Integration of design and control by nonlinear analysis

Bildea, C. S.

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Integration of Design and Control by Nonlinear Analysis

Costin Sorin Bildea
Integration of Design and Control by Nonlinear Analysis

ACADEMISCH PROEFSCHRIFT

ter verkrijging van de graad van doctor aan de Universiteit van Amsterdam, op gezag van de Rector Magnificus Prof. dr. J.J.M. Franse ten overstaan van een door het college voor promoties ingestelde commissie, in het openbaar te verdedigen in de Aula der Universiteit op 29 maart 2001 te 10.00 uur

door

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geboren te Hunedoara (Roemenie)
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Universiteit van Amsterdam

Cover: The Mandelbrot set, a nonlinear object embedding an infinite number of nonlinear objects.
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Introduction

Main contributions

This thesis brings the following contributions to the field of integration between design and control:

1. Systemic, two-level approach to integrated design and control of chemical processes. This involves: i) design of controllable basic flowsheet structures (BFS), the building blocks of chemical plants, and ii) coupling the BFS in a controllable flowsheet. The approach is compatible with the hierarchical methodology of conceptual design.

2. Guidelines for controllable design of a heat-integrated distillation system with prefractionator, recommending the forward heat-integrated alternative.

3. Nonlinear approach to problems where state multiplicity and instability limits the range of controllable designs. Desirable regions of operation and potential stability or operability problems are found after dividing the space of design parameters in regions with different steady-state and dynamic bifurcation diagrams. The approach is applied to heat-integrated chemical reactors, for which a design methodology is proposed.

4. Steady-state classification of ideal, binary distillation by rigorous application of the singularity theory. In this way, all possible bifurcation diagrams are identified. The effect of physical and design parameters on the location and extent of multiplicity region is presented.

5. Introduction of plant Damkohler number (Da) to study the nonlinear behaviour of the reactor separator-recycle systems. It is demonstrated that, for feasible operation, Da must exceed a critical value $Da^c$, corresponding to a bifurcation point of the mass balance equations. When Da is close to $Da^c$, control structures manipulating reaction conditions are recommended. For large values of Da, reaction conditions may be kept constant.
Motivation

In an early paper, Ziegler and Nichols (1943) remarked that...

"... it is important to realise that controller and process form a unit; credit or discredit for results obtained are attributable to one as much as the other. A poor controller is often able to perform acceptably on a process which is easily controlled. The finest controller made, when applied to a miserably designed process, may not deliver the desired performance."

This thesis addresses the problem of integrating conceptual design and plantwide control. To give a comprehensive presentation, the thesis may be divided into three parts. First, a systemic approach to integration between conceptual design and plantwide control is presented. Afterwards, design of controllable basic flowsheet structures (BFS) is considered. Finally, the nonlinear behaviour of reactor – separator – recycle systems is analysed, in order to account for the effect of BFS coupling on the overall controllability.

Linear approach

The most common approach to integration of process design and control is linear controllability analysis. Because all process controllers seek to invert the process they control, any feature of the process which inhibits this mathematical inversion is a fundamental limitation to control performance. Consequently, the controllability of a process design can be assessed without the need to design the control system and simulate closed-loop dynamic behaviour. Based on this ideas, many linear controllability indicators (Skogestad and Postlethwaite, 1996) have been proposed and employed with good results. In this thesis, this diagnosis-oriented approach is used to analyse the interaction between design and control a heat-integrated distillation system. Several designs alternatives are considered, their controllability properties are analysed and guidelines for controllable design are derived.

Nonlinear approach

Nevertheless, chemical plants are nonlinear systems. The nonlinearity of chemical processes manifests as parametric sensitivity, state multiplicity, instability, or oscillatory behaviour (Seider and Brengel, 1991). These features can be predicted by singularity (Golubitsky and Schaeffer, 1985) and bifurcation (Guckenheimer and Holmes, 1983) theories, which have been extensively applied in the field of chemical reaction engineering. However,
few applications to design are reported (Russo and Bequette, 1995, Heiszwolf and Fortuin, 1997, Khinast et al., 1998). The ideas behind the synthesis-oriented nonlinear approach to integrate design and control are presented below.

Generally, the dependence of system’s state vs. one distinguished operating variable (bifurcation parameter) is presented in bifurcation diagrams. Then, codimension-2 bifurcation varieties are computed. They divide the space of the remaining parameters into regions corresponding to qualitatively-different bifurcation diagrams. This way, desirable regions of operation and potential stability or operability issues are identified. Design alternatives near the bifurcation varieties generally should be avoided. The reason is that the uncertainty of the design parameters and the disturbances affecting the process may shift the operating point to a region where the qualitative behavior is different from the expected one. Then, undesirable phenomena may occur: loss of stability, reaction ignition or extinction, reverse sign of the gain in control loops, etc. Moreover, close to bifurcation points, high sensitivity to disturbances is expected.

In this thesis, design and control of heat-integrated plug-flow reactors, binary distillation, and reactor-separator-recycle systems are considered by a nonlinear approach.

**Thesis overview**

Each chapter is written in the form of an article, including abstract, conclusions, literature references and notation sections. Thus, the chapters may be read independently.

**Chapter 1. Integration of conceptual design and plantwide control**

*Previous work*

In the hierarchical approach to conceptual design (Douglas, 1988), the design process is viewed as a hierarchy of activities in stages of increasing complexity. Because each stage inherits information from its predecessors, the complexity is incremental and can be easily managed. This approach proved successful in a broad range of applications.

The plantwide control problem refers to design of the control loops needed to operate an entire process and achieve its design objectives. However, it is not concerned with tuning and behaviour of all control loops in a chemical plant, but rather with the control philosophy of the overall plant (Skogestad and Larsson, 1998), with emphasis on the structural decisions (i.e. what are the controlled outputs, manipulated inputs, measurements, control configuration and controller type).
Introduction

During the last decade, many articles focussing on plantwide control have been published. A partial list includes Price and Georgakis (1993), Ponton and Liang (1993), Price et. al, (1994), Luyben et al. (1997), Ng and Stephanopoulos (1998), Zheng et al. (1999).

Integration between plantwide control and conceptual design aims towards generation of controllable design alternatives. Nevertheless, the above-cited papers invariably consider fixed flowsheet structure and a fixed operating point. An exception is a study of reactor-separation-recycle processes performed by Lyman et al. (1996). The first step of their method was the identification of the design decisions, which are related to flowsheet configuration, unit sizing, and control structure. The authors recommend to keep the number of design decisions small, a limit of six being suggested. Then, the effect of process design, control structure and controller tuning on the dynamic performance was studied by designed experimentation and extensive dynamic simulation.

Original contribution

However, generating all design alternatives of a chemical process is a very difficult task, because the number of design decisions is very large. Moreover, designed experimentation requires huge modelling and simulation effort. Consequently, a better design methodology incorporating controllability aspects is needed. We remark that the difficulty of the problem stems from the limited information available at the early stages of design, in contrast with the considerable consequences of the decisions taken.

Chapter 1 addresses the problem of integrating conceptual design and plantwide control by a systemic approach. Process plants are represented as subsystems, called basic flowsheet structures (BFS), that interact through material and energy streams. Each BFS has associated control objectives that can be achieved by manipulating local variables. The task of the plantwide control system is to coordinate the BFS, by setting their control objectives. This representation reveals two steps for integrating conceptual design and plantwide control:

1. Design controllable BFS.
2. Couple the basic flowsheet structures in such a way that a controllable system is obtained.

This way, it is possible to consider the plantwide control at a fundamental level of the hierarchical procedure of conceptual design. More specifically, the recycle structure of the flowsheet, the reactor, and the performance of the separation units (product purity or recovery) establish the mass balance. The reactor and separation are assumed to be controllable, but this is set as an explicit task for their design. Plantwide control structures can
be proposed and their performance can be assessed based on steady state sensitivity analysis or on more advanced tools. The methodology, summarised in Table 0.1, was applied to toluene hydrodealkylation (HDA) plant.

**Table 0.1 Summary of integrating conceptual design and plantwide control.**

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Introduction

Chapter 2. Heat-integrated, complex distillation arrangements

A heat-integrated prefractionator / side-stream column configuration for ternary separation was chosen as a case study for the diagnosis-oriented, linear approach to integrate design and control of basic flowsheet structures.

Previous work

Due to the higher cost of energy during the last decade, there is an increased interest in heat-integrated, complex distillation arrangements. These alternatives have, for some range of feed composition, lower energy consumption than the conventional configurations (Petlyuk et. al, 1965, Alatiqi and Luyben, 1985, Doukas and Luyben, 1978a, Cheng and Luyben, 1985). However, the dynamics and control of these systems is as important as energy saving (Frey et. al, 1984, Doukas and Luyben, 1978b, Alatiqi and Luyben, 1986, Ding and Luyben, 1990).

Original contribution

For conventional distillation columns, rules endorsed by industrial experience lead to controllable designs. More decisions, for which no guidelines are available, are involved in the design of complex configurations. They affect the controllability properties, besides energy consumption, total annual cost or other economic index. However, a fixed design was assumed in the previous studies on controllability of heat-integrated complex distillation arrangements. Hence, interaction between design and control was not addressed.

Chapter 2 (Bildea and Dimian, 1999a) investigates the interaction between design and control of a heat-integrated prefractionator / side-stream column configuration for ternary separation (Figure 0.1). For this arrangement, an important design decision refers to the split between the light and heavy components to be performed in the prefractionator. According to this, several designs are possible. They are presented and discussed, for both the forward and reverse heat-integration alternatives. Multi-input multi-output (MIMO) controllability analysis is performed based on linear models.

For the forward heat-integration arrangement, the design with a small prefractionator performs better. It has excellent controllability properties and good composition control can be obtained using only temperature measurements. The reverse heat-integrated alternative is more interactive. In this case, good control can be achieved only if at least one composition analyser is available. Controllability is improved if a sharp split between the light and heavy components is performed in the prefractionator.
Fig. 0.1. Prefractionator / side stream column configuration for ternary separation

Chapters 3-4. Nonlinear approach to design of heat-integrated reactors

Previous work

From the rich literature about the nonlinear behaviour of reacting systems, only a few articles are dedicated to heat-integrated chemical reactors. Lovo and Balakotaiah (1992) computed the uniqueness-multiplicity boundary of the tubular reactor with internal or external heat exchange and CSTR with external heat exchange. For limiting cases, they presented analytical expressions of the ignition, extinction, and cusp points. Subramanian and Balakotaiah (1996) classified the steady-state and dynamic behaviour of several distributed reactor models, including the CSTR with external heat exchange and the tubular reactor with internal heat exchange.

Original contribution

When the work presented in this thesis was started, the author faced the unsuccessful simulations of the toluene hydrodealkylation plant. A close look to the convergence process revealed the cause: the heat-integrated reactor exhibited multiple steady states, and the ignited, middle or extinguished steady state was found, depending on the tear-streams initialisation. Then, the author realised the analogy between the dynamic behaviour of the
plant and converging its steady state simulation by the direct-substitution method. Hence, it became apparent that nonlinearities play an important role in operability. Nevertheless, from the previous studies it was not clear how the results of nonlinear analysis should be applied to design controllable plants. Later, when the nonlinear study of the heat-integrated PFR was completed, a methodology emerged: divide the space of the design parameters into regions with different steady-state and dynamic bifurcation diagrams by computing loci of codimension-2 singular points, and identify desirable regions of operation and potential stability or operability issues. Simultaneously with the work presented in this thesis, Khinast et al. (1998) applied the same methodology for the continuously stirred decanting reactor.

Chapter 3 (Bildea and Dimian, 1998) studies the steady-state and dynamic behaviour of the heat-integrated PFR. (Figure 0.2). A first-order, irreversible, exothermic reaction, and adiabatic reactor operation is considered.

![Diagram of Heat-integrated PFR](image)

**Figure 0.2. Heat-integrated PFR.**

The steady-state and dynamic behaviour is classified by computing the hysteresis, isola, boundary-limit, double-zero and double-Hopf varieties. State multiplicity, isolated solution branches and oscillatory behaviour are possible for realistic values of model parameters. The influence of reaction kinetics and thermodynamics and FEHE efficiency on the extent of multiplicity region is studied. Subsequently, it is discussed how the results can be used to avoid operational problems and a design methodology is proposed. Three reaction systems with different kinetic and thermodynamic characteristics are used as examples.
Chapter 4 (Bildea and Dimian, 1999b, Bildea et al, 2001) analyses a more complex system: first-order, reversible, exothermic reaction, and adiabatic operation in a two-bed tubular reactor. Again, steady-state and dynamic classification is achieved by computing the hysteresis, isola, boundary-limit, double-zero and double-Hopf varieties. State multiplicity, isolated branches and oscillatory behaviour are possible for realistic values of model parameters. Subsequently, it is shown how the results may be used to avoid operational problems.

Chapter 5. Control of heat-integrated reactors

Previous work

Control of heat-integrated reactors was considered mostly by linear techniques. Silverstein and Shinnar (1982) evaluated the stability of a heat-integrated reactor using the frequency response of the individual equipment components. They considered the multiplicity region and concentrated on the intermediate, open-loop unstable operating point. Tyreus and Luyben (1993) analysed a reactor / preheater process. Reactor dynamics (dead-time and inverse response) was captured by a transfer function containing a gain, a positive zero, dead-time and two first-order lags. Luyben et al. (1998) considered the control of the heat-integrated PFR, without steam-generator. Using nonlinear dynamic simulation, they showed that systems with low heat-integration (large furnace, small feed-effluent heat-exchanger) are easier to control.

Original contribution

However, the meaning of “large” or “small” units was unclear and extension to other reaction systems was not obvious, because the results were presented in terms of dimensional variables. Moreover, it was not evident how the designer can find the stabilisability limit in other way than performing extensive dynamic simulation.

