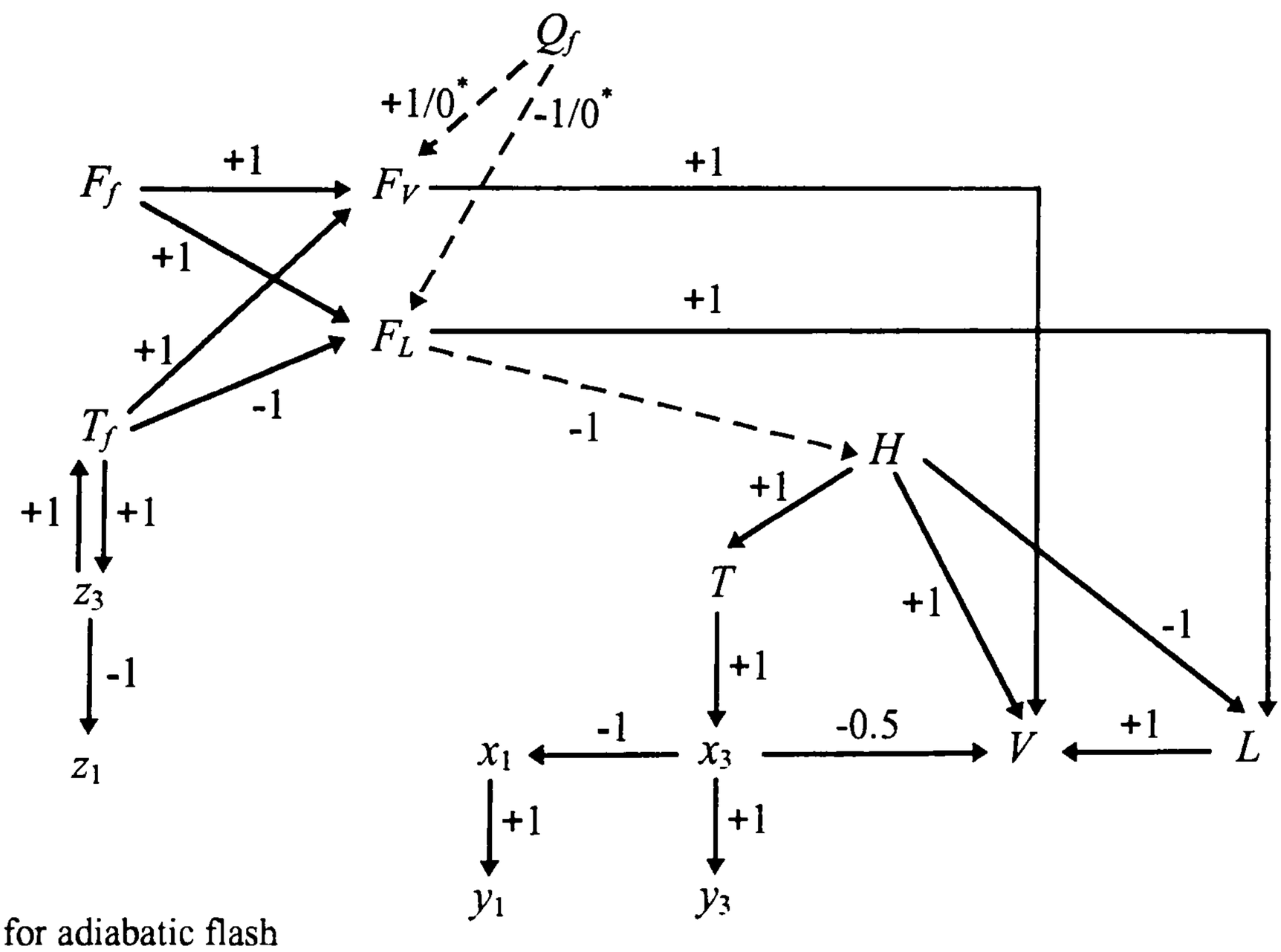
$$H_{out} = H_V\ V + H_L\ L \tag{4.48}$$

$$y_3 = K_3\ x_3 \tag{4.49}$$

(a)



(b)



N.B.* $w=0$, for adiabatic flash

Figure 4.17 Ternary flash unit: (a) schematic representation and (b) weighted digraph model.

Causal Relationships

Based on Eqs. (4.42) to (4.49) and the simplifying assumptions, the following causal relationships are obtained:

$$\frac{\partial V}{\partial F_f} = 1 \rangle 0 \quad \Rightarrow \quad F_f \xrightarrow{+w} V \tag{4.50}$$

$$\frac{\partial L}{\partial F_f} = 1 \rangle 0 \quad \Rightarrow \quad F_f \xrightarrow{+w} L \tag{4.51}$$

$$\frac{\partial y_3}{\partial z_3} = \frac{1}{\Psi} \rangle 0 \quad \Rightarrow \quad z_3 \xrightarrow{+w} y_3 \tag{4.52}$$

$$\frac{\partial x_3}{\partial z_3} = \frac{1}{1-\Psi} \rangle 0 \quad \Rightarrow \quad z_3 \xrightarrow{+w} x_3 \tag{4.53}$$

$$\frac{\partial y_3}{\partial x_3} = K_3 \rangle 0 \quad \Rightarrow \quad x_3 \xrightarrow{+w} y_3 \tag{4.54}$$

$$\frac{\partial x_3}{\partial T} \rangle 0 \quad \Rightarrow \quad T \xrightarrow{+w} x_3 \tag{4.55}$$

$$\frac{\partial x_1}{\partial x_3} \langle 0 \quad \Rightarrow \quad x_3 \xrightarrow{-w} x_1 \tag{4.56}$$

$$\frac{\partial H_{in}}{\partial Q_f} = 1 \rangle 0 \quad \Rightarrow \quad Q_f \xrightarrow{+w} H_{in} \tag{4.57}$$

$$\frac{\partial H_{in}}{\partial F_f} = H_f \langle 0 \quad \text{(liquid feed)} \quad \Rightarrow \quad F_f \xrightarrow{-w} H_{in} \tag{4.58}$$

$$\frac{\partial H_{in}}{\partial H_f} = F_f \rangle 0 \quad \Rightarrow \quad H_f \xrightarrow{+w} H_{in} \tag{4.59}$$

$$\frac{\partial H_f}{\partial T_f} \rangle 0 \quad \Rightarrow \quad T_f \xrightarrow{+w} H_f \tag{4.60}$$

$$\frac{\partial z_3}{\partial T_f} \rangle 0 \quad \Rightarrow \quad T_f \xrightarrow{+w} z_3 \tag{4.61}$$

$$\frac{\partial H_{out}}{\partial H_{in}} = 1 \rangle 0 \quad \Rightarrow \quad H_{in} \xrightarrow{+w} H_{out} \tag{4.62}$$

$$\frac{\partial H_V}{\partial H_{out}} = \frac{1}{V} \rangle 0 \quad \Rightarrow \quad H_{out} \xrightarrow{+w} H_V \tag{4.63}$$

$$\frac{\partial H_L}{\partial H_{out}} = \frac{1}{L} \rangle 0 \quad \Rightarrow \quad H_{out} \xrightarrow{+w} H_L \tag{4.64}$$

$$\frac{\partial T}{\partial H_V} \rangle 0 \quad \Rightarrow \quad H_V \xrightarrow{+w} T \tag{4.65}$$

$$\frac{\partial T}{\partial H_L} \rangle 0 \quad \Rightarrow \quad H_L \xrightarrow{+w} T \tag{4.66}$$

$$\frac{\partial V}{\partial H_{out}} = \frac{1}{H_V} \rangle 0 \quad \Rightarrow \quad H_{out} \xrightarrow{+w} V \tag{4.67}$$

$$\frac{\partial L}{\partial H_{out}} = \frac{1}{H_L} \langle 0 \quad \Rightarrow \quad H_{out} \xrightarrow{-w} L \tag{4.68}$$

$$\frac{\partial V}{\partial H_V} = -\frac{1}{H_V} \langle 0 \quad \Rightarrow \quad H_V \xrightarrow{-w} V \tag{4.69}$$

$$\frac{\partial L}{\partial H_L} = -\frac{1}{H_L} \rangle 0 \quad \Rightarrow \quad H_L \xrightarrow{+w} L \tag{4.70}$$

$$\frac{\partial V}{\partial L} = 1 \rangle 0 \quad (\text{if } T, z = \text{constant, and } F_f \neq \text{constant}) \quad \Rightarrow \quad L \xrightarrow{+w} V \tag{4.71}$$

Weighted Digraph Model

Based on relations (4.50) to (4.71) and the following considerations, the WDG model for a ternary flash, depicted in Fig. 4.17b, can be derived. Because the stages of the distillation column are considered adiabatic flash chambers, the WDG model for the ternary flash takes into account the most important variables which characterise the behaviour of a distillation column.

- Variables H_{in} and H_{out} are combined into one variable H representing the energy content of the flash unit;

- Influences on flash temperature T and composition x_3 are exerted through the variable H, which characterises that changes in the temperature and composition arise from changes in the energy level of the unit;

- Flow rates change faster than temperature and compositions, and therefore influences on H are represented by *temporal edges* and direct influences on L and V are represented by *ordinary* edges;

- Some influences are represented indirectly by a longer path. For example, influence (z_3, y_3) is represented by the path:

$$z_3 \rightarrow T_f \rightarrow F_L \rightarrow H \rightarrow T \rightarrow x_3 \rightarrow y_3 .$$

  This satisfies the need to ensure that all changes in y_3 necessarily imply changes in T when z_3 varies, if the feed F_f is always a saturated liquid;

- All *weights* are set equal to ±1, except for the negative compensatory influence (H, V) from Eq. (4.69), which follows the path including T and x_3 since it comes from the energy balance. From the observed behaviour of typical systems, it can be concluded that V increases with increase in

enthalpy, so the positive influence (H, V) from Eq. (4.67) must prevail over the negative influence from Eq. (4.69), which consequently should have a smaller *weight*;

- For an adiabatic flash the *weight* of the influence (Q_f, H) is zero;

- Influence (4.70) is ignored since an increase in H_L is always accompanied by a stronger increase in H_V, which in turn increases V and consequently decreases L;

- Dummy variables F_V and F_L are created to represent the vapour and liquid feed flow rates, respectively, before entering the flash drum but after the heater. Since the heat content in a flash drum or distillation tray is intrinsically related to the liquid hold-up, the influence in H is through F_L.

4.6.3 Distillation Column

The WDG model for the distillation column is based on the steady-state model for the ternary flash, to which information about dynamics and interactions of adjacent units (sections) are included. *Weights* are adapted to improve the description of the qualitative behaviour of each section of the column and the basic flash model is modified to represent the reflux drum and bottom sections, since these are slightly different from the rectifying, feed tray and stripping sections.

The appropriate modifications to the basic flash model are derived from the mathematical model and causal relationships between the column state variables.

Mathematical Model

Consider the schematic representation of a typical section, j, of the column shown in Fig. 4.18a. The dynamic mathematical models describing the section, based on the work by Gani *et al.* (1986), with some mathematical rearrangements, are as follows:

$$M_j = S_{D1}\ L_j \tag{4.72}$$

$$\delta L_j = \frac{dM_j}{dt} = F_{f_j} + V_{j-1} + L_{j+1} - V_j - L_j \tag{4.73}$$

$$\delta x_{3_j} = \frac{dx_{3_j}}{dt} = \frac{1}{M_j}\begin{bmatrix} F_{f_j}\left(z_{3_j} - x_{3_j}\right) + V_{j-1}\left(y_{3_{j-1}} - x_{3_j}\right) + \\ + L_{j+1}\left(x_{3_{j+1}} - x_{3_j}\right) - V_j\left(y_{3_j} - x_{3_j}\right) \end{bmatrix} \tag{4.74}$$

$$\delta H_j = \frac{dH_j}{dt} = \frac{1}{M_j}\begin{bmatrix} F_{f_j}\left(H_{f_j} - H_{L_j}\right) + V_{j-1}\left(H_{V_{j-1}} - H_{L_j}\right) + \\ + L_{j+1}\left(H_{L_{j+1}} - H_{L_j}\right) - V_j\left(H_{V_j} - H_{L_j}\right) \end{bmatrix} \tag{4.75}$$

The following inequalities can usually be assumed for a non-azeotropic system:

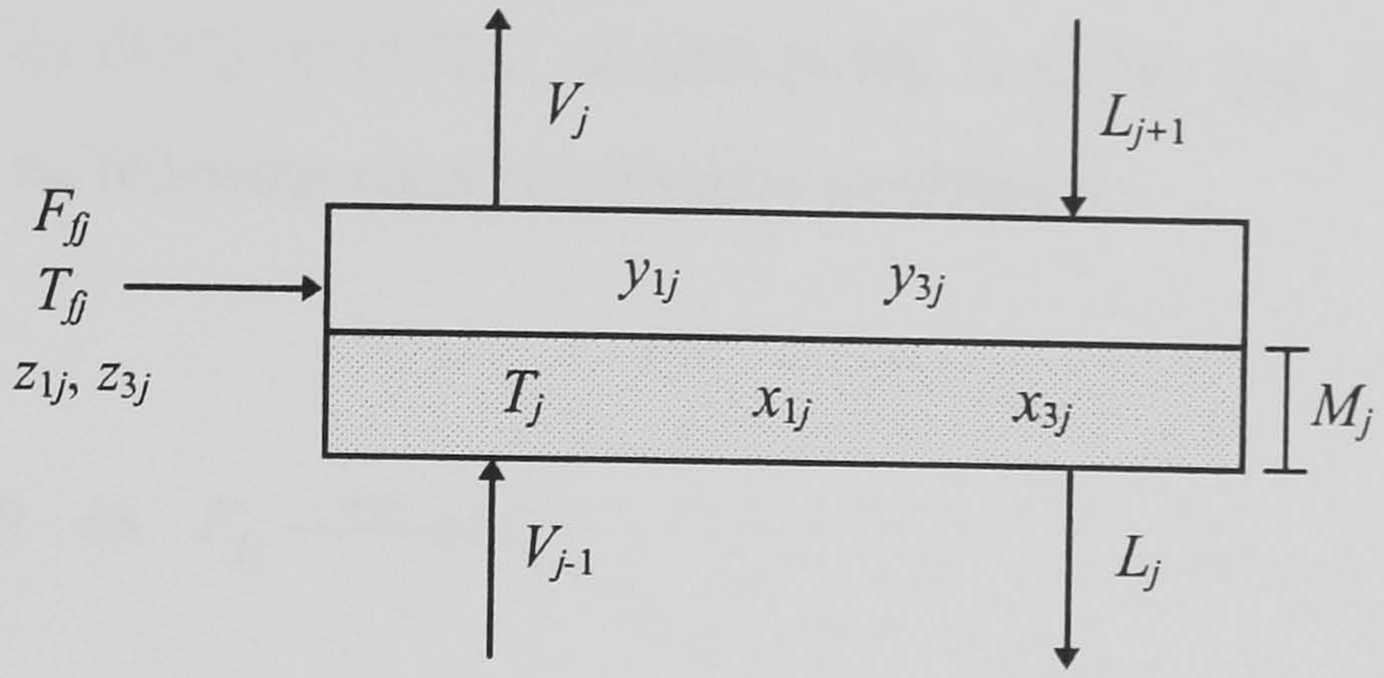
$$y_{3_j} \langle\ x_{3_j} \tag{4.76}$$

$$y_{3_{j-1}} \langle\ x_{3_j} \tag{4.77}$$

$$x_{3_{j+1}} \langle\ x_{3_j} \tag{4.78}$$

$$z_{3_j} \langle\ x_{3_j} \tag{4.79}$$

(a)



(b)

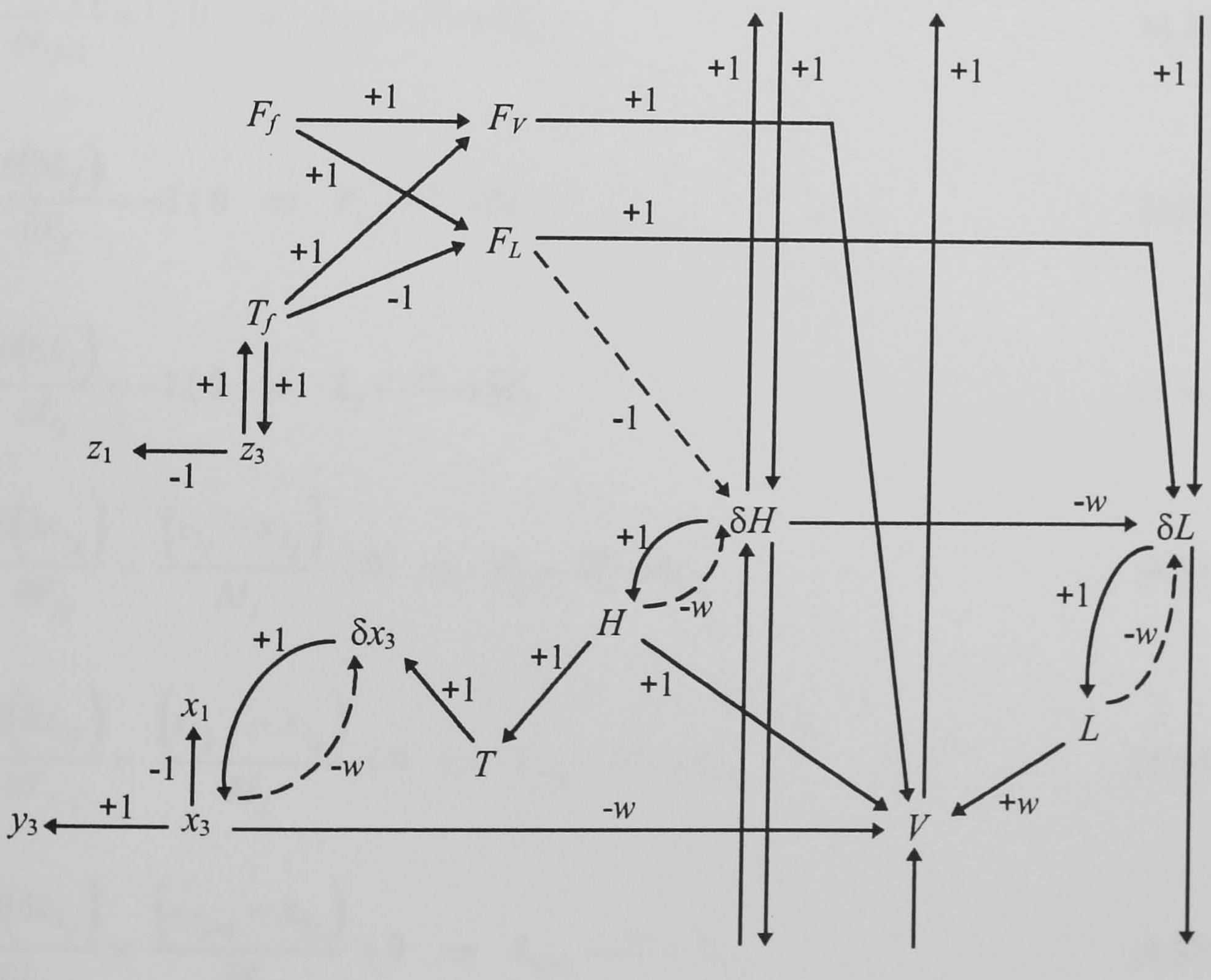


Figure 4.18 Generic section of a non-azeotropic ternary distillation column: (a) schematic representation and (b) weighted digraph model.

Causal Relationships

Based on Eqs. (4.72) to (4.75), relations (4.76) to (4.79) and the simplifying assumptions, the following causal relationships are obtained:

$$\frac{\partial(\delta L_j)}{\partial F_{f_j}} = 1 \rangle 0 \quad \Rightarrow \quad F_{f_j} \xrightarrow{+w} \delta L_j \tag{4.80}$$

$$\frac{\partial(\delta L_j)}{\partial V_{j-1}} = 1 \rangle 0 \quad \Rightarrow \quad V_{j-1} \xrightarrow{+w} \delta L_j \tag{4.81}$$

$$\frac{\partial(\delta L_j)}{\partial L_{j+1}} = 1 \rangle 0 \quad \Rightarrow \quad L_{j+1} \xrightarrow{+w} \delta L_j \tag{4.82}$$

$$\frac{\partial(\delta L_j)}{\partial V_j} = -1 \langle 0 \quad \Rightarrow \quad V_j \xrightarrow{-w} \delta L_j \tag{4.83}$$

$$\frac{\partial(\delta L_j)}{\partial L_j} = -1 \langle 0 \quad \Rightarrow \quad L_j \xrightarrow{-w} \delta L_j \tag{4.84}$$

$$\frac{\partial(\delta x_{3_j})}{\partial F_{f_j}} = \frac{(z_{3_j} - x_{3_j})}{M_j} \langle 0 \quad \Rightarrow \quad F_{f_j} \xrightarrow{-w} \delta x_{3_j} \tag{4.85}$$

$$\frac{\partial(\delta x_{3_j})}{\partial V_{j-1}} = \frac{(y_{3_{j-1}} - x_{3_j})}{M_j} \langle 0 \quad \Rightarrow \quad V_{j-1} \xrightarrow{-w} \delta x_{3_j} \tag{4.86}$$

$$\frac{\partial(\delta x_{3_j})}{\partial L_{j+1}} = \frac{(x_{3_{j+1}} - x_{3_j})}{M_j} \langle 0 \quad \Rightarrow \quad L_{j+1} \xrightarrow{-w} \delta x_{3_j} \tag{4.87}$$

$$\frac{\partial(\delta x_{3_j})}{\partial V_j} = -\frac{(y_{3_j} - x_{3_j})}{M_j} \rangle 0 \quad \Rightarrow \quad V_j \xrightarrow{+w} \delta x_{3_j} \tag{4.88}$$

$$\frac{\partial\left(\delta x_{3_j}\right)}{\partial z_{3_j}} = \frac{F_{f_j}}{M_j} \rangle 0 \quad \Rightarrow \quad z_{3_j} \xrightarrow{+w} \delta x_{3_j} \tag{4.89}$$

$$\frac{\partial\left(\delta x_{3_j}\right)}{\partial x_{3_j}} = \frac{1}{M_j}\left[-F_{f_j} - V_{j-1} - L_{j+1} + V_j\right] \langle 0 \quad \Rightarrow \quad x_{3_j} \xrightarrow{-w} \delta x_{3_j} \tag{4.90}$$

$$\frac{\partial\left(\delta x_{3_j}\right)}{\partial y_{3_{j-1}}} = \frac{V_{j-1}}{M_j} \rangle 0 \quad \Rightarrow \quad y_{3_{j-1}} \xrightarrow{+w} \delta x_{3_j} \tag{4.91}$$

$$\frac{\partial\left(\delta x_{3_j}\right)}{\partial x_{3_{j+1}}} = \frac{L_{j+1}}{M_j} \rangle 0 \quad \Rightarrow \quad x_{3_{j+1}} \xrightarrow{+w} \delta x_{3_j} \tag{4.92}$$

$$\frac{\partial\left(\delta x_{3_j}\right)}{\partial y_{3_j}} = -\frac{V_j}{M_j} \langle 0 \quad \Rightarrow \quad y_{3_j} \xrightarrow{-w} \delta x_{3_j} \tag{4.93}$$

$$\frac{\partial\left(\delta H_j\right)}{\partial F_{f_j}} = \frac{\left(H_{f_j} - H_{L_j}\right)}{M_j} \langle 0 \quad \Rightarrow \quad F_{f_j} \xrightarrow{-w} \delta H_j \tag{4.94}$$

(valid for saturated liquid feeds)

$$\frac{\partial\left(\delta H_j\right)}{\partial V_{j-1}} = \frac{\left(H_{V_{j-1}} - H_{L_j}\right)}{M_j} \rangle 0 \quad \Rightarrow \quad V_{j-1} \xrightarrow{+w} \delta H_j \tag{4.95}$$

$$\frac{\partial\left(\delta H_j\right)}{\partial L_{j+1}} = \frac{\left(H_{L_{j+1}} - H_{L_j}\right)}{M_j} \langle 0 \quad \Rightarrow \quad L_{j+1} \xrightarrow{-w} \delta H_j \tag{4.96}$$

$$\frac{\partial\left(\delta H_j\right)}{\partial V_j} = -\frac{\left(H_{V_j} - H_{L_j}\right)}{M_j} \langle 0 \quad \Rightarrow \quad V_j \xrightarrow{-w} \delta H_j \tag{4.97}$$

$$\frac{\partial\left(\delta H_j\right)}{\partial H_{f_j}} = \frac{F_{f_j}}{M_j} \rangle 0 \quad \Rightarrow \quad H_{f_j} \xrightarrow{+w} \delta H_j \tag{4.98}$$

$$\frac{\partial\left(\delta H_j\right)}{\partial H_{L_j}} = \frac{1}{M_j}\left[-F_{f_j} - V_{j-1} - L_{j+1} + V_j\right] \langle 0 \quad \Rightarrow \quad H_{L_j} \xrightarrow{-w} \delta H_j \tag{4.99}$$

$$\frac{\partial(\delta H_j)}{\partial H_{V_{j-1}}} = \frac{V_{j-1}}{M_j} \rangle 0 \quad \Rightarrow \quad H_{V_{j-1}} \xrightarrow{+w} \delta H_j \tag{4.100}$$

$$\frac{\partial(\delta H_j)}{\partial H_{L_{j+1}}} = \frac{L_{j+1}}{M_j} \rangle 0 \quad \Rightarrow \quad H_{L_{j+1}} \xrightarrow{+w} \delta H_j \tag{4.101}$$

$$\frac{\partial(\delta H_j)}{\partial H_{V_j}} = -\frac{V_j}{M_j} \langle 0 \quad \Rightarrow \quad H_{V_j} \xrightarrow{-w} \delta H_j \tag{4.102}$$

Weighted Digraph Model

The WDG model for the ternary flash is modified to represent the individual sections of the column. The modifications, which are based on relations (4.80) to (4.102) and the considerations listed below, result in the basic WDG model shown in Fig. 4.18b for a representative section. Figure 4.20 shows the complete model for the column, including the condenser and reboiler.

- In each section of the column, the fast dynamics are associated with the column hydraulics represented by the variable δL and the slow ones associated with energy flow represented by the variable δH. Changes in composition (δx_3) are considered to be a consequence of changes in the energy level in each section;

- Exchange of mass and energy between sections is made through the *differential nodes* δL and δH for liquid and energy flows, respectively, and the *algebraic node* V for vapour flow, since vapour hold-up is considered to be negligible, i.e. $\delta V = 0$;

- Relations (4.80), (4.83), (4.85), (4.88), (4.89), (4.94), (4.97) and (4.98) are essentially represented in the basic flash model if it is assumed that the

algebraic variables H, L and x_3 are replaced by their *differential* equivalents δH, δL and δx_3;

- H_V and H_L are grouped into a generic variable H;

- Relations (4.84), (4.90), (4.99) and (4.102) can be considered to be compensatory influences of the *self-regulating groups*: (δL, L); (δx_3, x_3) and (δH, H) and so are represented by *temporal edges* with *weights* different from ±1;

- Relations (4.81), (4.82), (4.87), (4.91), (4.92), (4.95), (4.96), (4.100) and (4.101) are indirectly represented through other influences so as to reduce the number of edges and so simplify the structure of the model;

- An order-of-magnitude analysis reveals that relations (4.85) to (4.88) are not important when compared to relations (4.89) to (4.93). This means that compositions are more strongly and directly affected by temperature and compositions of adjacent units than by flow rates;

- Relation (4.93) is indirectly represented by the *self-regulating* influence (4.90);

- For the top section (*RD*), schematically represented in Fig. 4.19a, a total condenser is assumed so the variable V and all *edges* related to it are eliminated from the basic model of the typical section (Fig. 4.18b). A variable R_f is included to represent the reflux flow rate, as shown in Fig. 4.20. Influences from R_f on the rectifying section (*RS*) are similar to those of the liquid portion of the feed, F_L, on the feed tray section (*FT*). The model for the heat-exchanger is coupled with the model of the *RD* section to provide the cooling water side (*CD*) contribution;

- For the bottom section (BT), schematically represented in Fig. 4.19b, the influence of L on V in the typical section (Fig. 4.18b) is eliminated, since the vapour boil-up (V_{BT}) can be considered as being mainly influenced by the reboiler duty (Q_{RB}) and the temperature of the upper section (T_{SS}), rather than by the liquid flow rate from the upper section (L_{SS}), which mainly influences the bottom product flow rate (B). A model for steam condensation is coupled with the bottom section model to provide the steam side contribution in the reboiler;

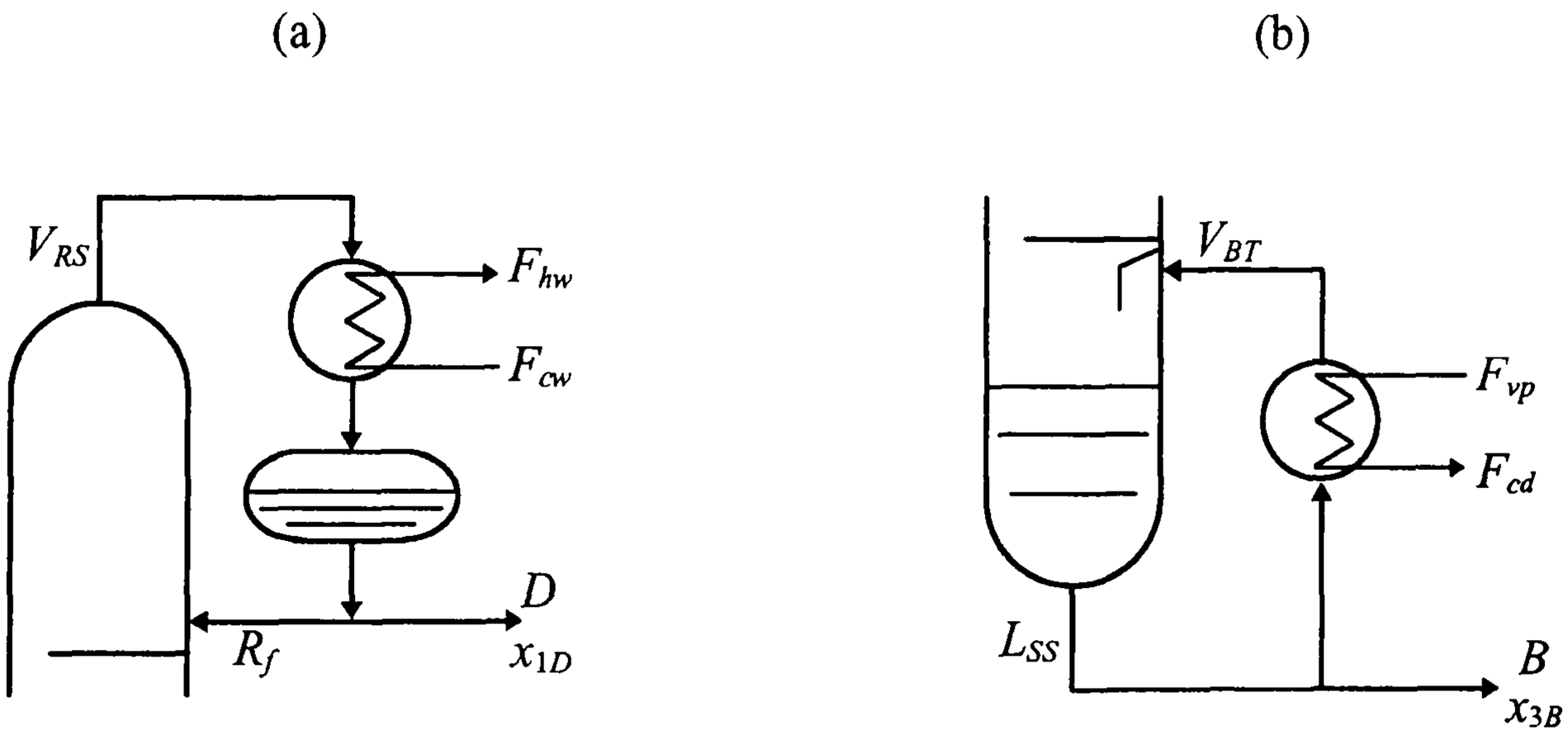


Figure 4.19 Schematic representation of (a) top and (b) bottom sections of a distillation column.

- The vapour flow rates (V) in the column are usually more strongly affected by temperature variations than by changes in the liquid flow rate (L). Therefore, the relation (L, V) must have a smaller *weight* than the relation (H, V);

- Further down the column, the influence (H, V) is relatively stronger than the influence (L, V), since V is affected mainly by the reboiler duty (Q_{RB}), which directly affects δH. Therefore, the *weight* of the influence (L, V)

must vary when moving down the column and will be smaller in the stripping section (*SS*);

- The liquid level in the bottom section (*BT*) is strongly affected by the reboiler duty, and therefore a high *weight* is attributed to the relation (δH, δL) in this section. In other sections of the column, the liquid level is mainly a function of the liquid flow rate from the upper section;

- Reactive *weights* of *self-regulating groups* are arbitrarily set at -0.5. Exceptions include:

  a) The liquid level at the bottom of the column (*BT*) and reflux drum (*RD*) are considered to vary quickly in relation to the overall column dynamics, and therefore high *weights* are attributed to the reactive influences (L, δL) in the *RD* and *BT* sections;

  b) Changes in the energy content (δH) affect the vapour flow rate (V) and liquid composition (x_3) of each section. After some time, changes in the composition influence the rate of change of V negatively. To reproduce this difference in speed of the influences, the reactive *weight* of the (H, δH) influence is set higher than that of (x_3, δx_3). This means that influences from H occur faster than those from x_3. Moreover, the compensatory influence (x_3, V) tends to be smaller in the lower section of the column, since the influence (H, V) is relatively stronger at the bottom of the column because of the influences from the reboiler.

The WDG model for the sections can be combined to represent the overall description of the column, and the result is shown in Fig. 4.20. The model is valid for any distillation column, with any number of trays separating a non-azeotropic ternary mixture composed of one feed, a total condenser and one thermosyphon reboiler. However, the fact that the influence of mechanical

construction details in the qualitative behaviour of the distillation column is not considered to impose limits to the generality of the model.

Qualitative Behaviour of the Column

Figures 4.21 to 4.39 compare results of the qualitative and quantitative (numerical) simulations for step disturbances in the feed flow rate (F_f), feed temperature (T_f), reflux flow rate (R_f) and reboiler duty (Q_{RB}), in turn. The aim is to analyse the adequacy of the WDG model, shown in Fig. 4.20, in describing the dynamic response of a column, i.e. its effectiveness in capturing the most important features of the dynamic behaviour of each section of the column.

Qualitative trajectories are obtained by applying the algorithm for inference of behaviour, described in Chapter 3, to the model shown in Fig. 4.20, for positive or negative step disturbances of ±1 to one of the input variables (F_f, T_f, R_f or Q_{RB}).

The numerical simulation is based on Eqs. (4.72) to (4.75) and the methodology described by Gani *et al.* (1986). The description of the ternary system and column parameters used are given in Table A.6 in Appendix A. A positive or negative step disturbance of 10% is considered for each input variable (F_f, T_f, R_f or Q_{RB}) at a particular time.

The behaviour of the rectifying (*RS*) and stripping (*SS*) sections in the qualitative simulation are compared to the behaviour of the top (number 8) and bottom (number 1) trays of the column described in Appendix A. The results are compared in terms of liquid (L) and vapour (V) flow rates in each section of the column, temperature (T) and composition of the most volatile component (x_1) of the top product (D), and temperature (T) and composition of the least volatile component (x_3) of the bottom product (B).

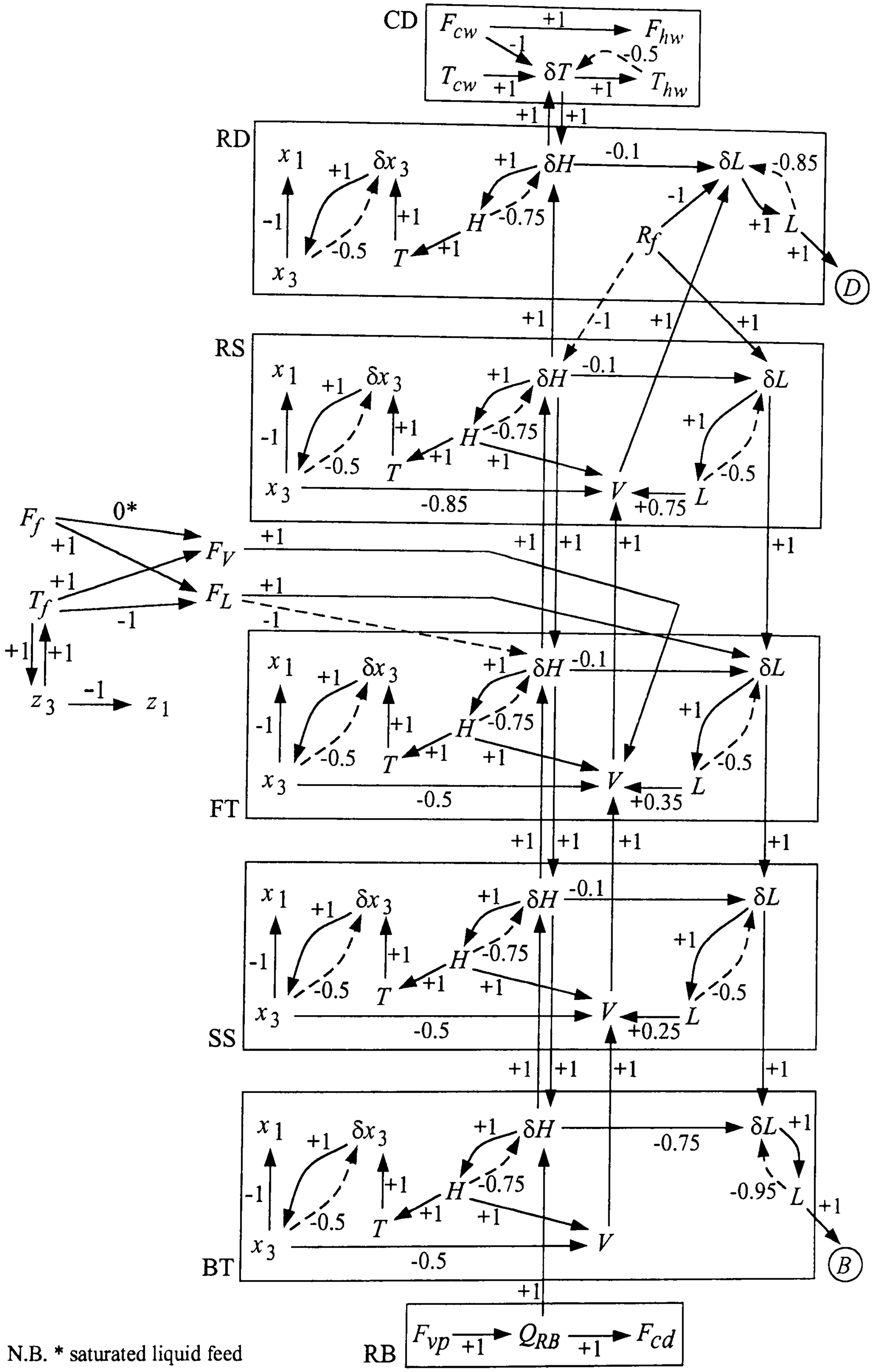


Figure 4.20 Weighted digraph model for a non-azeotropic ternary distillation column.