Chapter 5 (Bildea et al, 2000a) studies the interaction between design and control of a heat-integrated PFR. A nonlinear, dynamic model is developed. It is shown that two design parameters have to be set during conceptual design: FEHE efficiency and steam-generator duty. Four different design alternatives, for which controllability problems are expected, are investigated. Three different control structures are considered. Linear controllability analysis shows that systems with small steam-generator and large FEHE are difficult to stabilise. Bypass around the FEHE can not be used to reject disturbances and there is no incentive to control both FEHE and furnace outlet temperatures. Next, the nonlinear behaviour of the
Introduction

controlled system is analysed by bifurcation theory. The range of the dimensionless design parameters for which the system cannot be stabilised is detected. The operating points are classified according to their position relative to different bifurcation varieties (Figure 0.3). Doing this, the meaning of "small" or "large" units becomes clear. The results are confirmed by nonlinear dynamic simulation.

Figure 0.3. Phase diagram in the space of design parameters.
Cusp and Hopf-and-pitchfork varieties divide the parameters space into regions with different conversion vs. controller gain bifurcation diagrams.

Chapter 6. Multiple steady states in binary distillation

Previous work

The possibility of state multiplicity and instability in ideal distillation was simultaneously recognised by researchers at University of Trondheim and Technical University of Denmark. Jacobsen and Skogestad (1990, 1991) pointed out the following sources of multiplicity: 1) the nonlinear transformation between mass and molar flow rates, and 2) interaction between flows and composition due to energy balance. Nielsen (1990) presents experimental purity vs. volume reflux rate diagrams, that prove the existence of
multiple steady states. Kienle et al. (1995) and Køggersbol et al. (1996) found experimentally multiple steady states in the methanol-propanol separation. They computed the locus of limit points and presented different bifurcation diagrams.

Original contribution

However, previous studies considered only one column design and only one mixture. Moreover, they did not provide a complete classification of the steady state behaviour.

Chapter 6 (Bildea and Dimian, 1999c) analyses the multiplicity of states in binary distillation by rigorous application of the singularity theory. The mass reflux flow rate is considered as bifurcation parameter. Codimension-2 varieties (Figure 0.4), dividing the feed composition ($z_F$) – boilup ($V$) parameter space into regions with different types of bifurcation diagrams (Figure 0.5), are computed. Finally, the effect of physical and design parameters on the location and extent of the multiplicity regions is investigated.

Figure 0.4. Typical phase diagram for ideal, constant molar overflow, binary distillation. H – hysteresis; BL – boundary limit; DC – double cross; CL – cross-and-limit. Bold line represents the unicity-multiplicity boundary. Different types of bifurcation diagrams existing in regions I - V are presented in Figure 0.5.
Chapter 7. Nonlinear behaviour of reactor - separator - recycle systems

Previous work

The stand-alone reactor was the subject of most applications of nonlinear analysis in chemical reaction engineering. Although the effect of recycling a fraction of the reactor effluent has been occasionally studied (Pareja and Reilly, 1969, Recke and Jørgensen, 1999), the systems considered are not good examples for chemical reactors in recycle plants. Typically, the reactor effluent is processed by the separation section. Hence, the composition and temperature of the recycle stream are different from the reactor effluent. Moreover, often temperature controllers keep constant reaction temperature, or, for adiabatic reactors, constant temperature of the reactor feed.

Original contribution

The following points will argue that the nonlinear behaviour of reactor-separator-recycle systems is relevant for integrating conceptual design and plantwide control:

1. When integrating conceptual design and plantwide control, the need for quantitative information is in contradiction with the requirement of early consideration. These two
items can be reconciled at the "recycle structure of the flowsheet" stage in the hierarchical design methodology. At this point, the reactor is the first unit to be considered in detail because the chemical species present in the reactor effluent determine the separation section. Hence, because reactor modeling, sizing, and control are considered before separation is addressed, the first available quantitative model of the plant consists of black box separation and kinetic reactor, connected by recycle streams.

2. The author considers that the essential task of plantwide control is to maintain the material and energy balance of the whole process, while controlling the condition of the streams leaving the plant (products, by-products, emissions, etc.). In most cases, the energy balance can be controlled easily by diverting any imbalance towards the utility system. Controlling the mass balance is more difficult, because every component (even traces) fed into the plant or formed through a chemical reaction must leave the plant, as a product or through a chemical reaction. These considerations emphasise the important role played by reactor design in controllability properties of the process.

Chapter 7 (Bildea et. al, 2000b) addresses the nonlinear behaviour of certain reactor-separator-recycle systems (Figure 0.6).

**Figure 0.6. General structure of Reactor-Separator-Recycle systems.**
Each general stream (Plant feed; Reactor feed; Reactor effluent / Separation feed; Recycle; Products) represents an arbitrary number of real streams.
The dimensionless mole-balance equations of reactor-separator-recycle system are parameterized by the plant Damkohler number (Da) and the separation specifications. When one reactant is involved, it is demonstrated that reactant accumulation does not occur if $Da > Da^c$, where the critical value $Da^c$ depends on separation performance. For $Da = Da^c$, a transcritical bifurcation of the mole balance equations takes place. For high purity of product and recycle streams, $Da^c = 1$. These conclusions apply to the CSTR, PFR, first-order and n\textsuperscript{th}-order reactions.

Further, when two different control structures are compared, it is shown that their relative performance depends on the design. Close to $Da^c$ (small reactor or slow kinetics), it is necessary to change the reaction conditions when disturbances affect the process.

Finally, two different control structures for a second order reaction are discussed. In these cases, the critical value of the plant Damkohler number corresponds to a fold bifurcation of the mole balance equations.

Reviewing several plantwide control studies that reported bad controllability, it was found that the designs considered are close to the critical value of the plant Damkohler number.

**Literature cited**


Introduction


Introduction


Chapter 1 Systemic Approach to Integrating Conceptual Design and Plantwide Control

Abstract

The problem of integrating conceptual design and plantwide control is addressed by a systemic approach. Process plants are represented as subsystems, called basic flowsheet structures (BFS), that interact through material and energy streams. Each BFS has associated control objectives that can be achieved by manipulating local variables. The task of the plantwide control system is to coordinate the BFS, by setting their control objectives. This representation allows to consider the plantwide control at a fundamental level of the hierarchical procedure of conceptual design. More specifically, when the recycle structure of the flowsheet, the reactor, and the performance of the separation units (product purity or recovery) are specified, the mass balance is established. Then, control structures can be proposed and their performance can be assessed based on steady state sensitivity analysis or on more advanced tools. Toluene hydrodealkylation (HDA) plant is used as a case study.
Chapter 1. Integrating Conceptual Design and Plantwide Control

Introduction

In the traditional approach to process system design, the control engineer is involved only in the late design stages. At this point, as the result of economic optimisation, the flowsheet structure is established, most of the units are sized and the chemical engineer had defined the control objectives. Then, the control engineer applies some methodology to design the control structure. Finally, a nonlinear dynamic model of the plant is used to evaluate the performance of the whole system. If the operability objectives can not be met (the process is not controllable), then the design must be changed. In the fortunate case, changing only the equipment size or operating conditions restores controllability, but very often flowsheet structure modification is needed. In these cases, there is no guarantee that the new design is the optimal one. Moreover, the cost of the design changes quickly increases as the design progresses. Consequently, plantwide control should be considered during the early stages of design.

Luyben et al. (1999) define the plantwide control problem as "how to develop the control loops needed to operate an entire process and achieve its design objectives". One step in their design methodology is inventorying the control degrees of freedom, that is the control valves. Thus, they regard all control objectives as part of the plantwide control problem.

In this article, we consider that distinction should be made between "local" and "plantwide" control objectives. For example, controlling products purity of a distillation column is a local problem. However, the setpoints of the purity control structure can be changed. Thus, the objectives of local control may be manipulated variables of plantwide control structures. This point of view was also expressed by Skogestad and Larsson (1998), who pointed out that plantwide control is not concerned with tuning and behaviour of all control loops in a chemical plant, but rather with the control philosophy of the overall plant.

During the last decade, many articles focussing on plantwide control have been published. A partial list includes Luyben (1993), Price and Georgakis (1993), Ponton and Liang (1993), Downs and Vogel (1993), Lyman and Georgakis (1995), Luyben et al. (1997), Ng and Stephanopoulos (1998), Skogestad and Larsson (1998), Zheng et al. (1999). However, they invariably consider fixed flowsheet structure and a given operating point. Although the interaction between design and control is well recognised, there are few indications about how to design processes having good controllability properties. Lyman et al. (1996) used designed
Chapter 1. Integrating Conceptual Design and Plantwide Control

experimentation and extensive dynamic simulation to study how the process design, control structure and controller tuning affect the dynamic performance of several reactor-separation-recycle processes. The first step of the proposed method is the identification of the design decisions. They are related to flowsheet configuration, unit sizing, and control structure. The authors recommend to keep the number of design decisions small, a limit of six being suggested.

However, during the design of chemical plants, the number of design decisions is much larger. In this case, designed experimentation requires huge modelling and simulation effort. For this reason, a better methodology is needed. Rather than evaluating all possible alternatives, it is desirable to decompose the plantwide control problem into a hierarchy of decisions (Zheng et al., 1999). At each level of hierarchy, alternatives are generated and only economically attractive alternatives are kept for further consideration. This approach is motivated by the hierarchical procedure for conceptual process design (Douglas, 1988), which proved successful in a broad range of applications. A similar approach was used by Fisher et al. (1988), and Ponton and Liang (1993) to analyse the process (functional) controllability or to develop the control system.

This article presents a systemic methodology for integrating plantwide control and hierarchical conceptual design. Two ideas are central to our approach:

1. Process plants are represented as subsystems, called basic flowsheet structures (BFS), that interact through material and energy streams. This representation reveals two steps for integrating conceptual design and plantwide control: a) design controllable BFS and b) couple the BFS in such a way that a controllable system is obtained.

2. The essential task of plantwide control is to coordinate the BFS by setting their (local) control objectives, in order to maintain the material balance of the whole process.

In this article, we argue that controlling the mass balance is a key activity to develop successful plantwide control structures. Thus, any feasible control structure must maintain the material balance of all components, including impurities and traces. Because the chemical reactor is the place where chemical species are formed or destroyed, kinetic reactor modelling is the pre-requisite for quantitative analysis of plantwide control structures. Note that detailed separation modelling is not necessary, as usually we are able to design separation units with good controllability properties. However, analysis of the kinetic reactor – black box
Chapter 1. **Integrating Conceptual Design and Plantwide Control**

separation – recycle system can be used to test the validity of the well-controlled separation hypothesis.

Moreover, it is shown that interaction of the chemical reactor with the separation section through recycles and plantwide control 1) enhances the nonlinearity of the reactor and 2) the genericness of balance equations is lost (from a strictly mathematical point of view). Consequently, even simple reactors (like adiabatic PFR) may exhibit multiple steady states or bifurcations not manifested by the stand-alone reactors.

This article is organised as follows: next section presents a systemic view of integration between conceptual design and plantwide control: chemical plants can be decomposed into several sub-systems interacting through material and energy streams; control objectives assigned to each subsystem are achieved using only local measurements and manipulated variables; the plantwide control coordinates the subsystems, by setting their (local) control objectives. Then, the hierarchical approach to conceptual design is followed. Design decisions are presented briefly. Activities related to design of the plantwide control system and its interaction with conceptual design are discussed. Finally, the methodology is applied to the toluene hydrodealkylation plant (HDA).

**Basic idea of the systemic approach**

The goal of integrated conceptual design and plantwide control is invention of design alternatives with good controllability properties. This is in contrast with the classical approach, where the design alternatives are generated first, and their controllability is evaluated afterwards.

To provide a (partial) answer to this problem, we take a systemic approach. We consider that a process plant consists of several subsystems interconnected through material and energy streams (Figure 1.1). The subsystems will be called *basic flowsheet structures* (BFS), defined as parts of the plant for which (local) control objectives are assigned and can be achieved using only manipulated variables that are interior to the BFS. Unit operations are the simplest BFS. Often, some units interact so strongly that they must be treated as one entity. Examples are heat-integrated reactors, complex distillation arrangements, azeotropic distillation with solvent recycle, etc. Delimiting the BFS depends on what design / modelling details are available and how the control objectives are assigned. However, we consider that
Chapter 1. Integrating Conceptual Design and Plantwide Control

the basic flowsheet structures can be identified based on engineering judgement, and do not address this problem here.

Figure 1.1. Systemic representation of chemical plants.

The plant is decomposed into basic flowsheet structures, that interact through material and energy streams.

The task of plantwide control is to harmonise the BFS in such a way that the whole system operates in a required manner. This is achieved by changing the control objectives of the BFS. The controllability of the BFS is a necessary (but not sufficient) condition for the controllability of the entire plant. Consequently, our approach to integrate design and control consists of two steps:

1. Design basic flowsheet structures with good controllability properties. This is possible for unit operations, where a lot of industrial experience exists. However, it is an open field of research for more complex sub-systems. Chapters 2 – 6 of this thesis deal with design and control of some BFS (heat-integrated distillation, heat-integrated reactors, distillation).
2. Couple the BFS in such a way that a controllable system is obtained. Interaction between the chemical reactor and the separation section, due to material recycle and plantwide control, is the subject of Chapter 7 in this thesis.

**Integrating conceptual design and plantwide control**

In the following, the hierarchical approach to conceptual design will be followed, and control-related design activities will be discussed. Table 1.1 presents a summary of the proposed approach to integrating conceptual design and plantwide control.

**Initial data**

The initial design data concerns reactions chemistry, raw materials, production rate, product specifications and economic constraints. Control-related data must specify the required production range, product grades, variability of the raw material.