Figure 4.21 shows the behaviour of the top and bottom product flow rates for a negative step disturbance in the feed flow rate (F_f). It can be seen from Fig. 4.21a that the transient response of the distillate (dashed line) is quite complex. The WDG model is able to adequately capture this peculiar trajectory by qualitatively describing the two turning points and the approach to the steady-state, as can be seen in Fig. 4.21b. This means that first and second-order derivatives are described well, fast and slow dynamics are scheduled with respect to time and the strength of the influences are adequately represented. This is achieved by the proper use of *weights*, *temporal edges* and *differential nodes*.

Figures 4.22 and 4.23 illustrate the behaviour of the flows inside the column for a negative step disturbance in the feed flow rate. It is clear that, although the WDG model adequately captures the general qualitative behaviour of the variables, including the complex behaviour of the vapour flow rate, in some cases it may not be sensitive enough to distinguish small variations around the initial state, i.e. the model cannot determine if the final state will be slightly higher or lower than the initial state. For example, in the stripping section, the vapour flow oscillates and the final value (103.0 mol/s) is slightly lower than the initial one (103.1 mol/s). Although in qualitative terms this essentially means no real variation, the qualitative model predicts an increase in the vapour flow rate. This indicates that the *weights* should be better tuned to reduce the range of the positive influence and thus approximate the final to the initial state. However, it may still not be able to distinguish between small variations around a given value. It would only be possible to do this if more precise numerical information were provided to the qualitative model, which would obviously require considerable effort which would be disproportionate to the significance of the result.

Temperature, compositions and bottom vapour flow rate (V_{BT}) are very well described for the negative disturbance in feed flow rate (F_f). Figures 4.24 and 4.25

illustrate the behaviour of distillate composition and temperature, and bottom product composition and temperature.

For step disturbances in the feed temperature (T_f) and reflux rate (R_f) the WDG model describes the changes in all variables very well, including vapour and liquid flow rates inside the column. Figures 4.26 to 4.32 illustrate the behaviour of some of the variables for a negative step disturbance in the feed temperature (T_f) and a positive step disturbance in the reflux flow rate (R_f).

For a positive step in the reboiler duty (Q_{RB}) most of the qualitative results are very good, as can be seen by examining Figs. 4.33 to 4.39. However, the qualitative trajectories for the internal liquid flow rates of the column do not exactly match, in terms of main features, those of the quantitative simulation. This happens because for the quantitative simulation, the reflux flow rate has to be automatically adjusted with increase in the reboiler duty in order to avoid a low level at the bottom of the column, which would terminate the simulation.

Clearly, the WDG model for the distillation column (Fig. 4.20) is very effective in describing complex patterns of dynamic behaviour without generating ambiguous solutions. It is able to capture the differences in the qualitative behaviour of each section of the column, revealing its adequacy to represent distributed parameter systems. This is possible because of the use of *weights* which have to be tuned to reflect the strength of the influences so that the behaviour of each particular section can be described properly.

As expected, the WDG model for the distillation column is far more complex than those for the CSTR or heat-exchanger. This means that it is difficult to visualise

the flow of information and how this affects the variables and consequently to explain how solutions are generated over time. However, it can be used to understand how general behaviour patterns arise. For example, for a negative step disturbance in the feed flow rate (F_f) the two turning points of the distillate flow rate (Fig. 4.21) can be explained using the WDG model in Fig. 4.20. The initial decrease in the distillate flow rate is a direct consequence of the decrease in the internal vapour flow rate through the positive *ordinary edge* from F_f to F_L and then to L and V, which is then propagated to D. After the second time step, the energy content (δH) is affected through the negative *temporal edge* from F_L. Therefore, δH, T and V increase which causes D to increase, giving rise to the first turning point in the distillate curve. The second turning point arises from the decrease in V due to a combination of reactive influences from x_3, δH and changes in the vapour boil-up (V_{BT}). However, simply examining the model, it is not possible to determine which reactive influence prevails and generates the second turning point. By experience, it is known that it is mainly due to adjustments in the vapour boil-up, which is a consequence of changes in temperature and composition of the liquid from tray 1 (stripping section). The mathematical model shows that the steady-state is approached because of the reactive influences due to x_3 and H.

The visualisation of the flow of information in the distillation model can be made easier by creating a visualisation interface coupled with the qualitative simulation algorithm. This gives insight into the dynamic flow of information through the model structure and the dominant influences which characterise the system behaviour so that they can be traced over time.

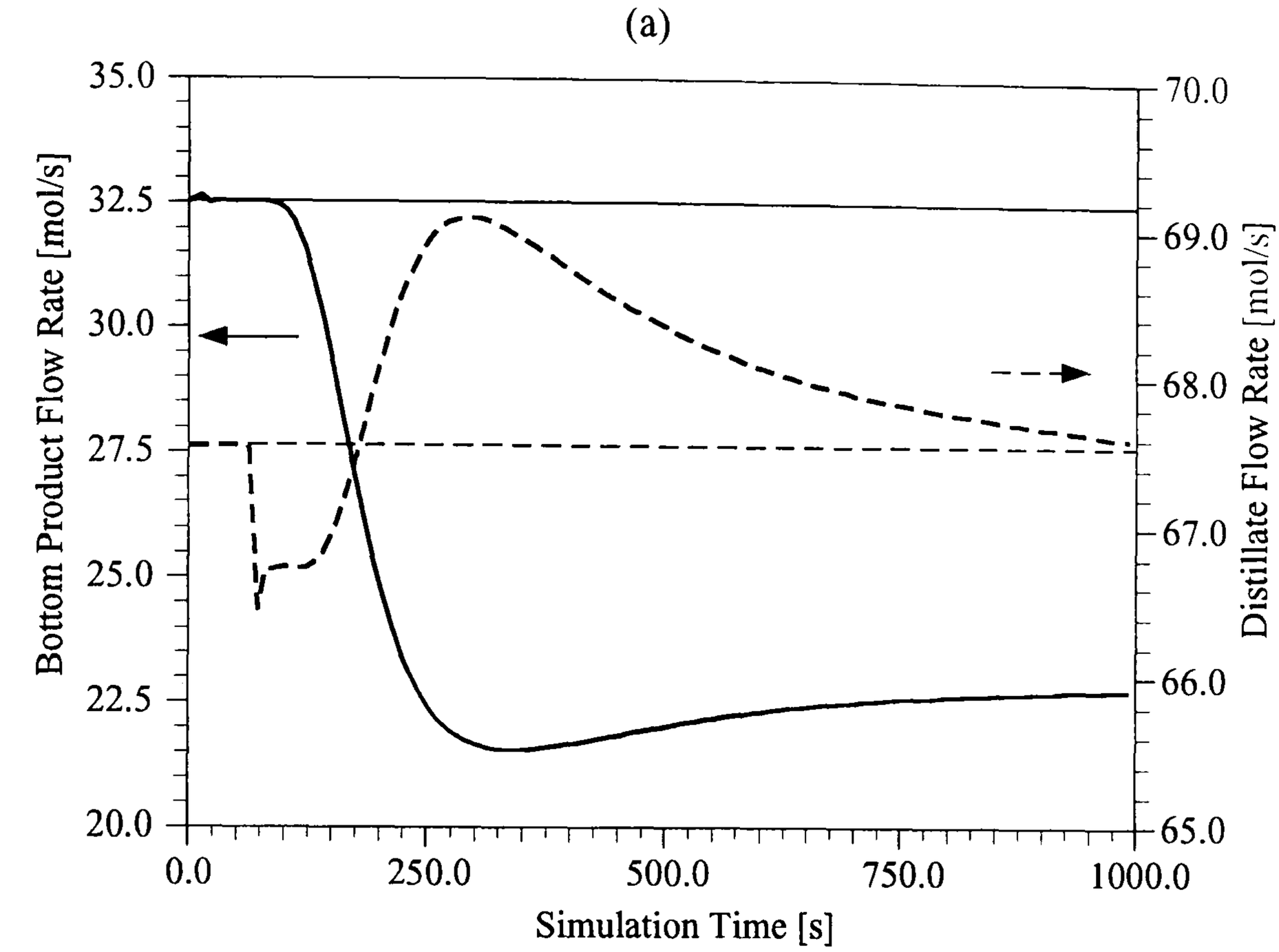
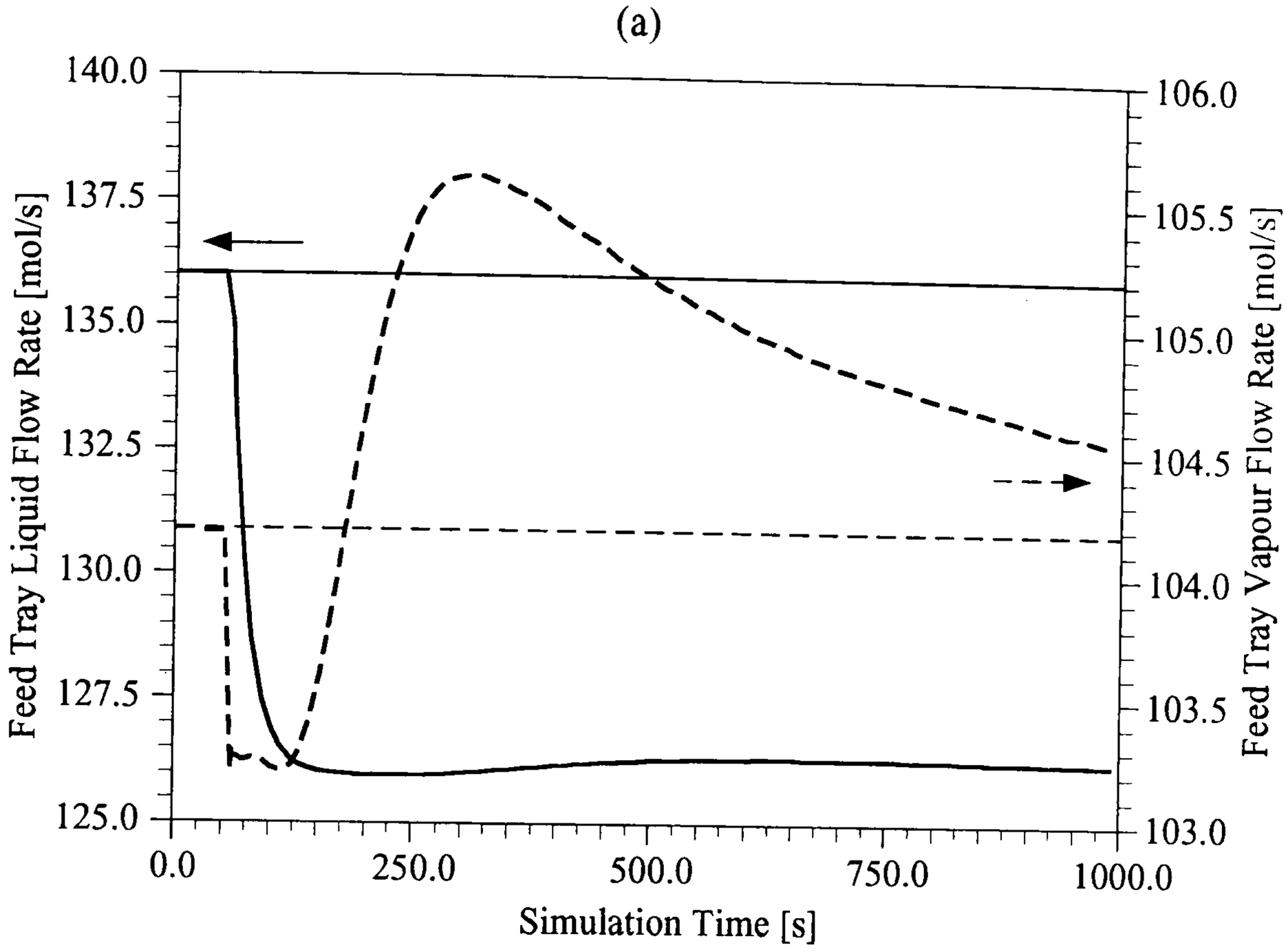


Figure 4.21 Distillation column - distillate and bottom product flow rate profiles for a negative step disturbance in feed flow rate: (a) numerical simulation; (b) qualitative simulation.



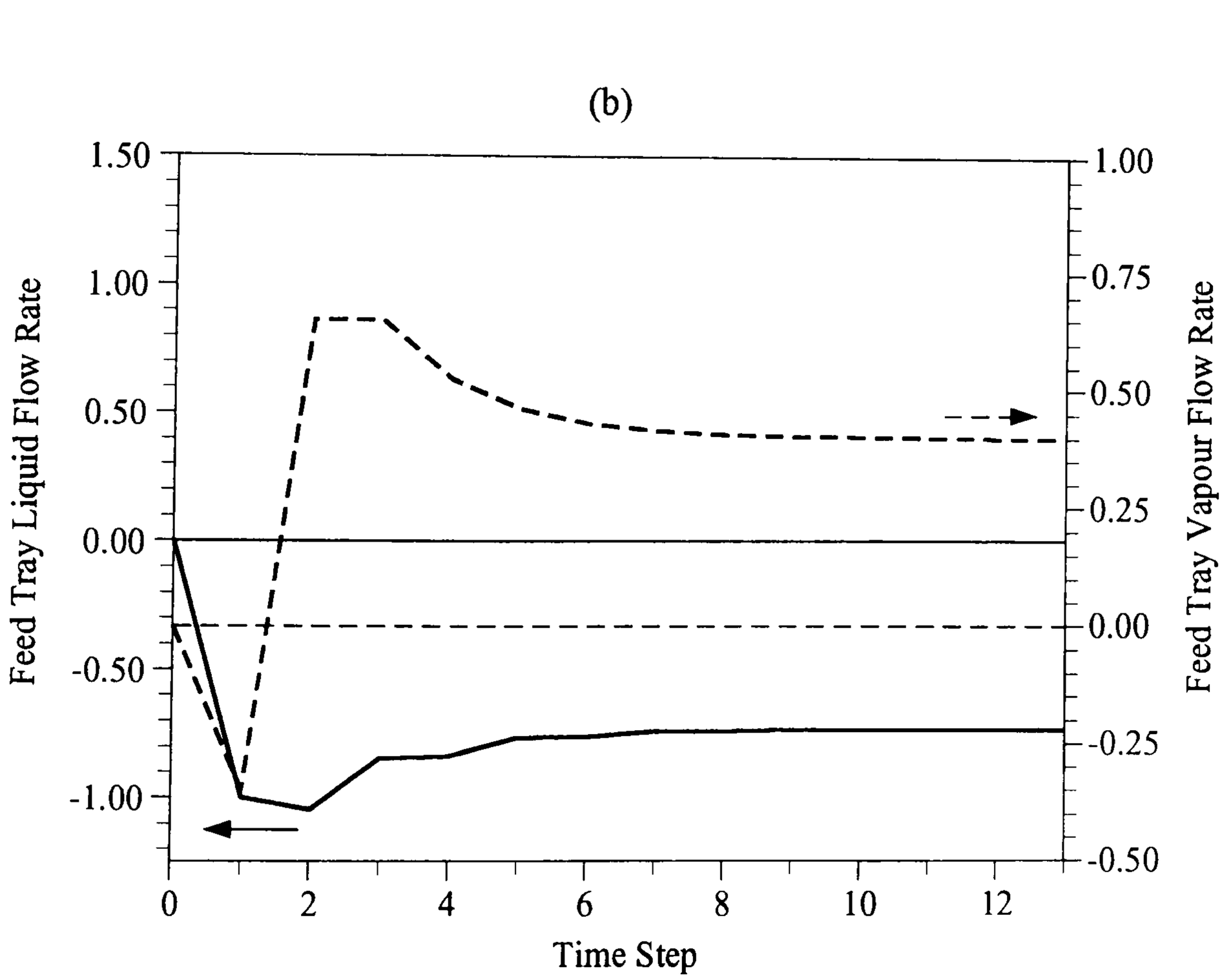


Figure 4.22 Distillation column - feed tray liquid and vapour flow rate profiles for a negative step disturbance in feed flow rate: (a) numerical simulation; (b) qualitative simulation.

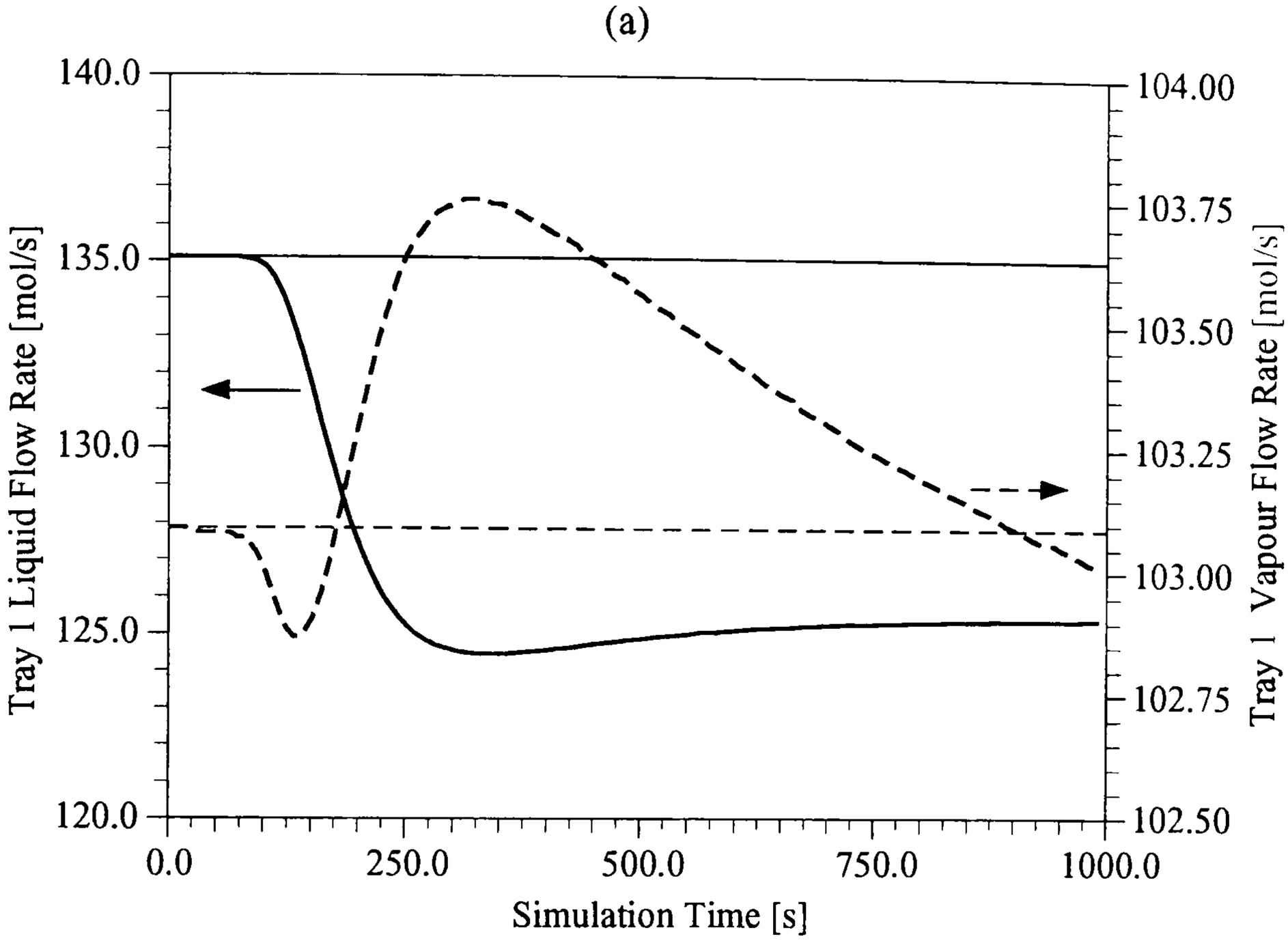


Figure 4.23 Distillation column - liquid and vapour flow rate profiles for a negative step disturbance in feed flow rate: (a) numerical simulation (tray 1); (b) qualitative simulation (stripping section).

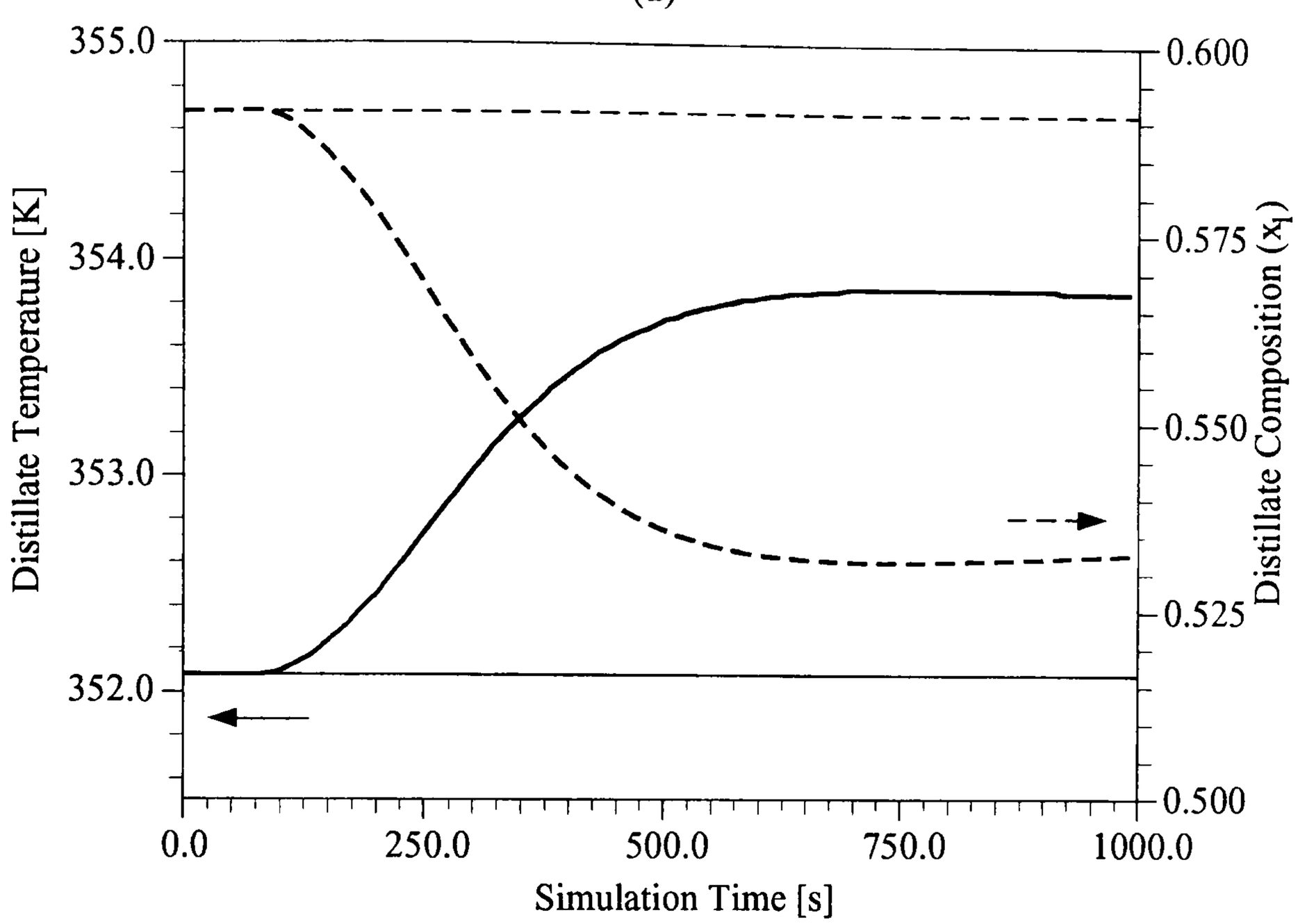
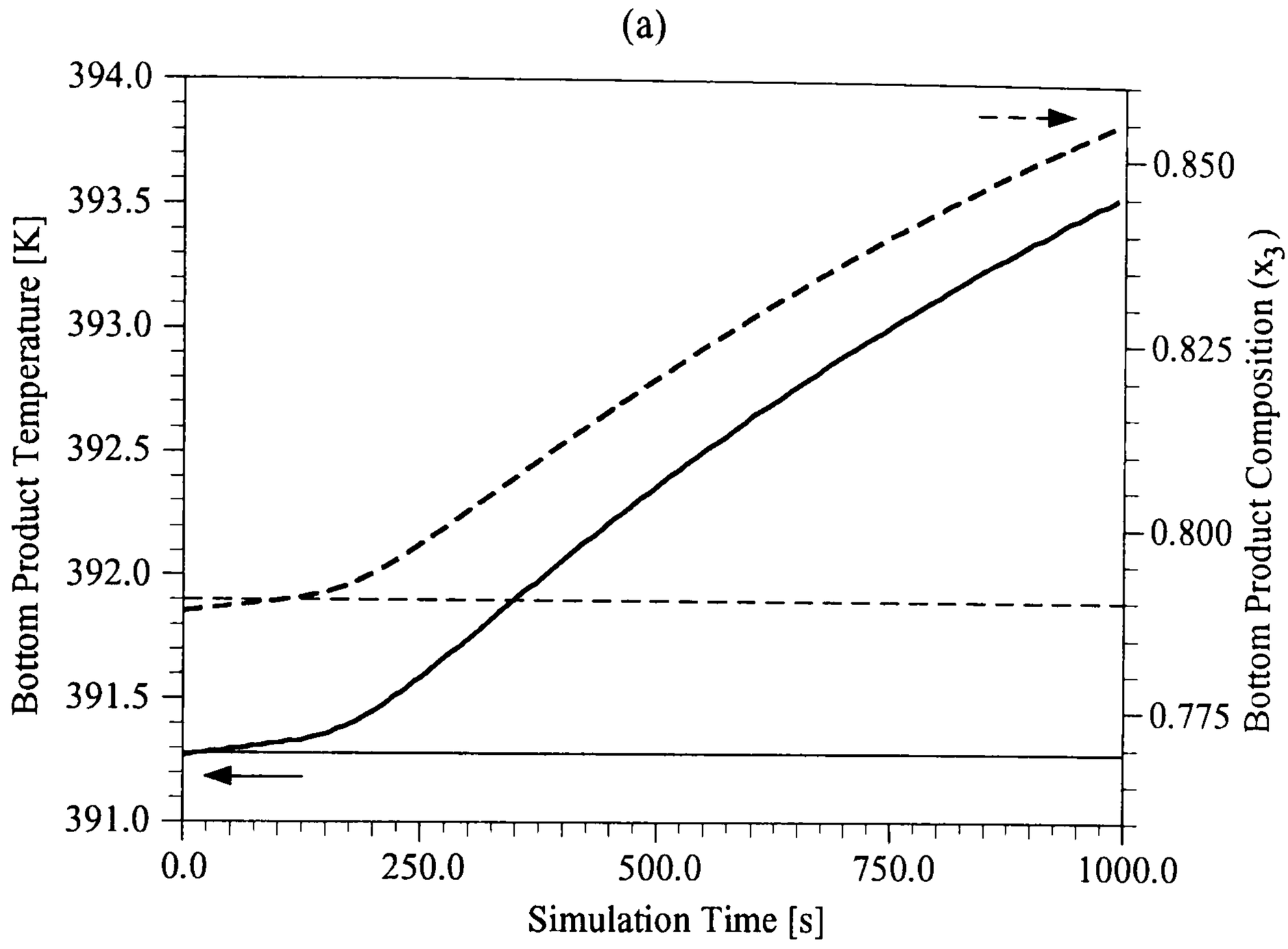


Figure 4.24 Distillation column - distillate temperature and composition profiles for a negative step disturbance in feed flow rate: (a) numerical simulation; (b) qualitative simulation.



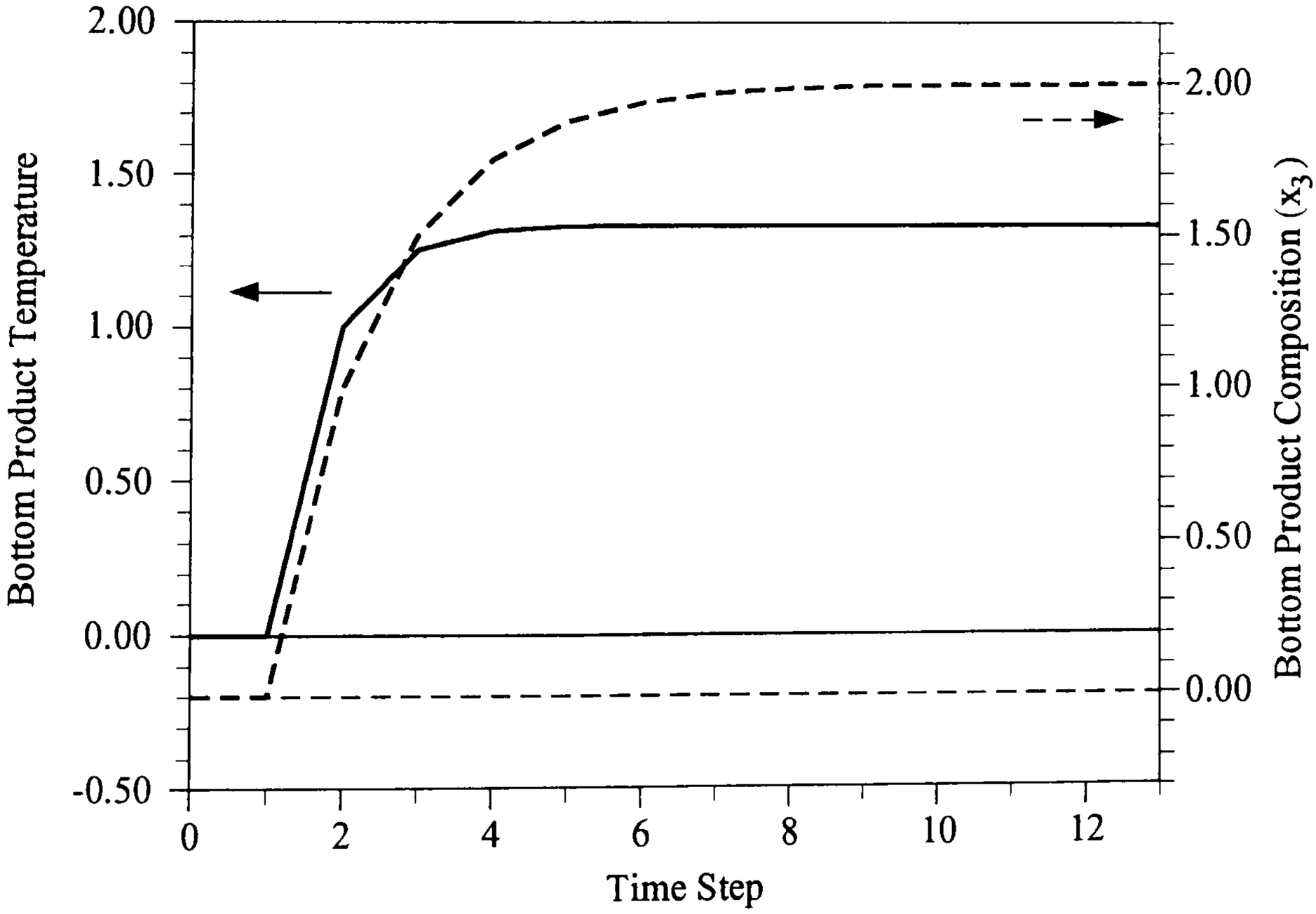
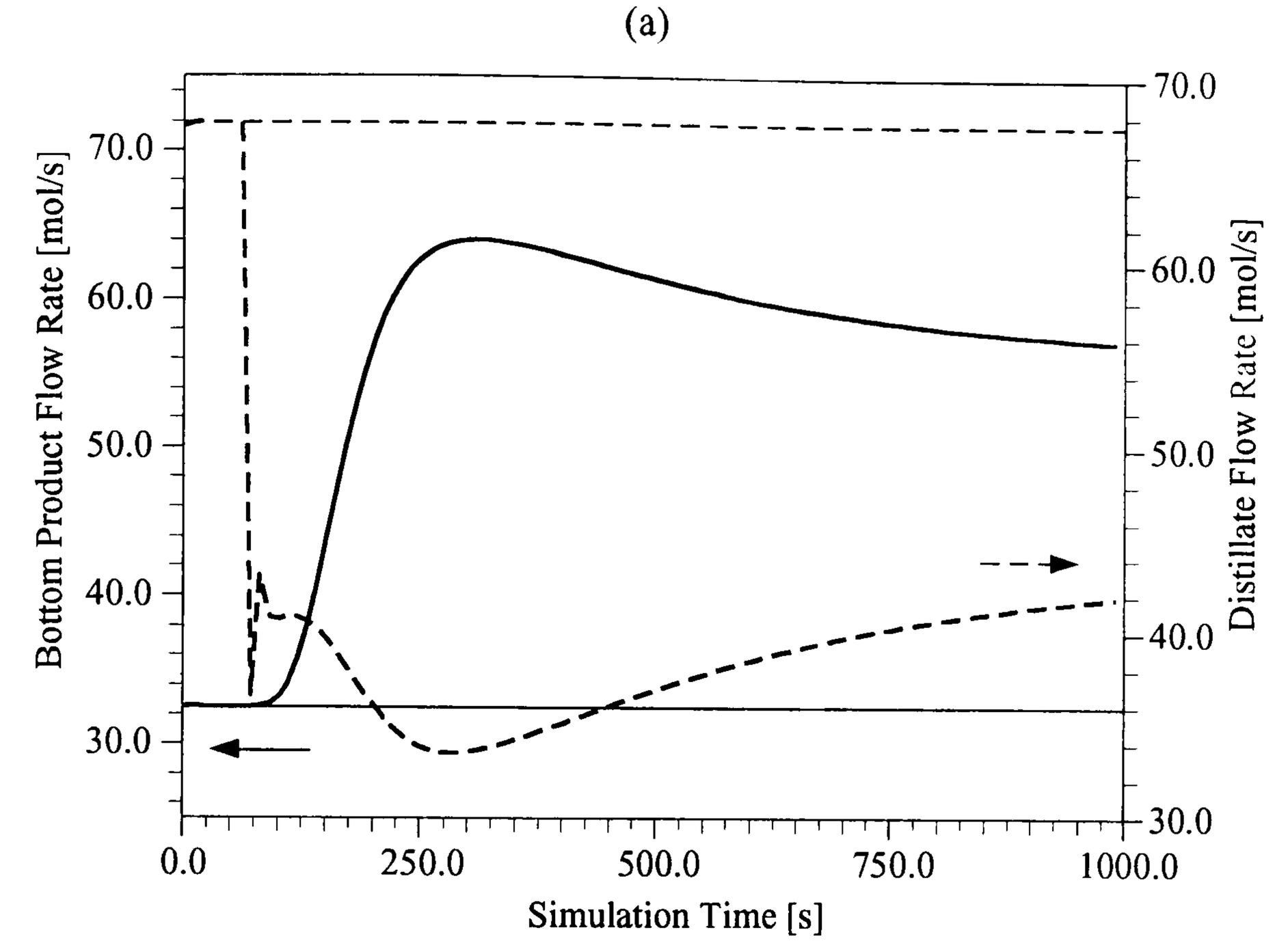


Figure 4.25 Distillation column - bottom product temperature and composition profiles for a negative step disturbance in feed flow rate: (a) numerical simulation; (b) qualitative simulation.



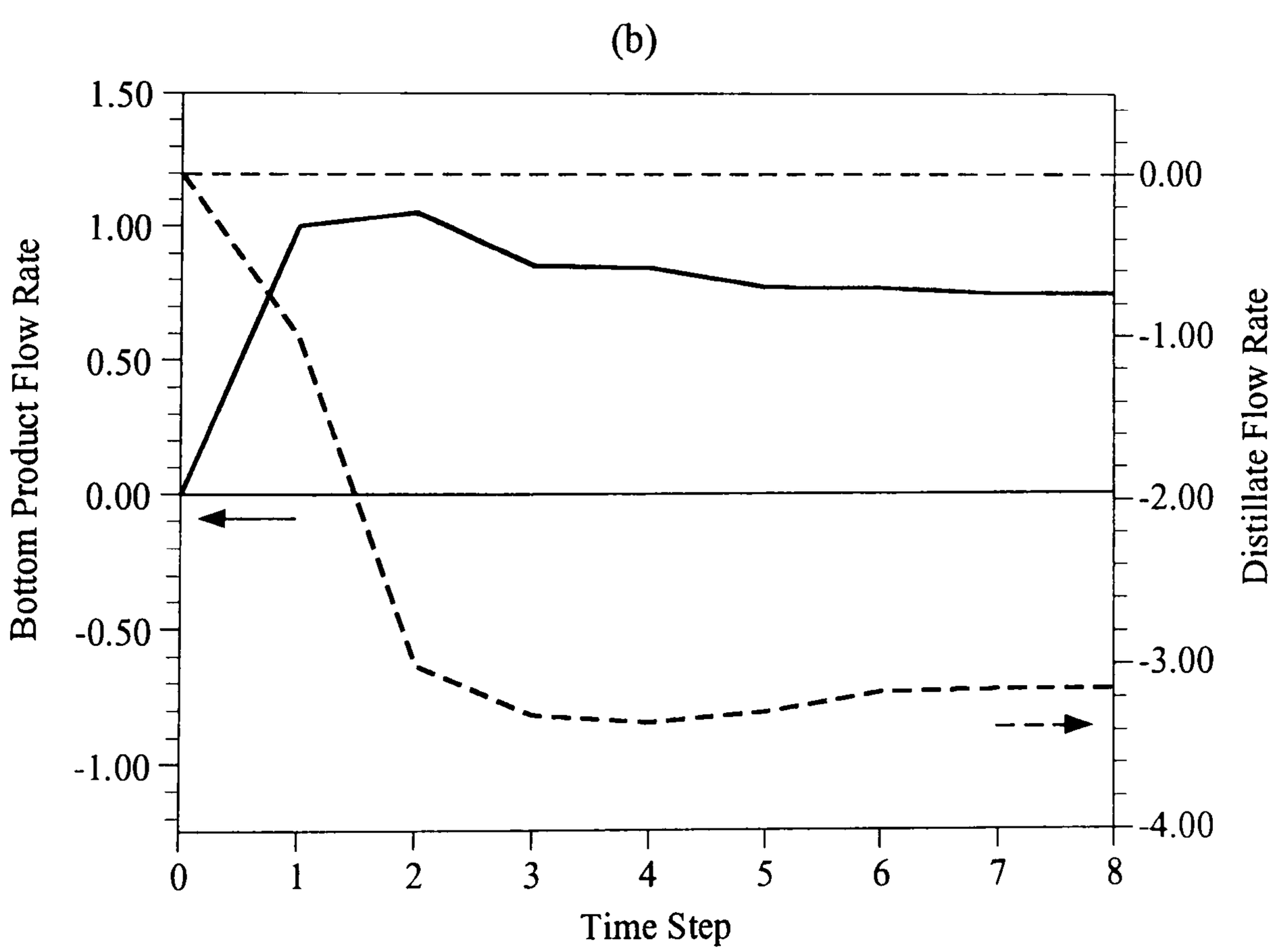


Figure 4.26 Distillation column - distillate and bottom product flow rate profiles for a negative step disturbance in feed temperature: (a) numerical simulation; (b) qualitative simulation.