**Input-output**

Typical design decisions taken at this level are related to feed purification, gas recycle and purge, and removal or recycle of reversible by-products. They establish the number and composition of product streams.

Ponton and Liang (1993) affirm that throughput manipulation can be decided now. They propose several alternatives:

1. Limiting reactant on flow control, inventory control in the direction of flow. This is the most appealing alternative, which works in many situations. However, when the limiting reactant is recycled, the reactor must convert the entire amount fed in the process. Failing this (due to large feed, small reactor volume, or slow kinetics), the reactant will accumulate in the system.

2. Product on flow control, inventory control in the direction opposite to flow. There are many circumstances in which this is not applicable. For example, if the product leaves the plant as a distillate, then the column feed should control the level in the condenser drum. Such control scheme is unusual for distillation columns.

3. A control loop manipulating limiting reactant to control production rate. This strategy may work for rather simple processes, but it will fail for large plants, because of the significant lag between feed and product streams.

Price and Georgakis (1993) pointed an additional alternative:
4. Use of an internal throughput manipulator (as reaction volume, temperature or pressure).

It should be noted that, in recycle systems, the production rate is strictly related to the reactant feed flow rate. For this reason, the feed stream cannot be on flow control, but must be used to keep constant reactant inventory.

Among the previously discussed alternatives, the first and the last ones are the most attractive. Their relative performance depends mainly on the reaction kinetics, reactor size and the desired production rate. However, because at this level the reactor is not designed, the quantitative information available does not suffice to take any plantwide control decision.

Recycle structure of the flowsheet

At this level, the number of reaction systems and recycle streams is established. Additional units (as compressor) may be also introduced. Separation sections are specified as product recovery or purity. Flow rate and composition of the streams are expressed in terms of several design variables, as reaction conversion, recycle flow rate or composition, etc. In the hierarchical procedure, the values of the design variables are found at the next level, when the cost of separation can be calculated.

Now, plantwide control can be considered.

First, several candidate controlled variables may be identified. They are not assigned to a particular basic flowsheet structure, hence they have a true plantwide character. Examples are production rate, recycle composition, ratio between reactants at reactor inlet, etc.

Secondly, candidate manipulated inputs can be identified. When they are flow rates connecting BFS, the choice affect the control of upstream and downstream BFS. As an example, it was proposed (Luyben et al., 1997) to keep reactant recycle on flow control and to change the setpoint of this loop when production changes are required. This implies that the inventory control of the upstream unit is in direction opposite to flow, while the inventory control of the downstream unit is in the direction of flow. Manipulated flows should be chosen with care in order to avoid over-specification with respect to plant mass balance. Ponton and Liang (1993) present a method to address this problem. Its application is not difficult, because the plant representation is simple and number of streams to be considered is small. A second category of plantwide manipulated variables is the setpoints of the BFS control. Examples are reaction conversion, separation performance, etc.

The choice of the controlled and manipulated variables can be based on engineering judgement or on systematic methods. We recommend to consider the material balance. This

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gives valuable insight into the number of control degrees of freedom, functional controllability, I/O pairing for decentralized control. As an example, when two reactants are recycled, the mass balance equations reveal that the recycle flow rate of the non-limiting component is not determined. Hence, it should be on flow control.

The proposed control structures may be analysed by steady state simulation. In commercial simulators, unit operation input data or flow rate and composition of feed streams can be varied in order to meet some “design specifications”. Although the design specifications are apparently similar to SISO feedback controllers, it does not mean that the final control structure has to be decentralized or the I/O pairing must be preserved. This approach is advantageous because the nonlinear character of the process is intact. Sensitivity analysis shows what plantwide manipulated inputs (setpoints of BFS control) are necessary. Variability of the streams connecting BFS indicate what disturbances must be rejected by BFS. This way, the designer can identify control objectives of the BFS, and set them as explicit targets for BFS design.

Besides sensitivity, other nonlinearities deserve a special discussion. In the field of chemical reaction engineering, state multiplicity, instability and oscillatory behaviour are well-known phenomena (Seider and Brengel, 1991). They can be analysed using bifurcation / singularity theories. Typically, bifurcation diagrams are used to represent system’s state vs. one distinguished operating parameter (called bifurcation variable), while the other operating parameters are constant. Thus, the bifurcation diagram can be regarded as a cross-section of the multi-dimensional state vs. parameters diagram, that is parallel to the bifurcation variable and orthogonal to the others parameters. The situation is different when a plantwide control system is in place. In this case, changing only the bifurcation variable is impossible, because the plantwide control system fulfils its objectives by changing additional operating parameters. Hence, the bifurcation diagrams are now cross-sections orthogonal to the control objectives. This can lead to state multiplicity and instability even if the uncontrolled system has a unique, stable state.

Bifurcation theory demonstrates that the number of steady states can change at fold, transcritical or pitchfork bifurcation points. These points are one-codimensional, that is the value of one parameter (the bifurcation parameter) is fixed. Fold points are generic for small changes of the additional parameters. In the general case (systems without special properties), the transcritical and pitchfork bifurcations are not generic, that is they disappear for small
Chapter 1. Integrating Conceptual Design and Plantwide Control

changes of the additional parameters. However, the plantwide control system achieves its objectives by changing the additional parameters. This brings special symmetry properties, the result being the genericness of the transcritical and pitchfork bifurcations.

Separation system

Because of problem complexity, the separation system is usually decomposed into sub-systems, as vapour, liquid and solid separation. For each sub-system, there are systematic procedures to generate design alternatives.

For many separation operations, there exist procedures, endorsed by industrial experience, that lead to designs that are economically optimal and have good controllability properties. However, there are also many cases where design procedures / guidelines are still needed. In Chapter 2 of this thesis, a heat-integrated complex distillation arrangement will be considered.

Heat integration

Pinch-analysis can be used to develop heat-integration schemes. However, although arrangements obtained by heat-integration are optimal from the viewpoint of investment and utility consumption, they may be barely operable. Reactor effluent travelling through the reboilers of several distillation columns and then pre-heating the reactor feed is unlikely to be found in any real plant.

We adopt the position that the heat-integration should be performed locally. When energy can not be recovered by integration in one plant section, it should be used to generate steam or power, and exported this way to other sections. In any case, the designer should check that heat-integration does not damage the process controllability.
Chapter 1. Integrating Conceptual Design and Plantwide Control

Table 1.1 Summary of integrating conceptual design and plantwide control.

<table>
<thead>
<tr>
<th>Conceptual design</th>
<th>Local control</th>
<th>Plantwide control</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>0. Initial information</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Chemistry</td>
<td>-</td>
<td>Production range and grades</td>
</tr>
<tr>
<td>Raw materials</td>
<td></td>
<td>Raw-materials variability</td>
</tr>
<tr>
<td>Product specification</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Economic constraints</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>1. Input-output</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Feed streams and purification.</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>2. Recycle structure</strong></td>
<td>Reactor control</td>
<td>Identify plantwide controlled and manipulated variables. Propose control structures, based on simplified material balance. Behaviour of the reactor-separator-recycle system: set constraints for the design variables; identify disturbances affecting the separation section.</td>
</tr>
<tr>
<td>Recycle streams.</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Material balance with recycles.</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Reactor design.</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>3. Separation system</strong></td>
<td>Control of the separation units</td>
<td>Evaluate control structures for economic optimum operating point.</td>
</tr>
<tr>
<td>General structure.</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Vapour recovery and gas separations. Solid recovery and separations. Liquid separation. Design variables by optimisation.</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>4. Energy integration</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Heat exchanger network.</td>
<td>Ensure that controllability is preserved.</td>
<td>local heat-integration and decoupling through the utility system improves controllability</td>
</tr>
<tr>
<td><strong>5. Implementation of process control</strong></td>
<td>Dynamic modelling, controller tuning, and dynamic simulation.</td>
<td></td>
</tr>
</tbody>
</table>
Case study: HDA plant

The toluene hydrodealkylation (HDA) plant has been used intensively as a reference case study both for process synthesis (Douglas, 1988) and plantwide control (Fisher et al., 1988, Ponton and Liang 1993, Luyben and Tyreus, 1997, Ng and Stephanopoulos, 1996). Several control configurations have been proposed, but they differ in the choice of the controlled outputs and manipulated inputs or they do not agree on the input-output pairing. Moreover, no design details, nor dynamic simulation results have been reported.

Initial data

The nominal production rate is 120 kmol/h benzene at a purity exceeding 99.95%. The feed streams are pure toluene, and technical-grade hydrogen. Production flexibility of ±25% is required. The following vapour-phase reactions are considered:

\[ C_6H_5 - CH_3 + H_2 \rightarrow C_6H_6 + CH_4 \]
\[ 2C_6H_6 \leftrightarrow C_{12}H_{10} + H_2 \]

The reactor is adiabatic. The reaction temperature must be kept below 980 K to prevent hydro-cracking reactions. The hydrogen / aromatics ratio at the reactor inlet must exceed 5/1 to minimize coking. For the same reason, the reactor effluent must be quenched rapidly to 900 K.

Input-output

At this level, the design decisions establish the number and composition of product streams. The components in the reactor effluent are ordered by their boiling point, and assigned to product (benzene, diphenyl) or recycle streams (toluene, hydrogen). Separation of the gaseous components (hydrogen, methane) seems difficult. For these reason, when the two components are recycled together, a purge stream is necessary to avoid methane buildup. The input-output structure of the HDA plant is presented in Figure 1.2.
Chapter 1. Integrating Conceptual Design and Plantwide Control

Recycle structure of the flowsheet

The difference of the boiling points of hydrogen and methane, and benzene, toluene and diphenyl is very large. Consequently, it is easy to separate the gaseous and liquid components from the reactor effluent by a simple flash. Then, two recycles are identified:

- Gas recycle. The overhead of the flash contains unreacted hydrogen and methane. A compressor is used to recycle it to the reaction section. A purge stream is necessary to prevent methane build-up.

- Liquid recycle. Separation of the benzene / toluene / diphenyl mixture by distillation is easy. Traces of methane that remain dissolved after gas-liquid separation can be also removed. Although recycling the diphenyl is a design option (because it is the product of a reversible reaction), its removal accounts for other heavy by-products.

At this level, the flowsheet is completed by inclusion of the adiabatic tubular reactor, and the furnace needed to bring the reactants to reaction temperature. The recycle structure of the HDA plant is presented in Figure 1.3.
Figure 1.3. Recycle structure of the HDA plant

Control objectives are attached to various streams. Potential manipulated variables are presented in *italics*.

To perform the mass balance, a simple plant model is considered: perfect separation in all units and negligible side reactions. The following mass-balance equations can be derived:

$$F_B = F_1$$  \hspace{1cm} (1.1)

$$y_{HP} = y_{H2} \cdot \frac{F_1}{F_2} = y_{H2} \cdot \frac{F_1}{F_p}$$  \hspace{1cm} (1.2)

$$\frac{y_{HR}}{y_{T3}} = \frac{X}{F_1} \cdot (F_R \cdot y_{H2} + F_2 \cdot y_{H0})$$  \hspace{1cm} (1.3)

$$F_T = F_1 \cdot \frac{1 - X}{X}$$  \hspace{1cm} (1.4)

$$F_p = F_2$$  \hspace{1cm} (1.5)

$F_j$ and $y_{kj}$ denote the flowrate of stream $j$ and the molar fraction of the $k$ component (H - hydrogen, T - toluene) in the stream $j$, respectively; $X$ is the reaction conversion.

If production rate and hydrogen/toluene ratio at the reactor inlet ($y_{H2}/y_{T3}$) are defined, Eqs. 1.1 to 1.5 have two degrees of freedom. Hence, the values of two design variables (not
any combination is feasible) must be specified. Douglas (1988) considers reactor conversion and purge composition as design variables and optimises the economic potential of the plant. We use his results \( (X = 0.78, Y_{RF} = 0.424) \) as a starting point in this study. Table 1.2 presents the flow rate, temperature, pressure and composition of various streams in the HDA plant. Although some variables were computed after sizing the flash and the distillation columns, approximate values can be assumed now and are presented here.