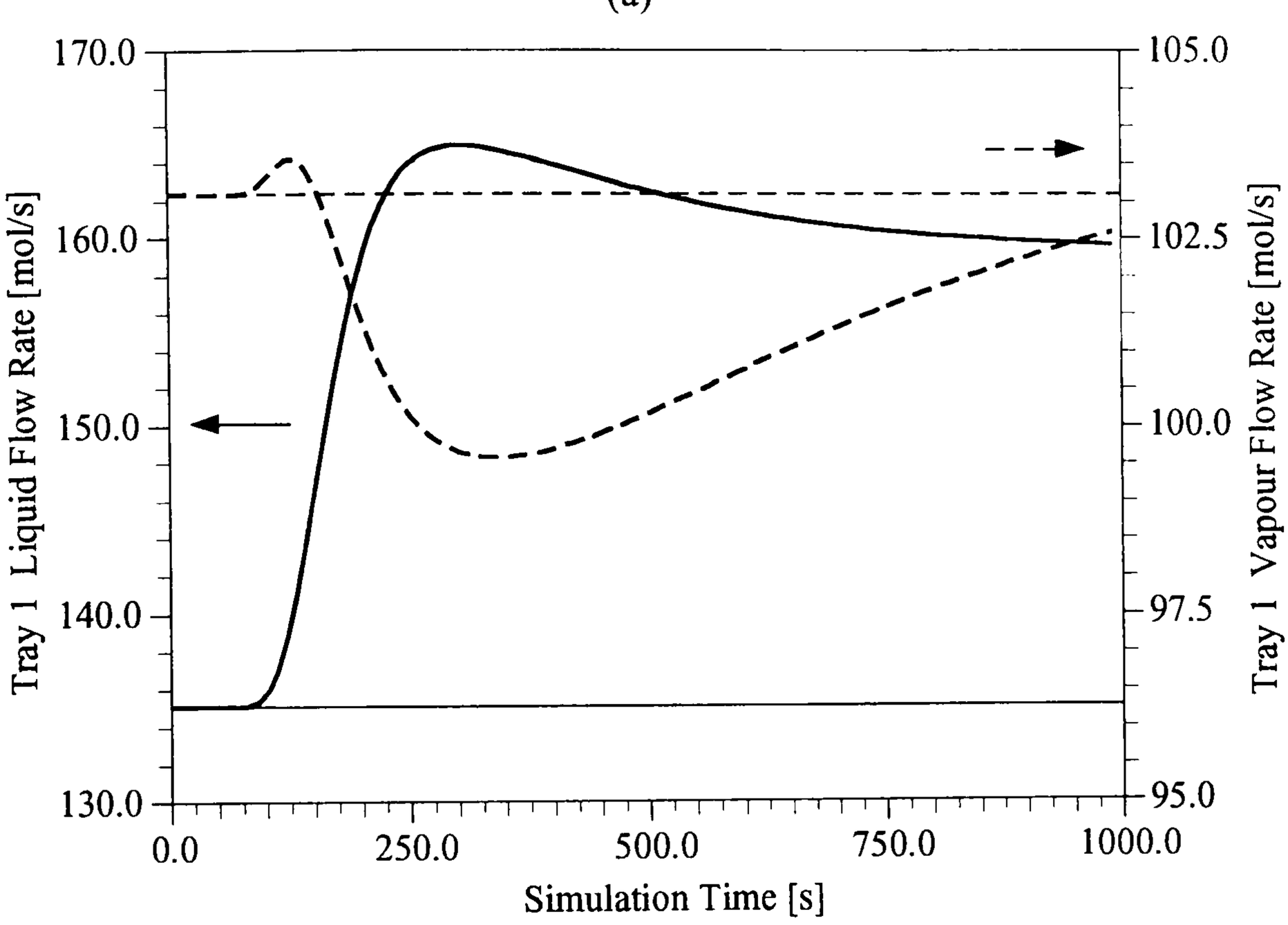
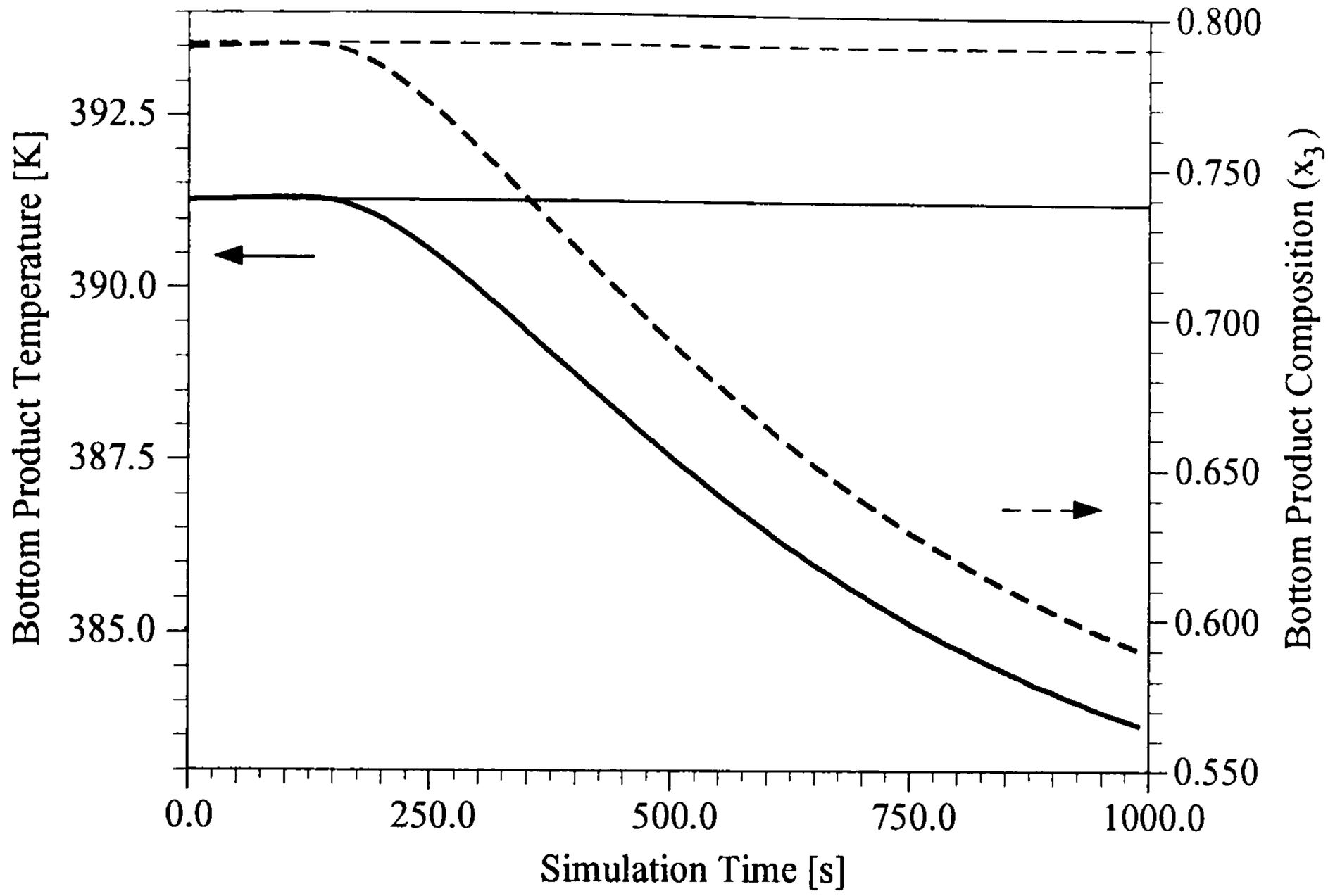


Figure 4.27 Distillation column - liquid and vapour flow rate profiles for a negative step disturbance in feed temperature: (a) numerical simulation (tray 1); (b) qualitative simulation (stripping section).

(a)



(b)

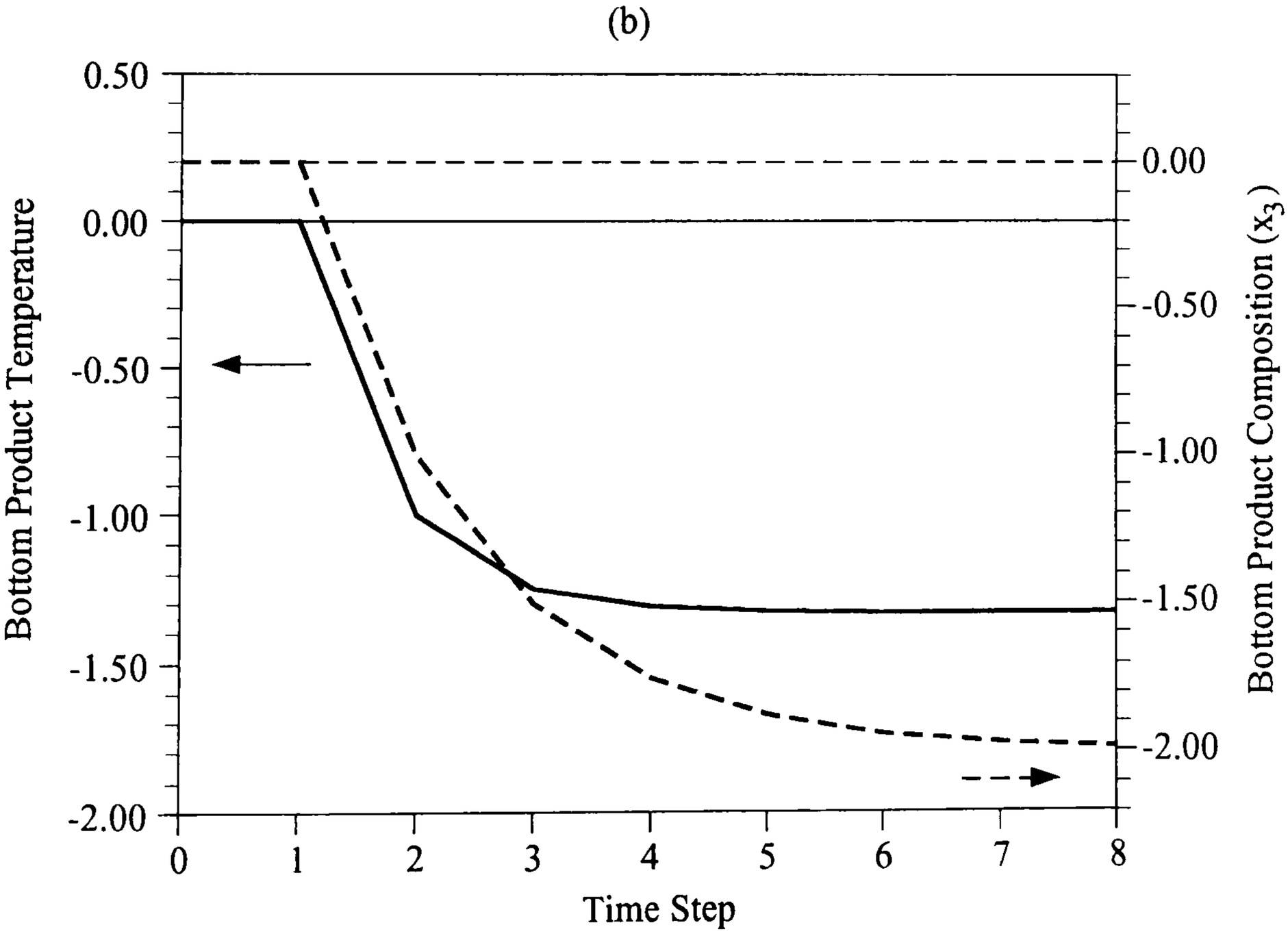


Figure 4.28 Distillation column - bottom product temperature and composition profiles for a negative step disturbance in feed temperature: (a) numerical simulation; (b) qualitative simulation.

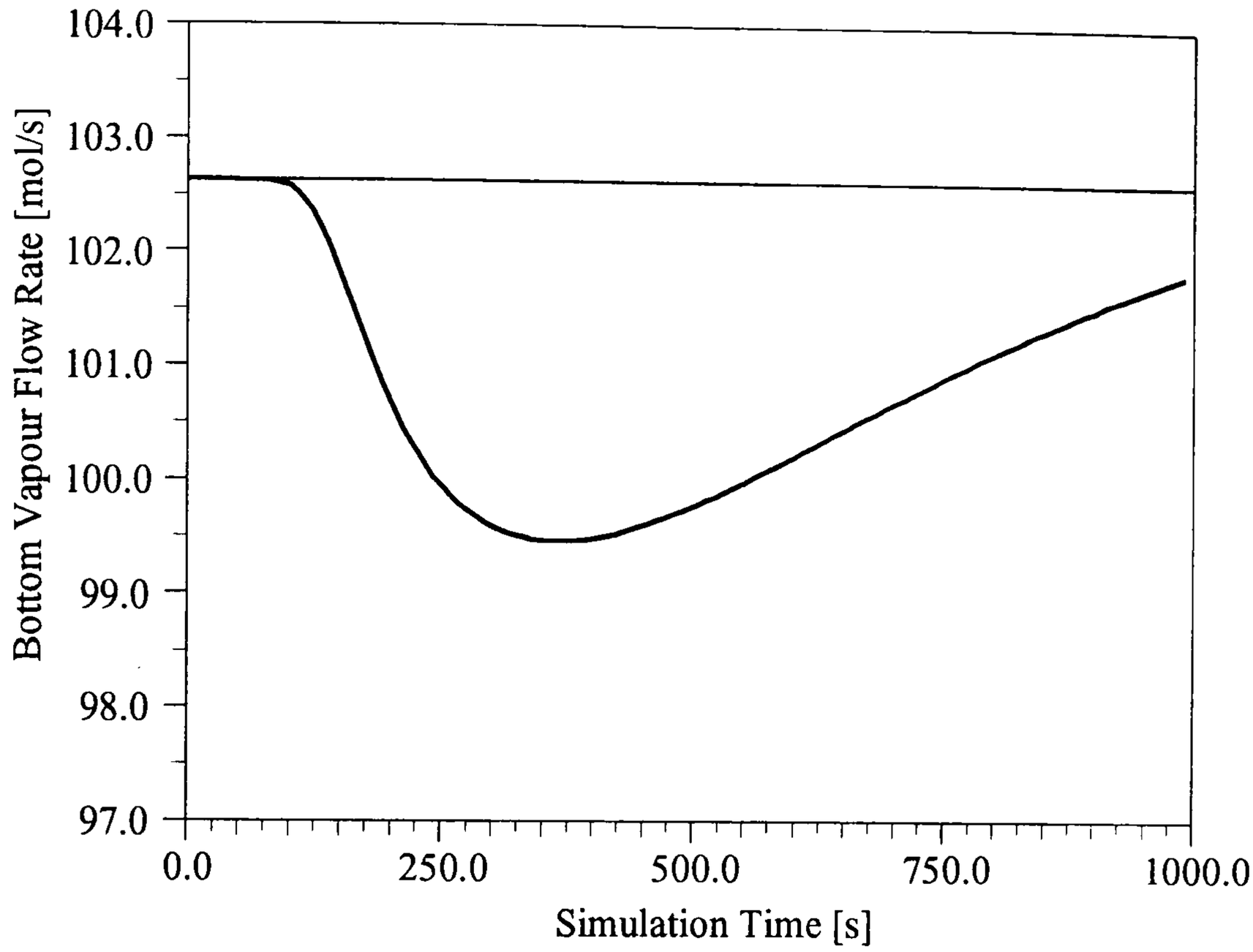
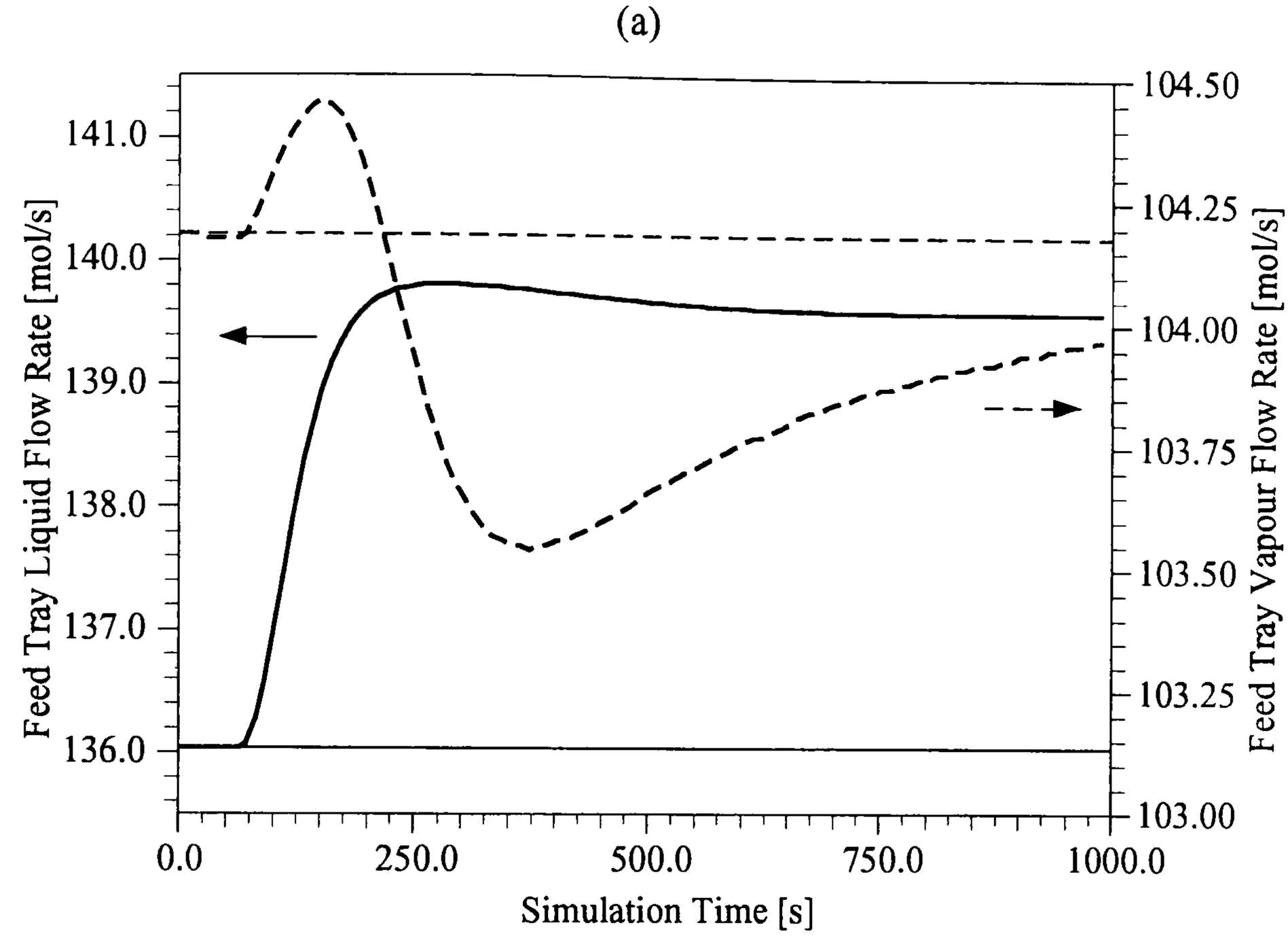


Figure 4.29 Distillation column - bottom vapour flow rate profile for a negative step disturbance in feed temperature: (a) numerical simulation; (b) qualitative simulation.



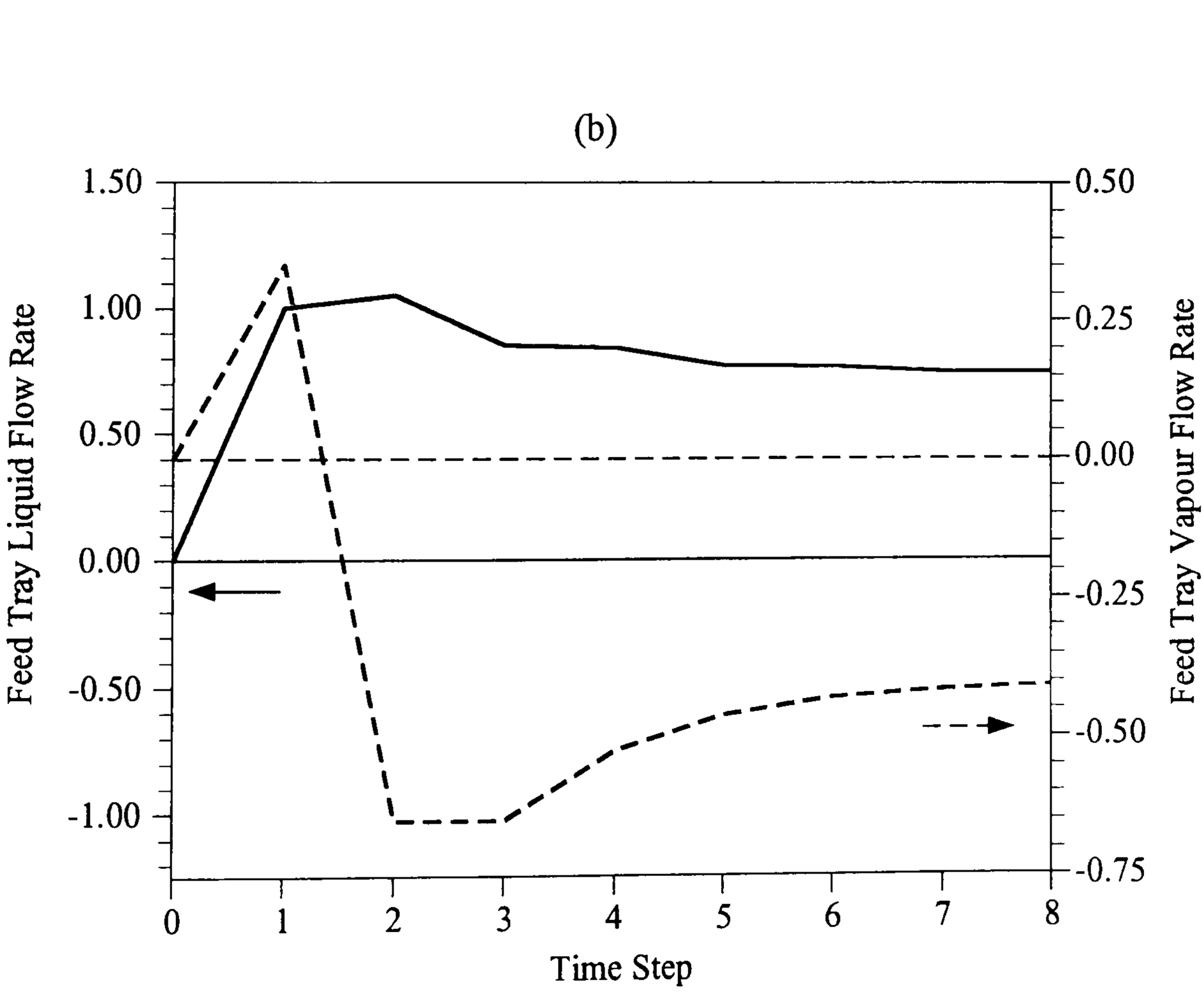


Figure 4.30 Distillation column - feed tray liquid and vapour flow rate profiles for a positive step disturbance in reflux flow rate: (a) numerical simulation; (b) qualitative simulation.

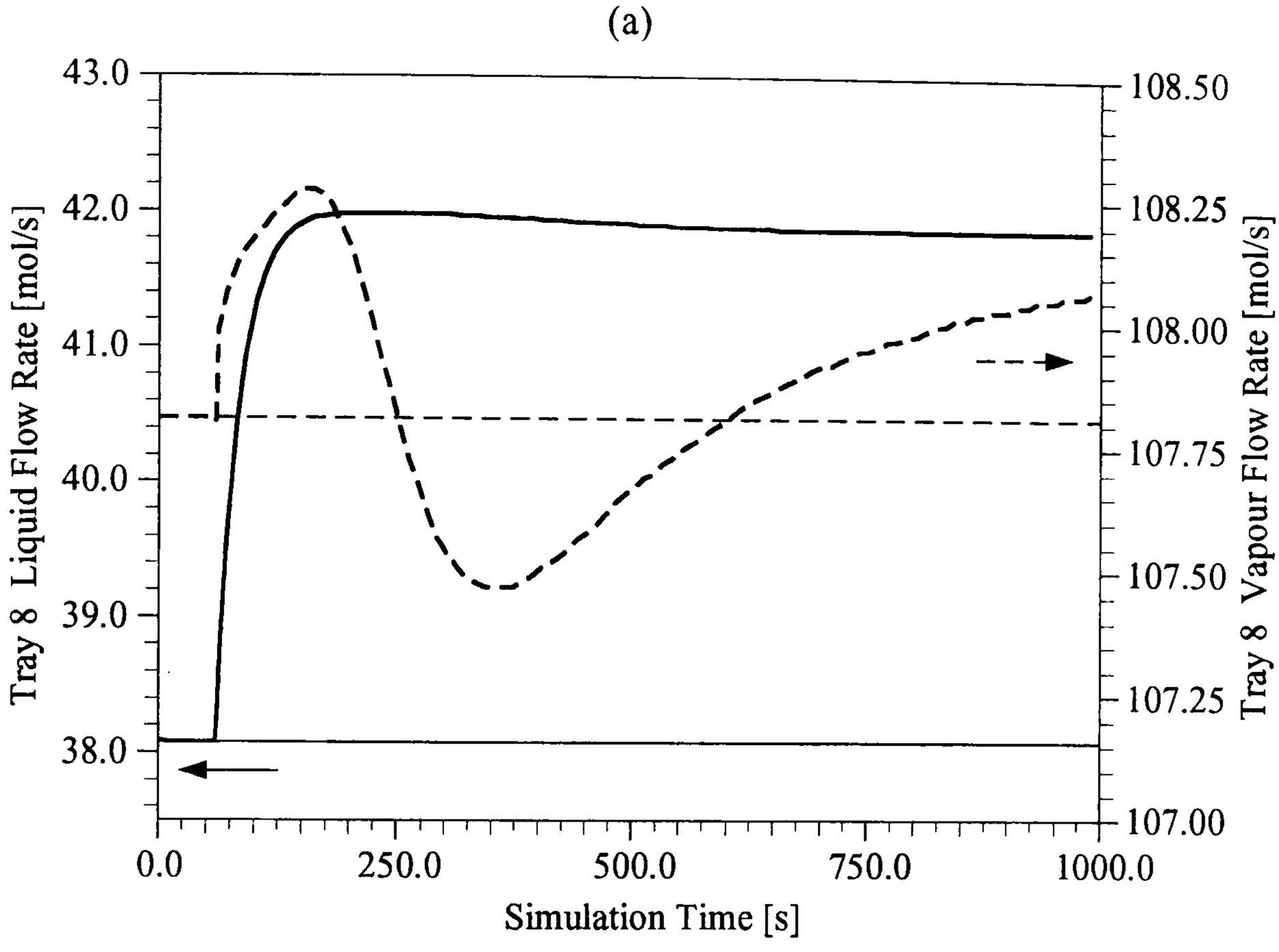
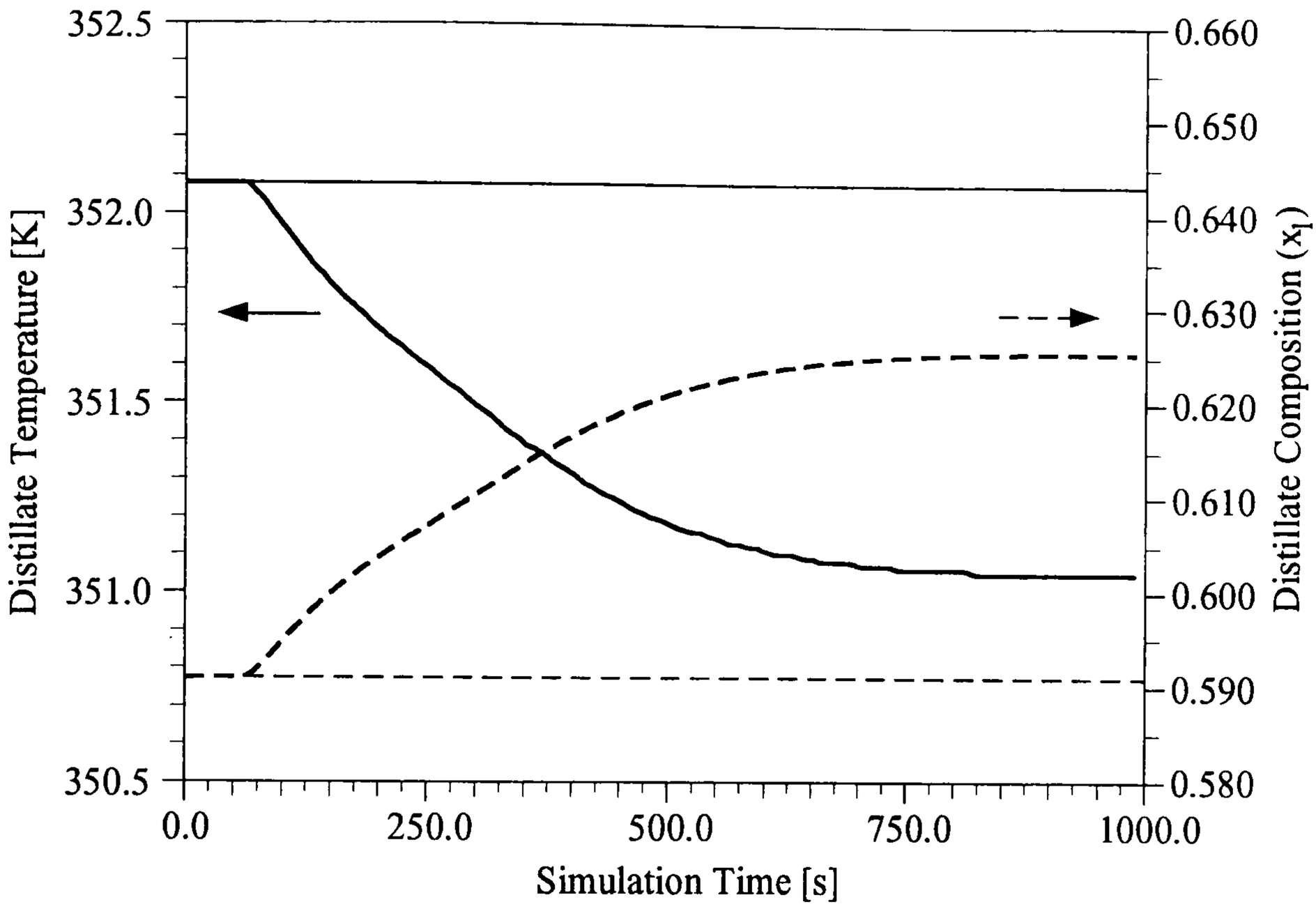


Figure 4.31 Distillation column - liquid and vapour flow rate profiles for a positive step disturbance in reflux flow rate: (a) numerical simulation (tray 8); (b) qualitative simulation (rectifying section).



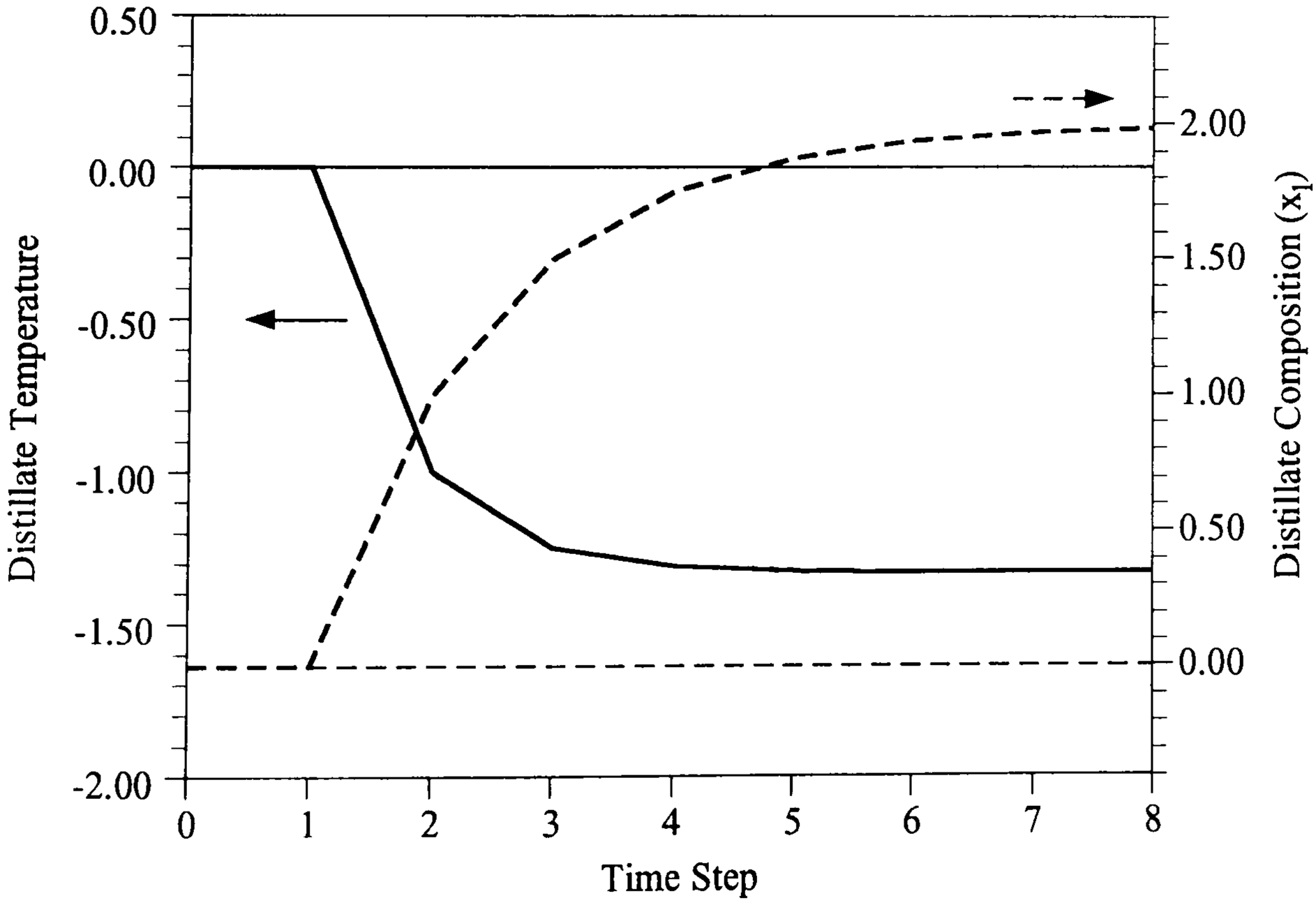
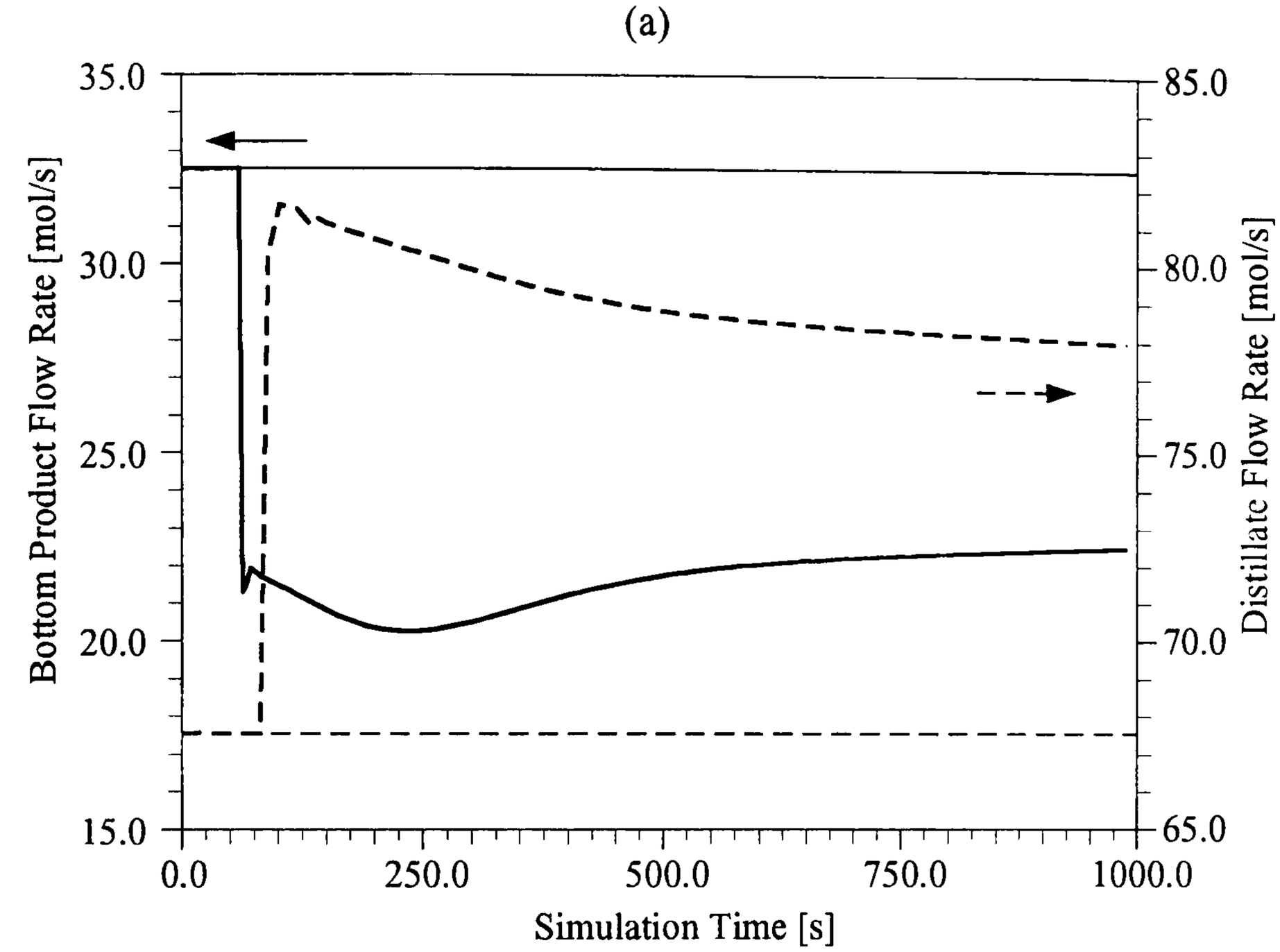


Figure 4.32 Distillation column - distillate temperature and composition profiles for a positive step disturbance in reflux flow rate: (a) numerical simulation; (b) qualitative simulation.