### Table 1.2. HDA plant: Mass and heat balance results.
The complete flowsheet is presented in Figure 1.10.

<table>
<thead>
<tr>
<th>Stream</th>
<th>Flow rate (kmol/h)</th>
<th>Temperature (°C)</th>
<th>Pressure (bar)</th>
<th>Toluene</th>
<th>Hydrogen</th>
<th>Benzene</th>
<th>Methane</th>
<th>Diphenyl</th>
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</thead>
<tbody>
<tr>
<td>1</td>
<td>125</td>
<td>50</td>
<td>1.2</td>
<td>1</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>2</td>
<td>229.5</td>
<td>38</td>
<td>37.5</td>
<td>0</td>
<td>0.95</td>
<td>0</td>
<td>0.05</td>
<td>0</td>
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<tr>
<td>8</td>
<td>157.1</td>
<td>199.9</td>
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<tr>
<td>9</td>
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<td>1.4</td>
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<td>0</td>
<td>0.770</td>
<td>0</td>
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<tr>
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<td>36.1</td>
<td>125.6</td>
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<td>0.963</td>
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<td>118.3</td>
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<td>0</td>
<td>170 PPM</td>
<td>0</td>
<td>0</td>
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<td>34</td>
<td>0</td>
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<tr>
<td>R</td>
<td>1350</td>
<td>20.8</td>
<td>34.5</td>
<td>0</td>
<td>0.424</td>
<td>0.004</td>
<td>0.572</td>
<td>0</td>
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<td>B</td>
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<td>85.8</td>
<td>1.2</td>
<td>500 PPM</td>
<td>0</td>
<td>0.9995</td>
<td>0</td>
<td>0</td>
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<td>D</td>
<td>1.6</td>
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<td>1.3</td>
<td>0.155</td>
<td>0</td>
<td>0</td>
<td>0</td>
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</tr>
<tr>
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<td>31.4</td>
<td>12</td>
<td>0</td>
<td>0.079</td>
<td>0.016</td>
<td>0.905</td>
<td>0</td>
</tr>
<tr>
<td>P</td>
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<td>34</td>
<td>0</td>
<td>0.424</td>
<td>0.004</td>
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</tr>
<tr>
<td>R1</td>
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<td>0.455</td>
<td>0.003</td>
<td>0.450</td>
<td>0</td>
</tr>
<tr>
<td>R2</td>
<td>1739</td>
<td>636.8</td>
<td>34.5</td>
<td>0.092</td>
<td>0.455</td>
<td>0.003</td>
<td>0.450</td>
<td>0</td>
</tr>
<tr>
<td>R3</td>
<td>1739</td>
<td>688.2</td>
<td>34</td>
<td>0.020</td>
<td>0.384</td>
<td>0.073</td>
<td>0.526</td>
<td>0.0008</td>
</tr>
<tr>
<td>R4</td>
<td>1803.9</td>
<td>620.0</td>
<td>34</td>
<td>0.027</td>
<td>0.370</td>
<td>0.097</td>
<td>0.504</td>
<td>0.001</td>
</tr>
<tr>
<td>R5</td>
<td>1803.9</td>
<td>550.0</td>
<td>34</td>
<td>0.027</td>
<td>0.370</td>
<td>0.097</td>
<td>0.504</td>
<td>0.001</td>
</tr>
<tr>
<td>R6</td>
<td>1803.9</td>
<td>111.4</td>
<td>34</td>
<td>0.027</td>
<td>0.370</td>
<td>0.097</td>
<td>0.504</td>
<td>0.001</td>
</tr>
</tbody>
</table>
Chapter 1. Integrating Conceptual Design and Plantwide Control

At this point, control issues can be considered. There are five potential controlled variables: production ($F_B$), hydrogen / toluene ratio ($y_{\text{H2}}/y_{\text{T}}$), pressure, purge composition ($y_{\text{HP}}$), and conversion ($X$). However, some of them might be left uncontrolled. For example, conversion control is difficult, because of the dead-time associated with the plug-flow reactor. Moreover, it requires composition analyser, which is expensive, require maintenance, and has unfavourable dynamics. For these reasons, it would be desirable to develop a control structure in which controlling reactor inlet temperature ensures almost constant reaction conversion.

The following manipulated variables are available: toluene feed ($F_1$), hydrogen feed ($F_2$), gas recycle ($F_R$), purge ($F_P$) and furnace duty ($q_B$). When furnace duty is used as manipulated variable, it controls the reactor inlet temperature. The setpoint of this control loop may be used, in a cascade manner, to control one of the previous variables.

Next, the reactor can sized. For the required conversion, mass balance equations 1.1-1.5 are solved, and the flow rate and composition of the reactor inlet is found (results for $X=0.78$ are presented in Table 1.2). Then, the value of the reactor inlet temperature is chosen, and the adiabatic PFR equations are solved. In this study, the following kinetics was used:

$$r_i = k_i \cdot c_T \cdot c_H^{0.5}$$

$$k_i = 5.943 \times 10^{14} \cdot \exp \left( -\frac{34138 \text{ K}}{T} \right) \quad (\text{m}^3/\text{kmol})^{1/2} \cdot \text{s}^{-1} \quad (1.6)$$

Figure 1.4 presents the conversion vs. reactor volume when the reactor inlet temperature was set to 910 K. It reveals that small reactors (say $V < 30 \text{ m}^3$) lead to unfeasible designs, because the mole balance equations do not have a feasible solution ($0 < X < 1$). Moderate conversion can be obtained, but for the same reactor volume high conversion is also possible. It should be remarked that the behaviour of the HDA reactor placed in the recycle system is very different from the behaviour of the stand-alone reactor, which can exhibit only a single, stable steady state. The state multiplicity identified here, which was not noticed by other researchers who studied the HDA plant, has deep implications on control. The unusual behaviour on the intermediate branch, where larger reactor gives smaller conversion, indicates that this steady state is unstable. Hence, any control structure that keeps constant reactor inlet temperature, reactor inlet hydrogen/toluene ratio, and purge composition, is unstable. When disturbances affect the process, small deviations from the steady state are unavoidable. Let us consider a conversion increase. Then, the recycle toluene flow rate decreases. The control structure adjusts the hydrogen feed and gas recycle in order to keep constant hydrogen/toluene
ratio and purge composition, leading to smaller reactor inlet flow rate, larger residence time, and further increase of conversion.

\[
\begin{align*}
F_1 &= 120 \text{ kmol/h} \\
y_{H_2} &= 0.95 \\
y_{HP} &= 0.4 \\
y_{H_3}/y_{T_3} &= 4.5 \\
y_{H_3}/y_{T_3} &= 5
\end{align*}
\]

**Figure 1.4. Conversion vs. reactor volume, for fixed purge composition and hydrogen / toluene ratio.**

The HDA plant exhibits state multiplicity. Moderate conversion can be obtained only at unstable steady state.

It should be remarked that it is difficult to manipulate the gas recycle flow rate using the compressor. Moreover, hydrogen loss is minimised when the flow rate of the gas recycle is at its maximum value. Because operating the compressor at a flow rate smaller than its maximum capacity requires recycling compressed gas back to the suction (Shinskey, 1988), there is no benefit in terms of energy saving. Since the compressor is already installed, the investment cost is not affected. For these reasons, only control structures keeping constant gas recycle will be considered further.

Inspection of the mass balance equations gives more insight into control issues:

- The only way in which fed toluene can leave the system is as benzene (Eq. 1.1). Hence, any strategy that will try to set the production rate without modifying the toluene feed will fail.
- Eq. 1.2 shows that the purge composition can be controlled using the hydrogen feed (\(F_2\)) or purge flow rate (\(F_p\)).
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- The pressure is constant if the gas holdup remains constant. Because the number of moles of gaseous components does not change in the reaction, hydrogen feed and purge flow rates must be equal (Eq. 1.5). Consequently, assigning one of them for purge composition control leaves the other one for pressure control.

- The remaining manipulated variable (furnace duty) can be used to control either the conversion or the hydrogen / toluene ratio. Both controlled variables seem to be important. Low conversion means more utility consumption for separation, while high conversion affects reaction selectivity. Low hydrogen / toluene ratio leads also to undesirable by-products.

This way, a large number of control structures have been excluded from further analysis. Table 1.3 presents five control structures that were considered, denoted CS1 to CS5.

<table>
<thead>
<tr>
<th>Control structure</th>
<th>Hydrogen/Toluene ratio</th>
<th>Purge composition</th>
<th>Conversion</th>
</tr>
</thead>
<tbody>
<tr>
<td>CS1</td>
<td>Hydrogen feed</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>CS2</td>
<td>Hydrogen feed</td>
<td>-</td>
<td>Setpoint of the furnace duty-reactor inlet temperature loop</td>
</tr>
<tr>
<td>CS3</td>
<td>-</td>
<td>Hydrogen feed</td>
<td>-</td>
</tr>
<tr>
<td>CS4</td>
<td>-</td>
<td>Hydrogen feed</td>
<td>Setpoint of the furnace duty-reactor inlet temperature loop</td>
</tr>
<tr>
<td>CS5</td>
<td>Setpoint of the furnace duty-reactor inlet temperature loop</td>
<td>Hydrogen feed</td>
<td>-</td>
</tr>
</tbody>
</table>

In all control structures, gas recycle flow rate is constant; toluene feed is used to set the production; purge flow rate is used to keep constant pressure.

The control structures were evaluated by steady state sensitivity analysis using ASPEN PLUS®, where SISO control loops can be simulated by design-specification blocks. Figure 1.5 presents the most important results.

- Without control, hydrogen / toluene ratio deviates considerably from the admissible range (Figure 1.5a). Controlling the purge composition (CS3, CS4) reduces the variability of the hydrogen / toluene ratio, but it is still low at high production rates. If this variable is left uncontrolled, the plant must be overdesigned, in order to meet the specification $\gamma_{\text{H}_2}/\gamma_{\text{T_3}} = 5$ under the worst-disturbance conditions.
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- For all control structures, the hydrogen lost with the purge is proportional with the production (Figure 1.5 b). When purge composition is controlled (CS3, CS4 and CS5), the hydrogen loss is less sensitive to the production change.
- The conversion (Figure 1.5c) and reactor outlet temperature (Figure 1.5d) strongly depend on the production when both \( \gamma_{\text{H}}/\gamma_{\text{T3}} \) and \( \gamma_{\text{H}} \) are controlled (CS5). This may affect negatively reaction selectivity.
- For the design considered (conversion=0.78), CS5 requires high flexibility of the separation section (Figure 1.5e). At the lowest production, the liquid separation section handles 90 kmol/h benzene, 55 kmol/h toluene and small amounts of diphenyl. At the highest production, the mixture consists of 150 kmol/h benzene, 15 kmol/h toluene and small amounts of diphenyl. However, the variability of the stream entering the separation section can be reduced if the system is designed for lower conversion, for example X=0.5, as illustrated in Figure 1.5f.

It may be concluded that, for the design considered, there is no incentive to control the reactor conversion. Either the purge composition or the hydrogen / toluene ratio (but not both!) should be controlled. In the first case, the whole plant must be overdesigned to work at hydrogen / toluene ratio higher than necessary. In the second case, increased hydrogen loss at high production must be accepted. To control both variables, the plant must be designed for a lower conversion, with a higher load of the liquid separation section. Taking into account these results, control structure CS1 presents a minimum of disadvantages and will be further considered.

The operating point used during the previous sensitivity analysis was found by Douglas (1988) after the liquid separation had been considered. However, one could arrive at control structure CS1 based only on the mass balance equations and the fact that conversion control is difficult. In this case, the requirement of good plantwide controllability is a constraint for further design optimisation. To illustrate this idea, the dependence of the conversion versus reactor volume is presented in Figure 1.6, for fixed values of the gas recycle flow rate and hydrogen / toluene ratio.
Figure 1.5. HDA plant. Control structure evaluation by steady state simulation
Control structures 1-5 are presented in Table 1. There is no incentive to control reactor conversion. For the design considered, control of both hydrogen/toluene ratio and purge composition requires high flexibility of the separation section.
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Figure 1.6. Conversion vs. reactor volume, for fixed values of the gas recycle flow rate and hydrogen / toluene ratio.

For small gas recycle, the operating point is stable only at high conversion.

Again, the system exhibits state multiplicity. The turning points of the volume-conversion diagrams represent the stability limit. Hence, it is possible to have moderate conversion at a stable operating point only if the gas recycle is large enough. When the operating point is chosen by optimisation, the locus of the turning point must be considered as a feasibility limit.

It should be remarked that designs near the turning points are dangerous. Consider the stable operating point $X=0.55$, $F_R=700 \text{ kmol/h}$. The reactor volume can be calculated, but the kinetic data is always uncertain. When the reaction rate is over-estimated, the real operating point falls at the left of the volume-conversion line, in the region where no feasible operating point exists. Then, although the control system does a perfect job, toluene accumulation occurs, leading to infinite recycle.

Results in Figure 1.6 assumed that all the unreacted toluene is recovered and recycled. The effect of losing a small fraction $(1-\alpha)$ of toluene is presented in Figure 1.7. For $\alpha > 0.9$, state multiplicity is preserved.
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Figure 1.7. Conversion vs. reactor volume, for fixed values of the gas recycle flow rate and hydrogen / toluene ratio, when a fraction $1-\alpha$ of the unreacted toluene is lost.

Separation system

The synthesis of the liquid separation section (Figure 1.7) is easy. A stabilizer is necessary to remove methane traces. Then, the benzene – toluene – diphenyl mixture is separated in a light-first, two-columns sequence. Experience-endorsed guidelines are available to design the distillation columns: calculate the minimum reflux and the minimum number of trays by the Underwood-Fenske method, set the number of trays $N=2N_{\min}$ and the reflux ratio $R=1.2R_{\min}$, then use rigorous simulation to adjust $N$, $R$, and feed tray location, until the desired purity is achieved. The results are presented in Table 1.4.

Control of the distillation column is also easy. It should be remarked that the range of feed disturbances affecting the separation section was obtained as a result of the sensitivity analysis performed at the previous design level. This way, disturbance rejection can be set as a distinct design specification.
Chapter 1. Integrating Conceptual Design and Plantwide Control

**Table 1.4. Design results for the liquid separation section.**

<table>
<thead>
<tr>
<th>Distillation columns</th>
<th>STABIL</th>
<th>PRODCOL</th>
<th>RECCOL</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of stages (including condenser and reboiler)</td>
<td>7</td>
<td>28</td>
<td>6</td>
</tr>
<tr>
<td>Feed tray</td>
<td>2</td>
<td>14</td>
<td>4</td>
</tr>
<tr>
<td>Distillate vapour fraction</td>
<td>1</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Distillate flow (kmol/h)</td>
<td>8.925</td>
<td>120.98</td>
<td>-</td>
</tr>
<tr>
<td>Bottoms flow (kmol/h)</td>
<td>-</td>
<td>-</td>
<td>1.583</td>
</tr>
<tr>
<td>Reflux ratio</td>
<td>0.75</td>
<td>1.8</td>
<td>0.6</td>
</tr>
<tr>
<td>Condenser pressure (bar)</td>
<td>12</td>
<td>1.2</td>
<td>1.2</td>
</tr>
<tr>
<td>Reboiler pressure (bar)</td>
<td>12.45</td>
<td>1.4</td>
<td>1.3</td>
</tr>
<tr>
<td>Column diameter (m)</td>
<td>0.894</td>
<td>1.574</td>
<td>0.634</td>
</tr>
</tbody>
</table>

**Heat-integration**

In the HDA plant, energy recovery is possible by using the reactor effluent to preheat the reactor feed, in a feed-effluent heat exchanger (FEHE). Apparently, there is no need for a furnace. However, the furnace is required for start-up, and the purge is available as fuel. When the furnace is included and a FEHE of reasonable size is used, there is heat excess that
must be removed. The exergetic advantageous solution is to place a steam generator before the FEHE. The resulting heat-integration arrangement is presented in Figure 1.8.

![Diagram of a heat-integrated reactor](image)

**Figure 1.8. Heat-integrated HDA reactor.**

During conceptual design, furnace and steam generator duties, and FEHE efficiency are of interest. Because one heat-balance constraint, two design decisions should be taken, for example FEHE efficiency and steam generator duty. They can be found optimising according to some economic criteria. However, it is known that auto-thermal reactors have complex, nonlinear behaviour, including state multiplicity, isolated solution branches and sustained oscillations. Indeed, a plot of the reactor conversion versus dimensionless furnace duty (Figure 1.9) reveals state multiplicity, with the required operating point on the middle, unstable branch. Consequently, the design and control of the heat-integrated PFR should be analysed in more detail.
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In order to assess the performance of the plantwide control structure, we developed a SPEEDUP® model, including reactor dynamics and rigorous distillation, in which all control loops were implemented (Figure 1.10). The following scenario was used to evaluate the performance of the control system: Initially, set the production rate to 120 kmol/h. After 5 hours, increase to 150 kmol/h. Later, reduce in two steps (at time 15, and 25 hours) to 90 kmol/h.

Figure 1.11. present the results of dynamic simulation. Production change can be easily achieved. The product purity is held on specification. Reaction selectivity remains high. The constraint related to hydrogen / toluene ratio is satisfied most of the time.
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Figure 1.10. Plantwide control of the HDA plant
Figure 1.11. Dynamic simulation results.
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Conclusions

- The systemic approach reveals two steps for integration of conceptual design and plantwide control:
  1. Design controllable basic flowsheet structures.
  2. Couple the basic flowsheet structures in such a way that a controllable system is obtained.
- Controlling the mass balance is a key activity to develop successful plantwide control structures. Any feasible control structure must maintain the material balance for all components, including impurities.
- In the systemic approach, the effect of plantwide disturbances on the locally controlled basic flowsheet structures can be assessed by nonlinear sensitivity analysis. Because it depends on the plantwide control structure, this has to be specified before completing the design.
- Disturbance rejection can be set as a distinct specification for BFS design. If the target of controllable BFS appears unattainable, design modifications should be considered.
- The reactor placed in recycle systems and the stand-alone reactor have very different behaviour. The nonlinear behaviour of reactor – separator – recycle systems should be further investigated.

Literature cited


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Chapter 2
Interaction between Design and Control of a Heat-Integrated Distillation System with Prefractionator

Abstract

The relationship between the design and the control of a heat-integrated distillation set-up, consisting of a prefractionator and a side stream main column, is analysed. The separation of a pentane-hexane-heptane mixture with moderate purity requirements is considered. Both forward and reverse heat-integration schemes are investigated. Different designs are possible, depending on the light/heavy split in the prefractionator and the heat-integration scheme. They are similar with respect to energy consumption, but very different with respect to dynamic behaviour. The differences are studied using frequency-dependent controllability analysis, as well as carrying out closed-loop simulation, in the presence of large feed composition disturbances. Thus, it is found that the forward heat-integration is much easier to control, and with only temperature measurements. The low-cost design with a small prefractionator has the best closed-loop performance. The reverse heat-integration can be controlled only if a composition analyser of the side stream is available; sharp light/heavy split in the prefractionator gives better disturbance rejection. The superior dynamic behaviour of the forward heat-integration scheme was confirmed by the study of a high purity separation of the benzene-toluene-xylene mixture.
Chapter 2. Design and Control of a Distillation System with Prefractionator

**Introduction**

The direct and indirect sequences are well-known conventional configurations for separation of ternary mixtures by distillation. Due to the higher cost of energy during the last decade, there is an increased interest in other alternatives. They include heat integration (Rathore et al., 1974, Morari and Faith, 1980), thermal coupling (Petlyuk et al., 1965, Tedder and Ruud, 1985, Alatiqi and Luyben, 1985, Nikolaides and Malone, 1988), and complex configurations (Doukas and Luyben, 1978, Cheng and Luyben, 1985). These alternatives have, for some range of feed composition, lower energy consumption than the conventional configurations. As an example (Cheng and Luyben, 1985), for a 25/50/25 benzene – toluene – xylene mixture, the prefractionator / side-stream column (Figure 2.1) with reverse heat integration and low operating pressure consumed 45% less energy than the direct sequence.

![Diagram](attachment:image.png)

**Figure 2.1. Prefractionator / side stream column configuration for ternary separation.**

The dynamics and control of these systems is as important as energy saving (Tyreus and Luyben, 1978, Frey et al., 1984, Chiang and Luyben, 1985, Mizsey et al., 1998, Doukas and Luyben, 1978, Alatiqi and Luyben, 1986). Ding and Luyben (1990) studied the controllability of the prefractionator / side stream column with reverse heat integration. The control structure used concentration measurements and single-input single-output (SISO) loops, including split-range control. For the low purity separation, the system was found controllable. For the high-purity case, the system could handle only small disturbances.
However, they did not study the controllability of the forward heat-integration arrangement. Moreover, the relationships between the design and controllability properties were not analysed.

The fact that the design of a process determines its controllability is well recognised (Luyben and Luyben, 1997). However, no applications to complex distillation configurations are reported. In the previous studies, heuristic rules (endorsed by industrial experience) were applied to obtain an approximate design, which was later refined by rigorous methods. Afterwards, the controllability analysis was performed.

For conventional distillation columns, there are few design decisions to be taken and the design is almost established once the product purity is specified. However, more decisions (for which no guidelines are available) are involved in the design of complex configurations. They will affect the controllability properties, besides energy consumption, total annual cost or other economic index.

This work investigates the interaction between design and control of the heat integrated prefractionator / side-stream column configuration for ternary separation. For this arrangement, an important design decision refers to the split between the light and heavy components to be performed in the prefractionator. According to this, several designs are possible. They are presented and discussed, for both the forward and reverse heat-integration arrangements. Multi-input multi-output (MIMO) controllability analysis is performed based on linear models. For the particular mixture considered in this work and forward heat-integration, the design with a small prefractionator performs better. It has excellent controllability properties and good composition control can be obtained using only temperature measurements. The reverse heat-integrated alternative is more interactive. In this case, good control can be achieved only if at least one composition analyser is available. Controllability is improved if a sharp split between the light and heavy components is performed in the prefractionator. Finally, the performance of the proposed control structures is evaluated by dynamic simulation.

Throughout this work, A, B and C will denote the light, intermediate and heavy components, respectively. For the particular separation investigated, they are pentane, hexane and heptane, respectively.
Design of the prefractionator / side stream column configuration

The prefractionator / side stream column configuration is presented in Figure 2.1. Typically, the design specifications are expressed as the purity of the product streams. Design decisions refer to the range of operating pressures and the quality of the separation to be performed in the prefractionator.

For each column, a pressure profile, which is related to the heat-integration scheme, is assumed. For forward heat-integration, heat transfer is possible when the distillate of the prefractionator is hotter than the bottom of the second column. This can be achieved if the pressure in the prefractionator is high enough. If reverse heat-integration is of interest, the second column is the one to be operated at high pressure. Several design iterations may be necessary because the pressure profile depends on the design of the columns.

To design the prefractionator, the fractional recoveries of the light and heavy components, $F_{AB} \cdot z_{ABA}/F_{ABC \cdot AB CA}$ and $F_{BC} \cdot z_{BCC}/F_{ABC \cdot ABC C}$, respectively, are specified (the first and second subscripts denotes the stream and the component, respectively). A binary A/C separation is considered and the Underwood-Fenske method is used to find the minimum reflux and minimum number of trays. Then the reflux ratio and the number of trays are chosen (for example $1.2 \cdot R_{\text{min}}$ and $2 \cdot N_{\text{min}}$). This gives an approximate design, due to the presence of the intermediate component. Consequently, a rigorous method is used to adjust the product and reflux flow rates until the desired A/C separation is achieved. At the same time, the top and bottom concentrations of the intermediate component are obtained.

The main column may be treated as two pseudo-conventional sections. The mass balance equations give the flow rates of the product streams. The controlling feed (the one requiring the largest reflux, Nikolaides and Malone, 1987) is found and the reflux ratio is set accordingly. Then, an approximate design is found by a short-cut method. Finally, a rigorous method is used to refine the number of trays, feed and side stream locations.

Depending on the specified recoveries for the light and heavy components in the prefractionator, different designs are arrived at. Consider a 150 mol/s equimolar ABC feed. Let the desired purity of the A, B and C products be 0.99, 0.98 and 0.99, respectively. Assume that the B product should contain equal amounts of A and C, i.e. 0.5 mol/s. Then, the whole amount of A (or C) that leaves the prefractionator with the bottom (or top) stream will end in the side stream of the main column. Consequently, the maximum amount of A (or C) in the prefractionator bottom (top) is 0.5 mol/s.
To design the prefractionator, several specifications are possible. Table 2.1 displays four design alternatives. In the first one, a sharp A/C separation is performed in the prefractionator. Alternatively (Design II), the quantities of light in bottoms and heavy in distillate are close to their maximum allowed values, and sharp A/B and B/C split is required in the main column. Other options investigated are: (III) high recovery of the heavy component, low recovery of the light component and (IV) low recovery of the heavy component, high recovery of the light component.

The alternatives have different operating parameters (reflux, condenser and reboiler duty, etc.) and columns' size (number of trays, diameter). In addition, different controllability properties are expected.

**Heat integration considerations**

The forward heat-integration will be discussed. The same considerations apply to the reverse heat-integration.

Usually, the heat-integration is attempted by combining in one heat exchanger the condenser of the first column and the reboiler of the second column. In most of the cases, the duties do not match. First solution is to install auxiliary equipment. If there is a heat surplus ($Q_{c1} > Q_{w2}$), a condenser is added to the first column. If there is a heat deficiency ($Q_{c1} < Q_{w2}$), a reboiler is added to the second column. In both cases, the capital cost is increased because additional equipment, but one variable (duty) is preserved to be manipulated in the control structure.
An alternative solution is to change the columns' design to match the duties. For heat surplus, the second column reflux may be increased and the number of trays decreased. This requires higher reboiler duty. Because the additional duty is attained by heat integration, there is no penalty in terms of energy consumption. Note that the option of decreasing $Q_{c1}$ may not work due to the minimum-reflux constraint. If there is heat deficiency, $Q_{c1}$ may be modified by increasing the reflux in the first column. The number of trays is decreased, but the reboiler duty ($Q_{w1}$) must be increased. Because this does not come by heat integration, there is a penalty in terms of energy consumption. As in the previous case, we remark that decreasing $Q_{c2}$ may not always work.

The systems investigated in this work are based on the perfect-match alternative. This way one manipulated variable (the duty of the additional condenser or reboiler) is missing. The inclusion of the auxiliary heat exchanger should lead to better controllability. However, the improvement might be minimal because only small duty change will be available.

Additional energy can be recovered if various hot streams are used to preheat the prefractionator or main column feed streams. Because this article concentrates on the controllability analysis, we included in our flowsheet only heat integration between the feed and product streams.

**Design results**

The general design specifications were:

a) Feed: 41.66 mol/s, equimolar pentane / hexane / heptane mixture, at 25 °C and 1 bar.

b) Product specification: pentane, 99%; hexane, 98%, impurified with equal amounts of pentane and heptane; heptane, 99%; 25 °C and 1 bar.

The following assumptions were made during the design: ideal vapour-liquid equilibrium; 100% tray efficiency; saturated liquid reflux; columns feed: saturated liquid, 0.1 bar pressure difference between the feed and the feed tray; 10 °C pinch temperature difference for feed preheating; 10 °C temperature difference in heat-integrated reboiler/condenser; pressure drops: 0.01 bar/tray, 0.05 bar in condenser; residence times of 5 min in reflux drums and 10 min in reboilers; heat transfer coefficients of 500 W/(m²K) in the heat exchangers.
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Figure 2.2. Prefractionator / side stream column with forward heat integration.

Figure 2.3. Prefractionator / side stream column with reverse heat integration.
Chapter 2. *Design and Control of a Distillation System with Prefractionator*

Steady state design was performed in ASPEX PLUS® (ASPEX Technology Inc, 1997a), using RADFRAC™ models for the distillation columns. When pinch analysis was applied, the feed preheating layout presented in Figures 2.2 and 2.3 was obtained. Table 2.2 summarises the results of column sizing. Table 2.3 presents the flow rate and composition of intermediate streams.