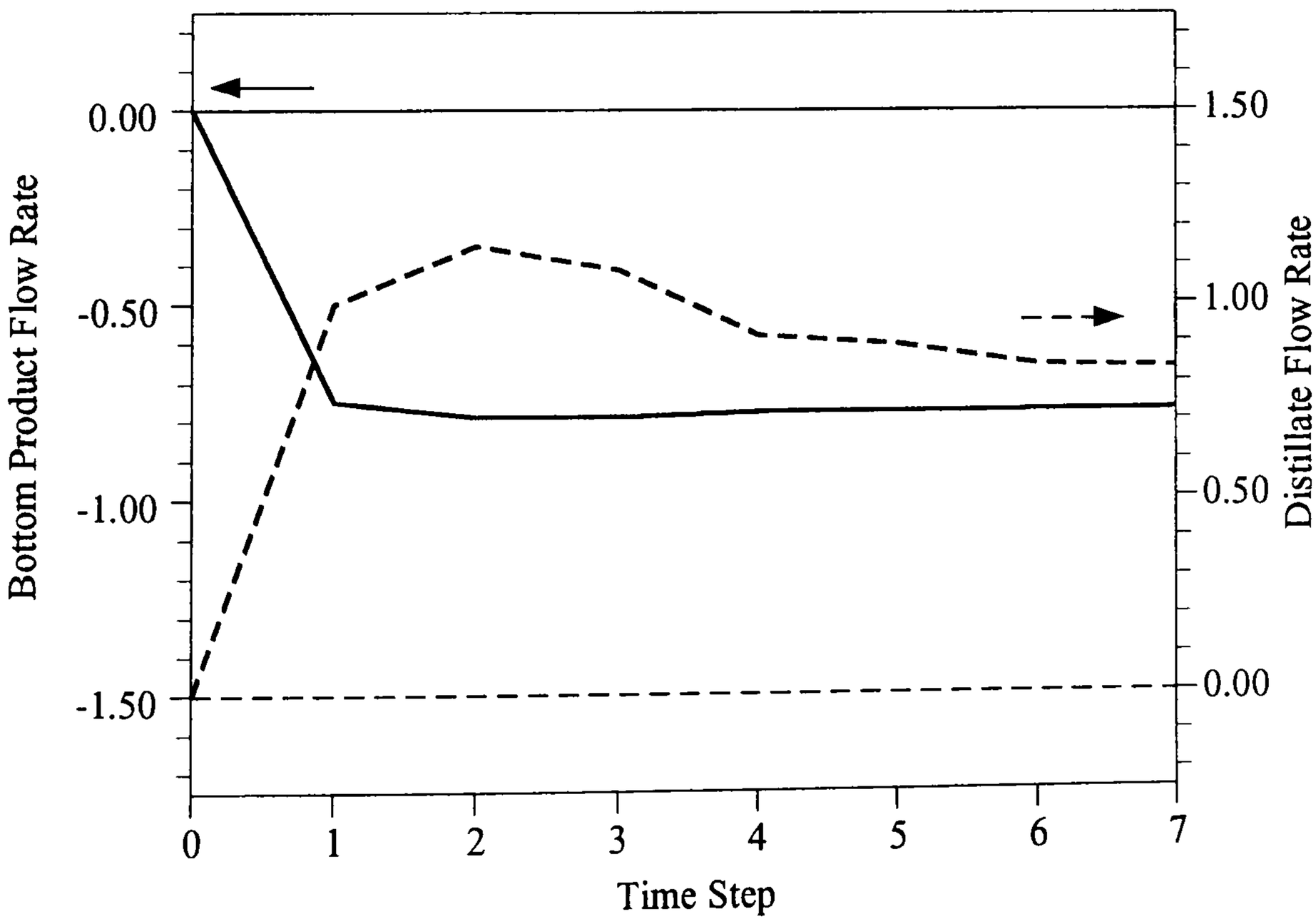
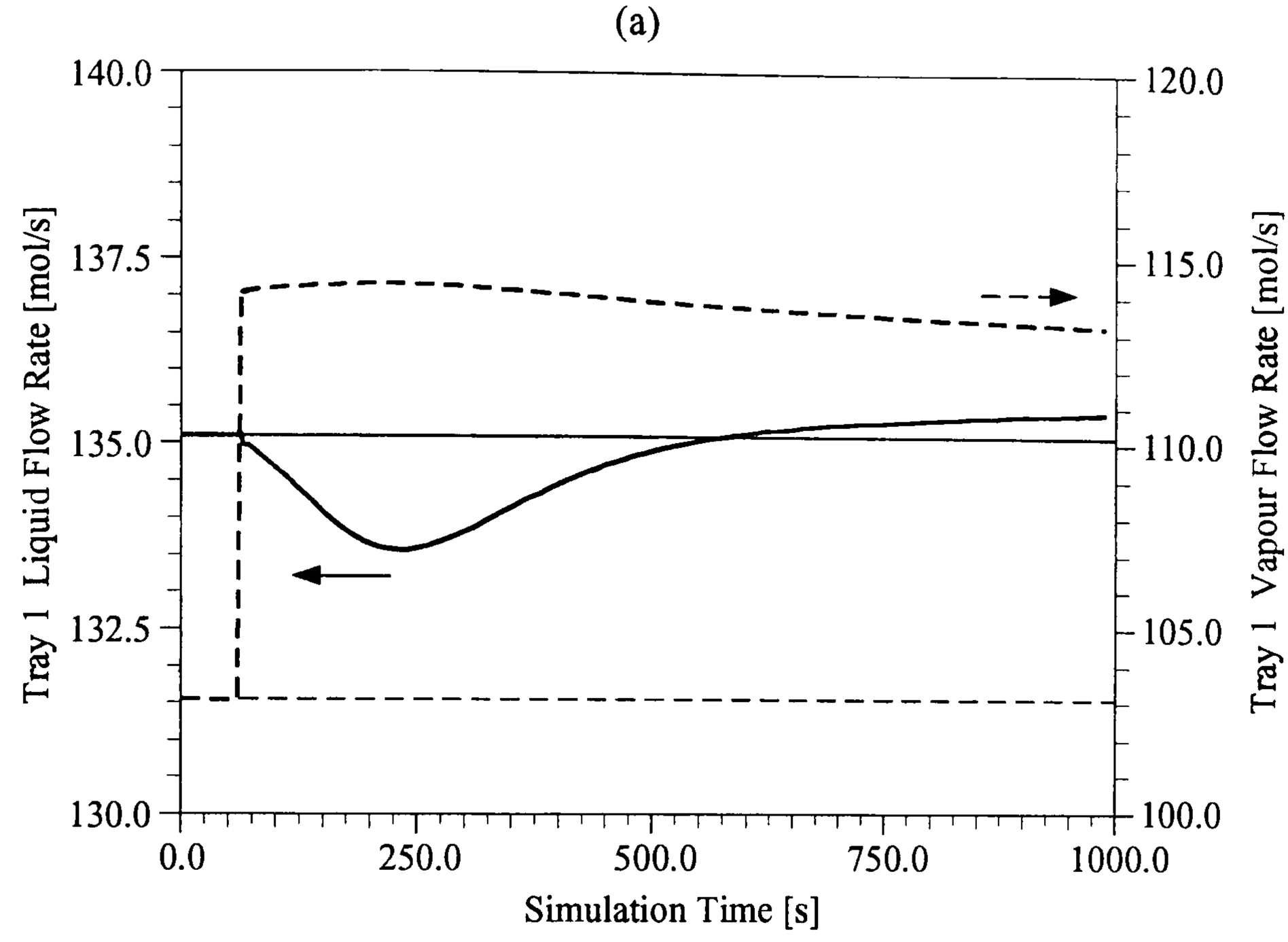


Figure 4.33 Distillation column - distillate and bottom product flow rate profiles for a positive step disturbance in reboiler duty: (a) numerical simulation; (b) qualitative simulation.



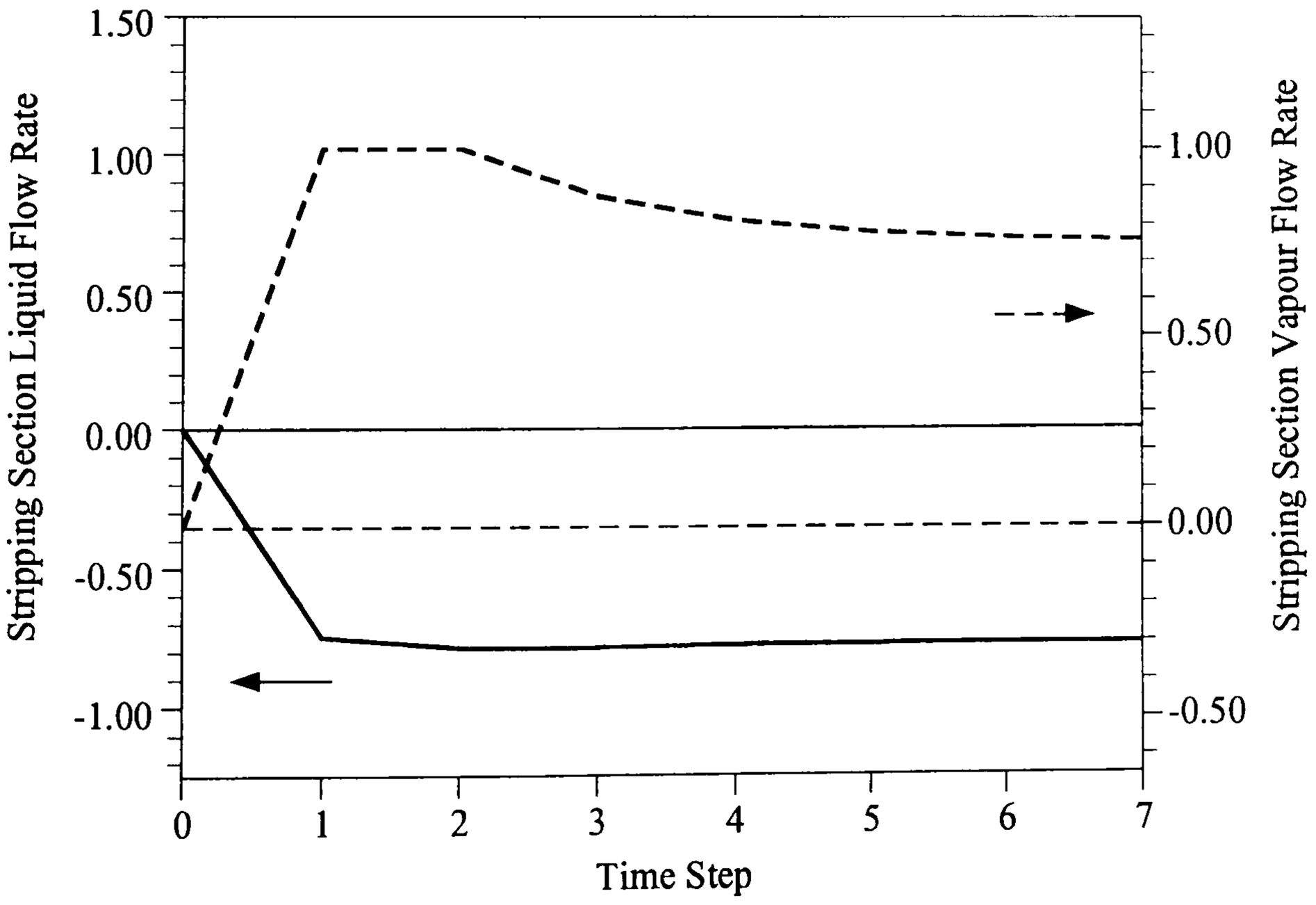


Figure 4.34 Distillation column - liquid and vapour flow rate profiles for a positive step disturbance in reboiler duty: (a) numerical simulation (tray 1); (b) qualitative simulation (stripping section).

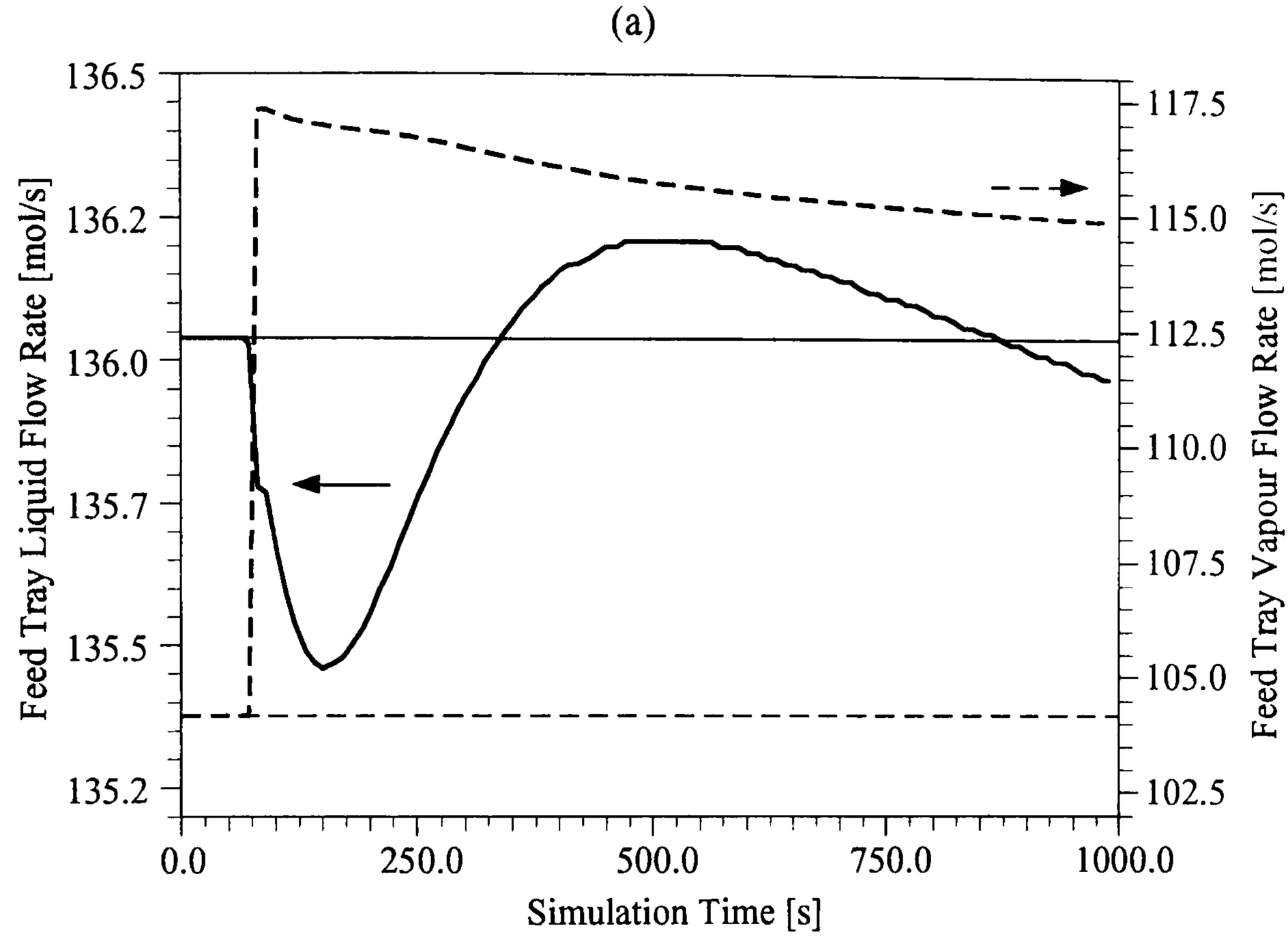


Figure 4.35 Distillation column - feed tray liquid and vapour flow rate profiles for a positive step disturbance in reboiler duty: (a) numerical simulation; (b) qualitative simulation.

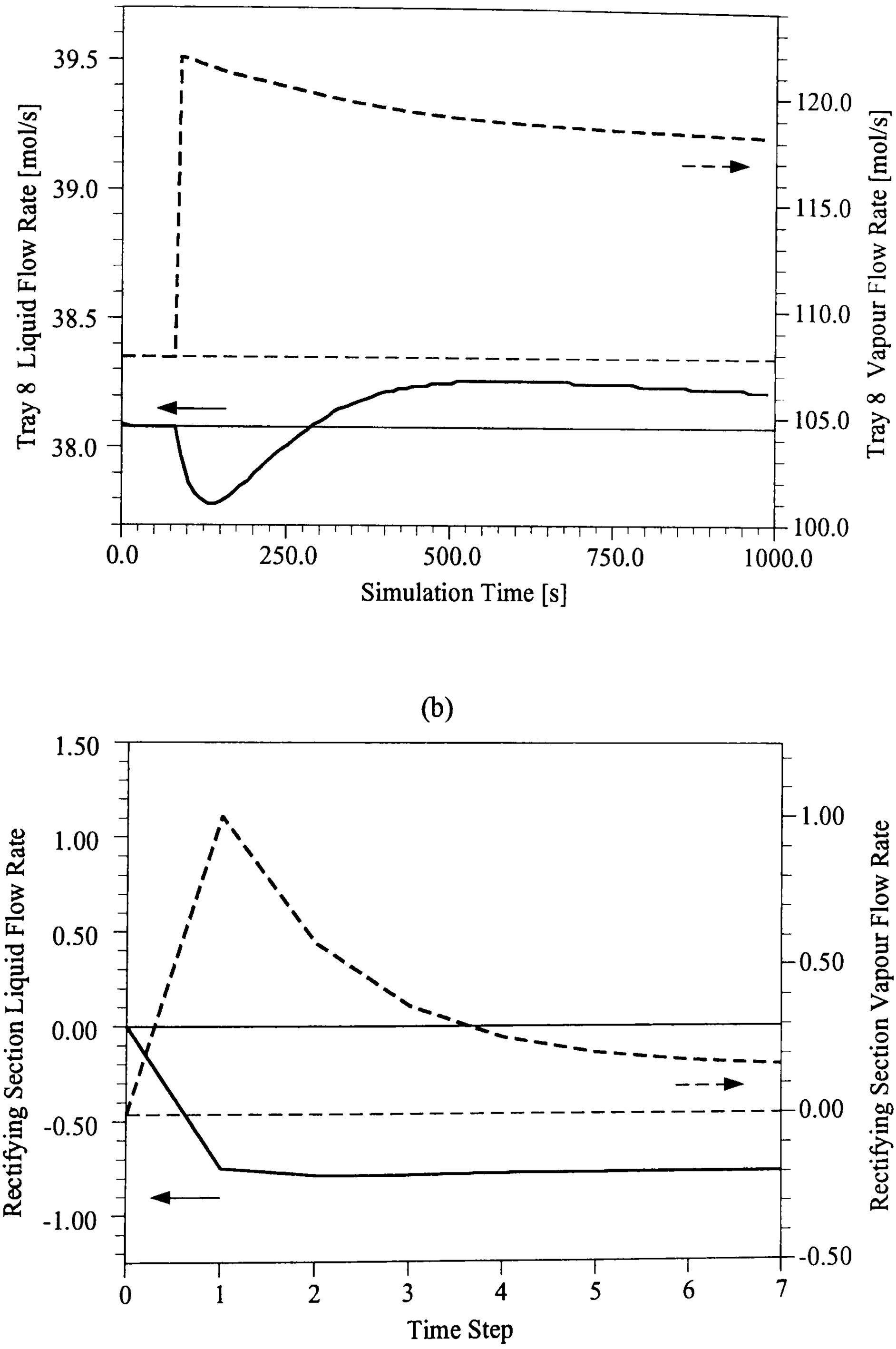


Figure 4.36 Distillation column - liquid and vapour flow rate profiles for a positive step disturbance in reboiler duty: (a) numerical simulation (tray 8); (b) qualitative simulation (rectifying section).

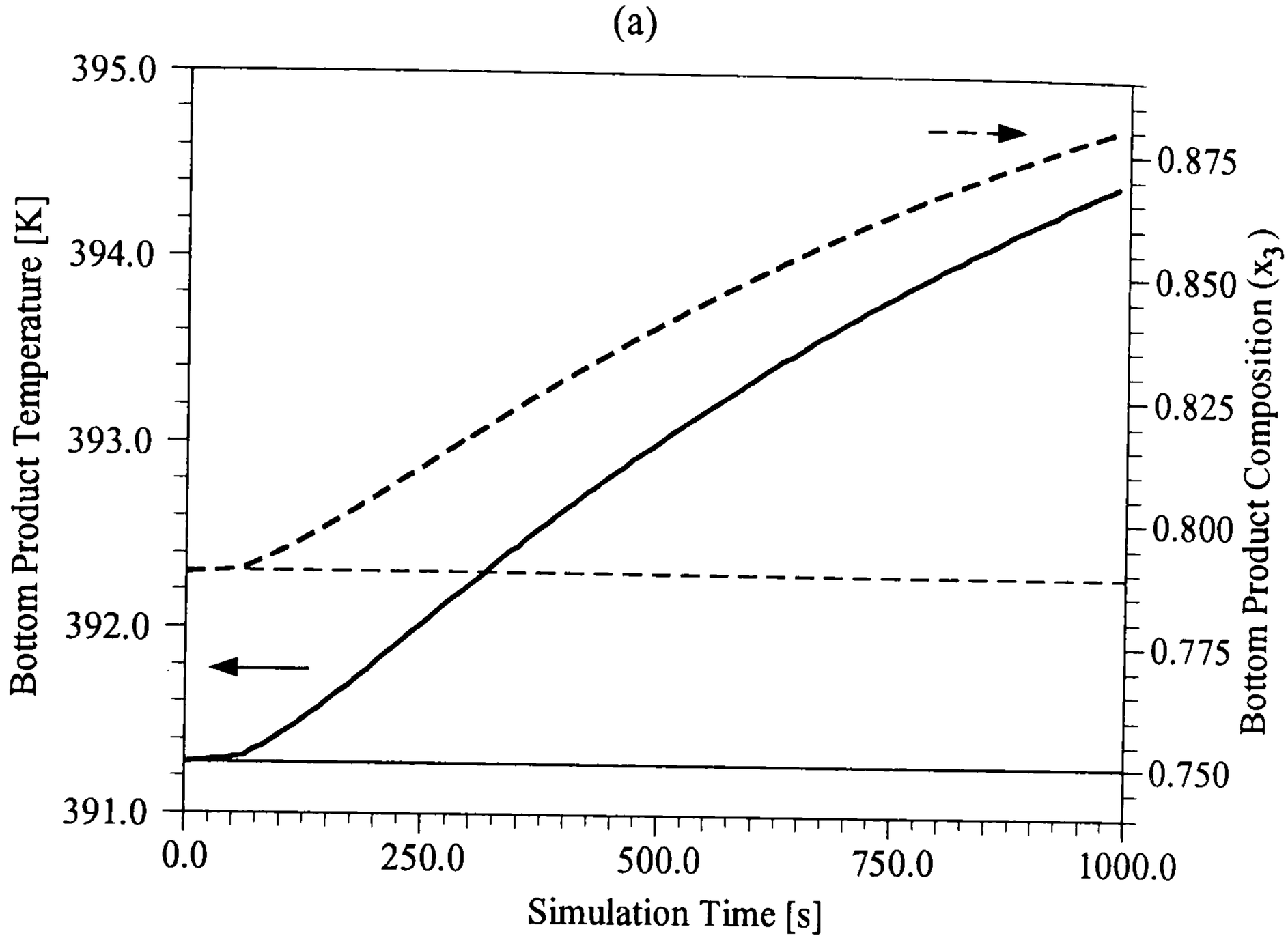


Figure 4.37 Distillation column - bottom product temperature and composition profiles for a positive step disturbance in reboiler duty: (a) numerical simulation; (b) qualitative simulation.

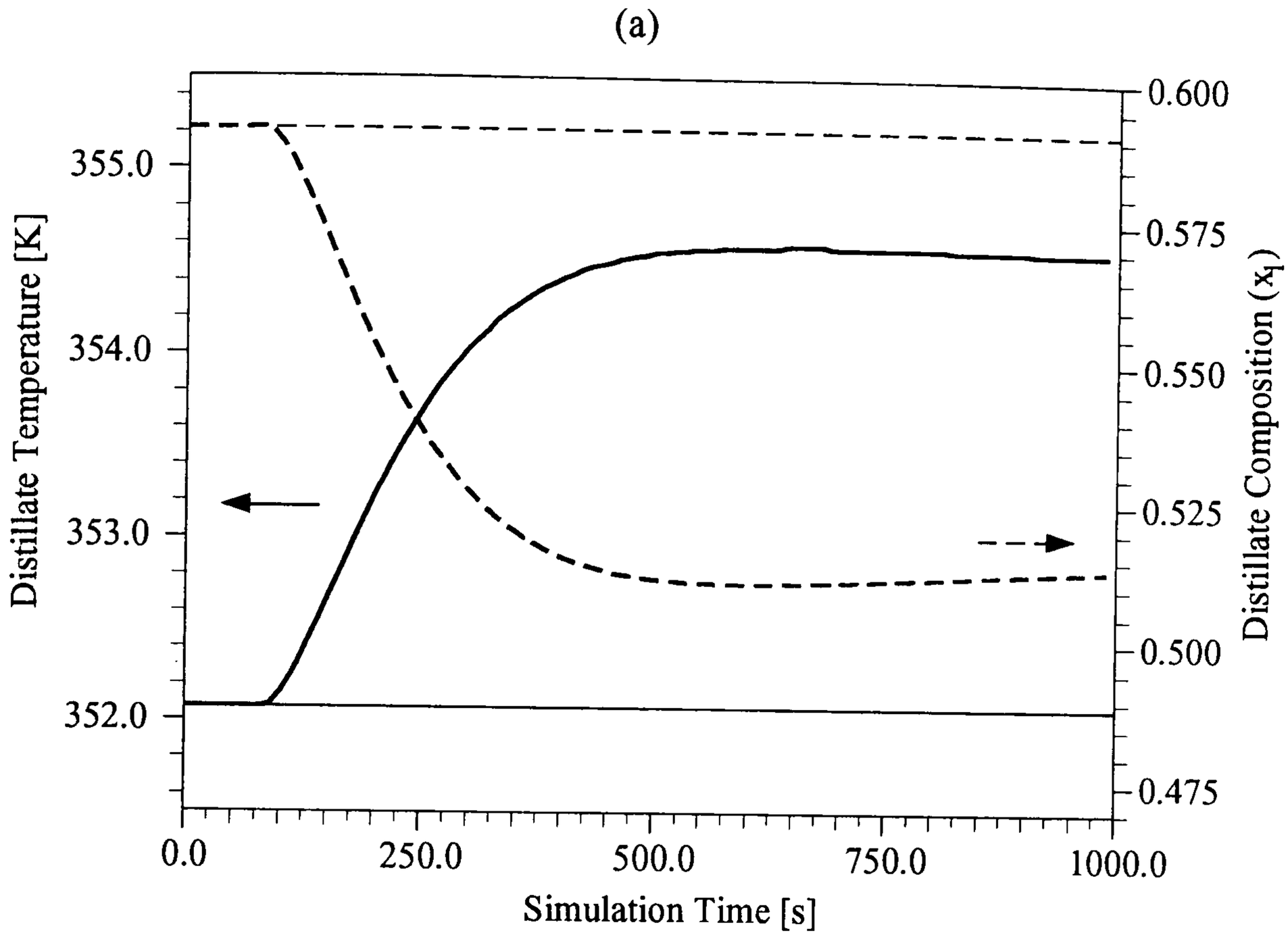


Figure 4.38 Distillation column - distillate temperature and composition profiles for a positive step disturbance in reboiler duty: (a) numerical simulation; (b) qualitative simulation.

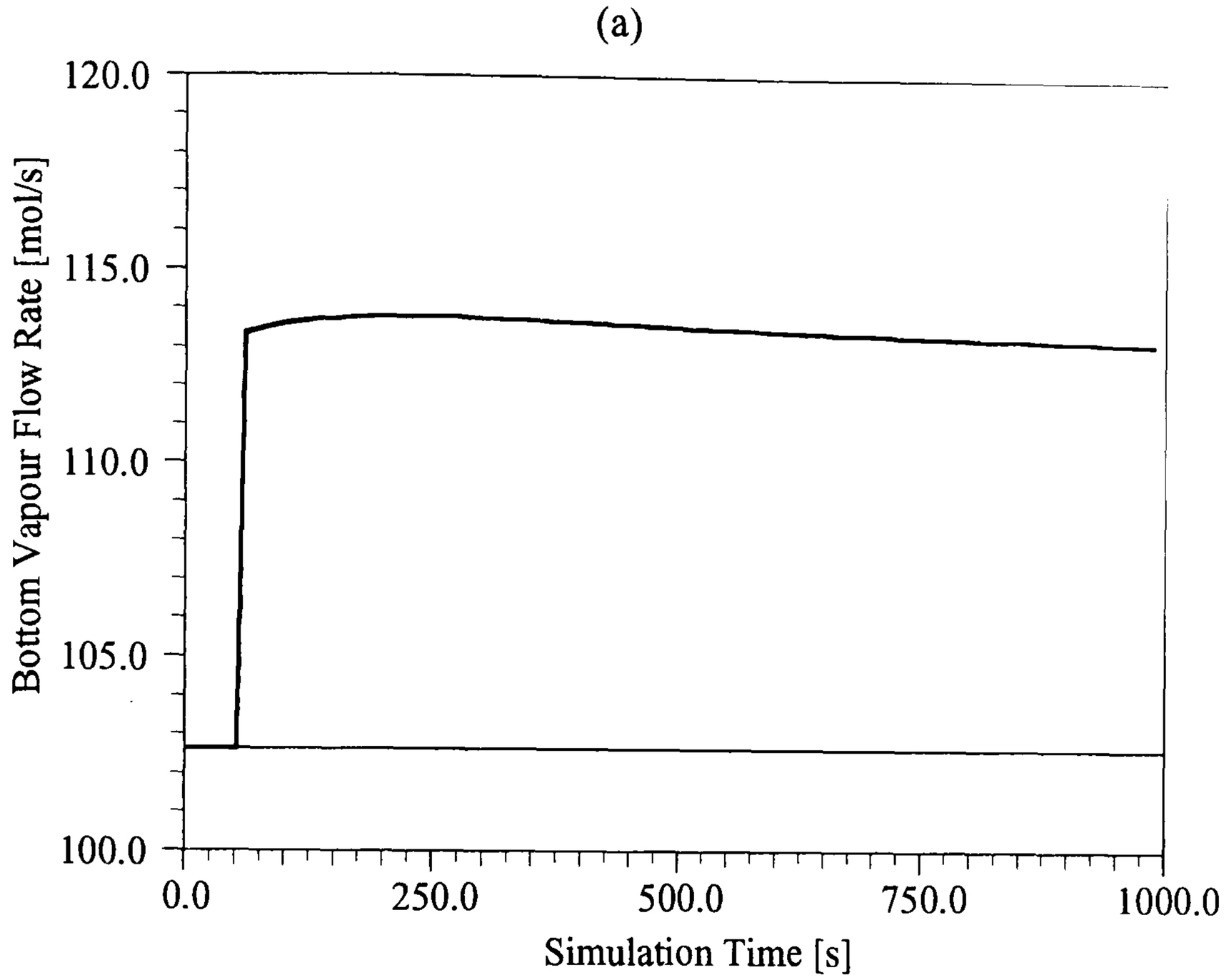


Figure 4.39 Distillation column - bottom vapour flow rate profile for a positive step disturbance in reboiler duty: (a) numerical simulation; (b) qualitative simulation.

4.7 Main Characteristics of the WDG Approach with Respect to the Structural Elements

From the previous sections it can be concluded that the WDG approach is able to represent complex features of dynamic behaviour and describe distributed parameter systems without generating spurious solutions. This has been made possible by the new features of the weighted digraph approach, as described below:

- **Representation of process dynamics** - The explicit consideration of *differential nodes* in the digraph structure, associated with *temporal edges*, provide the necessary information about system dynamics, while constraining the solution space to avoid inappropriate solutions. The enhanced qualitative state descriptor, which assigns to variables and *weights* real values, gives flexibility to the procedure and allows the description of complex patterns of behaviour;

- **Avoiding ambiguous solutions** - The use of *temporal edges* and functional weighting enables simplification of the digraph structure by avoiding conflicts and breaking loops and cycles, which would tend to give rise to ambiguous solutions and spurious behaviour. The breaking of loops and cycles also eliminates problems of convergence during the inferring with respect to process behaviour;

- **Description of functions with different shapes** - Functional weighting enhances knowledge representation and formalises quantitative knowledge about the relative strength of the influences. These are associated with *temporal* and *multiple edges*, to allow the description of the shape of monotonic and non-monotonic functions;

- **Ability to represent distributed parameter systems** - The multi-layer approach associated with the use of *weights* allows the description of

changes in space, with *temporal edges* and *differential nodes* accounting for changes in time. Together, these allow the description of distributed parameter systems.

4.8 Concluding Remarks

The generation of operating procedures for a chemical plant requires models which can adequately represent the underlying process behaviour of combinations of plant units.

In this chapter, WDG models for heat-exchangers, CSTRs and distillation columns have been developed and their performance tested for start-up or disturbances in the inlet conditions. All models are capable of adequately describing dynamic patterns of process behaviour without generating spurious solutions. The complex dynamic behaviour of a non-azeotropic ternary distillation column is well represented in terms of flow rates, temperatures and compositions, showing that the WDG methodology is also able to deal with distributed parameter systems.

Although the procedure represents oscillations and turning points quite well, it does not have enough sensitivity to distinguish between very small variations around a given state. This means that there is a dead zone for small qualitative states.

The effectiveness in representing complex processes, such as distillation, unfortunately results in a more complex model structure which makes difficult the prediction of the flow of information and so provide a simple explanation of the system responses. But of course it is an intrinsic feature of this problem, which can be assisted by computer based reasoning.

The ability to describe process dynamics, including *inverse responses* and oscillations, shows that the WDG methodology holds promise of supporting tasks

based on analyses of process dynamics, such as synthesis of control and operating strategies. It is particularly appropriate for generating operating procedures, since it may be used as a means of propagating goals and identifying situations which may result in violation of operating constraints.

WDG models embed useful knowledge about causal relationships between system variables and provide a platform for systematically representing intuitive knowledge from operators and engineers.

Chapter 5

Generation of Process Plant Operating Procedures

5.1 Introduction

It is now recognised that the analysis of the life-cycle performance of a process plant must be considered in the preliminary design stage in order to ensure high quality and efficient designs. In particular, the analysis of operating procedures at early stages in process design can lead to a less costly and less failure prone plant, since a process flowsheet consistent with the plant operations can be developed from the beginning, so avoiding later alterations which call for intermediate storage and start-up auxiliary equipment. It is also the time to ensure that the operating conditions are safe. O'Shima (1983) points out that inefficient operating procedures account for a large portion of the total causes of accidents which happen in chemical plants. He reports that in Japan, in 1981, 112 out of 482 operational failures that occurred in petrochemical complexes were due to errors in operating procedures.

Operating procedures establish a sequence of actions to be performed by plant operators or computers, in order to bring the system from an initial to a desired goal and maintain that state subject to constraints imposed by safety, environmental

regulations, process chemistry, equipment material and construction details. For plants of medium and high levels of complexity, there may be a large number of possible alternative paths leading from the initial to the goal state and that, depending on the nature of the constraints, make the synthesis of operating procedures a task for experts.

Generating operating procedures requires a deep understanding of the interactions between process variables and the underlying mechanisms that determine process behaviour. It is based on a decision-making process which involves knowing how variables must be manipulated in order to achieve desired process conditions, while avoiding hazardous situations. This process tends to be based on extensive experience, intuitive knowledge and inductive reasoning. Because of this, qualitative reasoning techniques are appealing support tools. Attempts have been made (Fusillo and Powers, 1987, 1988; Lakshmanan and Stephanopoulos, 1988a, b, 1990; Hangos *et al.*, 1991) in developing methodologies for the systematic synthesis of operating procedures based on artificial intelligence methods. However, these approaches present limitations related to the use of steady-state or very complex dynamic models, which are difficult to simulate and prone to generate ambiguous solutions, compromising the effectiveness of the methodology.

This chapter presents a critical review of existing approaches for synthesis of operating procedures and proposes a strategy for generating start-up procedures based on weighted digraph (WDG) models. These models are able to efficiently represent process dynamics and describe distributed parameter systems without generating ambiguous solutions, and therefore are adequate in dealing with critical operations, such as start-up. The proposed strategy focuses on the sequencing of operations and dynamic qualitative simulation of process behaviour to allow the identification of potential operational problems that can prevent the start-up or lead to hazardous conditions. The aim is to demonstrate the suitability of the WDG

models to support the generation of operating procedures at early stages of process design, as one component of a process engineering design environment.

5.2 Review of Prior Work on Generation of Operating Procedures

Despite the importance and difficulties involved in the systematic synthesis of operating procedures, little formal work has been reported in the literature. As pointed out by Tomita *et al.* (1989), one of the main reasons for that is the fact that this task is heavily based on the heuristic knowledge of operators and engineers, which is not easily programmable. As a consequence, current industrial practice relies on recommended procedures compiled in operations manuals and the experience of operating personnel (Lakshmanan and Stephanopoulos, 1988a).

Despite the pioneering paper by Rivas and Rudd (1974) addressing the problem of automatic generation of operating procedures, no significant progress was made until the end of the 1980s, mainly due to the restrictive computing environments available. However, the huge development of computer technology during the last decade and the growing interest in the area of artificial intelligence, associated with the trend of increasingly automated plants, have stimulated research interest in the automatic synthesis of operating procedures.

In their pioneering work, Rivas and Rudd (1974) model the chemical processing system as a network of valves and connectors. The synthesis procedure involves the sequencing of valve openings and closings to reach the operational objectives while avoiding hazards. Synthesis is carried out by formulating goals (input by the user) which identify critical operations and the order in which they are to be performed. Goals are expressed as Boolean statements in symbolic logic and are used by a computer program to generate sequences of valve operations, which are tested by

numerical simulation to ensure that the safety criteria are satisfied. A batch catalyst regeneration system is used to illustrate the procedure. The main drawback of this work is the need to specify the tree of goals which have to be satisfied, including the order in which they are to be achieved. The goals include not only the overall operating objective, e.g. "start-up of the regeneration system", but also more detailed actions, such as "stop air flow", "align nitrogen flow", etc. For complex processes the foresight and specification of all operating goals which might have to be faced is not a feasible task.

O'Shima (1978, 1983) presents a method for automatically determining a sequence of valve operations to establish mass flow between two locations in a chemical plant. Using the valves and connectors approach by Rivas and Rudd (1974), the author develops an algorithm for path finding and for discovering orderings of valve operations based on the depth-first search method (Tarjan, 1972). The main disadvantage of this approach is the use of an intricate system of flow states ("flow", "block", "trap" and "branch") applied to each valve of the system. Since these devices are present in large numbers in chemical plants, this poses a problem in understanding how the procedure is generated. Another problem is related to the inability to deal with loops that frequently occur in integrated topologies.