### Table 2.2. Results of the design of prefractionator / side stream column.
Numbers in bold represent heat supplied / removed by heat integration

<table>
<thead>
<tr>
<th></th>
<th>Design I</th>
<th>Design II</th>
<th>Design III</th>
<th>Design IV</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>PF</td>
<td>C</td>
<td>PF</td>
<td>C</td>
</tr>
<tr>
<td><strong>Forward heat integration</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>reflux ratio</td>
<td>2.007</td>
<td>2.393</td>
<td>1.958</td>
<td>2.355</td>
</tr>
<tr>
<td>total stages</td>
<td>24</td>
<td>28</td>
<td>12</td>
<td>30</td>
</tr>
<tr>
<td>feed tray (from top)</td>
<td>12</td>
<td>6, 20</td>
<td>6</td>
<td>6, 23</td>
</tr>
<tr>
<td>side-stream tray</td>
<td>12</td>
<td>12</td>
<td>12</td>
<td>12</td>
</tr>
<tr>
<td>top stage pressure (bar)</td>
<td>7.3</td>
<td>7.1</td>
<td>7.4</td>
<td>1</td>
</tr>
<tr>
<td>reboiler duty, $10^6$ W</td>
<td>1.323</td>
<td><strong>1.223</strong></td>
<td>1.337</td>
<td><strong>1.242</strong></td>
</tr>
<tr>
<td>condenser duty, $10^6$ W</td>
<td><strong>1.223</strong></td>
<td>1.223</td>
<td><strong>1.242</strong></td>
<td><strong>1.257</strong></td>
</tr>
<tr>
<td>feed preheat duty, $10^6$ W</td>
<td>0.529</td>
<td>0.514</td>
<td>0.573</td>
<td>0.502</td>
</tr>
<tr>
<td>total duty, $10^6$ W</td>
<td>1.852</td>
<td>1.851</td>
<td>1.909</td>
<td>1.674</td>
</tr>
</tbody>
</table>

|                      |          |           |            |           |          |           |          |           |
| **Reverse heat integration** |          |           |            |           |          |           |          |           |
| reflux ratio         | 0.891    | 2.588     | 0.980      | 2.568     | 1.016    | 2.568     | 0.828    | 2.5       |
| total stages         | 20       | 34        | 10         | 35        | 15       | 35        | 15       | 35        |
| feed tray (from top) | 10       | 7, 25     | 5          | 6, 26     | 9        | 6,25      | 6        | 7,26      |
| side-stream tray     | 15       | 15        | 15         | 15        |          |           |          |           |
| top stage pressure (bar) | 1        | 5.95      | 1          | 5.4       | 1        | 5.6       | 1        | 5.75      |
| reboiler duty, $10^6$ W | **1.059** | 1.812     | **1.072**  | 1.793     | **1.065** | 1.794     | **1.041** | 1.790     |
| condenser duty, $10^6$ W | 0.998    | **1.059** | 1.030      | **1.072** | 1.010    | **1.065** | 0.992    | **1.041** |
| total duty, $10^6$ W  | 1.812    | 1.793     | 1.794      | 1.794     |          |           |          |           |
Table 2.3. Flow rate and composition of the streams leaving the prefractionator.

<table>
<thead>
<tr>
<th></th>
<th>Design I</th>
<th>Design II</th>
<th>Design III</th>
<th>Design IV</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Forward heat integration</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Distillate (mol/s)</td>
<td>18.47</td>
<td>18.76</td>
<td>18.01</td>
<td>20.33</td>
</tr>
<tr>
<td>Pentane (%)</td>
<td>75.16</td>
<td>73.27</td>
<td>75.72</td>
<td>68.28</td>
</tr>
<tr>
<td>Hexane (%)</td>
<td>24.81</td>
<td>25.91</td>
<td>24.21</td>
<td>31.17</td>
</tr>
<tr>
<td>Heptane (%)</td>
<td>0.03</td>
<td>0.82</td>
<td>0.07</td>
<td>0.55</td>
</tr>
<tr>
<td>Bottoms (mol/s)</td>
<td>23.19</td>
<td>22.91</td>
<td>23.66</td>
<td>21.33</td>
</tr>
<tr>
<td>Pentane (%)</td>
<td>0.02</td>
<td>0.62</td>
<td>1.07</td>
<td>0.03</td>
</tr>
<tr>
<td>Hexane (%)</td>
<td>40.11</td>
<td>39.41</td>
<td>40.28</td>
<td>35.39</td>
</tr>
<tr>
<td>Heptane (%)</td>
<td>59.86</td>
<td>59.97</td>
<td>58.65</td>
<td>64.58</td>
</tr>
<tr>
<td><strong>Reverse heat integration</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Distillate (mol/s)</td>
<td>18.89</td>
<td>18.54</td>
<td>18.06</td>
<td>19.31</td>
</tr>
<tr>
<td>Pentane (%)</td>
<td>73.50</td>
<td>73.95</td>
<td>76.31</td>
<td>71.88</td>
</tr>
<tr>
<td>Hexane (%)</td>
<td>26.46</td>
<td>25.22</td>
<td>23.46</td>
<td>27.43</td>
</tr>
<tr>
<td>Heptane (%)</td>
<td>0.04</td>
<td>0.83</td>
<td>0.05</td>
<td>0.69</td>
</tr>
<tr>
<td>Bottoms (mol/s)</td>
<td>22.78</td>
<td>23.13</td>
<td>23.61</td>
<td>22.36</td>
</tr>
<tr>
<td>Pentane (%)</td>
<td>0.03</td>
<td>0.77</td>
<td>0.47</td>
<td>0.05</td>
</tr>
<tr>
<td>Hexane (%)</td>
<td>39.03</td>
<td>39.84</td>
<td>40.74</td>
<td>38.43</td>
</tr>
<tr>
<td>Heptane (%)</td>
<td>60.94</td>
<td>59.39</td>
<td>58.79</td>
<td>61.52</td>
</tr>
</tbody>
</table>

Controllability analysis

The main control objective is to maintain the products' concentration at their setpoints. The composition of the feed stream is considered as disturbance. Because distributed control systems implementing PID controllers are widespread in the chemical industry, decentralised control is of interest. In addition, an attempt is made to develop the control system using temperature measurements to infer the concentrations because composition analysers are expensive, require maintenance and have unfavourable dynamics. The temperature-control trays were selected to be close enough to product withdrawal, but still sensitive. Typical temperature profiles in the side stream column are presented in Figure 2.4.
The performance of the control system can be improved if concentration controllers are added. They may give, in a cascade manner, the setpoint of the temperature control loops. Therefore, the analysis will try to assess both disturbance rejection and set point tracking properties of different system designs. To accomplish this task, the Closed Loop Disturbance Gain (CLDG) and the Performance Relative Gain Array (PRGA), respectively, will be calculated. For an excellent presentation of the controllability analysis tools, we refer to the book of Skogestad and Postlethwaite (1996).
Chapter 2.  Design and Control of a Distillation System with Prefractionator

The controllability properties of Design I and III (and Design II and IV) are similar, for both forward and reverse heat integration schemes. For this reason, the results for Design I and II will be presented comparatively.

SPEEDUP® (ASPEN Technology Inc, 1997b) was used to simulate the dynamic behaviour of the different configurations. In all cases, levels were controlled by P-only algorithm. The maximum allowed control error ($\varepsilon_{\text{max}}$) and the maximum control action ($u_{\text{max}}$) were chosen as 50% of the steady state value of the controlled and manipulated variables, respectively. The gain of the controllers was set to $K_p = u_{\text{max}} / \varepsilon_{\text{max}}$. For pressure controllers, the maximum allowed control error was set to 0.1 bar and integral action ($T_i=0.2$ h) was used. Small adjustments of one of the operating parameters (reboiler duty) were necessary to get the same stationary state as the one obtained by steady state simulation in ASPEN PLUS®. (The differences may be due to the local thermodynamic model used by SPEEDUP®).

SPEEDUP® allows two different types of dynamic simulation. In pressure-driven simulations, the pressure-drop across the valves determines the flow rates. In the flow-driven case, the flow rates are specified. The latter alternative assumes fast flow controllers and was used in this study.

The state space linear model of the process was obtained using the CDI (Control Design Interface) facility offered by SPEEDUP®:

$$\frac{dx(t)}{dt} = A \cdot x(t) + B \cdot u(t) + B_d \cdot d(t)$$
$$y(t) = C \cdot x(t) + D \cdot u(t) + D_d \cdot d(t)$$
$$e(t) = r(t) - y(t)$$

(2.1)

$x$, $u$, $d$, $y$, $e$ and $r$ are the vectors of the state variables, manipulated inputs, disturbances, controlled outputs, control errors and setpoints, respectively. $A$, $B$, $B_d$, $C$, $D$ and $D_d$ are matrices of appropriate dimensions. In order to obtain meaningful controllability results, the inputs, disturbances and outputs were scaled. In terms of scaled variables, the control objective is to keep $|e(t)| < 1$, using $|u(t)| < 1$, when disturbances $|d(t)| < 1$ affect the process.

In terms of transfer functions, the linear model of the process is given by:

$$y(s) = G(s) \cdot u(s) + G_d(s) \cdot d(s)$$

(2.2)

where:

$$G(s) = C \cdot (sI - A)^{-1} \cdot B + D$$

(2.3a)
$G_d(s) = C \cdot (sI - A)^{-1} \cdot B_d + D_d \quad (2.3b)$

Linear models were used only for controllability analysis. The full nonlinear model was used to evaluate the performance of the control system.

**Prefractionator / side stream column with forward integration**

It is necessary to control four concentrations in the product streams: $B$ in top, $A$ and $C$ in side stream, and $B$ in bottom. If inferential control is used, it is necessary to control four temperatures in the main column, corresponding to the four sections. Additionally, large amounts of light/heavy component going in the bottom/distillate of the prefractionator makes the separation in main column very difficult. Consequently, it may be desirable to control the composition of the streams leaving the prefractionator, controlling two temperatures in the stripping and rectifying sections.

However, after closing the inventory control loops, only four manipulated variables are left: prefractionator reflux flow rate and reboiler duty, main column reflux and side draw flow rate. Moreover, dynamic considerations discourage the use of prefractionator manipulated variables to control temperatures in main column.

It was found that acceptable control could be achieved when only one temperature is controlled in the prefractionator (Figure 2.5). Because the vapour dynamics are faster than the liquid dynamics, the reflux ratio is kept constant and one temperature in the prefractionator bottom is controlled by the reboiler duty. This loop gets the setpoint from a controller regulating the temperature in the bottom of the main column.

The main column reflux and side draw flow rates are used to control temperature in the top and between the side stream and the second feed, respectively.

Table 2.4 gives the nominal values and scaling factors for the manipulated inputs, controlled outputs and disturbances. The maximum change of the set point in temperature control loops was assumed to be 5 °C.

When decentralised control is of interest, the input-output pairing may be evaluated using the Relative Gain Array, defined as:

$$\Lambda = G \otimes (G^{-1})^T \quad (2.4)$$
Chapter 2. Design and Control of a Distillation System with Prefractionator

Figure 2.5. Forward heat-integration. Control structure.
Standard notation is used. YC denotes ratio controller.

Table 2.4. Control of prefractionator / side stream column with forward heat integration. Nominal values and scaling factors of outputs, inputs and disturbances.

<table>
<thead>
<tr>
<th>Variable</th>
<th>Nominal value</th>
<th>Scaling factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>y₁ (PF stage 22 temperature, °C)</td>
<td>167.3</td>
<td>1</td>
</tr>
<tr>
<td>y₂ (C stage 4 temperature, °C)</td>
<td>40.3</td>
<td>0.5</td>
</tr>
<tr>
<td>y₃ (C stage 17 temperature, °C)</td>
<td>81.8</td>
<td>0.5</td>
</tr>
<tr>
<td>u₁ (PF reboiler duty, 10⁶ W)</td>
<td>1.337</td>
<td>0.674</td>
</tr>
<tr>
<td>u₂ (C reflux flow rate, mol/s)</td>
<td>33.22</td>
<td>16.66</td>
</tr>
<tr>
<td>u₃ (C side draw flow rate, mol/s)</td>
<td>13.89</td>
<td>6.94</td>
</tr>
<tr>
<td>Design II</td>
<td></td>
<td></td>
</tr>
<tr>
<td>y₁ (PF stage 10 temperature, °C)</td>
<td>161.2</td>
<td>1</td>
</tr>
<tr>
<td>y₂ (C stage 4 temperature, °C)</td>
<td>40.7</td>
<td>0.5</td>
</tr>
<tr>
<td>y₃ (C stage 17 temperature, °C)</td>
<td>78.6</td>
<td>0.5</td>
</tr>
<tr>
<td>u₁ (PF reboiler duty, 10⁶ W)</td>
<td>1.337</td>
<td>0.674</td>
</tr>
<tr>
<td>u₂ (C reflux flow rate, mol/s)</td>
<td>32.69</td>
<td>16.66</td>
</tr>
<tr>
<td>u₃ (C side draw flow rate, mol/s)</td>
<td>13.89</td>
<td>6.94</td>
</tr>
<tr>
<td>Disturbances</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Light in feed</td>
<td>0.333</td>
<td>0.1</td>
</tr>
<tr>
<td>Intermediate in feed</td>
<td>0.333</td>
<td>0.1</td>
</tr>
<tr>
<td>Heavy in feed</td>
<td>0.333</td>
<td>0.1</td>
</tr>
</tbody>
</table>
For both designs analysed, the chosen pairing corresponds to positive diagonal elements in the RGA matrix.

\[
\Lambda_1(0) = \begin{bmatrix}
1.483 & -0.492 & 0.009 \\
0.019 & 1.090 & -0.109 \\
-0.502 & 0.402 & 1.100 \\
\end{bmatrix}
\]

\[
\Lambda_2(0) = \begin{bmatrix}
2.308 & -1.326 & 0.19 \\
0.082 & 1.155 & -0.163 \\
-1.316 & 1.171 & 1.145 \\
\end{bmatrix}
\]

Close-to-one diagonal elements denote little interaction. Small values in the RGA matrix also indicate that model uncertainty is not a problem.

The RGA_number, defined as:

\[
RGA\_number = \| I - \Lambda(j\omega) \|_{\text{num}}
\]  

has small value and drops to zero for high frequency (Figure 2.6), showing that good control performance is possible.

**Figure 2.6. Forward heat-integration. RGA_number.**

The RGA_number has small values and drops to zero at high frequencies. Design I is less interactive.
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Design I is (slightly) less interactive. However, because interactions can help the disturbance rejection, it cannot be concluded that it is also better.