Kinoshita *et al.* (1982) manually divide a plant into sub-systems consisting of small groups of connected units ("key" equipment and their peripherals). The procedure is based on the generation of a sequence of state transitions for leading the process from initial to final states for each subsystem, without considering the connectivity between sub-systems. Operations are then timed to ensure consistency between sub-units. The authors recognise the role of constraints in limiting the search space to carry out the synthesis procedure. The input to the program requires the identification of all individual operations for each sub-system, i.e. a tree of all possible sequences of operations, maximum allowable time for each operation and

information about the constraints on the states of adjacent sub-systems. Although this approach yields efficient schedules, the overhead involved in setting up the input to the program is too great for practical use.

Ivanov and co-workers (1980) have developed algorithms for generating optimal start-up sequences starting from a "transition network" representation of the chemical plant (input by the user), composed of nodes and arcs. The nodes represent possible transitions and the arcs (edges) represent possible ways of carrying the system between states. Each arc receives a weighting factor based on some pre-determined optimality criterion. The input of the "transition network" and the *a priori* assignment of weighting factors limit the practical applicability of the method.

The work by Fusillo and Powers (1987, 1988) represents the first attempt to define a formal theory for planning process operations. They were also the first to employ qualitative models, in terms of signed digraphs, to propagate effects through process topology and identify unit manipulations. Qualitative descriptions are far more expressive than the primitive Boolean formulations used by previous researchers. The method allows for the use of global and local design constraints, and introduces the concept of "stationary states", in which the system does not change over time and steady-state models apply. The method is very effective in scheduling valve operations and dealing with operating constraints. However, it is limited by the use of steady-state qualitative models and the need for numerical simulation to verify the effects of process manipulations and determine the feasibility of the operating procedure. Another drawback is related to the fact that the qualitative models do not contemplate distributed parameter systems, such as distillation columns, which have to be treated as "black-boxes", so changes inside the systems are ignored.

Lakshmanan and Stephanopoulos (1988a, b, 1990) present a framework for the automatic synthesis of operating procedures based on a general object-oriented modelling structure and a non-linear planning approach. Partial plans (non-linear)

admit information to be left unspecified or allow the existence of a set of operators for executing a single task. This results in a more efficient planning procedure, when compared with other approaches. The main drawback of the method is the use of steady-state models to describe the system behaviour based on the concept of "stationary states" developed by Fusillo and Powers (1987).

Tomita *et al.* (1989) have developed a computer-based system for generating sequences of operations for starting up a chemical plant. The method is based on logic statements that contain information about paths, valves (closed/open) and arcs (edges) states (live/dead), process conditions in each arc (phase, temperature, pressure and compositions) and operating constraints. It is very efficient as a planning tool, but is limited in terms of dealing with unexpected situations, since the knowledge base is built by using heuristic knowledge from human operators. There is no attempt to use qualitative models to describe the internal behaviour of process units.

Csáki *et al.* (1991) and Hangos *et al.* (1991) extend the qualitative simulation technique (QSIM), developed by Kuipers (1986), to include actions from operators and automatically generate operating advice. These works were the first attempts to use dynamic information in terms of qualitative models to constrain the search space, making more efficient the search method for generating start-up procedures. The methodology is illustrated by the start-up of a distillation column with energy feedback. The main limitation of the approach is related to the complexity of the qualitative simulation method (QSIM), which requires very detailed non-easy-to-handle models. The authors do not attempt to describe the physical internal dynamic behaviour of the distillation column.

Rotstein *et al.* (1992) and Crooks *et al.* (1994) present studies on synthesis of operating procedures for batch plants. The method by Rotstein *et al.* (1992) is based on the qualitative process theory, developed by Forbus (1984), which contemplates

only steady-state models. Crooks *et al.* (1994) generate control sequence specifications for multi-purpose batch and semi-continuous processes.

Naka and McGreavy (1994) describe a method for establishing the relation between the topological structure of a chemical plant and operating actions in terms of valve manipulations. Particular attention is directed to presenting the information visually to allow a better understanding of the procedure. A computer integrated engineering environment is provided so that the start-up sequences can be generated based on process flow diagrams (PFD) and preliminary piping and instrumentation diagrams (P&ID). The method requires detailed dynamic numerical simulation to determine reliable values of hold-ups and rates of mass and heat transfer, in order to solve local conflicts in terms of valve manipulation strategies.

From the above review it is clear that prior work in the area has mainly concentrated on developing automatic planning methodologies for generating operating procedures. Apart from the work by Csáki *et al.* (1991) and Hangos *et al.* (1991) based on the complex QSIM method, there is no attempt to employ dynamic information to constrain the solution space and to explore the use of qualitative dynamic simulation to verify the feasibility of the procedures generated by the planning methodologies. So there is considerable scope for work in the area.

5.3 Generation of Start-up Procedures Based on Weighted Digraph Models

As pointed out by Stephanopoulos and Han (1994), the generation of operating procedures involves sequencing of operations while contemplating operational constraints, such as:

- Allowed temporal sequences of primitive operations, e.g. valves or pumps on or off;

- Disallowed mixtures of chemicals for safety and other reasons;
- Bounds on the values of operating variables, e.g. maximum allowed reactor temperature;
- Desired sequences of operations, e.g. start recovery section before reaction section; and
- Process behaviour.

The movement between the initial and final states is carried out through a series of intermediate states which must be consistent not only with the above operational constraints, but also with physical and chemical constraints, such as conservation of mass, energy and momentum, in order to be considered physically feasible. For underconstrained problems more than one feasible sequence may exist and performance criteria must be used to select the most efficient one.

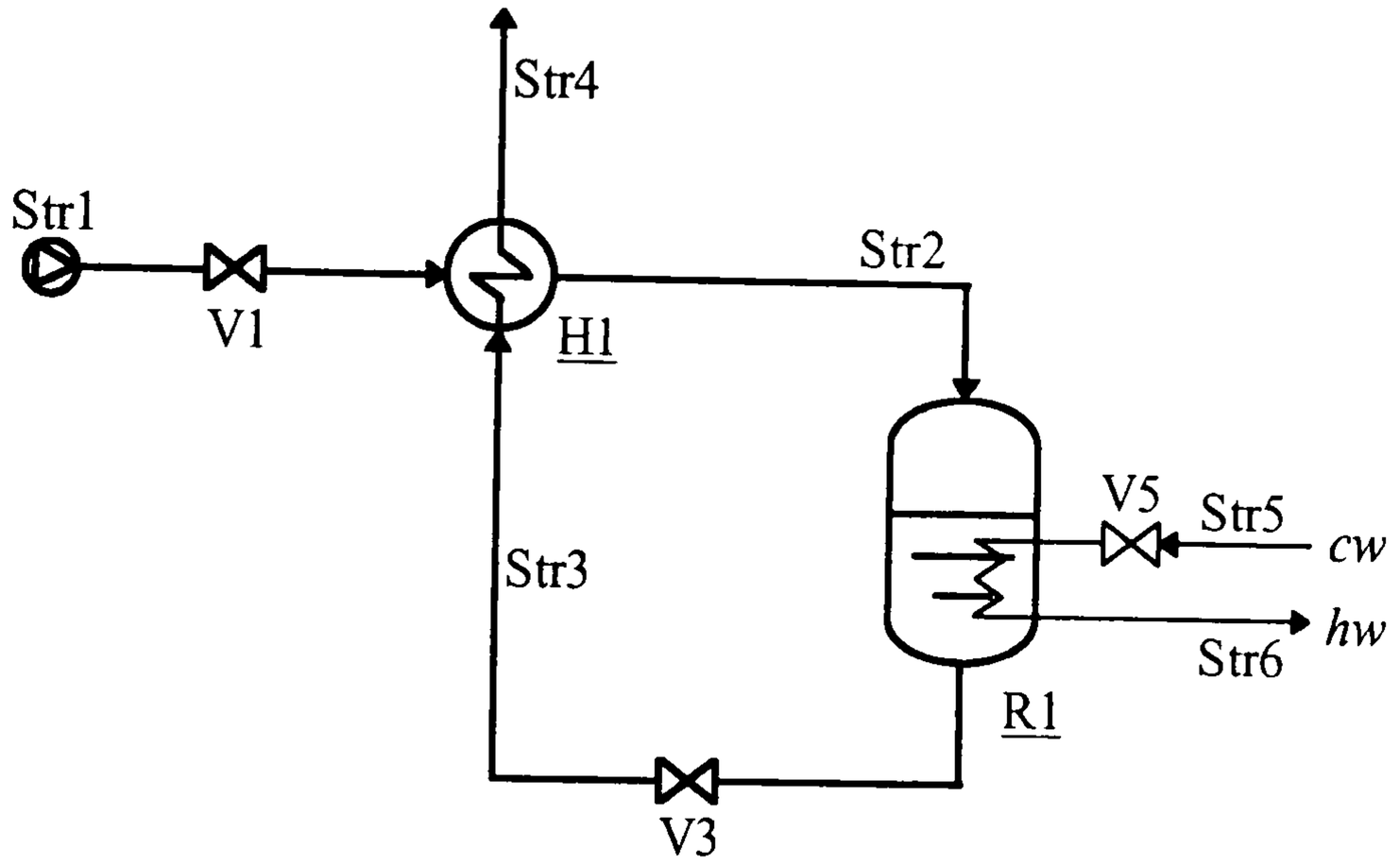
The strategy for generating start-up procedures based on weighted digraph (WDG) models focuses on sequencing valve manipulations and equipment operations. The sequencing procedure is subject to physical and chemical constraints, described in terms of weighted digraph models, and explicit operational constraints in terms of qualitative bounds on the operating conditions.

5.3.1 Process Representation

The process flow diagram (PFD) of a chemical plant is translated into a directed graph (process flowsheet graph - PFG) as illustrated in Fig. 5.1. Nodes represent process units (equipment), except valves and pumps, while edges represent pipelines, i.e. mass and energy flows between process units. Nodes and edges are sequentially numbered. Pumps and valves are considered as part of the pipeline system. In this

preliminary study, pumps are omitted from the PFD, since the effects of system pressure variations are not being considered.

(a)



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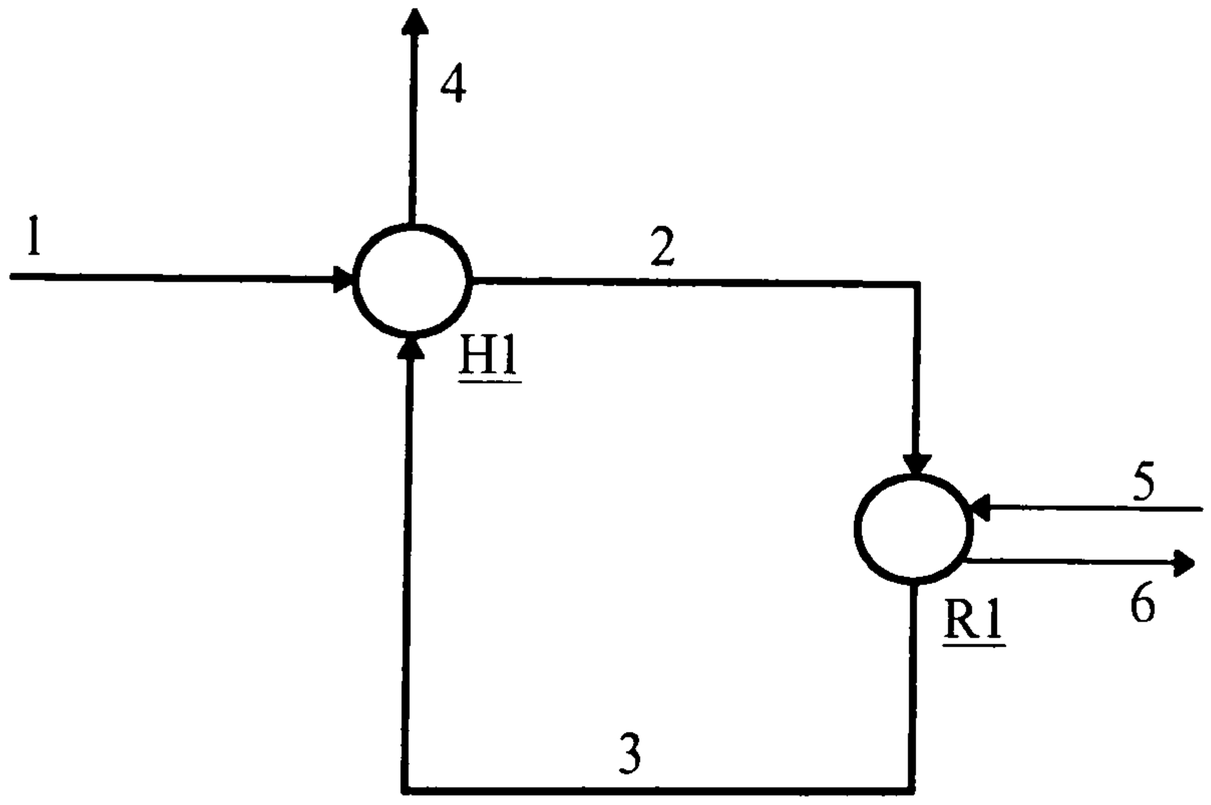


Figure 5.1 Schematic process representation: (a) process flow diagram and (b) process flowsheet graph.

The dynamic behaviour of equipment (nodes) are described by previously developed weighted digraph (WDG) models. These models are introduced in the algorithm as modules that are called every time the simulation needs to evaluate the state or dynamic behaviour of a node. There is a module for each type of

equipment (node), e.g. heat-exchanger, CSTR and distillation column. Nodes are identified by the following attributes:

- **Type** - Describes the type of the equipment represented by the node, i.e. heater, CSTR, etc.;
- **State** - The state of the node can be *ON* or *OFF* to indicate if the equipment is operating or not.

Edges are characterised by a group of attributes as follows:

- **Head** - number of the node to which the edge points at, i.e. number of the equipment to which the pipeline heads for;
- **Tail** - number of the node from which the edge originates;
- **Type** - The edge can be related to the *FEED*, product (*PROD*) or utility (*UTIL*), or can be an intermediate (*INTM*) edge;
- **Valve** - Identifies if a valve or group of valves is present or not in the pipeline, {0} or {1}, respectively;
- **Valve condition** - The state of the valve (or group of valves) can be: *OPEN* or *CLOSED*;
- **Condition** - If all valves present in the edge are *OPEN* and any *tail node* is *ON* the edge is *ACTIVE*, otherwise it is *INACTIVE*. Feed and utilities streams are always activated by valves;
- **State** - The state of the edge is characterised by a vector with 5 components: *FLOW*, *TEMP*, *PRES*, *CONA*, *CONB* and *CONC*, which describe the qualitative values of the variables: flow, temperature, pressure and compositions of components A, B and C (ternary system).

respectively. Normal operating conditions are represented by the qualitative value state: {0}. Positive and negative qualitative states represent numerical values above and below normal operating conditions, respectively. States can be *initial*, *final* or *intermediate*;

- **Constraints** - Identifies the existence, or not, of operational constraints associated to the edge, {1} or {0}, respectively;

- **Qualitative constraint value** - Represents the maximum (max) or minimum (min) allowed qualitative value for the constrained system variable, in terms of flow rate, temperature, pressure or compositions. Qualitative bounds are arbitrary values that intend to translate the existence of quantitative bounds on the operating conditions of the system. For example, if a reactor has a maximum limit on the operating temperature, this can be translated as the qualitative value: $T_{max} = +1.5$. The exact value of the qualitative limit does not matter, since what is important is to show that there is a maximum (or minimum) bound for the system variable;

5.3.2 Algorithm for the Generation of Start-up Procedures

The algorithm for the generation of start-up procedures, shown in Fig. 5.2, is made up of two modules. The first is concerned with operations sequencing, while the second involves the dynamic simulation of the start-up of the plant based on the proposed sequence. The second module is directed to the verification of the feasibility of the procedure and identification of potential bottlenecks related to the process topology.

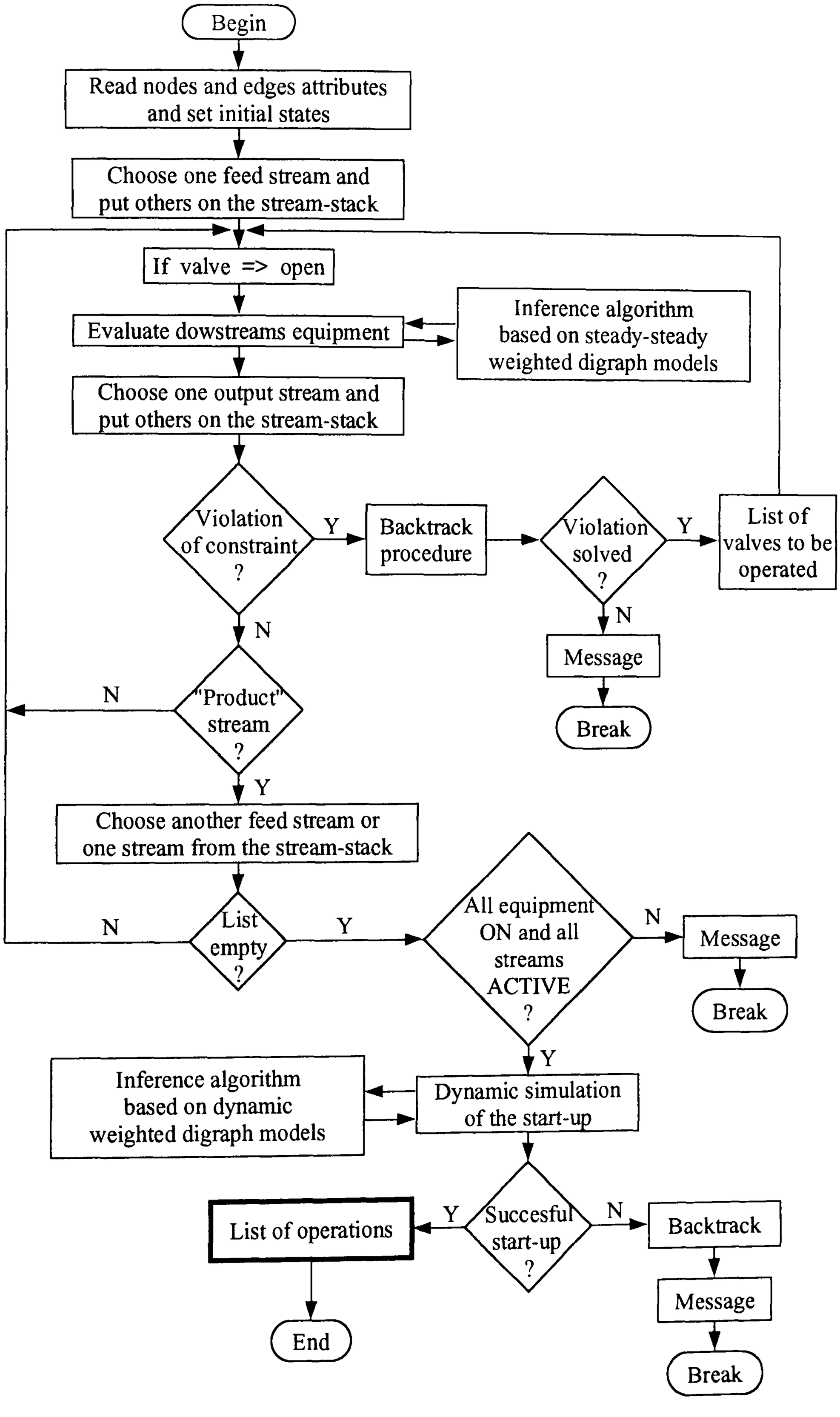


Figure 5.2 Algorithm for the generation of start-up procedures.

Sequencing Operations

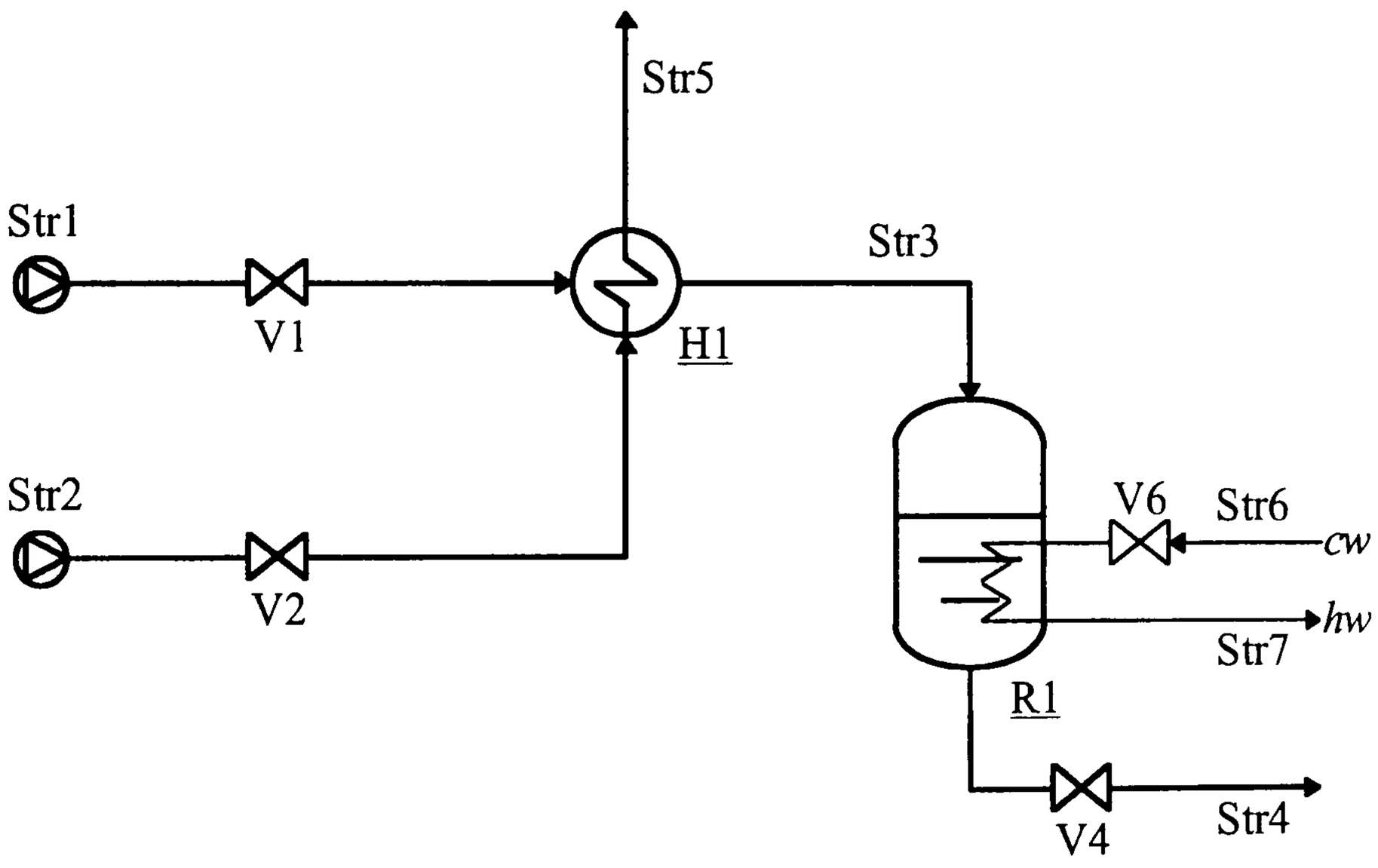
Attributes that describe nodes and edges, including initial states of valves and goal states (normal operating conditions), are input to the algorithm. Initial states for all variables are set as zero, negative or positive values, whether they are at, below or above normal operating conditions, respectively. The initial state of input edges (feed streams) is compared with their goal state. The algorithm chooses one of the input edges whose state is different from the goal state, and put it on the *operations list*, while the remaining ones are put on the stack for later assessment. The feed valve of the chosen stream is *opened* and its flow state is set equal to zero (operating condition). The valve manipulation introduces a step disturbance in the system that is calculated as the difference between the new flow state (zero) and the initial state of the feed stream. The disturbance is propagated forwards through the process flowsheet graph (PFG) by applying a search procedure based on the depth-first search algorithm (Tarjan, 1972). The determination of the effects of valve manipulations through process variables is made by using the steady-state version of the WDG models, where *temporal edges* are deactivated and *differential nodes* are treated as *algebraic nodes*, and the inference of behaviour algorithm, described in Chapter 3. By knowing the states of the edges that enter in each node, the WDG model is simulated and the states of the output edges of the node are determined. If all input edges of the node are *active* (flow state ≥ 0), the equipment is set *ON* and added to the *operations list*. If the equipment is set *ON*, all the output edges are *activated* and existing valves are *opened* and also added to the *operations list*. Each time a node (equipment) is evaluated, a check for violation of constraints in the output edges is performed. If a constraint is violated the algorithm *backtracks*.

Backtracking - Backtracking is performed in the same way as the forward propagation of effects, but considers output edges of the nodes as input, and input edges as outputs. In backtracking mode the disturbance propagated is the qualitative

difference between the value (minimum or maximum) of the constraint and the instantaneous value of the variable. WDG models are used to guide the search for variables that can be manipulated in order to avoid the violation. For example, if the cold stream of a heat-exchanger has a temperature of -1.5, that violates the qualitative constraint: T_{min} = -1, the difference ΔT=+0.5 is propagated backwards in the PFG, using the search procedure and WDG models, until either an *inactive* stream is found which when activated is able to provide at least the +0.5 qualitative value needed to avoid the violation of the temperature constraint, or a cycle or dead-end is reached. By analysing the WDG model of the heat-exchanger shown in Fig. 4.2b backwards, it can be seen that to increase the outlet cold stream temperature in +0.5, one of the following disturbances must be introduced: (a) increase in the inlet cold stream temperature; (b) increase in the inlet hot fluid temperature; (c) increase in the inlet hot fluid flow rate; or (d) decrease in the inlet cold fluid flow rate. Variables that are already on the normal operating condition (design value) are not taken into account. Those outside the design conditions, and which are able to potentially solve the violation, are followed in the search for the "root cause" of the problem. When the cause is found action is taken, e.g. a valve is *opened* or an equipment started-up. If the violation cannot be overcome due to cycles or other reasons, e.g. dead-ends, a message containing a list of all variables that need to be altered in order to potentially solve the violation is generated. Besides, a possible "root cause" of the problem is suggested based on the value of the disturbance needed to lift the violation, e.g. "a source of heat is needed" would be the message delivered by the algorithm if a ΔT = +0.5 was needed. Chapter 6 illustrates the problem of constraints violations and cycles (integrated processes).

After the backtracking is successfully finished, a new list of valves to be operated is generated and the forward search works through the list until all valves are opened or constraints violated. Afterwards, control is transferred to the list of valves previously put in the stack. A valve is chosen and the procedure is repeated until all

streams are *active* and equipment operating. Figures 5.3 and 5.4 illustrate the procedure applied to a PFD composed of a heat-exchanger and a CSTR, and which is based on the WDG models described in Chapter 4.



Initial conditions: V1, V2, V4 - closed ⇒ Fstr1, Fstr2, Fstr4 = -1 (below design conditions)

V6 - open ⇒ Fstr6 = 0

Tstr1, Tstr2, Tstr3, Tstr4, Tstr5 = 0 (at design conditions).

Constraint: Tstr3 ≥ -0.3 (in order to align Str3 to the reactor).

Figure 5.3 Schematic representation of a process flow diagram, with initial conditions and constraints, for illustrating the procedure for sequencing operations.

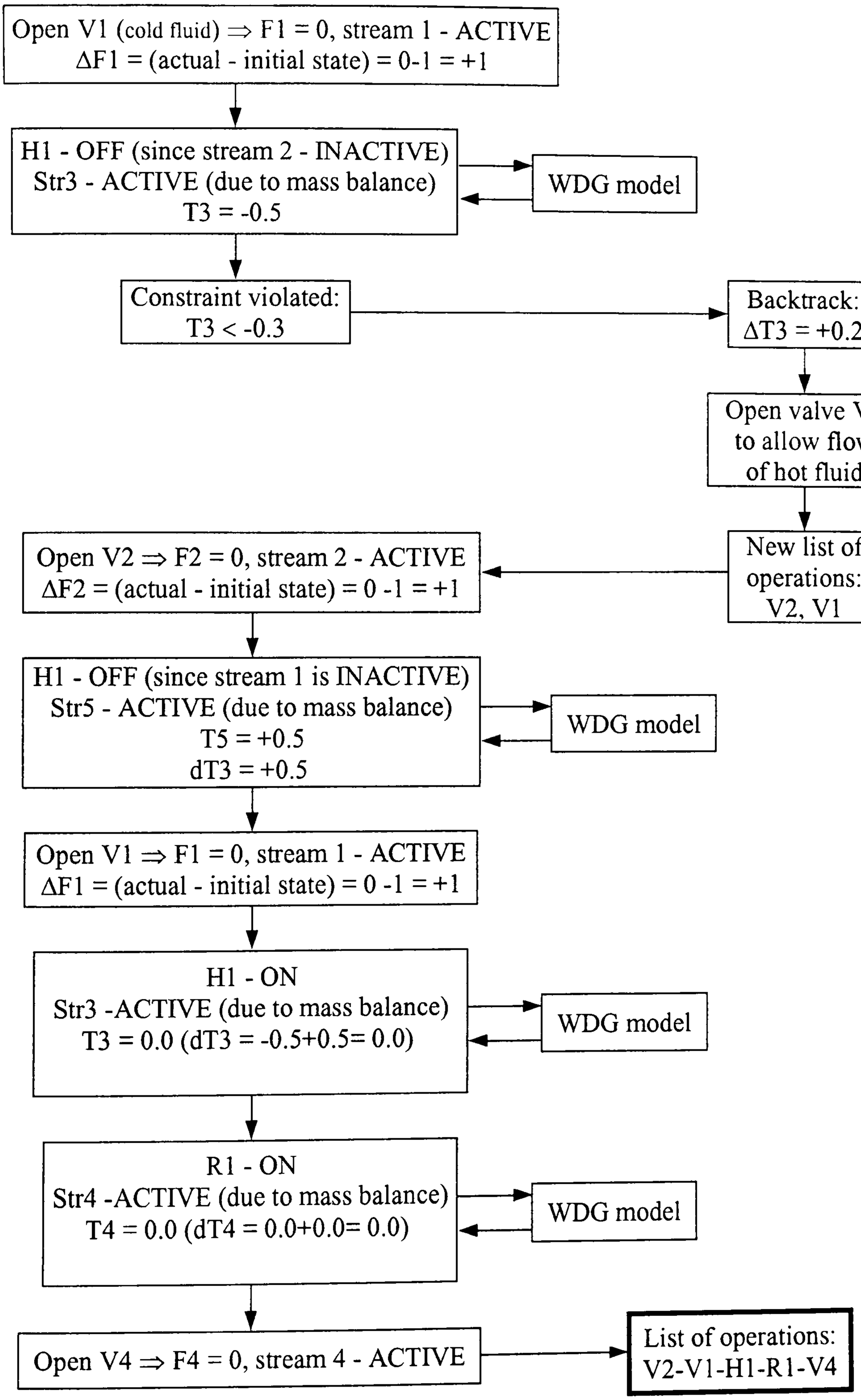


Figure 5.4 Example of the procedure for sequencing operations.

Dynamic Simulation of a Plant Start-up

By following the sequence of operations previously determined, the algorithm simulates the start-up of the plant using the inference of behaviour procedure described in Chapter 3, which is based on WDG models for describing the qualitative dynamic behaviour of each unit of equipment. The plant is considered successfully started up if the qualitative state of all streams are close to *NIL* (zero) at the end of the simulation. This means that all variables have reached their normal operating conditions. Otherwise a detailed list of the state of all variables during the start-up is generated, which can be used to carry out an analysis of what is possibly wrong with the procedure or the plant topology. The algorithm also traces back through the network of weighted digraphs for the possible "root cause" of the problem and delivers a short message describing the most probable cause, e.g. "low inlet temperature of reactor-R1".

5.4 Concluding Remarks

The generation of operating procedures for the transient operation of chemical plants, such as start-up, is a non-trivial multi-objective task. The increasing complexity of modern plants, which are highly integrated to minimise energy use and waste of raw materials, has given rise to the need for methodologies for the automatic generation of operating procedures. The intention is to systematise the procedures aiming at relying less in experience-based approaches and allowing potential operating problems to be identified at an early stage of design, with the objective of improving overall efficiency and safety. At this stage, information is mainly qualitative, which makes qualitative simulation an adequate tool in supporting synthesis of operating procedures.

In this chapter a strategy for generating start-up procedures for chemical plants based on weighted digraph models has been proposed. The aim is to demonstrate that weighted digraph models are well suited to be used in a process engineering design environment to support generation of operating procedures. The strategy is concerned not only with sequencing operations based on the process flow diagram of the plant and operational constraints, but also with testing the feasibility of the procedure and identifying potential bottlenecks related to the process topology. This is achieved by dynamically simulating the start-up of the plant based on the proposed sequence of operations.

The effectiveness of this approach is analysed in the following chapter by reference to two case studies, based on a heat-exchanger network and a process sub-system composed of a CSTR and a feed/effluent heat-exchanger.

Chapter 6

Start-up Procedures Based on Weighted Digraph Models

6.1 Introduction

Operating procedures during start-up and shut-down involve a set of complex and coordinated actions which need to be considered during the design stage of a process plant. The existence of operational constraints, recycles and coupled systems makes the synthesis of operating procedures more difficult, because certain sequences of operations may have to be avoided in order to prevent violation of constraints, or auxiliary equipment may be needed to allow start-up of the plant.

This chapter is concerned with the evaluation of the performance and limitations of the strategy for generation of start-up procedures based on weighted digraphs, proposed in the previous chapter. The suitability of the strategy in sequencing operations in the presence of operational constraints is analysed by reference to a case study of a network of heat-exchangers. The effectiveness of the approach in describing start-up dynamic trajectories and the ability to identify the need for auxiliary equipment to allow the start-up are also evaluated, based on an integrated

process flow diagram for energy recovery composed of a CSTR and a feed/effluent heat exchanger.

6.2 Comparison of Qualitative and Quantitative Trajectories

The ability of the qualitative procedure to describe dynamic trajectories during the start-up of process plants can be determined by comparing qualitative and quantitative dynamic trajectories. Quantitative trajectories are obtained from numerical simulation of the start-up of the plant, using the sequence of operations derived from the qualitative approach.

The comparison of results is made in terms of an analysis of the shape of the dynamic trajectories, taking into account the description of the most important features of the system, such as turning points, tendency to approach the steady-state and large deviations from the design conditions.

The numerical simulation is used as a reference to test the effectiveness of the qualitative algorithm in:

- Generating operating procedures which, when used by the numerical simulator to start-up the plant, do not violate operational numerical constraints;
- Identifying situations in which the plant cannot be started up without process modifications, in terms of the need for start-up auxiliary equipment.

6.3 Start-up of a Network of Heat-Exchangers

Consider the network of heat-exchangers schematically represented in Fig. 6.1, which is based on the modification of a case study presented by Fusillo and Powers (1988). The streams leaving the network are fed to other sub-systems and therefore some are subject to operational constraints. Stream number 10 (Str10) has a maximum temperature limit, while the temperature of stream number 13 (Str13) cannot fall below a minimum limit. The initial system temperatures and flow rates are set below the design conditions, and therefore negative qualitative values are assumed as the initial states. All valves are initially closed.

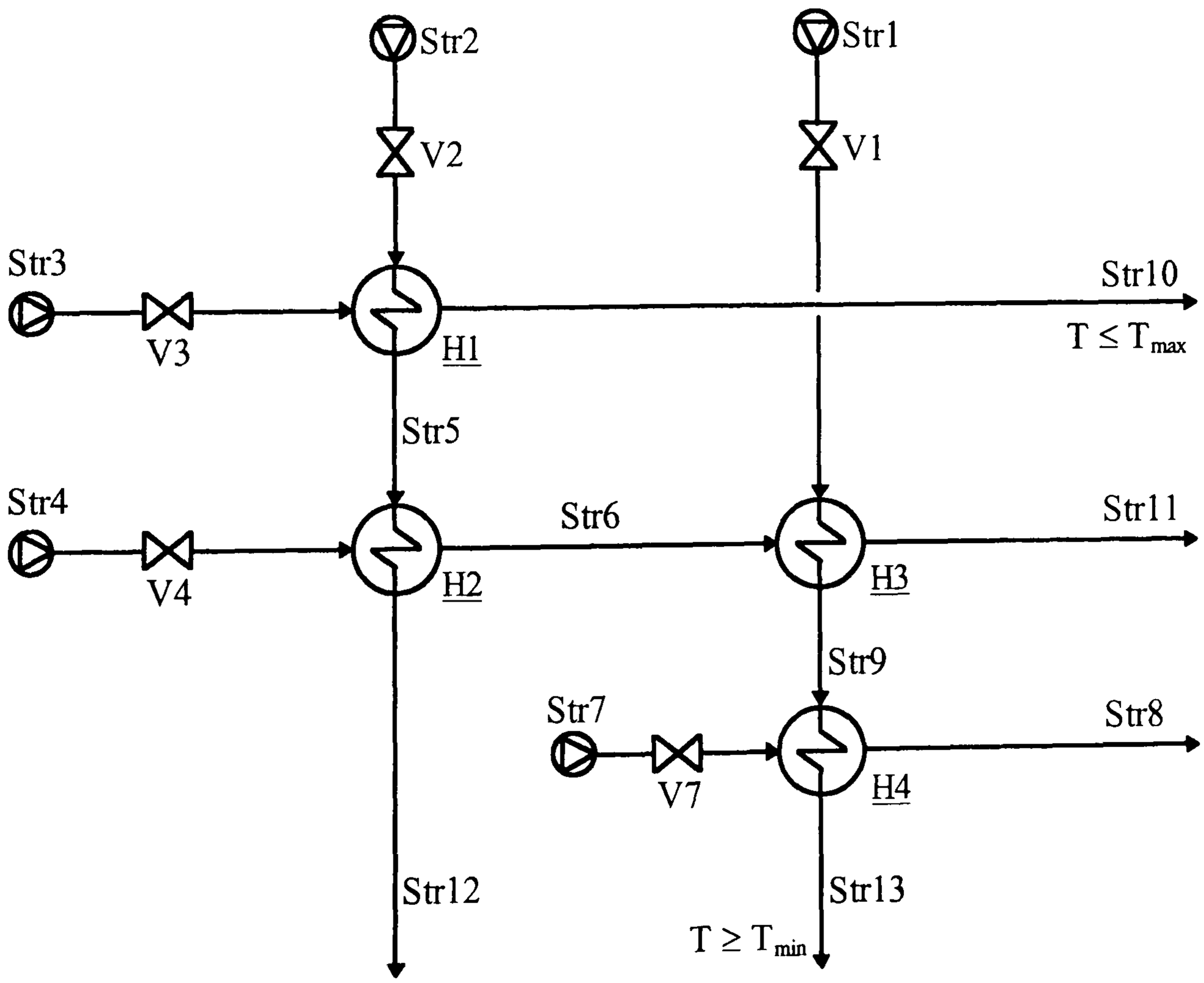


Figure 6.1 Schematic representation of the network of heat-exchangers.

Numerical values of parameters and design conditions used in the numerical simulations are listed in Tables A.7 to A.10 in Appendix A. Table 6.1 lists the quantitative values of the constraints and their equivalent qualitative translations.