To analyse the disturbance rejection properties, the Closed Loop Disturbance Gain (CLDG) is calculated:

$$\tilde{G}_d = \tilde{G} \cdot G^{-1} \cdot G$$  \hspace{1cm} (2.6)

where $\tilde{G}$ is a matrix consisting of diagonal elements of $G$.

Its elements, $\tilde{g}_{ik}$, give the apparent gain of the $k^{th}$ disturbance on the $i^{th}$ output under decentralised control. The necessary condition to avoid inputs constraints is:

$$\left| \tilde{g}_{ik} \right| > \left| \tilde{g}_{ik} \right| \quad \forall k$$ \hspace{1cm} (2.7)

Although in both cases the inputs are powerful enough to reject disturbances (Figure 2.7), the second design is clearly better.

The ability of the system to follow set point changes can be analysed using the Performance Relative Gain Array:

$$\Gamma = \tilde{G} \cdot G^{-1}$$  \hspace{1cm} (2.8)

Its elements, $\gamma_{ij}$, show how the $i^{th}$ manipulated input must change when the $j^{th}$ setpoint is changed. More precisely, input saturation is not a problem if:

$$\left| \tilde{g}_{ii} \right| > \left| \gamma_{ij} \right| \cdot \left| R_k \right| \quad \forall k$$ \hspace{1cm} (2.9)

where $R_k$ is the scaling factor of the $k^{th}$ setpoint.

Figure 2.8 shows the frequency dependent PRGA for the second design (the results for the first design are similar). To allow comparison with the loop gain, they are multiplied by the scaling factors for setpoint changes.

The following conclusions can be drawn:

- Fast change of the prefractionator bottom temperature ($y_1$) cannot be achieved with the available manipulated inputs (at high frequency, condition 2.9 is not satisfied for $k=1$).
- Input saturation may occur in the third loop (at high frequency, condition 2.9 is not satisfied for $i=3$).

However, there are few negative implications on the controllability properties. If temperatures are used to infer concentration, fast setpoint tracking is not a concern. When composition controllers provide the setpoint for the temperature control loops, the system will
be almost all the time close to the desired operating point; hence, only small adjustments of the temperature setpoints will be necessary.

**Figure 2.7.** Forward heat-integration. Frequency dependent loop gain, $g_u$ (♦) and CLDG elements, $\tilde{g}_{dk}$ (■ $k=1$, ▲ $k=2$, × $k=3$).

Design II has better disturbance rejection properties.
Figure 2.8. Forward heat-integration, Design II. Frequency dependent loop gain $g_{ii}(\bullet)$ and PRGA elements $\gamma_k R_k$ (■ $k=1$, ▲ $k=2$, × $k=3$).

Fast setpoint change may cause input saturation.

In conclusion, the prefractionator / side stream column with forward heat integration has very good controllability properties. The practice of a small prefractionator (doing just the necessary separation) is justified (at least for the case analysed) by its better disturbance rejection properties.
Prefractionator / side stream column with reverse integration

After closing the inventory control loops, four manipulated variables are left for temperature (composition) control: prefractionator reflux flow rate, main column reflux and side stream flow rate, and reboiler duty. There are six temperatures it may be necessary to control, corresponding to the two sections of the prefractionator and the four sections of the main column.

A control structure, similar to the one developed for the forward integration scheme, can be imagined (Figure 2.9). The reflux flow rate controls one temperature in the top of the prefractionator (no input is available to control the temperature in the bottom). In the main column, the reflux ratio is kept constant and reboiler duty is used to control the bottom temperature. One temperature located either above or below the side stream is controlled by the side stream flow rate.

However, strong interactions make it unfeasible. A typical steady-state RGA matrix is:

\[ \Lambda_1(0) = \begin{bmatrix} 1.144 & 0.008 & -0.153 \\ -0.164 & -0.001 & 1.165 \\ 0.019 & 0.990 & -0.012 \end{bmatrix} \]

Figure 2.9. Reverse heat-integration. Unfeasible control structure using only temperature measurements.

Standard notation is used. YC denotes ratio controller.
The small values of the last two diagonal elements show strong interaction. Moreover, their negative sign means that the system is not decentralised integral controllable (Skogestad and Postlethwaite, 1996), i.e. there exists no diagonal controller with integral action such that both of the following conditions are true: a) The closed loop system is stable. b) The gains of any subset of loops can be arbitrarily reduced without causing system instability.

Adding a fourth control loop, in which the main column reflux flow rate controlled top temperature, did not help. Several other control structures were tried and it was found that the control of the temperature in the middle of the main column is responsible for the negative diagonal elements in the RGA matrix. Hence, if only temperatures are measured, the side stream flow can not be the manipulated variable in a control loop. However, keeping the side stream constant is unacceptable, because any change of the feed flow rate of the intermediate component will result in impure products.

A successful control structure (Figure 2.10) can be developed if the flow rate of the side stream is used to control its composition. The nominal and scaling values of the controlled outputs, manipulated inputs and disturbances are presented in Table 2.5.

![Diagram](image)

**Figure 2.10.** Reverse heat-integration. Feasible control structure using side stream composition analyzer.


Chapter 2.  

Design and Control of a Distillation System with Prefractionator

Table 2.5. Control of prefractionator / side stream column with reverse heat integration. Nominal values and scaling factors of outputs, inputs and disturbances.

<table>
<thead>
<tr>
<th>Variable</th>
<th>Nominal value</th>
<th>Scaling factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>Design I</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$y_1$ (PF stage 4 temperature, °C)</td>
<td>56.7</td>
<td>1</td>
</tr>
<tr>
<td>$y_2$ (C stage 4 temperature, °C)</td>
<td>104.1</td>
<td>0.5</td>
</tr>
<tr>
<td>$y_3$ (C side stream purity, °C)</td>
<td>0.98</td>
<td>0.02</td>
</tr>
<tr>
<td>$y_4$ (C stage 27 temperature, °C)</td>
<td>168</td>
<td>0.5</td>
</tr>
<tr>
<td>$u_1$ (PF reflux flow rate, mol/s)</td>
<td>16.83</td>
<td>8.33</td>
</tr>
<tr>
<td>$u_2$ (C reflux flow rate, mol/s)</td>
<td>35.94</td>
<td>10</td>
</tr>
<tr>
<td>$u_3$ (C side draw flow rate, mol/s)</td>
<td>13.89</td>
<td>6.94</td>
</tr>
<tr>
<td>$u_4$ (C reboiler duty, $10^6$ W)</td>
<td>1.814</td>
<td>0.907</td>
</tr>
<tr>
<td>Design II</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$y_1$ (PF stage 4 temperature, °C)</td>
<td>61.2</td>
<td>1</td>
</tr>
<tr>
<td>$y_2$ (C stage 3 temperature, °C)</td>
<td>98.7</td>
<td>0.5</td>
</tr>
<tr>
<td>$y_3$ (C side stream purity, °C)</td>
<td>0.98</td>
<td>0.02</td>
</tr>
<tr>
<td>$y_4$ (C stage 28 temperature, °C)</td>
<td>165.6</td>
<td>0.5</td>
</tr>
<tr>
<td>$u_1$ (PF reflux flow rate, mol/s)</td>
<td>18.16</td>
<td>8.33</td>
</tr>
<tr>
<td>$u_2$ (C reflux flow rate, mol/s)</td>
<td>35.66</td>
<td>10</td>
</tr>
<tr>
<td>$u_3$ (C side draw flow rate, mol/s)</td>
<td>13.89</td>
<td>6.94</td>
</tr>
<tr>
<td>$u_4$ (C reboiler duty, $10^6$ W)</td>
<td>1.790</td>
<td>0.907</td>
</tr>
<tr>
<td>Disturbances</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Light in feed</td>
<td>0.333</td>
<td>0.1</td>
</tr>
<tr>
<td>Intermediate in feed</td>
<td>0.333</td>
<td>0.1</td>
</tr>
<tr>
<td>Heavy in feed</td>
<td>0.333</td>
<td>0.1</td>
</tr>
</tbody>
</table>

For both designs analysed, the chosen pairing correspond to positive diagonal RGA elements:

$$\Lambda_1(0) = \begin{bmatrix} 1.824 & 0.028 & 0.001 & -0.853 \\ 2.758 & 11.90 & 0.002 & -13.66 \\ 0.036 & 1.273 & 0.445 & -0.755 \\ -3.618 & -12.20 & 0.552 & 16.27 \end{bmatrix}, \quad RGA\_number(0) = 63.3$$

$$\Lambda_2(0) = \begin{bmatrix} 1.265 & -1.625 & -0.05 & 1.364 \\ -0.283 & 23.15 & 0.050 & -21.92 \\ -0.052 & -18.66 & 0.151 & 19.56 \\ 0.068 & -1.868 & 0.803 & 1.997 \end{bmatrix}, \quad RGA\_number(0) = 90.5$$

For high frequencies, both RGA_numbers drop to zero, showing that control performance is possible.
Figure 2.11. Reverse heat-integration. Frequency dependent loop gain, $g_{ii}$ (●) and CLDG elements, $g_{di}$ (■ $k=1$, ▲ $k=2$, × $k=3$).

In Design II, the second manipulated input is not strong enough to reject the disturbances.
Figure 2.11 presents the closed loop disturbance gain. The main difference between Design I and Design II is in the disturbance rejection properties of the second control loop (main column top temperature – reflux flow rate). In the first case, all disturbances can be properly rejected. In the second case, the maximum allowed change of the reflux flow rate is not big enough to reject any of the three disturbances. There are also some problems with the first control loop (Design II performs slightly better). However, this loop is not directly related to product purity.

The frequency-dependent PRGA is similar to the forward heat-integration case. Input saturation may occur in some control loops, if fast tracking is required for certain setpoints. However, as previously discussed, this will rarely be the case.

We conclude that the controllability properties of the prefractionator / side stream configuration with reverse heat-integration are worse than for the forward integration scheme. If only temperature measurements are available, good control is impossible due to strong interactions. A feasible control structure, in which the composition of the side stream is measured, was developed. The controllability properties are better if the prefractionator performs a sharp A/C split.

We acknowledge the anonymous reviewer who suggested to ratio the side stream to the reflux flow rate. The closed-loop performance of this strategy was investigated. It achieves good control of the top and bottom purity. However, there is a rather large (about 2%) steady state error of the side stream composition. Hence, when tight control of the intermediate product purity is not required, this control scheme is also a good option.

**Closed loop dynamic simulation**

The performance of the proposed control configurations was tested using the SPEEDUP® nonlinear model. A typical run required about 80 seconds, on a Pentium II 350 MHz computer.

Design I was analysed for the forward heat-integration configuration. For the reverse heat-integration arrangement, Design II was investigated. A lag time of 1 min was assumed for temperature sensors. A dead time of 3 min was assumed for composition analysers. Control structures presented in Figures 2.5 and 2.10 were used. The parameters of the PI controllers were found by trial-and-error. They are presented in Table 2.6.
Table 2.6. Controller tuning.

<table>
<thead>
<tr>
<th></th>
<th>$K_p$</th>
<th>$T_1$</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Forward heat integration</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Loop 1</td>
<td>$0.0139 \times 10^6$ W/K</td>
<td>0.2 h</td>
</tr>
<tr>
<td>Loop 2</td>
<td>$16.66$ (mol/s) / K</td>
<td>0.2 h</td>
</tr>
<tr>
<td>Loop 3</td>
<td>$6.94$ (mol/s) / K</td>
<td>0.2 h</td>
</tr>
<tr>
<td>Loop 4</td>
<td>(Column tray 27 temperature – Loop 1 setpoint)</td>
<td>5 K/K</td>
</tr>
<tr>
<td><strong>Reverse heat integration</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Loop 1</td>
<td>$2.77$ (mol/s) / K</td>
<td>-</td>
</tr>
<tr>
<td>Loop 2</td>
<td>$3.61$ (mol/s) / K</td>
<td>0.2 h</td>
</tr>
<tr>
<td>Loop 3</td>
<td>$1.39$ mol/(s %)</td>
<td>0.2 h</td>
</tr>
<tr>
<td>Loop 4</td>
<td>$0.907 \times 10^6$ W/K</td>
<td>0.2 h</td>
</tr>
</tbody>
</table>

The control system performance, as deviation of the products' concentration from the design values, is presented in Figure 2.12. Disturbances $D_1$, $D_2$ and $D_3$ correspond to increase of the feed concentration of pentane, hexane and heptane, respectively. In each case, one concentration was increased from 33.3% to 43.3%, while the other two were decreased to 28.3%.

If the magnitude of the disturbances is taken into account, the performance of the control system seems acceptable. The purity deviation is less than 0.5 % for all streams. The settling time is less than 0.5 hours for forward heat-integration and less than 2 hours for reverse heat-integration. It is noted that the steady state error in concentration can be removed if concentration controllers are used.
Forward heat-integration. Design II

Reverse heat-integration. Design I

![Graphs showing control system performance](image)

**Figure 2.12. Control system performance.**

- ♦ pentane, ■ hexane, ▲ heptane. Disturbances D₁, D₂, D₃, correspond to increase of the feed concentration of pentane, hexane and heptane, respectively, from 0.33 to 0.43. Control structures are presented in Figures 2.5 and 2.10 (the composition-temperature cascades were not used). Controller tuning is presented in Table 2.6.
Conclusions

The relationship between the design and the control of a heat-integrated distillation set-up, consisting of a preflasher and a main column with side stream, was analysed. Both forward and reverse heat-integration schemes were investigated. Detailed results are presented for the separation of an equimolar pentane-hexane-heptane mixture, with moderate purity requirements. These results were confirmed by the high purity separation of the benzene-toluene-xylene mixture.