Table 6.1 Quantitative and qualitative operational constraints.

Stream number	Constraints	
	Quantitative	Qualitative
10	$T \leq 340$ K	$T \leq +0.2$
13	$T \geq 315$ K	$T \geq -0.2$

The strategy for generation of start-up procedures, described in Chapter 5, is applied to the network of heat-exchangers. The trace of the sequencing procedure applied by the algorithm is depicted in Fig. 6.2. The algorithm tentatively opens valve 1 and follows the flow of mass simulating heaters H3 and H4. As all other valves are still closed the constraint related to stream 13 is violated. The *backtrack* mode is activated and valve 7 is put on the stack to be opened before valve 1. The algorithm continues backtracking until all valves have been assessed and put on the valves-stack. At the end of the *backtrack* procedure a sequence of valve manipulations is proposed as follows:

V3 - V2 - V4 - V7 - V1.

The algorithm tests the above sequence in *forward* mode and the temperature constraint of stream 10 is violated. The *backtrack* mode is activated again and the

sequence of manipulations of valves 2 and 3 is inverted and a new list of operations is generated:

V2 - V3 - V4 - V7 - V1.

The simulation of the above sequence (**seq23471**) in *forward* mode is successful, since no constraint is violated. The algorithm then proceeds to the second module and performs the dynamic simulation of the start-up of the system by using the proposed sequence. Table 6.2 shows the sequence of operations generated by the simulation algorithm and Fig. 6.3 shows the qualitative dynamic trajectories for heat-exchangers H1 and H4. It can be seen that the system reaches the steady state without violating the temperature constraints of streams 10 and 13. Valve 2 (cold fluid) is opened before valve 3 (hot fluid), and this guarantees that the temperature of stream 3 is lowered before it leaves the heat-exchanger H1, and consequently the violation of the constraint of stream 10 is avoided. Valves 4 and 7 (hot fluid) are opened before valve 1 (cold fluid), which guarantees that the temperature of stream 13 never falls below the minimum limit.

In order to verify the reliability and efficiency of the qualitative procedure in dealing with operational constraints, the network of heat-exchangers has been numerically simulated for three different start-up sequences:

- The first is the one generated by the qualitative algorithm, for which a successful start-up is obtained: V2-V3-V4-V7-V1, referred to as: **seq23471**;
- The second sequence has been rejected as inadequate by the qualitative algorithm: V1-V3-V2-V4-V7, referred to as: **seq13247**;
- An optional sequence (V4-V2-V7-V1-V3) proposed by Fusillo and Powers (1988) has also been tested, and is referred to as: **seq42713**.

Figure 6.4 shows the quantitative dynamic trajectories for heat-exchangers H1 and H4 for **seq23471**. In general, it can be seen that the results qualitatively agree with those generated by the qualitative algorithm (Fig. 6.3) and no constraint is violated, which demonstrates the reliability of the method. By qualitatively comparing Figs. 6.3 and 6.4 it can be seen that the qualitative behaviour of stream 8 is not well described by the qualitative simulation. This is due to the fact that during the start-up variables are subject to multiple disturbances of different intensity, e.g. simultaneous disturbances from inlet flow rate and temperature, and the weighted digraph models were previously tested for single disturbances and in stand-alone mode, i.e. interactions between equipment were not considered. In order to make weighted digraphs more adequate for describing all dynamic features of systems during start-up, a systematic procedure for determining *weights* of interacting processes is needed. For the time being the type of dynamic trajectories generated by the qualitative procedure is adequate, since it is meant to be a rough estimate of the general behaviour of the system for the proposed sequence of operations.

Figure 6.5 depicts the dynamic trajectories of the system for **seq13247**, rejected by the qualitative algorithm. Clearly this was a correct decision, since both constraints of streams 10 and 13 are violated in this start-up procedure. Valve 1 is opened before valves 4 and 7 causing the violation of the constraint of stream 13 ($T \geq 315$ K), while the opening of valve 3 before valve 2 causes a peak of temperature in stream 10, which is above the maximum limit of 340 K.

The optional operating sequence proposed by Fusillo and Powers (1988) also leads to a successful start-up procedure, as shown in Fig. 6.6. So the system is underconstrained and there is more than one feasible operating path that leads the system from the initial to the goal state.

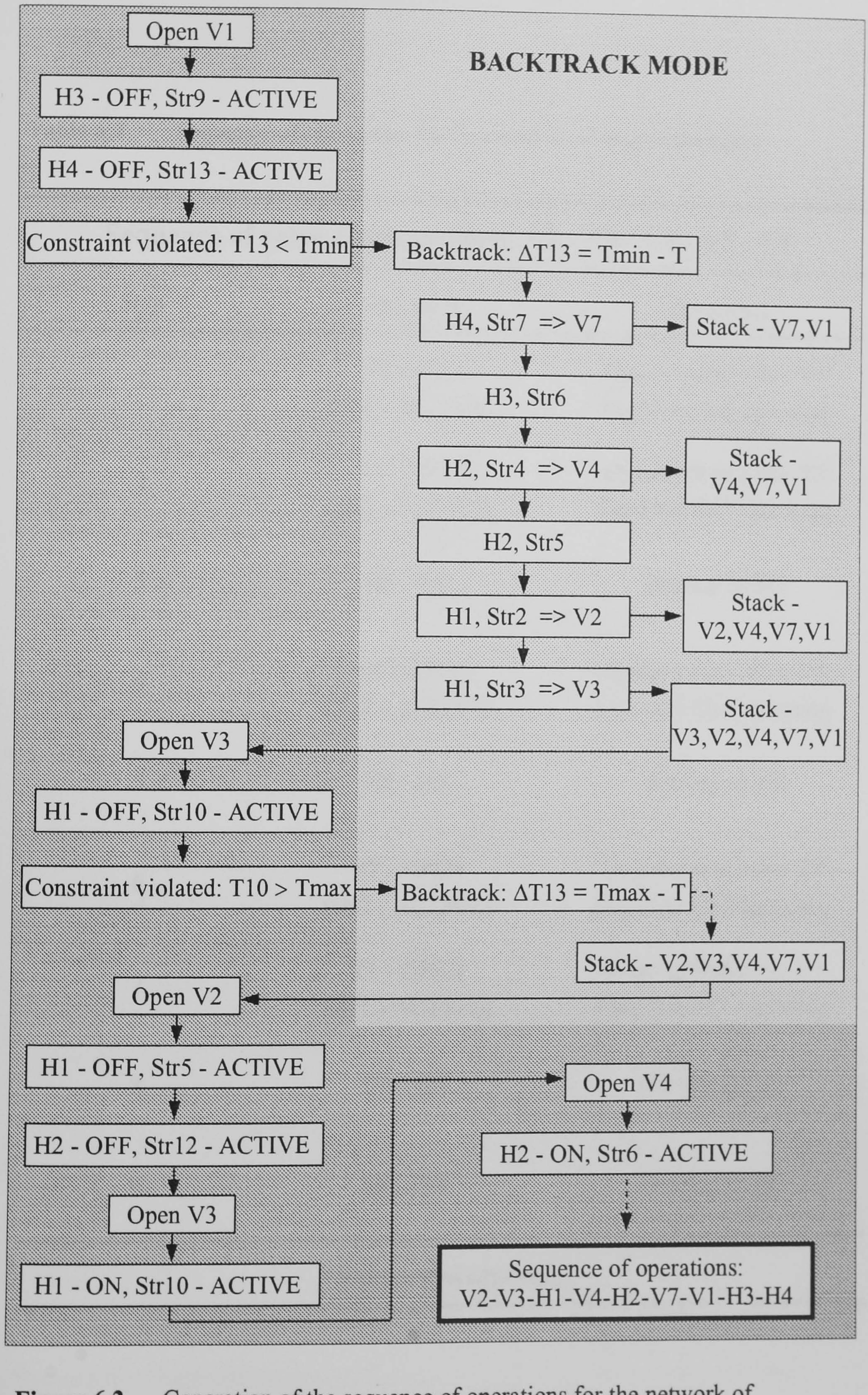
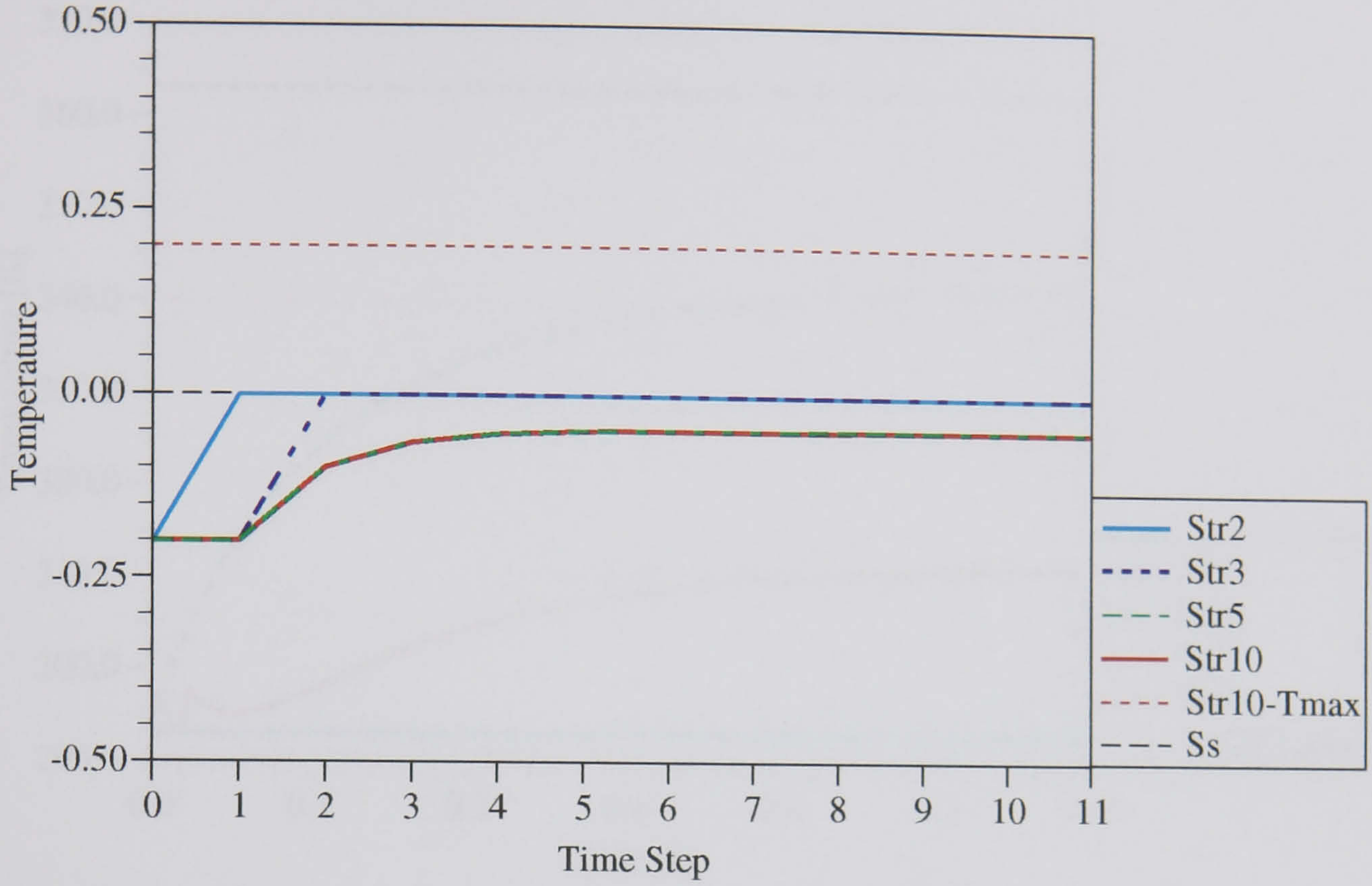


Figure 6.2 Generation of the sequence of operations for the network of heat-exchangers.

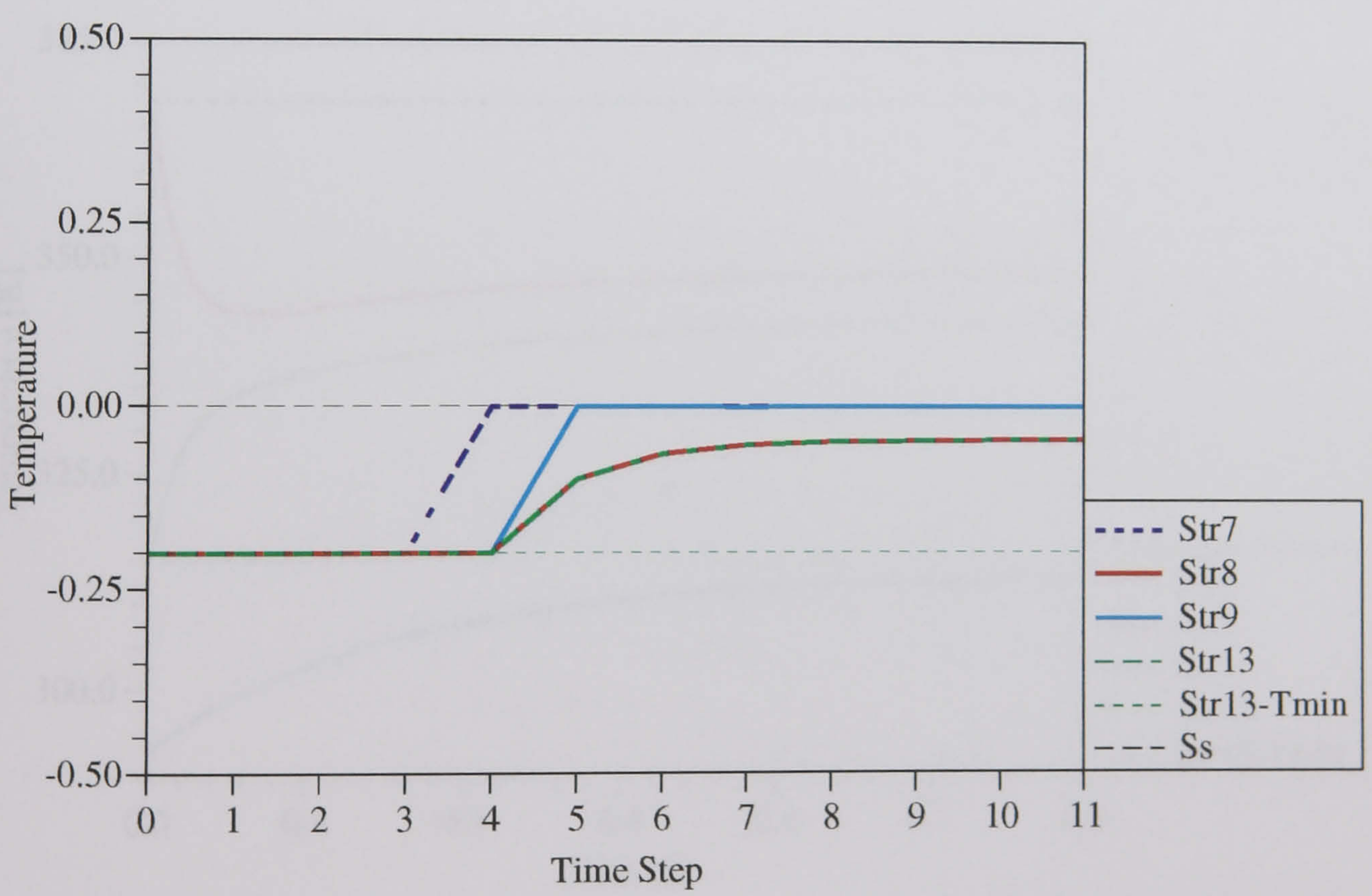
Table 6.2 Sequence of operations for the network of heat-exchangers.

Sequence of valve manipulations: V2 - V3 - V4 - V7 - V1		
Step	Operation	Comments
1	V2 OPEN (Str2 - ACTIVE)	Opening feed valve V2 Feed line Str2 operating
2	V3 - OPEN (Str3 - ACTIVE)	Opening feed valve V3 Feed line Str3 operating
3	H1 - ON	Starting up H1
4	V4 - OPEN (Str4 - ACTIVE)	Opening feed valve V4 Feed line Str4 operating
5	H2 - ON	Starting up H2
6	V7 - OPEN (Str7 - ACTIVE)	Opening feed valve V7 Feed line Str7 operating
7	V1 - OPEN (Str1 - ACTIVE)	Opening feed valve V1 Feed line Str1 operating
8	H3 - ON	Starting up H3
9	H4 - ON	Starting up H4
***Successful start-up**		

(a)



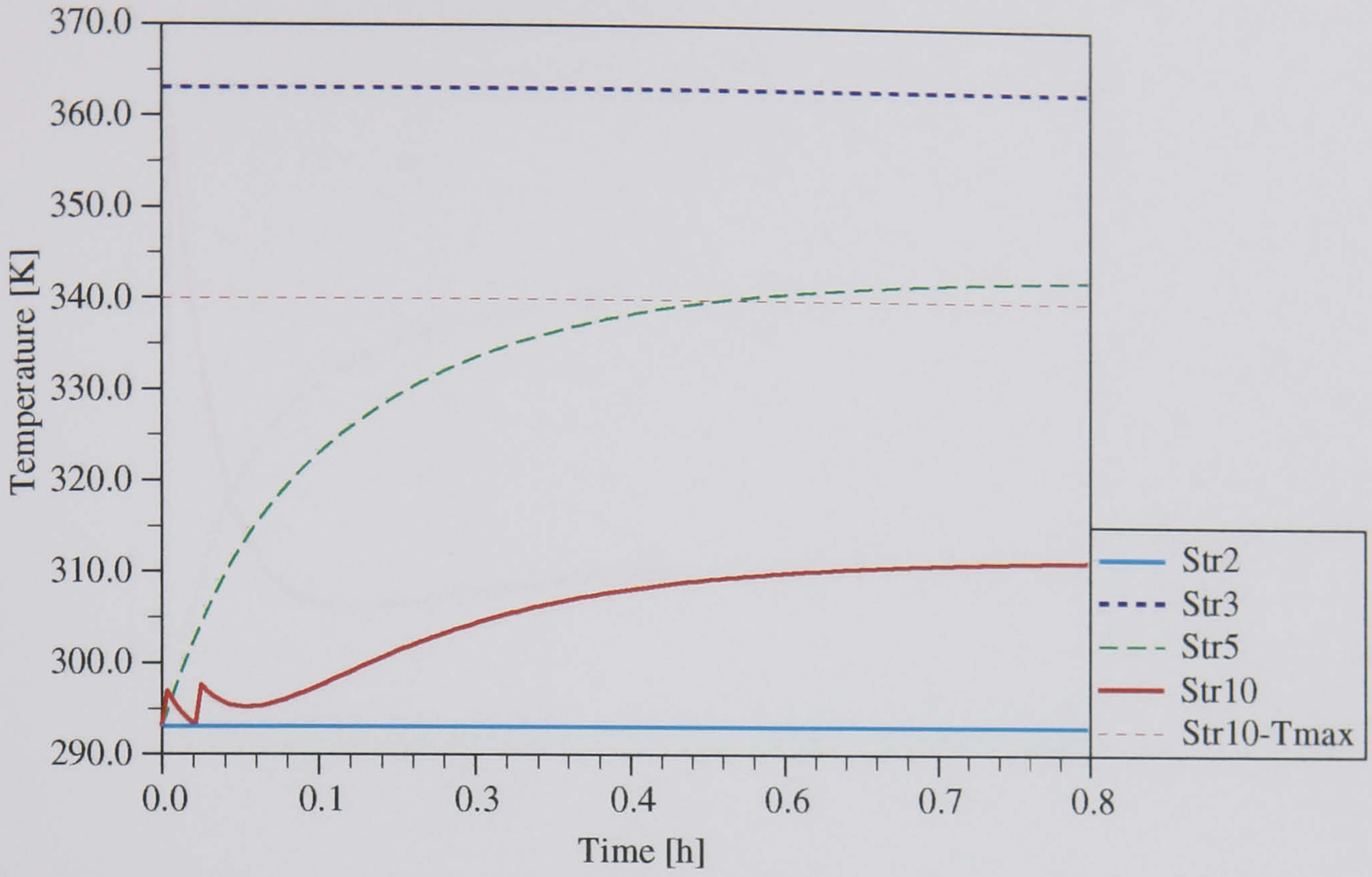
(b)



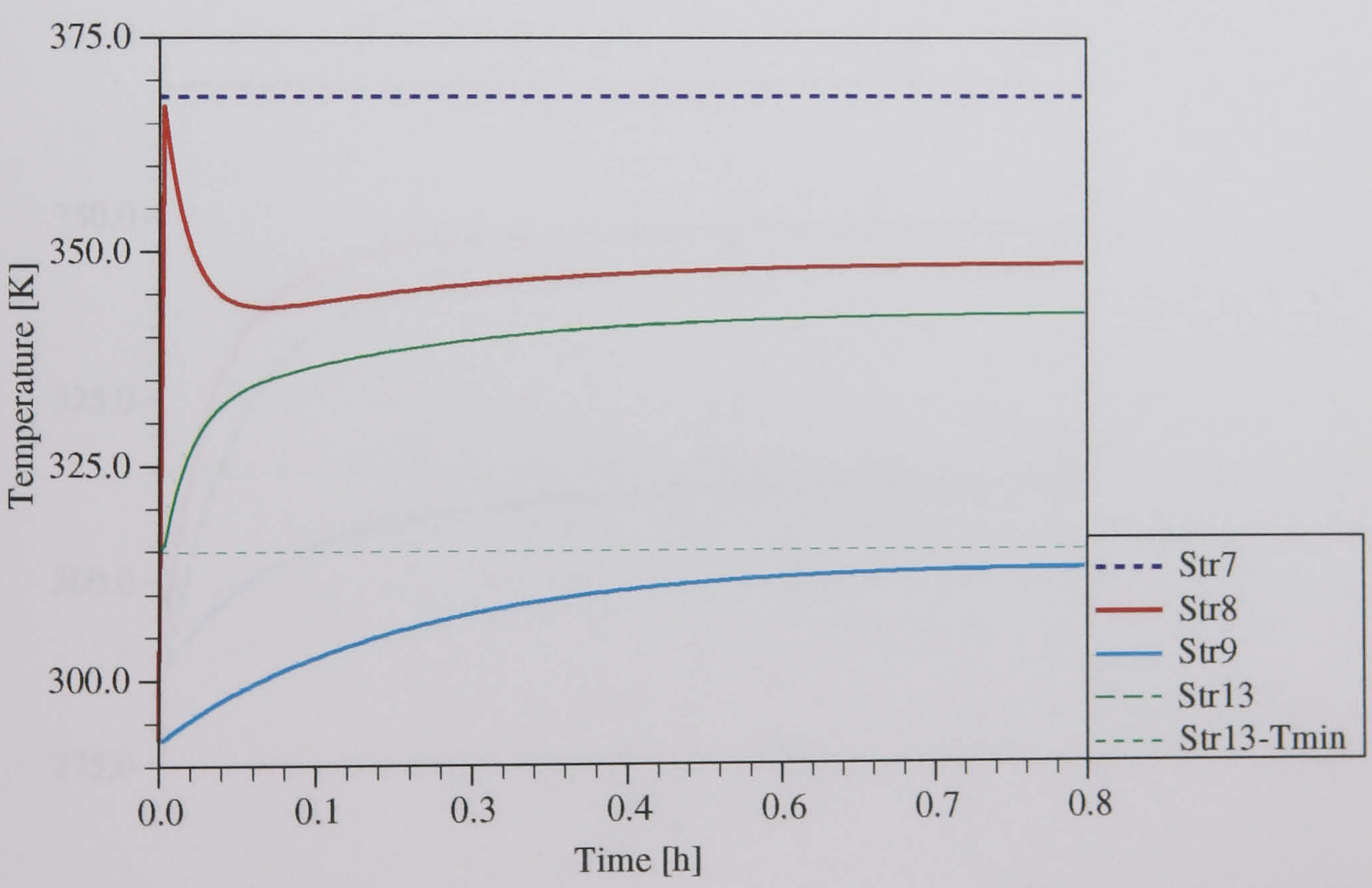
NB: Ss = design steady-state condition
Str = stream identifier

Figure 6.3 Network of heat-exchangers - qualitative temperature profiles for **seq23471**: (a) heater H1; (b) heater H4.

(a)



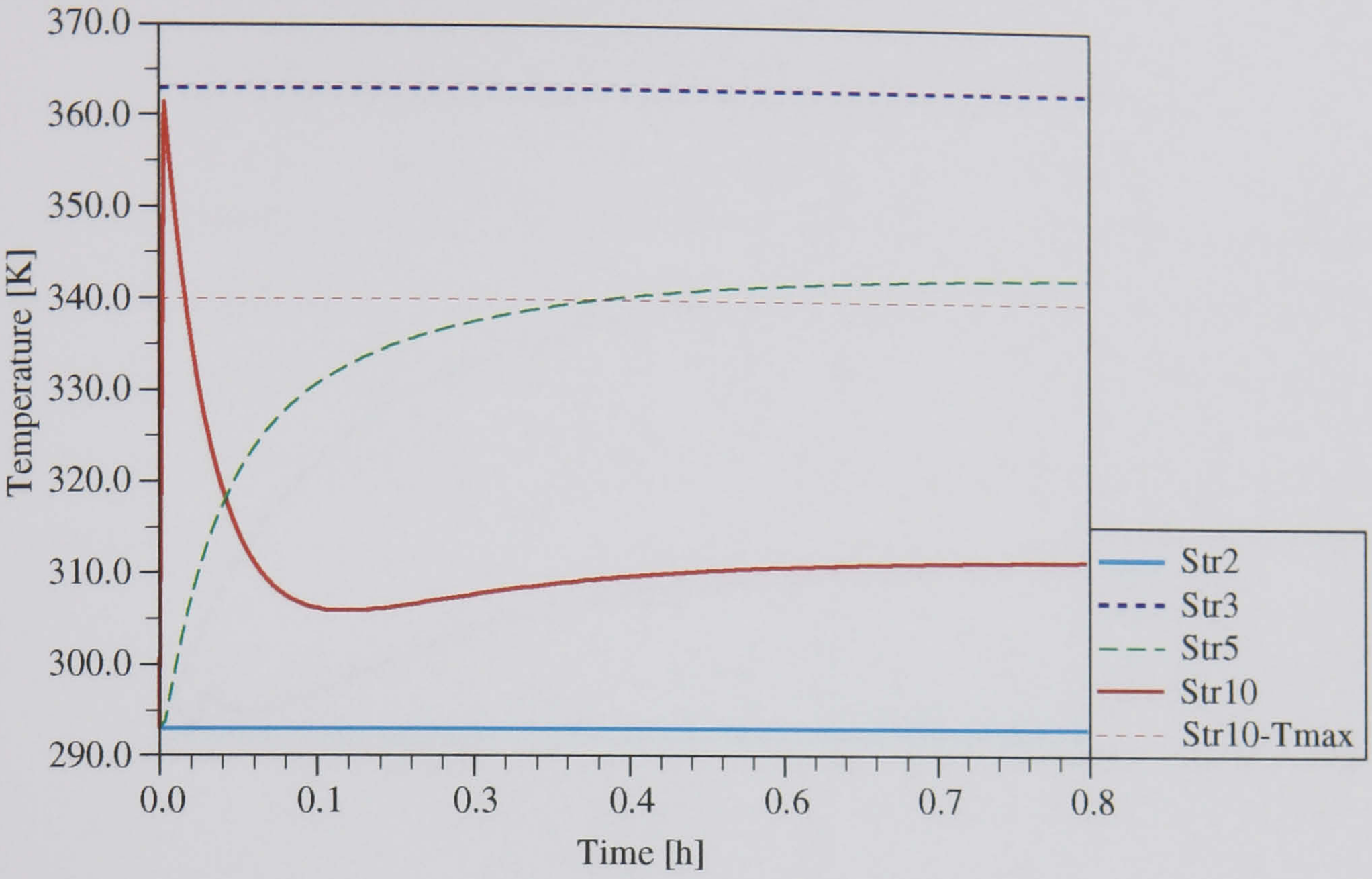
(b)



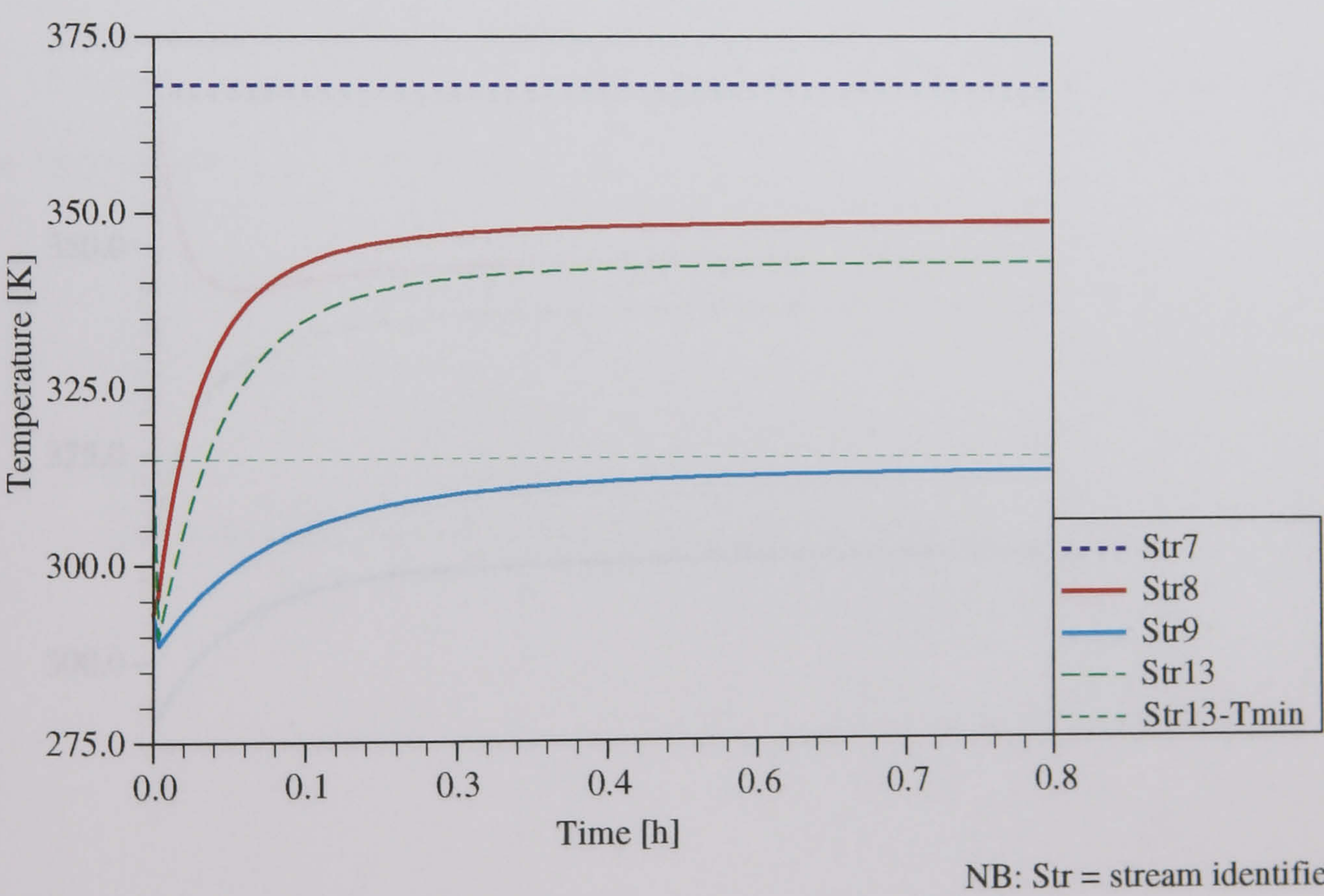
NB: Str = stream identifier

Figure 6.4 Network of heat-exchangers - quantitative temperature profiles for **seq23471**: (a) heater H1; (b) heater H4.

(a)



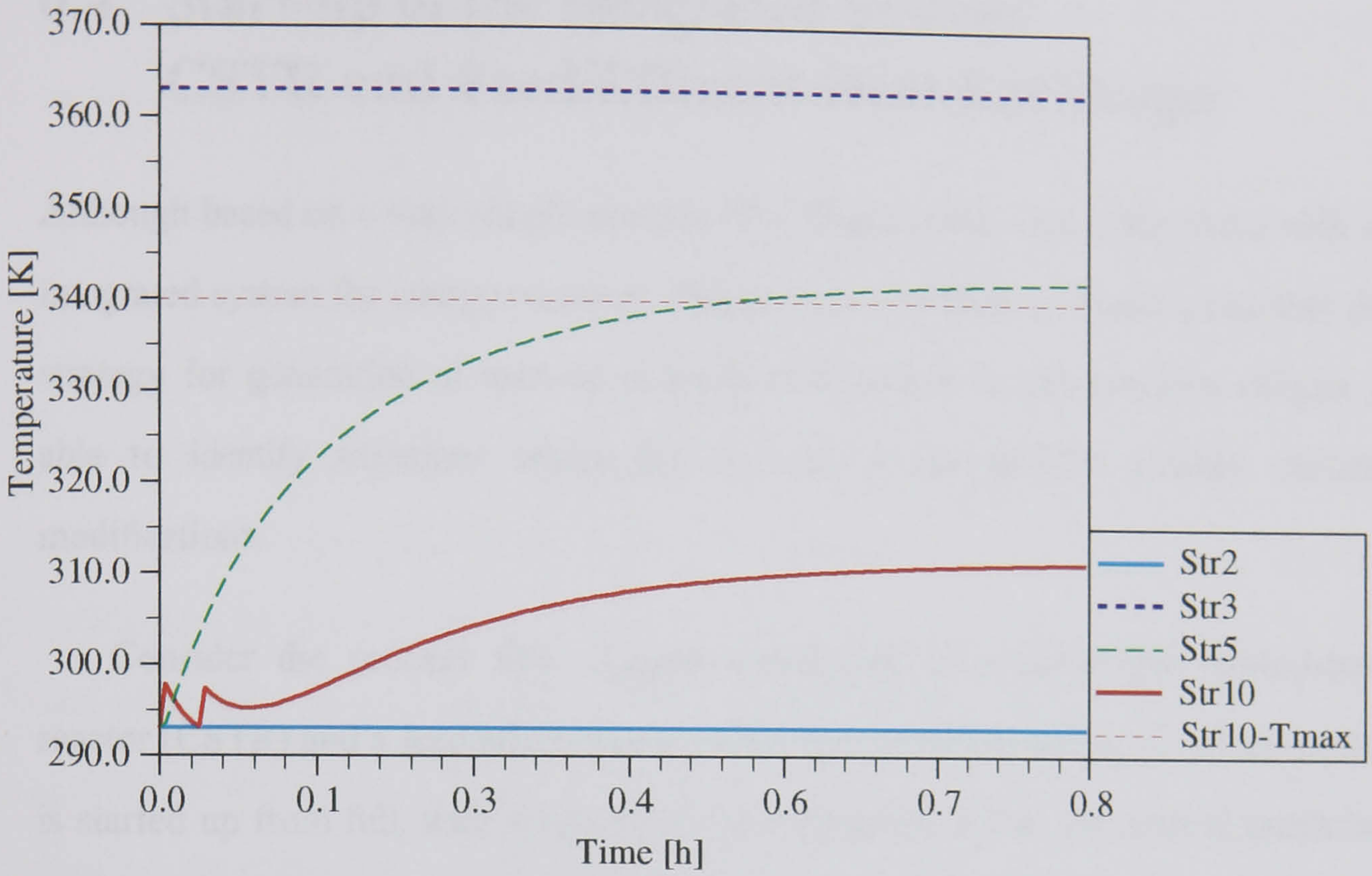
(b)



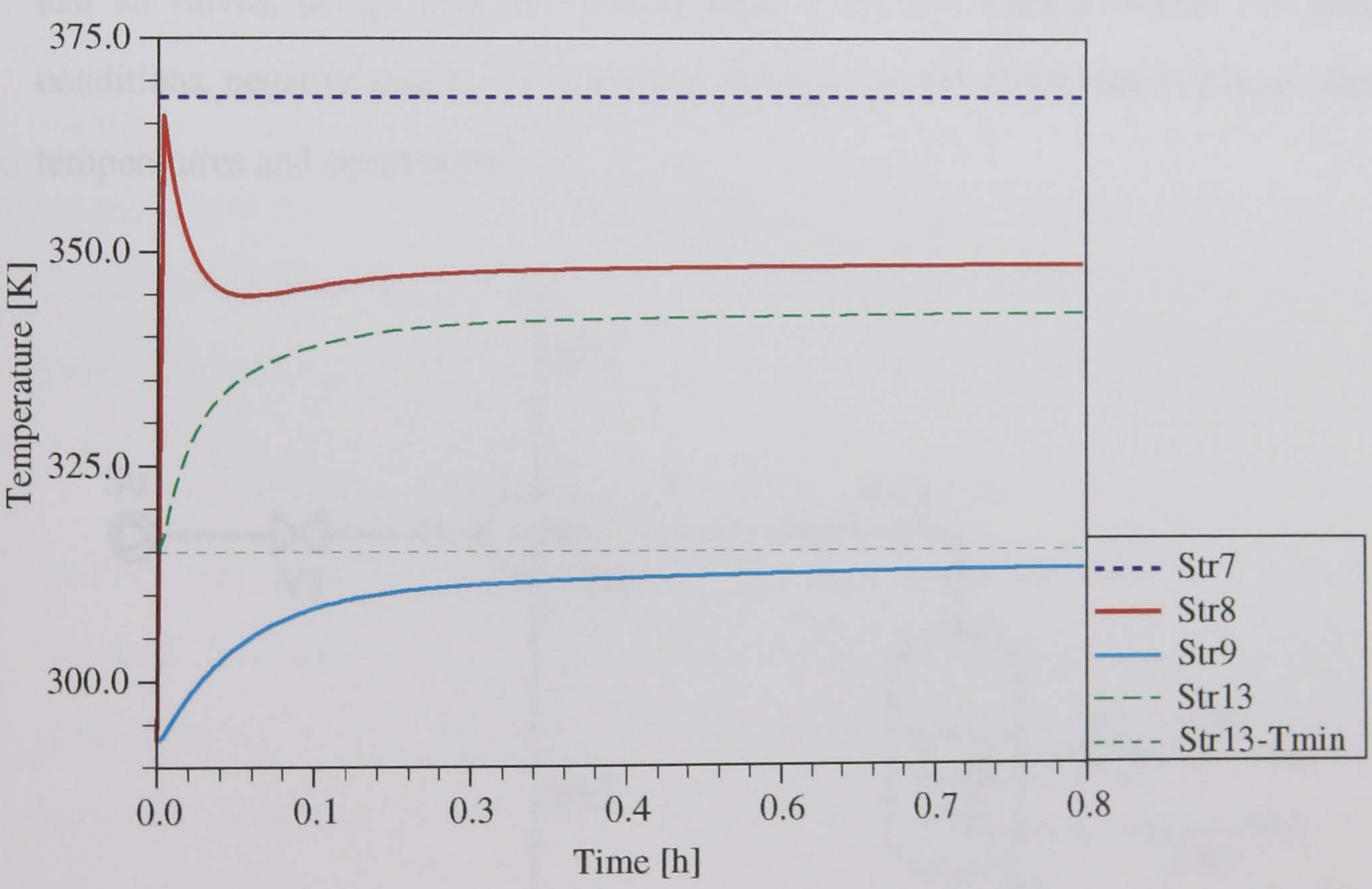
NB: Str = stream identifier

Figure 6.5 Network of heat-exchangers - quantitative temperature profiles for **seq13247**: (a) heater H1; (b) heater H4.

(a)



(b)



NB: Str = stream identifier

Figure 6.6 Network of heat-exchangers - quantitative temperature profiles for **seq42713**: (a) heater H1; (b) heater H4.

6.4 Start-up of the Integrated System: CSTR and Feed/Effluent Heat-Exchanger

Although based on a very simple process flow diagram this case study deals with an integrated system for energy recovery. This makes it possible to demonstrate that the strategy for generation of start-up procedures described in the previous chapter is able to identify situations where the start-up is not feasible without process modifications.

Consider the process flow diagram composed of a continuous stirred-tank reactor (CSTR) and a feed/effluent heat-exchanger, as shown in Fig. 6.7. The reactor is started up from full, with reactants at concentrations below the normal operating conditions. All initial temperatures and flow rates are set below the design conditions, and all valves, except that for cooling water (V5), are initially closed. For these conditions, negative qualitative values are attributed to the initial states of flow rates, temperatures and concentrations.

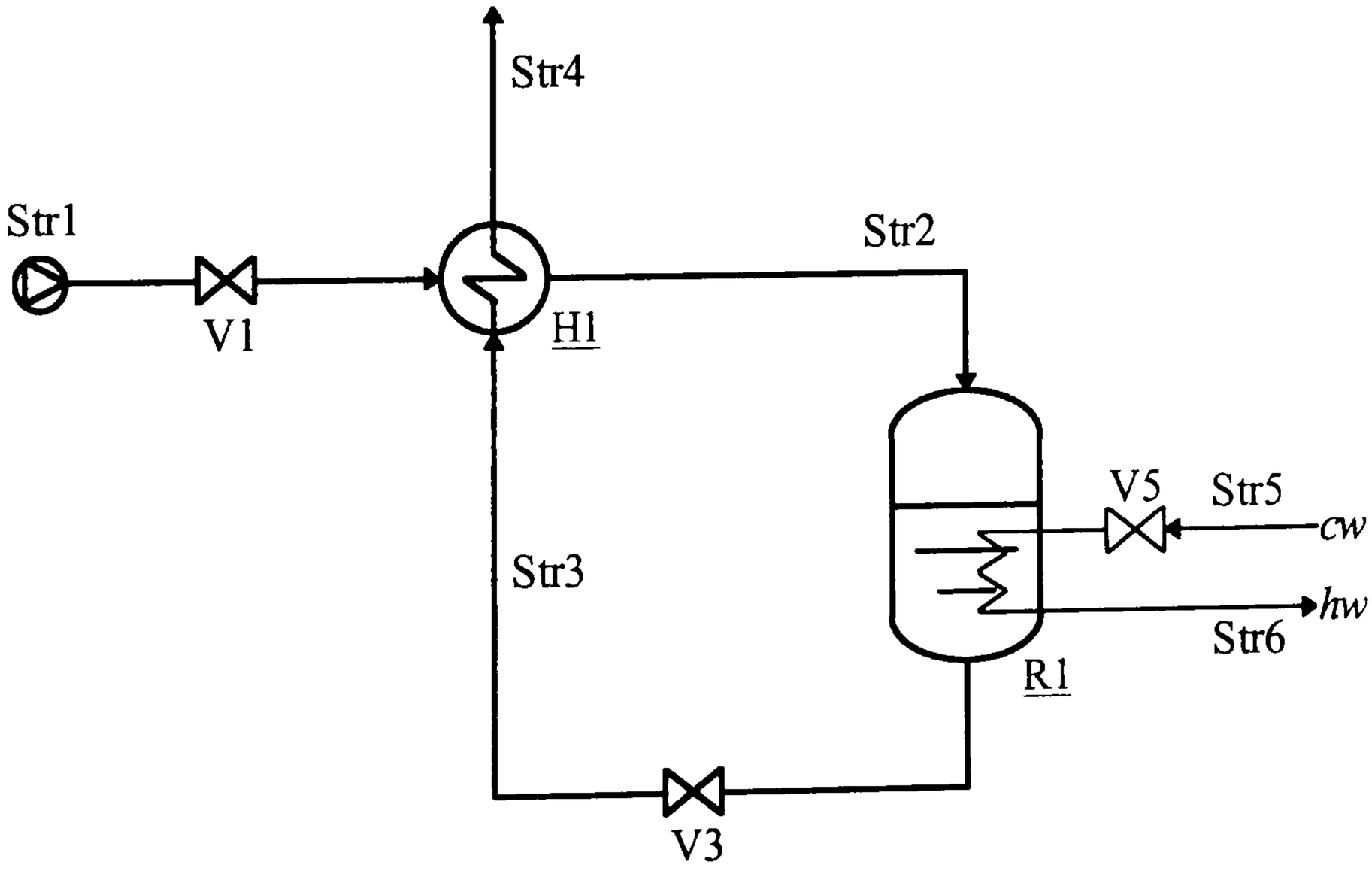


Figure 6.7 Schematic representation of the CSTR and feed/effluent heat-exchanger system.

Numerical values of the design conditions and parameters of this system, for use in numerical simulations, are listed in Tables A.11 to A.12 in Appendix A.

The process flowsheet graph (PFG), based on the process flow diagram (PFD) of the system (Fig. 6.7), is input to the algorithm for generation of start-up procedures. Table 6.3 summarises the sequence of operations and comments generated by the algorithm, which includes a statement about the impossibility of starting up the system. As can be seen in the comments of Table 6.3, the algorithm traces back to find out the "root cause" of the low temperature in the reactor feed and identifies a cycle involving stream 2. Despite this, the algorithm generates the qualitative dynamic trajectories of the system for the sequence: V1-V3, depicted in Fig. 6.8, to allow further analysis, since no other successful list of operations could be generated. This figure shows that the temperature of the system tends to settles at a level below the normal operating condition, which is not enough to allow the reactor to reach the desired steady-state. The concentration of component *A* in the reactor outlet stream settles at a level above the normal operating condition because of the low reaction temperature. These general qualitative features of the system are confirmed by the quantitative simulation of the start-up, as shown in Fig. 6.9.

The qualitative algorithm does not provide any support for reasoning with the results, and therefore cannot suggest ways of solving the problems detected. However, by analysing the system behaviour and qualitative results it is clear that the cycle involving stream 2 can be broken by including an auxiliary pre-heater for raising the reactor feed temperature and so allowing the start-up.

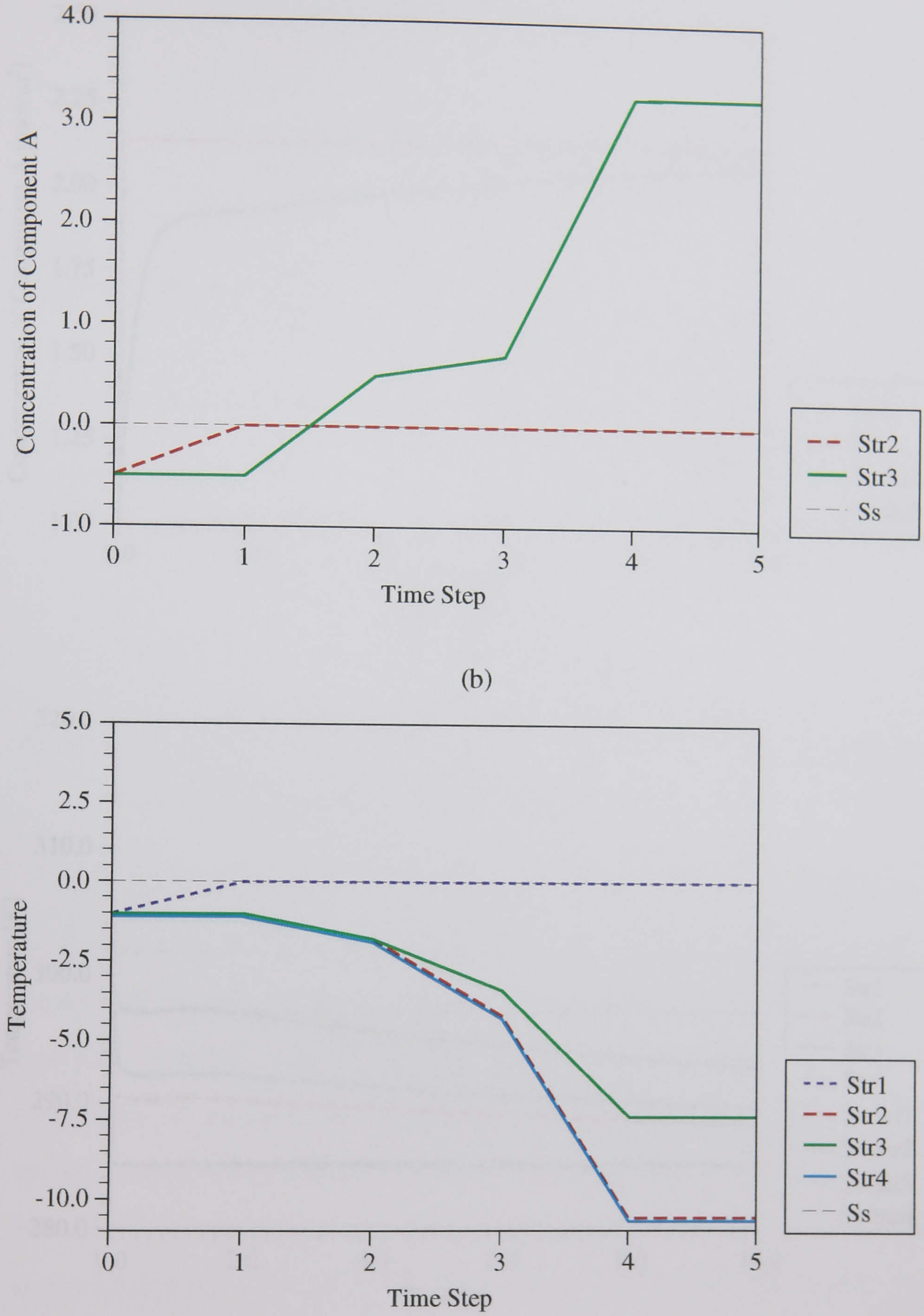
The process flow diagram (PFD) of the CSTR and heat-exchanger system is modified to include the start-up auxiliary pre-heater (H2) that uses steam as the hot fluid, as shown in Fig. 6.10.

Table 6.3 Sequence of operations for the CSTR and feed/effluent heat-exchanger system.

Sequence of valve manipulations: V1 - V3		
Step	Operation	Comments
1	V1 OPEN (Str1 - ACTIVE)	Opening feed valve V1 Feed line Str1 operating
2	R1 - ON	Starting up R1 Str2 - low T; Str3 - low T; Str3 - high C_a
3	V3 - OPEN (Str3 - ACTIVE)	Opening valve V3 Flow through Str3
4	H1 - ON	Starting up H1 Str3 - low T; Str2 - low T; Str4 - low T
***Problem during start-up: reactor R1 → low inlet temperature**		
***System cannot be started up;**		
***Cycle involving Str2 → low T_2 → low T_3 → low T_2**		

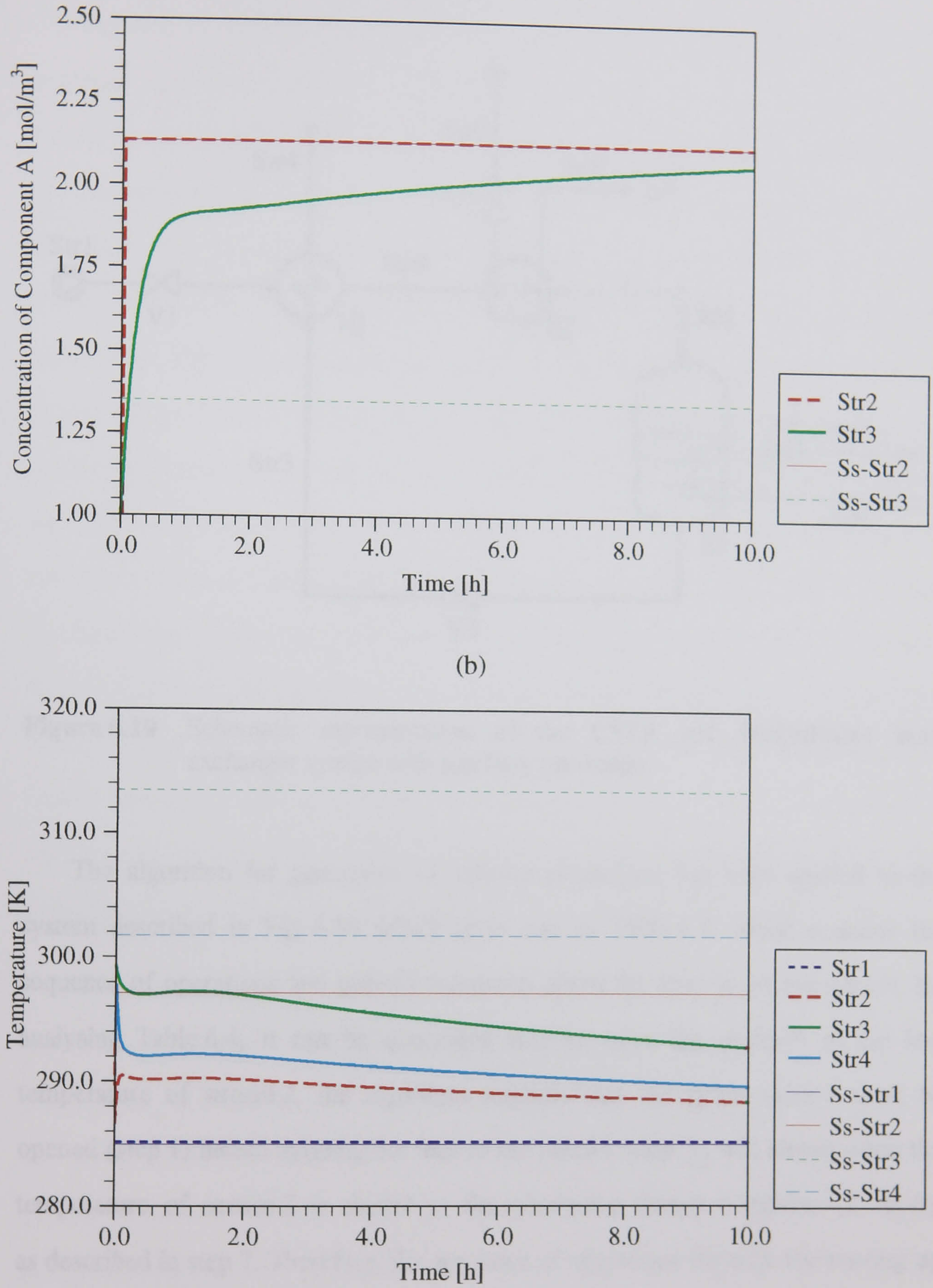
The initial conditions of heater H1 and reactor R1 are the same as for the basic PFD, and, additionally, the steam valve (V7) is set initially as closed. As this equipment is not intended to be operated during normal plant operation, valve 7 is not included in the list of valves to be normally manipulated by the algorithm of generation of start-up procedures, i.e. valve 7 can only be assessed in *backtrack* mode.

Numerical values of parameters and design conditions of the pre-heater (H2), used in numerical simulations, are listed in Table A.13 in Appendix A.



NB: Ss = design steady-state condition
Str = stream identifier

Figure 6.8 Qualitative simulation of the start-up of the CSTR and feed/effluent heat-exchanger system: (a) concentration profiles of component *A*; (b) temperature profiles.



NB: Ss = design steady-state condition
Str = stream identifier

Figure 6.9 Numerical simulation of the start-up of the CSTR and feed/effluent heat-exchanger system: (a) concentration profiles of component *A*; (b) temperature profiles.

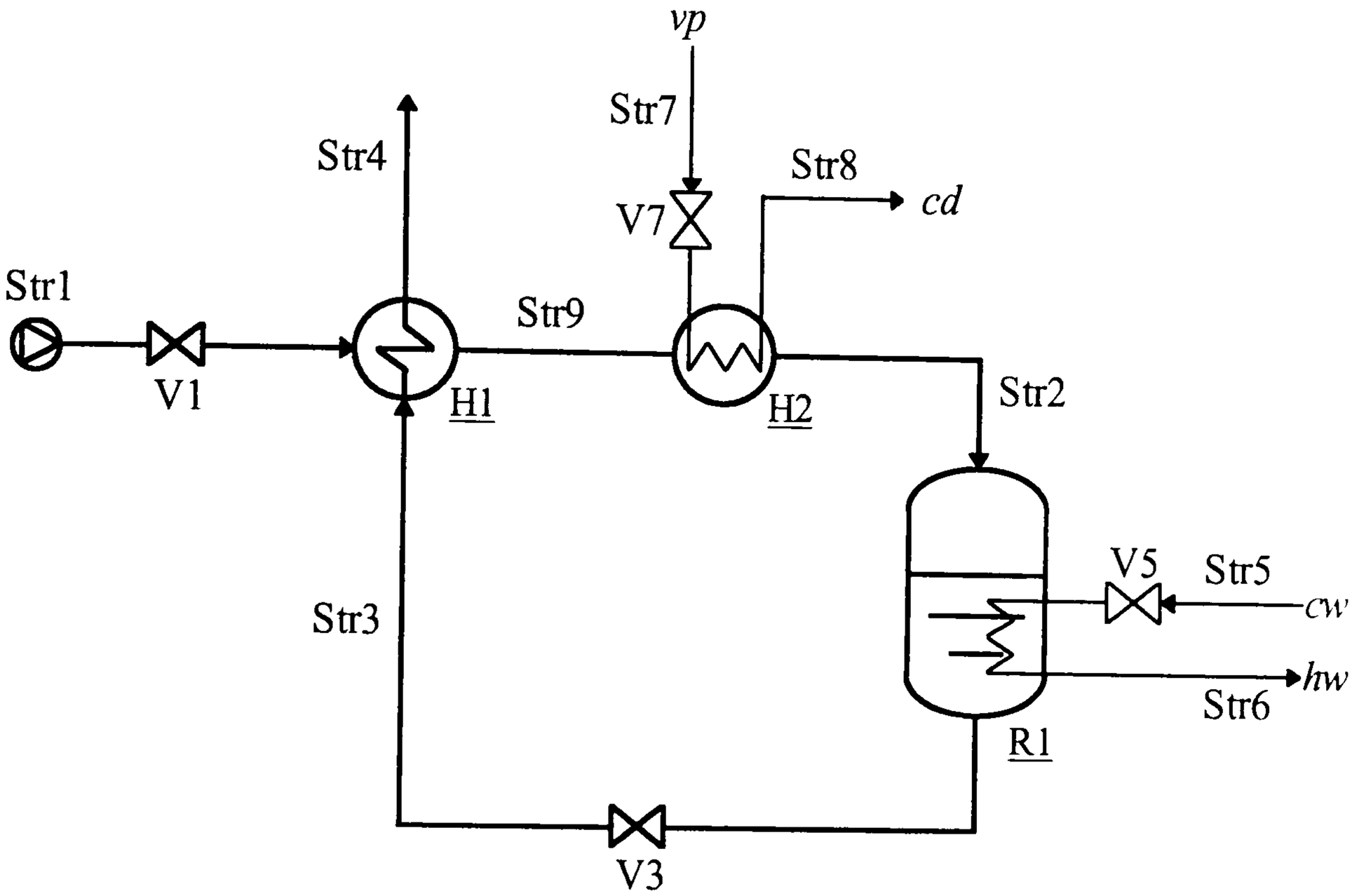


Figure 6.10 Schematic representation of the CSTR and feed/effluent heat-exchanger system with auxiliary pre-heater.

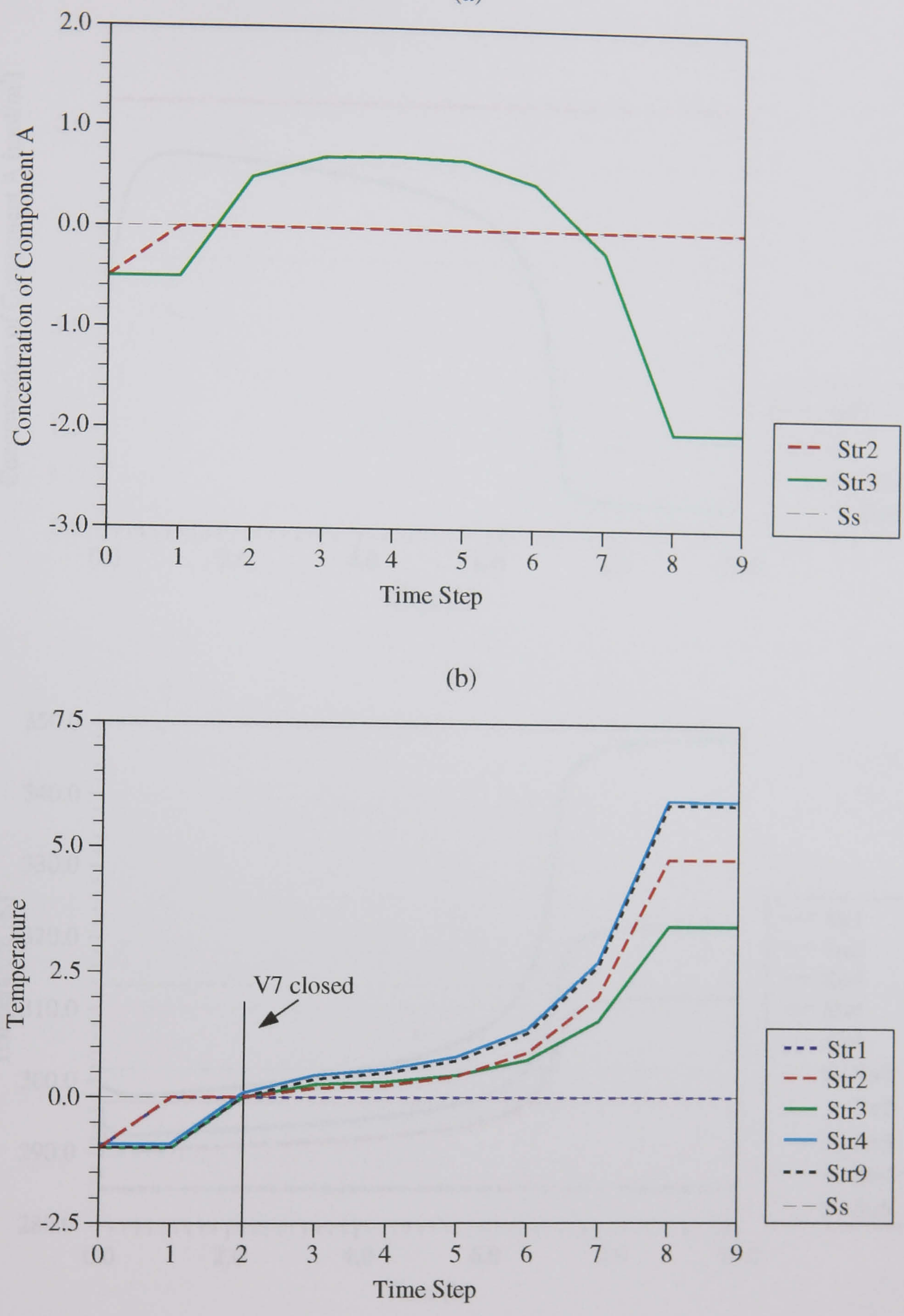
The algorithm for generation of start-up procedures has been applied to the system described in Fig. 6.10 which gives rise to Table 6.4, which contains the sequence of operations and general comments about the start-up of the system. By analysing Table 6.4, it can be concluded that to solve the problem of the low temperature of stream 2, the algorithm requires that the steam valve 7 must be opened (step 1) before opening the feed to the reactor (step 2), and closed when the temperature of stream 2 is almost at the qualitative design condition ($T_2 \cong 0$), as described in step 7. Therefore, the sequence of operations includes the starting up and shutting off of the auxiliary heater H2. Despite the successful generation of the operating procedure, the algorithm delivers a warning message, pointing out that a high inlet reactor temperature is to be expected.

Figure 6.11 describes the qualitative dynamic behaviour of the system for the proposed operating procedure of Table 6.4. It can be seen that the system can be started up, but both the reaction temperature and the conversion of component A in the reactor are higher than the desired steady-state values (qualitative value = 0). This behaviour is also confirmed by the numerical simulation of the start-up, as shown in Fig. 6.12. The analysis of the results indicates that the time for closing the steam valve 7 may be critical for the integrated system. If it is closed before the inlet reactor temperature (T_2) reaches an adequate value, the reactor may not start-up due to the temperature being too low. If it is closed after (T_2) has reached the design conditions, the period of time between detecting T_2 and closing valve 7 may be long enough to result in high temperatures (sometimes undesired) being reached. In fact, the qualitative simulation generates possible behaviour patterns, based on system characteristics, i.e. the algorithm detects the possibility of high system temperatures being reached, but this is not guaranteed since it depends on specific values of the system parameters. The importance of the time of closure of the steam valve (V7) suggests that the use of a temperature controller, which manipulates the steam flow rate (F_7) to lead the system to the desired steady-state, will be necessary.

Figure 6.13 shows the results of the numerical simulation of the system when a proportional controller based on the inlet temperature of the reactor (T_2), and manipulating the steam flow rate (F_7) is added to the basic PFD of Fig. 6.10. The numerical simulation is based on the sequence of operations proposed by the qualitative procedure (Table 6.4). Results show that the system reaches the desired steady-state values of temperatures and concentrations.

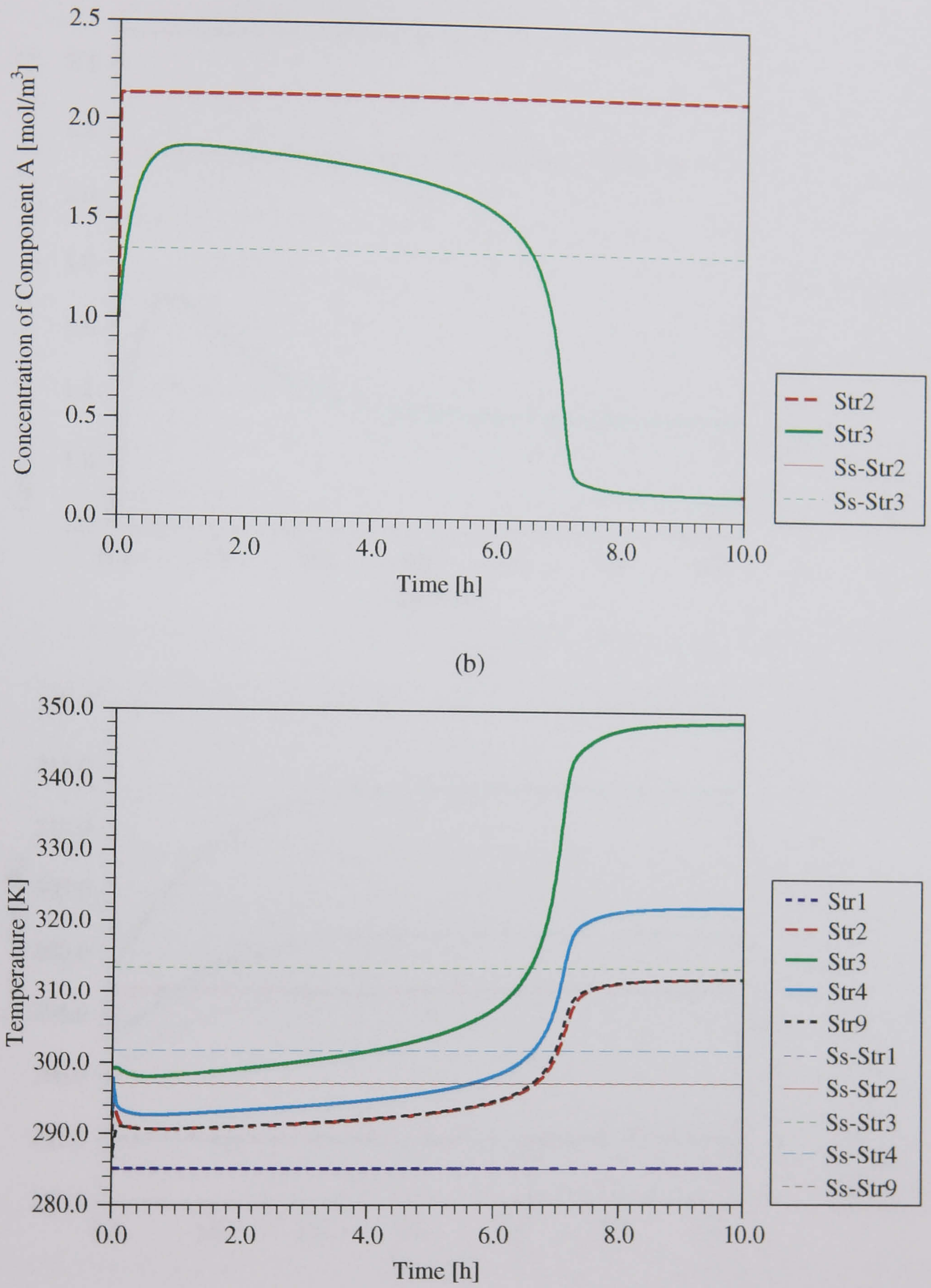
Table 6.4 Sequence of operations for the CSTR and feed/effluent heat-exchanger system with auxiliary pre-heater.

Sequence of valve manipulations: V7 - V1 - V3 - V7		
Step	Operation	Comments
1	V7 OPEN (Str7 - ACTIVE)	Opening steam valve V7 Steam line Str7 operating
2	V1 - OPEN (Str1 - ACTIVE)	Opening feed valve V1 Feed line Str1 operating
3	H2 - ON	Starting up H2 Str9 - low T
4	R1 - ON	Starting up R1
5	V3 - OPEN (Str3 - ACTIVE)	Opening valve V3 Flow through Str3
6	H1 - ON	Starting up H1
7	V7 - CLOSE (Str7 - INACTIVE)	$T_2 \cong 0$, closing valve V7 Steam line Str7 inactive
9	H2 - OFF	Shutting off H2 Str2 - high T
***System started up**		
Warning: Reactor R1 → high inlet temperature		



NB: Ss = design steady-state condition
Str = stream identifier

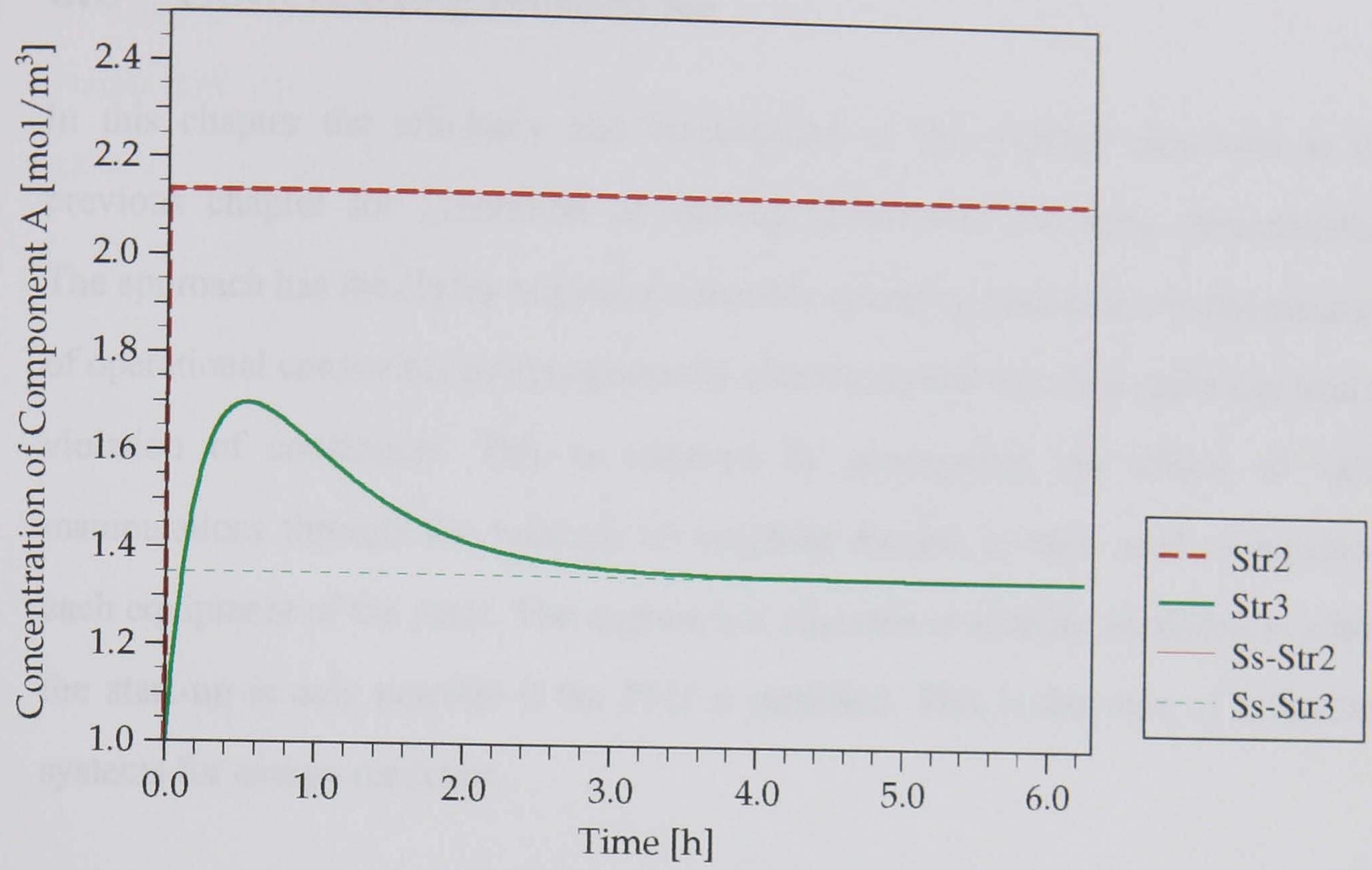
Figure 6.11 Qualitative simulation of the start-up of the CSTR and feed/effluent heat-exchanger system with auxiliary pre-heater: (a) concentration profiles of component *A*; (b) temperature profiles.



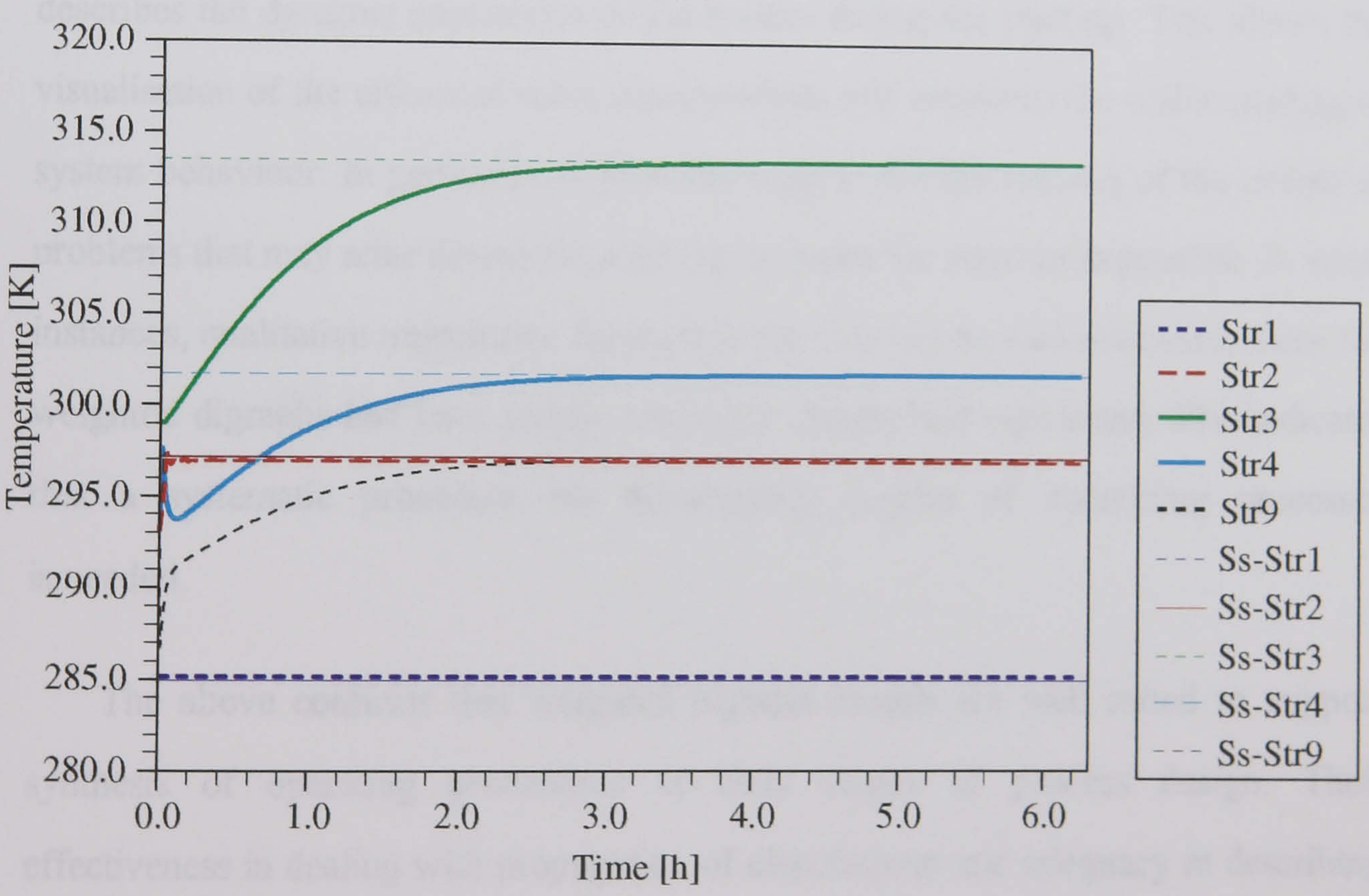
NB: Ss = design steady-state condition
Str = stream identifier

Figure 6.12 Numerical simulation of the start-up of the CSTR and feed/effluent heat-exchanger system with auxiliary pre-heater: (a) concentration profiles of component *A*; (b) temperature profiles.

(a)



(b)



NB: Ss = design steady-state condition
Str = stream identifier

Figure 6.13 Numerical simulation of the start-up of the CSTR and feed/effluent heat-exchanger system with auxiliary pre-heater and temperature control: (a) concentration profiles of component *A*; (b) temperature profiles.

6.5 Concluding Remarks

In this chapter the efficiency and functionality of the strategy described in the previous chapter for generation of start-up procedures has been demonstrated. The approach has the ability to generate feasible operating procedures in the presence of operational constraints by systematically identifying and rejecting paths that lead to violation of constraints. This is achieved by propagating the effects of valve manipulations through the network of weighted digraph models used to represent each equipment of the plant. The approach is also able to identify situations in which the start-up is only possible if the PFD is modified. This is the case of integrated systems for energy recovery.

The procedure not only generates sequences of operations but also qualitatively describes the dynamic trajectories of the system during the start-up. This allows the visualisation of the effects of valve manipulations and improves the understanding of system behaviour. In particular, it provides support for the analysis of the causes of problems that may arise during the start-up, or make the start-up impossible. In some instances, qualitative trajectories during start-up may not be well described, since the weighted digraphs had been mainly tested for stand-alone equipment. This indicates that a systematic procedure for determining *weights* of interacting processes is needed.

The above confirms that weighted digraph models are well suited to support synthesis of operating procedures at early stages of process design. Their effectiveness in dealing with propagation of disturbances and adequacy in describing dynamic trajectories make them powerful tools to be used as a platform to support generation of process plant operating procedures in a process engineering design environment.

There is clearly a need for an interactive interface to help with the analysis of results and reasoning about possible solutions for problems that arise during the generation of operating procedures. These problems are usually related to process topology, strategies of control and operational constraints. It must be able to reveal the way in which disturbances flow through weighted digraph models and be supported by a knowledge base in terms of weighted digraphs and heuristic rules to help with reasoning about the system response characteristics.

Chapter 7

Concluding Remarks and Suggestions for Future Work

7.1 Overview of the Study

Modern plants tend to have strong interactions between process units and are therefore more difficult to control and operate. This, associated with the need to meet much tighter operational and safety criteria, makes it important to consider operating procedures in the early stages of design to ensure safer and better operated plants.

Synthesis of operating procedures is usually based on heuristic and empirical knowledge, with procedures often being based on previous experience and/or similar processes. This involves intuitive knowledge, inductive reasoning and working with non-quantitative information based on heuristic knowledge from operators and engineers. Such information is difficult to represent in terms of mathematical procedures which can be easily translated into computer programs.

It is now possible to make use of computer based techniques based on the concept of an integrated concurrent process engineering environment which seeks to

develop an "intelligent" system to support synthesis of operating procedures. The creation of such a support system requires a qualitative reasoning methodology robust enough to describe complex patterns of process behaviour.

Qualitative reasoning techniques derived from artificial intelligence are available and are well suited to deal with non-numerical information, incomplete knowledge and cognitive tasks and to assist in synthesising operating procedures. However, the existing techniques often generate spurious (non-real) behaviour patterns and need to be extended to describe dynamic behaviour of distributed parameter systems.

This study has concentrated on providing a suitable framework for representing qualitative information and describing dynamic trajectories met with in chemical processes. It is based on a modification of the signed digraph approach (Iri *et al.*, 1979) and includes several new features (*differential nodes*, *temporal edges*, *functional weighting*, a multi-layer representation of the system and a comprehensive qualitative state descriptor). These features allow the description of different functional shapes and complex system behaviour. In particular, it is possible to describe distributed parameter systems without generating spurious solutions. This new approach is referred to as weighted digraph (WDG) and is able to solve the problems not handled by conventional methods. This makes it possible to apply qualitative reasoning to a wide variety of chemical engineering problems.

The weighted digraph approach has been applied to the description of the dynamic trajectories of several chemical processes used as case studies: heat-exchanger, continuous stirred-tank reactor (CSTR) with and without temperature control and a tray distillation column. In all cases it proved to be very effective in describing dynamic patterns of process behaviour. In particular, the problem of the distillation column shows that the methodology is able to deal with the distributed parameter system adequately, by describing the qualitative internal behaviour of the column without missing transient responses or the complex patterns

of internal flows. The CSTR with temperature control has been used to show that the approach is able to deal with oscillations and the complex patterns of behaviour that arise due to the controller.

Although the WDG approach qualitatively represents the general dynamic behaviour of processes quite well, it does have limitations. The procedure is not sensitive enough to distinguish between small variations around a given state. For complex processes, such as distillation, the resulting model structure has many elements, so the visualisation of the flow of information between process variables without the assistance of a computer based reasoning interface is difficult.

It has been shown that weighted digraphs are very effective in synthesis of start-up procedures. This was illustrated by two case studies, which examined the start-up of a network of heat-exchangers and of an integrated system composed of a CSTR and a feed/effluent heat-exchanger. The procedure has the ability to generate feasible operating procedures in the presence of operational constraints by systematically identifying and rejecting paths that lead to violation of the constraints. It is achieved by propagating the effects of valve manipulations through the network of process variables represented in the weighted digraph structures. The approach is also able to identify situations in which the process topology has to be modified in order to make start-up possible. This is relevant to the case of the integrated system for energy recovery, where an auxiliary heater is needed to allow the start-up of the reactor.

Weighted digraph structures are used not only for generating sequences of operations but also for describing the dynamic responses of the system. Qualitative dynamic trajectories provide an insight into the effects of valve manipulations, which allows the assessment of the feasibility of the proposed sequence of operations, helps the understanding of system behaviour and provides a support for reasoning about problems that may arise during start-up.

The approach is well suited to support generation of process plant operating procedures. However, further improvement is still needed to determine the *weights* of the WDG models and the initial qualitative conditions of the process flow diagram. At present, these parameters are determined empirically so may lead to limitations in terms of the description of dynamic trajectories of the start-up of interacting systems.

The importance of the method is due to its ability to represent qualitative information, describe dynamic trajectories of complex processes and formalise the way in which information flows between process variables. These features indicate that it can assist reasoning about behavioural characteristics of processes in a design environment and can be of considerable help when carrying out process analysis and data interpretation, especially in the early stages of process design where most of the data and information are qualitative. It also provides an effective platform for explaining behaviour, so that the underlying physical process can be understood better, and therefore decisions can be made about improvements in process performance, control and generation of operating strategies.

7.2 Suggestions for Future Work

The current work has provided a promising qualitative reasoning methodology that has the ability to describe dynamic trajectories of complex processes and potential in supporting synthesis of operating procedures for a process plant. Because it is a prototype, it needs further development both in terms of theory and applications. In particular, the following need to be addressed in more detail:

- Improvement of the qualitative description of dynamic trajectories by further developing the methodology of *weight* determination, replacing the empirical approach. A methodology for scaling inputs is also needed in order to allow better performance in cases with multiple disturbances.

At present, the relative magnitude of the input variables is only taken into account by the *weights* of the WDG model;

- Explicit consideration of the effects of pressure and mechanical design details of equipment in the weighted digraph models, since it is known that these may affect process behaviour;

- Creation of a computer based reasoning interface to allow the visualisation of the flow of information through the model structure of complex processes and improve the understanding of the interactions between process variables and the way solutions are generated;

- Extend the WDG approach for handling discontinuous transitions, such as the dry-up of a distillation column tray. At present, different models can be used to describe different solution spaces, but the modelling of transitions, or situations that lead to transitions, cannot be handled;

- Start-up of process flow diagrams with process control. Control loops represent a great challenge to qualitative reasoning methodologies, since they usually change the system behaviour to a more complex pattern, because of the increase in interactions between process variables;

- Development of a systematic methodology for determining the initial qualitative states of variables for the simulation of start-up of process flow diagrams, instead of the empirical approach presently used.

Appendix A

Supplementary Information for Numerical Simulations

A.1 Heat-Exchanger

Table A.1 Design conditions and parameters.

Variable/Parameter		Description	
		Hot side	Cold side
Fluid		hot water	cooling water
Flow rate, [m^3/h]		0.833	0.774
Inlet temperature, [K]		363.15	293.15
Pressure, [kPa]		101.325	101.325
Density, [kg/m^3]	in:	973.261	1047.851
	out:	1027.157	993.911
Heat capacity, [kJ/kg K]	in:	3.834	3.517
	out:	3.596	3.737
*U*A*, [kJ/h K]		7754.4	
Tube diameter, [m]		0.025	
Length, [m]		2.0	

Table A.2 Description of the constants used in the mathematical model.

Constant	Description
S_{E1} (hot fluid), S_{E3} (cold fluid)	$\frac{1}{\rho C_p A_{cs} l}$
S_{E2} (hot fluid), S_{E4} (cold fluid)	$\frac{1}{A_{CS} l}$
S_{E5}	$U A$

A.2 Non-isothermal CSTR

Table A.3 Parameter values.

Parameter	Value
λ, [kJ/kgmol A]	69778.9
ρ, [kg/m^3]	800.93
C_p, [kJ/kg K]	3.140
U, [kJ/h K m^2]	3066.22
A, [m^2]	23.23
V_R, [m^3]	1.3592
ρ_j, [kg/m^3]	997.96
C_{pj}, [kJ/kg K]	4.187
V_j, [m^3]	0.109
E/R, [K]	8392.94
k_o, [h^{-1}]	7.0×10^{10}

N.B. Source: Luyben (1990), pp 125.

Table A.4 Inlet conditions.

Variable	Value
F_o, [m^3/h]	1.1327
C_{ao}, [kgmol/m^3]	8.0
T_o, [K]	294.44
T_{jo}, [K]	294.44

N.B. Source: Luyben (1990), pp 125.

Table A.5 Steady-state conditions and parameters for the reactor with temperature control.

Variable	Value
C_{ass}, [kgmol/m^3]	3.9
T_{ss}, [K]	333.33
T_{jss}, [K]	330.33
F_{jss}, [m^3/h]	1.4130
F_{joss}, [m^3/h]	1.4130
T_{sp}, [K]	333.33
K_{ft}, [m^3/h K]	0.2039

N.B. Source: Luyben (1990), pp 125.

A.3 Non-azeotropic Ternary Distillation Column

Table A.6 Design data for the system: n-hexane (1); n-heptane (2); and n-octane (3).

Variable/Parameter	Value
Number of trays	8
Column diameter, [m]	2.134
Weir height, [m]	0.04
Weir length, [m]	0.25543
Hole/tray active area	0.12
Dry hole tray pressure drop, [Pa]	1.0×10^{-6}
Feed tray	6
Murphree tray efficiency	1.0
Reflux flow rate, [mol/s]	40.26
Reboiler duty, [J/s]	$3.59\times10^{+6}$
Feed flow rate, [mol/s]	100.0
Feed temperature, [K]	361.19
Feed pressure, [Pa]	$1.0133\times10^{+5}$
Feed composition: $z_1/z_2/z_3$	0.40/0.30/0.30

A.4 Network of Heat-Exchangers

Table A.7 Design data for heat-exchanger **H1**.

Parameter/Variable		Value/Description	
		Hot side (Str3/10)	Cold side (Str2/5)
Fluid		hot water	cooling water
Flow rate, [m^3/h]		0.833	0.774
Temperature, [K]	in:	363.15	293.15
	out:	313.15	344.36
Pressure, [kPa]		101.325	101.325
Density, [kg/m^3]	in:	973.261	1047.851
	out:	1027.157	993.911
Heat capacity, [kJ/kg K]	in:	3.834	3.517
	out:	3.596	3.737
*U*A*, [kJ/h K]		7754.4	
Tube diameter, [m]		0.025	
Length, [m]		2.0	

Table A.8 Design data for heat-exchanger **H2**.

Parameter/Variable		Value/Description	
		Hot side (Str4/6)	Cold side (Str5/12)
Fluid		hot water	cooling water
Flow rate, [m^3/h]		0.838	0.774
Temperature, [K]	in:	368.15	344.36
	out:	359.54	353.15
Pressure, [kPa]		101.325	101.325
Density, [kg/m^3]	in:	967.678	993.911
	out:	977.269	984.314
Heat capacity, [kJ/kg K]	in:	3.861	3.737
	out:	3.814	3.781
*U*A*, [kJ/h K]		1774.8	
Tube diameter, [m]		0.025	
Length, [m]		2.0	

Table A.9 Design data for heat-exchanger **H3**.

Parameter/Variable		Value/Description	
		Hot side (Str6/11)	Cold side (Str1/9)
Fluid		hot water	cooling water
Flow rate, [m^3/h]		0.838	1.736
Temperature, [K]	in:	359.54	303.15
	out:	338.15	313.27
Pressure, [kPa]		101.325	101.325
Density, [kg/m^3]	in:	977.269	1037.560
	out:	1000.625	1027.031
Heat capacity, [kJ/kg K]	in:	3.814	3.556
	out:	3.707	3.597
$U*A$, [kJ/h K]		1616.4	
Tube diameter, [m]		0.025	
Length, [m]		2.0	

Table A.10 Design data for heat-exchanger **H4**.

Parameter/Variable		Value/Description	
		Hot side (Str7/8)	Cold side (Str9/13)
Fluid		hot water	cooling water
Flow rate, [m^3/h]		2.793	1.736
Temperature, [K]	in:	368.15	313.27
	out:	349.00	343.15
Pressure, [kPa]		101.325	101.325
Density, [kg/m^3]	in:	967.678	1027.031
	out:	988.857	995.222
Heat capacity, [kJ/kg K]	in:	3.861	3.597
	out:	3.760	3.731
$U*A$, [kJ/h K]		6562.8	
Tube diameter, [m]		0.025	
Length, [m]		2.0	

A.5 CSTR and Feed/Effluent Heat-Exchanger System

Table A.11 Design data for the CSTR (R1).

Variable/Parameter	Value/Description	
	Feed (Str2)	Product (Str3)
Flow rate, [m^3/h]	9.137	9.212
Concentration of *A*, [kgmol/m^3]	2.1342	1.3466
Temperature, [K]	297.04	313.33
Flow rate, [kgmol/h]:		
Propylene oxide	19.5005	12.4060
Methanol	36.6055	36.6055
Water	364.1639	357.0695
Propylene glycol		7.0945
λ, [kJ/kgmol *A*]	-92395.52	
ρ, [kg/m^3]	966.4	
C_p, [kJ/kg K]	3.496	
$U*A$, [kJ/h K]	18667.0	
V_R, [m^3]	1.1355	
ρ_j, [kg/m^3]	999.8	
C_{pj}, [kJ/kg K]	3.604	
V_j, [m^3]	0.02	
E/R, [K]	9063.76	
k_o, [h^{-1}]	16.96×10^{12}	

N.B. 1) Reaction:

$$\text{Propylene oxide} + \text{Water} \xrightarrow{H_2SO_4} \text{Propylene glycol}$$

2) Propylene oxide is dissolved in methanol;

3) The reaction is first-order in propylene oxide (*A*) concentration and apparent zero-order in excess of water with the specific reaction rate: $k = k_0\, e^{-E/RT}$, h^{-1};

4) Source: Fogler (1992), pp 400.

Table A.12 Design data for the heat-exchanger (H1).

Parameter/Variable		Value/Description	
		Hot side (Str3/4)	Cold side (Str1/2 or 9)
Fluid		propylene oxide/ methanol/water/ propylene glycol	propylene oxide/ methanol/water
Mole fraction		0.0300/ 0.0886/ 0.8642/ 0.0172	0.0464/ 0.0871/0.8665
Flow rate, [m^3/h]		9.212	9.014
Temperature, [K]	in:	313.33	285.15
	out:	301.79	297.04
Pressure, [kPa]		101.325	101.325
Density, [kg/m^3]	in:	962.462	983.617
	out:	975.412	970.310
Heat capacity, [kJ/kg K]	in:	3.549	3.402
	out:	3.506	3.443
*U*A*, [kJ/h K]		22214.95	
Tube diameter, [m]		0.02	
Length, [m]		1.5	

Table A.13 Design data for the auxiliary heater (H2).

Parameter/Variable		Value/Description	
		Hot side (Str7/8)	Cold side (Str9/2)
Fluid		steam/condensate	propylene oxide/ methanol/water
Mole fraction			0.0464/ 0.0871/0.8665
Flow rate, [m^3/h]		645.0 / 150.5	9.014
Temperature, [K]	in:	393.38	285.15
	out:	393.38	297.04
Pressure, [kPa]		200.00	101.325
Density, [kg/m^3]	in:	1.1177	983.617
	out:	1.1177 / 938.884	970.310
Heat capacity, [kJ/kg K]	in:		3.402
	out:		3.443
Enthalpy of vaporisation, [kJ/kg]		2237.62	
*U*A*, [kJ/h K]		3554.56	
Tube diameter, [m]		0.025	
Length, [m]		1.0	

Bibliography

Andow, P. K. and F. P. Lees (1975). Process Computer Alarm Analysis: Outline of a Method Based on List Processing, *Trans. Inst. Chem. Engrs.*, **53**, 195-208.

Chang, C. C. and Yu, C.-C. (1990). On-line Fault Diagnosis Using the Signed Directed Graph, *Ind. Eng. Chem. Res.*, **29**, 1290-1299.

Crawford, J., A. Farquhar and B. Kuipers (1990). QPC: A Compiler from Physical Models into Qualitative Differential Equations, *Proc. 8th Natl. Conf. on Artif. Intell - AAAI-90*, 365-372.

Crooks, C. A., S. F. Evans and S. Macchietto (1994). An Application of Automated Operating Procedure Synthesis in the Nuclear Industry, *Comp. & Chem. Engng.*, **18, Suppl.**, S385-S389.

Csaki, Zs., K. M. Hangos, L. Hallager and S. B. Jørgensen (1991). Qualitative Simulation for Start-up a Distillation Column, *Proc. IFAC Workshop Comp. Soft. Struc. Integrating AI/KBS Sys. in Proc. Control*, Bergen, Norway, 81- 87.

Dague, P. (1993). Numeric Reasoning with Relative Orders of Magnitude, *Proc. 11th Natl. Conf. on Artif. Intell. - AAAI'93*, Washington, D.C., U.S.A., 541-547.

Dalle Molle, D. (1989). Qualitative Simulation of Dynamic Chemical Processes, PhD Thesis, University of Texas at Austin, Austin.

Dalle Molle, D. and T. F. Edgar (1989). Qualitative Simulation for Process Modeling and Control, *Proc. Amer. Control Conf.*, 1360-1367.

Dalle Molle, D. and T. F. Edgar (1990). Qualitative Modeling of Chemical Reaction Systems. In: *Artificial Intelligence in Process Engineering*, (M. L. Mavrovouniotis, ed.), Academic Press, London, **1**, 1-35.

Dalle Molle, D., B. J. Kuipers and T. F. Edgar (1988). Qualitative Modeling and Simulation of Dynamic Systems, *Comp. & Chem. Engng.*, **12**, 853-866.

de Kleer, J. (1990). Qualitative Physics. In: *Encyclopedia of Artificial Intelligence*, (S. C. Shapiro, ed.), Wiley, New York, **2**, 807-814.

de Kleer, J. and J. S. Brown (1984). A Qualitative Physics Based on Confluences, *Artif. Intell.*, **24**, 7-83.

Doherty, M. F. and J. D. Perkins (1978a). On the Dynamics of Distillation Processes - I: The Simple Distillation of Multicomponent Non-reacting, Homogeneous Liquid Mixtures, *Chem. Eng. Sci.*, **33**, 281-301.

Doherty, M. F. and J. D. Perkins (1978b). On the Dynamics of Distillation Processes - II: The Simple Distillation of Model Solutions, *Chem. Eng. Sci.*, **33**, 569-578.

Doherty, M. F. and J. D. Perkins (1979). On the Dynamics of Distillation Processes - III: The Topological Structure of Ternary Residue Curve Maps, *Chem. Eng. Sci.*, **34**, 1401-1414.

Dohnal, M. (1987). Naive Models in Chemical and System Engineering, *Proc. Chem. Eng. Fund. - CEF'87, The Use of Computers in Chemical Engineering*, Giardini, Italy, 650-654.

Dohnal, M. (1989). Methodology of Qualitative Modelling, *Proc. 2nd Scand. Conf. Artif. Intell. - SCAI'89*, Tampere, 484-500.

Dohnal, M. (1991a). A Methodology for Common-sense Model Development, *Comp. in Ind.*, **16**, 141-158.

Dohnal, M. (1991b). Large Qualitative Models of Complex Chemical and Bioengineering Processes, *Collec. Czech. Chem. Comm.*, **56**, 2107-2141.

Dvorak, D. and B. Kuipers (1989). Model-Based Monitoring of Dynamic Systems, *Proc. 11th Int. Joint Conf. Artif. Intell. - IJCAI'89*, 1238-1243.

Farquhar, A. (1994). A Qualitative Physics Compiler, *Proc. 12th Natl. Conf. Artif. Intell. - AAAI'94*, Seattle, U.S.A., 1168-1174.

Féray-Beaumont, S. E., F. M. R. Corea, M. T. Tham and A. J. Morris (1991). Qualitative Modelling of Distillation Columns, *Proc. IFAC Int. Symp. Adv. Cont. Chem. Proc. - ADCHEM'91*, Toulouse, France, 75-80.

Fogler, H. S. (1992). Elements of Chemical Reaction Engineering, 2nd ed., Prentice-Hall, London.

Forbus, K. D. (1984). Qualitative Process Theory, *Artif. Intell.*, **24**, 85-168.

Forbus, K. D. (1990). The Qualitative Process Engine. In: *Readings in Qualitative Reasoning About Physical Systems*, (D. S. Weld and J. de Kleer, eds.). Morgan Kaufmann, San Mateo, 220-235.

Forbus, K. D. and B. Falkenhainer (1990). Self-Explanatory Simulations: An Integration of Qualitative and Quantitative Knowledge, *Proc. 8th Natl. Conf. on Artif. Intell - AAAI-90*, 380-387.

Forbus, K. D. and B. Falkenhainer (1992). Self-Explanatory Simulations: Scaling up to Large Models, *Proc. 10th Natl. Conf. on Artif. Intell - AAAI-92*, 685-690.

Forbus, K. D. and P. B. Whalley (1994). Using Qualitative Physics to Build Articulate Software for Thermodynamics Education, *Proc. 12th Natl. Conf. on Artif. Intell - AAAI-94*, 1175-1182.

Fusillo, R. H. and G. J. Powers (1987). A Synthesis Method for Chemical Plant Operating Procedures, *Comp. & Chem. Engng.*, **11**, 369-382.

Fusillo, R. H. and G. J. Powers (1988). Operating Procedure Synthesis Using Local Models and Distributed Goals, *Comp. & Chem. Engng.*, **12**, 1023-1034.

Gani, R., C. A. Ruiz and I. T. Cameron (1986). A Generalized Model for Distillation Columns - I: Model Description and Applications, *Comp. & Chem. Engng.*, **10**, 181-198.

Gazi, E., W. D. Seider and L. H. Ungar (1994). Control of Nonlinear Processes Using Qualitative Reasoning, *Comp. & Chem. Engng.*, **18, Suppl.**, S189-S193.

Govind, R. and G. J. Powers (1982). Control System Synthesis Strategies, *AIChE J.*, **28**, 60-73.

Grantham, S.D. (1990). Automated Reasoning about Chemical Plants from First Principles: Application to Troubleshooting and Design, PhD Thesis, University of Pennsylvania, Philadelphia.

Grantham, S. D. and L. H. Ungar (1990). A First Principles Approach to Automated Troubleshooting of Chemical Plants, *Comp. & Chem. Engng.*, **14**, 783-798.

Guimarães, P. R. B. (1992). Artificial Neural Networks for Chemical Process Characterisation, Internal Report, Dept. of Chem. Engng., University of Leeds, Leeds.

Guimarães, P. R. B. and C. McGreavy (1995). Flow of Information Through an Artificial Neural Network, *Comp. & Chem. Engng.*, **19, Suppl.**. S741-S746.

Hangos, K. N. (1991). Qualitative Process Modelling. In: *Chemical Process Control - CPC IV, Proc. 4th Int. Conf. Chem. Proc. Control*, (Y. Arkin and W. H. Ray, eds.), 209-236.

Hangos, K. N., L. Hallager, Zs. Csaki and S. B. Jørgensen (1991). A Qualitative Model for Simulation of the Start-up of a Distillation Column with Energy Feedback. In: *Computer-Oriented Process Engineering*, (L. Puigjaner and A. Espuña, eds.), Elsevier, Amsterdam, 87-92.

Hashimoto, Y., T. Katoh, A. Yoneya, Y. Togari and C. McGreavy (1994). Neural Network Controller Improving its Own Reliability, *Proc. 5th Int. Symp. Proc. Sys. Eng. - PSE'94*, Kyongju, Korea, 1087-1092.

Hashimoto, Y., K. Kawahara, Y. Tanaka, A. Yoneya and Y. Togari (1991). Fault Diagnosis Utilizing a Three-Layer Directed Graph, , *Proc. 4th Int. Symp. Proc. Sys. Eng. - PSE'91*, Montebello, Canada, II.10.1-II.10.7.

Huang, Y. L. and L. T. Fan (1993). A Fuzzy-Logic-Based Approach to Building Efficient Fuzzy Rule-Based Expert Systems, *Comp. & Chem. Engng.*, 17, 181-192.

Hunt, K. J. and D. Sbarbaro (1991). Neural Networks for Nonlinear Internal Model Control, *IEE Proc.-D*, **138**, 431-438.

Hurme, M. (1992). A Review of Qualitative Reasoning Including Applications to Process Engineering and Plant Design, Technical Report, Dept. of Chem. Engng., Helsinki University of Technology, Helsinki.

Hurme, M. and M. Järveläinen (1991). Semiqualitative and Order of Magnitude Reasoning and Optimisation of Chemical Engineering Processes. In: *Computer-Oriented Process Engineering*, (L. Puigjaner and A. Espuña, eds.), Elsevier, Amsterdam, 93-98.

Iri, M., K. Aoki, E. O'Shima and H. Matsuyama (1979). An Algorithm for Diagnosis of System Failures in Chemical Processes, *Comp. & Chem. Engng.*, **3**, 489-493.

Iri, M., K. Aoki, E. O'Shima and H. Matsuyama (1980). A Graphical Approach to the Problem of Locating the Origin of the System Failure, *J. Opers. Res. Soc. Jap.*, **23**, 295-311.

Ivanov, V. A., V. V. Kafarov, V. L. Perov and A. A. Reznichenko (1980). On Algorithmization of the Start-up of Chemical Productions, *Eng. Cyber.*, **18**, 104-110.

Iwasaki, Y. and H. A. Simon (1986). Causality in Device Behaviour, *Artif. Intell.*, **29**, 3-32.

Jiang, Z. (1994). Interactive Graphics-Based Design of Multistage Multicomponent Distillation Processes, PhD Thesis, University of Leeds, Leeds.

Kinoshita, A., T. Umeda and E. O'Shima (1982). An Approach for Determination of Operational Procedure of Chemical Plants, *PSE*, **2**, 114-120.

Kramer, M. A. (1987). Malfunction Diagnosis Using Quantitative Models with Non-Boolean Reasoning in Expert Systems, *AIChE J.*, **33**, 130-140.

Kuipers, B. (1984). Commonsense Reasoning about Causality: Deriving Behavior from Structure, *Artif. Intell.*, **24**, 169-203.

Kuipers, B. (1986). Qualitative Simulation, *Artif. Intell.*, **29**, 289-338.

Kuipers, B. (1987). Abstraction by Time-Scale in Qualitative Simulation, *Proc. 5th Natl. Conf. on Artif. Intell - AAAI-87*, 621-625.

Kuipers, B. (1989). Qualitative Reasoning: Modelling and Simulation with Incomplete Knowledge, *Automatica*, **25**, 571-585.

Kuipers, B. (1992). QSIM and Partial Differential Equations, *personal communication*.

Kuipers, B. and D. Berleant (1988). Using Incomplete Quantitative Knowledge in Qualitative Reasoning, *Proc. 6th Natl. Conf. on Artif. Intell - AAAI-88*, 324-329.

Kuipers, B. and C. Chiu (1987). Taming Intractable Branching in Qualitative Simulation, *Proc. Int. Joint Conf. Artif. Intell. - IJCAI'87*, 1079-1085.

Kuipers, B., C. Chiu, D. T. Dalle Molle and D. R. Throop (1991). Higher-order Derivative Constraints in Qualitative Simulation, *Artif. Intell.*, **51**, 343-379.

Lakshmanan, R. and G. Stephanopoulos (1988a). Synthesis of Operating Procedures for Complete Chemical Plants - I. Hierarchical, Structured Modelling for Nonlinear Planning, *Comp. & Chem. Engng.*, **12**, 985-1002.

Lakshmanan, R. and G. Stephanopoulos (1988b). Synthesis of Operating Procedures for Complete Chemical Plants - II. A Nonlinear Planning Methodology, *Comp. & Chem. Engng.*, **12**, 1003-1021.

Lakshmanan, R. and G. Stephanopoulos (1990). Synthesis of Operating Procedures for Complete Chemical Plants - III. Planning in the Presence of Qualitative, Mixing Constraints, *Comp. & Chem. Engng.*, **14**, 301-317.

Laroche, L., N. Bekiaris, H. W. Andersen and M. Morari (1992). The Curious Behavior of Homogeneous Azeotropic Distillation - Implications for Entrainer Selection, *AIChE J.*, **38**, 1309-1328.

Lee, W. W. and B. Kuipers (1988). Non-Intersection of Trajectories in Qualitative Phase Space: A Global Constraint for Qualitative Simulation. *Proc. 6th Natl. Conf. on Artif. Intell - AAAI-88*, 286- 290.

Lin, L. (1991). Concurrency in Process Engineering Design, PhD Thesis, University of Leeds, Leeds.

Lu, M. L., X. Z. Wang, M. Benayoune, E. K. T. Kam, Y. Hashimoto, S. A. Yang and C. McGreavy (1994). A Computer Integrated Concurrent Process Engineering Environment, *Proc. 5th Int. Symp. Proc. Sys. Eng. - PSE'94*, Kyongju, Korea, 809-815.

Luyben, W. L. (1990). Process Modeling, Simulation and Control for Chemical Engineers, 2nd ed., McGraw-Hill, London.

Maciel, M. R. W. (1989). Interactive Design of Multicomponent Distillation Columns, PhD Thesis, University of Leeds, Leeds.

Mah, R. S. H. (1990). Graphs and Digraphs. In: *Chemical Process Structures and Information Flows*, Butterworths, London, 17-69.

Mavrovounitis, M. L. and G. Stephanopoulos (1987). Reasoning with Orders of Magnitude and Approximate Relations, *Proc. 5th Natl. Conf. on Artif. Intell - AAAI-87*, 626-630.

Mavrovouniotis, M. L. and G. Stephanopoulos (1988). Formal Order-of-Magnitude Reasoning in Process Engineering, *Comp. & Chem. Engng.*, **12**, 867-880.

McGreavy, C. (1985). Prospects for Computer Aided Process Engineers, *I. Chem. E. Symp. Series No. 94*, 353-366.

McGreavy, C., M. L. Lu, X. Z. Wang and E. K. T. Kam (1994). Characterisation of the Behaviour and Product Distribution in Fluid Catalytic Cracking using Artificial Neural Networks, *Chem. Eng. Sci.*, **49,** 4717-4724.

Mohindra, S. and P. A. Clark (1993). A Distributed Fault Diagnosis Method Based on Digraph Models: Steady-State Analysis, *Comp. & Chem. Engng.*, 17, 193-209.

Muratet, G. and P. Bourseau (1993). Artificial Intelligence for Process Engineering - State of the Art, *Comp. & Chem. Engng.*, **17, Suppl.**, S381-S388.

Naka, Y. and C. McGreavy (1994). Modular Approach for Start-up Operational Procedures of Chemical Plant, *Proc. 5th Int. Symp. Proc. Sys. Eng. - PSE'94*, Kyongju, Korea, 1007-1013.

Németh, T., E. I. Varga, K. M. Hangos and S. B. Jørgensen (1992). A Tool for Development of Qualitative and Approximate Quantitative Process Models, *Comp. & Chem. Engng.*, **16, Suppl.**, S491-S498.

O'Shima, E. (1978). Safety Supervision of Valve Operations, *J. Chem. Eng. Japan*, **11**, 390-395.

O'Shima, E. (1983). Computer Aided Plant Operation, *Comp. & Chem. Engng.*, 7, 311-329.

Oyeleye, O. O. and M. A. Kramer (1988). Qualitative Simulation of Chemical Process Systems: Steady-State Analysis, *AIChE J.*, **34**, 1441-1454.

Raiman, O. (1986). Order of Magnitude Reasoning, *Proc. 4th Natl. Conf. on Artif. Intell - AAAI-86*, 100-104

Raiman, O. (1991). Order of Magnitude Reasoning, *Artif. Intell.*, **51**, 11-38.

Rich, S. and V. Venkatasubramanian (1987). Model-Based Reasoning in Diagnostic Expert Systems for Chemical Process Plants, *Comp. & Chem. Engng.*, **11**, 111-122.

Richards, B. L., Kraan, I. and B. Kuipers (1992). Automatic Abduction of Qualitative Models, *Proc. 10th Natl. Conf. Artif. Intell. - AAAI-92*, 723-728.

Rivas, J. R. and D. F. Rudd (1974). Synthesis of Failure-Safe Operations, *AIChE J.*, **20**, 320-325.

Rotstein, G. E., R. Lavie and D. R. Lewin (1992). A Qualitative Process-Oriented Approach for Chemical Plant Operations - The Generation of Feasible Operation Procedures, *Comp. & Chem. Engng.*, **16, Suppl.**, S337-S344.

Rumelhart, D. E. and J. L. McClelland (1986). Parallel Distributed Processing: Explorations in the Microstructure of Cognition, **I & II**, MIT Press, Cambridge.

Savkovic-Stevanovic, J. (1992). A Framework for Qualitative Simulation in Process Engineering, *Comp. & Chem. Engng.*, **16, Suppl.**, S127-S134.

Savkovic-Stevanovic, J. (1995). Qualitative Modeling and Simulation of a Complete Chemical Plant Behaviour, *Acta Chim. Slovenica*, **42**, 63-68.

Scott, D. and F. Crawley (1992). Start up and Shut down of a Plant. In: *Process Plant Design and Operation*, IChemE, Rugby, 78-91.

Shak, K. F. A. (1994). Interactive Design of Non-Ideal Multicomponent Distillation Columns, PhD Thesis, University of Leeds, Leeds.

Shen, Q. and R. R. Leitch (1992). Multiple Models Based on Fuzzy Qualitative Modelling, *IFAC-Artif. Intell. in Real-Time Control*, Delft, The Netherlands, 365-370.

Shiozaki, J., H. Matsuyama, E. O'Shima and M. Iri (1985a). An Improved Algorithm for Diagnosis of System Failures in the Chemical Process, *Comp. & Chem. Engng.*, **9**, 285-293.

Shiozaki, J., H. Matsuyama, K. Tano and E. O'Shima (1985b). Fault Diagnosis of Chemical Processes by the Use of Signed, Directed Graphs. Extension to Five-Range Patterns of Abnormality, *Int. Chem. Eng.*, **25**, 651-659.

Skorstad, G. (1992). Towards a Qualitative Lagrangian Theory of Fluid Flow, *Proc. 10th Natl. Conf. Artif. Intell. - AAAI-92*, 691-696.

Stephanopoulos, G. (1983). Synthesis of Control Systems for Chemical Plants - A Chalenge for Creativity, *Comp. & Chem. Engng.*, **7**, 331-365.

Stephanopoulos, G. (1984). Chemical Process Control - An Introduction to Theory and Practice, Prentice-Hall, London.

Stephanopoulos, G. (1987). The Scope of Artificial Intelligence in Plant-wide Operations. In: *Foundations of Computer-Aided Process Operation - FOCAPO'87*, 505-555.

Stephanopoulos, G. and C. Han (1994). Intelligent Systems in Process Engineering: A Review, *Proc. 5th Int. Symp. Proc. Sys. Eng. - PSE'94*, Kyongju, Korea, 1339-1366.

Tarjan, R. (1972). Depth-first Search and Linear Graph Algorithms, *SIAM J. Comp.*, **1**, 146-159.

Tomita, S., K.-S. Hwang, E. O'Shima and C. McGreavy (1989). Automatic Synthesizer of Operating Procedures for Chemical Plant by Use of Fragmentary Knowledge, *J. Chem. Eng. Japan*, **22**, 364-372.

Tsuge, Y., H. Matsuyama and C. McGreavy (1989). Qualitative Simulator for Estimation of Plant Behaviour During Abnormal Situation, *I. Chem. E. Symp. Series No. 114*, 293-302.

Tsuge, Y., Shiozaki, J., H. Matsuyama and E. O'Shima (1985a). Fault Diagnosis Algorithms Based on the Signed Directed Graph and Its Modifications, *I. Chem. E. Symp. Series No. 92*, 133-144.

Tsuge, Y., Shiozaki, J., H. Matsuyama, E. O'Shima, Y. Iguchi, M. Fuchigami and M. Matsushita (1985b). Feasibility Study of a Fault-Diagnosis System for Chemical Plants, *Int. Chem. Eng.*, **25**, 660-667.

Umeda, T., T. Kuriyama, E. O'Shima and H. Matsuyama (1980). A Graphical Approach to Cause and Effect Analysis of Chemical Processing Systems, *Chem. Eng. Sci.*, **35**, 2379-2388.

Vianna, R. F. (1992). Qualitative Reasoning Methodology for Chemical Processes, Internal Report, Dept. of Chem. Engng., University of Leeds, Leeds.

Vianna, R. F. and C. McGreavy (1995a). Qualitative Modelling of Chemical Processes - A Weighted Digraph (WDG) Approach, *Comp. & Chem. Engng.*, **19, Suppl.**, S375-S380.

Vianna, R. F. and C. McGreavy (1995b). Qualitative Modelling of Distributed Parameters Systems, *Proc. IChemE Research Event*, Edinburgh, UK, 107-109.

Wang, X. Z., M. L. Lu, E. K. T. Kam and C. McGreavy (1994). Concurrent Engineering Application in Fluid Catalytic Cracking Process Design, *Proc. 5th Int. Symp. Proc. Sys. Eng. - PSE'94*, Kyongju, Korea, 381-386.

Wang, X. Z., S. A. Yang, E. Veloso and C. McGreavy (1995). Qualitative Process Modelling - A Fuzzy Signed Directed Graph Method, *Comp. & Chem. Engng.*, **19, Suppl.**, S735-S740.

Weld, D. S. and J. de Kleer (1990). Readings in Qualitative Reasoning About Physical Systems, Morgan Kaufmann, San Mateo.

Wilcox, N. A. and D. M. Himmelblau (1994a). The Possible Cause and Effect Graphs (PCEG) Model for Fault Diagnosis - I. Methodology, *Comp. & Chem. Engng.*, **18**, 103-116.

Wilcox, N. A. and D. M. Himmelblau (1994b). The Possible Cause and Effect Graphs (PCEG) Model for Fault Diagnosis - II. Applications, *Comp. & Chem. Engng.*, **18**, 117-127.

Yip, K. M.-k. (1993). Model Simplification by Asymptotic Order of Magnitude Reasoning, *Proc. 11th Natl. Conf. Artif. Intell. - AAAI'93*, 634-640.

Yu, C.-C. and C. Lee (1991). Fault Diagnosis Based on Qualitative/Quantitative Process Knowledge, *AIChE J.*, **37**, 617-628.