1. Four different designs were investigated, both for forward and reverse heat-integration. Energy integration took into account a total match of the reboiler and condenser of the first/second and second/first columns, respectively, as well as feed preheating with products’ excess enthalpy. Design I considered a tall preflasher for sharp light/heavy split, while Design II considered a shorter preflasher for a moderate light/heavy split. In the other cases, the preflasher was designed for high recovery of the light, or of the heavy component, respectively. In all designs the number of stages varied significantly for the preflasher, but remained practically constant for the main column.

2. In all cases, the total energy consumption varied only slightly. Consequently, energy recovery cannot be a selection criterion between alternatives. However, the dynamic behaviour showed significant differences.

3. MIMO linear controllability analysis in the frequency domain was performed to evaluate the dynamic properties of alternative designs. Large disturbances in feed concentration of the three components were considered. Among controllability indices, Closed Loop Disturbance Gain (CLDG) and Performance Relative Gain Array (PRGA) were calculated. The controllability analysis predicts, in all situations, better dynamic properties for the forward heat-integration scheme compared with the reverse one. This behaviour was tested by closed loop simulation with the full non-linear model.

4. For the preferred forward integration scheme, an efficient control structure, using only temperature measurements, was developed. The design with a short preflasher has, by far, the best performance.

5. The control of the reverse heat-integration scheme does not work with only temperature measurements. The system is controllable only if the concentration of the side stream can be measured. Moreover, better disturbance rejection is possible with a sharp light/heavy split in the preflasher.
6. The behaviour of a high purity separation of a benzene-toluene-xylene mixture was tested, similarly to the work of Ding and Luyben\textsuperscript{15}, who developed a quite complex control structure for the reverse heat-integration scheme. For forward heat-integration, the simple temperature-based control structure works well. This confirms the better controllability of the forward heat-integration arrangement.

7. It may be concluded that in general the forward heat-integration scheme is easier to control. A possible explanation may be the absence of a positive feedback of energy, which is very likely in the reverse heat-integration scheme.

**Literature cited**


Chiang, T.P. and W.L. Luyben, “Incentives for Dual Composition Control in Single and Heat-
Chapter 2. Design and Control of a Distillation System with Prefractionator


Chapter 3 Stability and Multiplicity Approach to the Design of Heat-Integrated PFR

Abstract

The non-linear behaviour of the heat-integrated plug flow reactor, consisting of feed-effluent heat exchanger (FEHE), furnace, adiabatic tubular reactor and steam-generator is studied, considering a first order, irreversible, exothermic, adiabatic reaction. Bifurcation theory is used to analyse the relationships among design, reaction thermodynamics and kinetics, and state multiplicity and stability. Hysteresis, isola and boundary limit varieties are computed, and the influence of the activation energy, reaction heat and FEHE efficiency on the multiplicity region is studied. The double-Hopf and double-zero bifurcation points divide the parameter space in regions with different dynamic behaviour. State multiplicity, isolated branches and oscillatory behaviour may occur for realistic values of model parameters. A design procedure is proposed to ensure a desired multiplicity pattern and a stable point of operation and to avoid high sensitivity. The procedure was applied to three reaction systems with different kinetic and thermodynamic characteristics.
Chapter 3. Design of Heat-Integrated PFR

Introduction

In the autothermal PFR, heat exchange between the effluent and the feed stream is used to preheat the feed. Apparently, for an exothermic reaction and adiabatic reactor operation, there is no need of an additional heat source. However, there are several reasons to include a heater (normally a furnace) and a cooler (normally a steam-generator) in the system:

1. A heater (furnace) is always required for startup. It may be placed upstream or downstream of the feed-effluent heat exchanger (FEHE). It is favourable to place the FEHE before the furnace, as it will work at lower temperatures. Steam can be used to generate power and/or be exported to drive separation units. This site integration solution is more efficient than process-process heat integration, as proposed by Douglas (1988).

2. Often, a purge stream is available as fuel for the furnace. Consequently, a steam-generator has to remove the heat excess. Placing the steam-generator before the FEHE allows heat recovery at higher temperature and is therefore preferable in view of exergetic considerations.

3. In the autothermal PFR, multiple steady-states may exist. Operation in the multiplicity region may be desirable or unavoidable. The classical analysis of van Heerden (1958) shows that the middle steady-state is unstable. Some states may also lose stability due to Hopf bifurcation and the system may exhibit oscillatory behaviour. In such cases, stable operation is possible only if a control system is in place. If the furnace and steam-generator are included, their duties are available as manipulated variables in temperature control loops.

The inclusion of furnace and steam-generator leads to the structure presented in Figure 3.1. This structure will be called heat-integrated PFR. It should be pointed out that other series of circumstances could easily lead to other equipment arrangements.

Typical initial design data of the heat-integrated PFR are reactor type (empty tube or packed bed), feed flow rate, reactor inlet temperature, and required conversion. Reaction kinetics and thermodynamics are also known. These data suffice to design the reactor. Next, the designer has to size the furnace, steam-generator, and FEHE. This can be done by optimising some steady-state related economic criteria and/or considering the plantwide energy balance.
However, the operating conditions may be different. There is always uncertainty of the kinetic parameters. Feed flow rate or concentration may deviate from the design values. Disturbances in feed temperature, furnace and steam-generator duties are unavoidable. Heat-exchange efficiency may change due to fouling. Therefore, the operating point may become unstable and/or sustained oscillations may occur. Also, the system may exhibit high sensitivity, which renders control difficult. Consequently, the design also must be assessed with regard to steady-state multiplicity, stability and sensitivity issues.

Many articles on non-linear behaviour of reacting systems have been published. The first-order reaction in CSTR, axial-dispersion tubular reactor, or catalyst particle are classic problems in chemical-reaction engineering. Only a few articles are, however, dedicated to heat-integrated chemical reactors. Lovo and Balakotaiah (1992) computed the uniqueness-multiplicity boundary of the tubular reactor with internal or external heat exchange and CSTR with external heat exchange. For limiting cases, they presented analytical expressions of the ignition, extinction and cusp points. Subramanian and Balakotaiah (1996) classified the steady-state and dynamic behaviour of several distributed reactor models, including the CSTR with external heat exchange and the tubular reactor with internal heat exchange.

Although singularity theory (Balakotaiah and Luss, 1982, 1984; Golubitsky and Schaeffer, 1985) and bifurcation theory (Iooss and Joseph, 1981; Scott, 1993) are proven

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**Figure 3.1. Heat-integrated PFR**

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Chapter 3. Design of Heat-Integrated PFR
tools, few applications to design are reported. Heiszwolf and Fortuin (1997) presented an application-oriented design procedure for unique and stable operation of first-order reaction systems in a CSTR. Morud and Skogestad (1998) discussed the dynamics of an industrial (multibed) ammonia reactor, where positive feedback due to heat integration led to oscillatory behaviour. They showed that instability occurred at a Hopf bifurcation point. Silverstein and Shinnar (1982) evaluated the stability of the heat-integrated PFR using the frequency response of individual equipment components. They did not, however, address the relationship between reaction kinetics and thermodynamics and state multiplicity and stability.

The goal of this work is to study the steady-state and dynamic behaviour of the heat-integrated PFR. We consider a first-order, irreversible, exothermic reaction, and adiabatic reactor operation. We concentrate on the relationship between reaction kinetics and thermodynamics, equipment design, and state multiplicity and stability. To achieve this goal, we use elements of bifurcation theory. Moreover, we show how the results can be applied during the design, in order to ensure a desired multiplicity pattern, a stable operating point, and to avoid high sensitivity. In the next section, we present the heat-integrated PFR model equations and the types of bifurcation of interest. Afterwards, we classify the steady-state and dynamic behaviour by computing the hysteresis, isola, boundary-limit, double-zero and double-Hopf varieties. State multiplicity, isolated branches and oscillatory behaviour are possible for realistic values of model parameters. We investigate the influence of reaction kinetics and thermodynamics and FEHE efficiency on the extent of multiplicity region. Subsequently, we discuss how the results can be used to avoid operational problems and propose a design procedure. Three reaction systems with different kinetic and thermodynamic characteristics are used as examples. The main ideas are reviewed in the last section. Details of the computation of the bifurcation points are given in Appendix.

Mathematical model

The dynamics of equipment in the heat-integrated PFR may be described by plug-flow, pseudohomogeneous models. The dimensionless equations are given below:
Chapter 3. Design of Heat-Integrated PFR

FEHE (tube and shell-side):

\[ M_i \frac{\partial \theta_i}{\partial \tau} = -\frac{\partial \theta_i}{\partial \xi} + NTU \cdot (\theta_i - \theta_i) \]  
(3.1a)

\[ M_i \frac{\partial \theta_i}{\partial \tau} = \frac{\partial \theta_i}{\partial \xi} - NTU \cdot (\theta_i - \theta_i) \]  
(3.1b)

Furnace:

\[ M_b \frac{\partial \theta_b}{\partial \tau} = -\frac{\partial \theta_b}{\partial \xi} + Q_b \]  
(3.1c)

Reactor:

\[ \frac{\partial \theta_r}{\partial \tau} = -\frac{\partial \theta_r}{\partial \xi} + Da \cdot \exp \left( \frac{\gamma \cdot \theta_i}{1+\theta_i} \right) \cdot (1-X) \]  
(3.1d)

Steam-generator:

\[ M_c \frac{\partial \theta_c}{\partial \tau} = -\frac{\partial \theta_c}{\partial \xi} - Q_c \]  
(3.1e)

with the boundary conditions:

\[ \theta_i(\tau,0) = \theta_i(\tau,0) \]
\[ \theta_i(\tau,1) = \theta_i(\tau,0) \]
\[ \theta_b(\tau,0) = \theta_b(\tau,0) \]
\[ \theta_r(\tau,1) = \theta_r(\tau,0) \]
\[ \theta_c(\tau,1) = \theta_c(\tau,1) \]
\[ X(\tau,0) = 0 \]

(3.2)

where \( \tau \) and \( \xi \) are dimensionless time and coordinate, respectively.

The model variables are dimensionless temperatures in the shell and tube-side of the FEHE (\( \theta_b \), \( \theta_i \)), furnace (\( \theta_b \)), reactor (\( \theta_r \)) and steam-generator (\( \theta_c \)), and conversion (\( X \)). The dimensionless parameters represent reactor residence time (\( Da \)), FEHE area (\( NTU \)), activation energy (\( \gamma \)), adiabatic temperature rise (\( B \)), furnace and steam-generator duties (\( Q_b \), \( Q_c \)), feed temperature (\( \theta_b \)), residence time ratios (\( M_b \), \( M_o \), \( M_b \), \( M_s \)), and heat capacity ratio (\( Le \)).

The choice of reference temperature \( \bar{T} \) deserves a special discussion. Most of the previous studies considered the Damkohler number (\( Da \)) as bifurcation parameter, and investigated the effect of feed flow rate change. However, feed flow rate can be easily controlled during the operation; hence, variation of this parameter is unlikely. Uncontrolled
changes of feed temperature, furnace and steam-generator duties, and FEHE efficiency are more probable. In the previous studies the feed temperature was chosen as reference \( \bar{T} = T_0 \), becoming part of several dimensionless numbers. In this way, the dimensionless feed temperature was set to zero \( \theta_0 = 0 \). When studying the steady-state behaviour with feed temperature as a variable, only one dimensionless parameter must depend on it; hence \( \bar{T} = T_0 \) is not a good choice.

If we consider isothermal operation of a fixed-size PFR, the reactant is almost completely converted \( (X=99.33\%) \) for \( Da = -\ln(1 - X) = 5 \). We choose the corresponding reaction temperature as reference. This is not restrictive. The reactor can be sized when the reactor inlet temperature, desired conversion, reaction kinetics, and thermodynamics are known; hence, the residence time is known. Then the reference temperature \( T \), for which \( Da = 5 \), can be calculated and used for dimensionless numbers. This way, changes of furnace or steam-generator duty, feed temperature and concentration, and FEHE efficiency are reflected by independent changes of the model parameters: \( Q_h \), \( Q_c \), \( \theta_b \), \( B \) and \( \varepsilon \), respectively.

Heiszwoolf and Fortuin (1997) and Chen and Luss (1989) used a similar procedure setting \( \gamma = 25 \) and \( Da = 1 \), respectively.

To study the influence of flow rate on steady-state multiplicity and stability, its change must be concentrated in one parameter. This can be achieved by introducing new dimensionless variables:

\[
H = \frac{K \cdot A_T}{\rho_c V_k (T)}
\]  

(3.3)

\[
H_h = \frac{q_h}{\rho_c T V_k (T)}
\]  

(3.4)

\[
H_c = \frac{q_c}{\rho_c T V_k (T)}
\]  

(3.5)

and replacing in Eq. 3.1 \( NTU \), \( Q_h \), and \( Q_c \) by \( Da \cdot H \), \( Da \cdot H_h \) and \( Da \cdot H_c \), respectively.

The steady-state model is obtained by dropping the time derivatives in Eq. 3.1. After analytical integration of the FEHE, furnace, and steam-generator equations, the following boundary value problem is obtained:

\[
\frac{d\theta_t}{dz} = B \cdot Da \cdot \exp \left[ \frac{\gamma \cdot \theta_t}{1 + \theta_t} \right] \left( 1 - \frac{\theta_t - \theta_t(0)}{B} \right)
\]  

(3.6a)
Chapter 3. Design of Heat-Integrated PFR

\[ f(\gamma, 0, Q_h, Q_c, \varepsilon, B, \gamma, Da) = (1 - \varepsilon) \cdot \gamma + (Q_h - \varepsilon Q_c) + \varepsilon \cdot \gamma(1) - \gamma(0) = 0 \]  \hspace{1cm} (3.6b)

where

\[ \varepsilon = \frac{NTU}{1 + NTU} \]  \hspace{1cm} (3.7)

is the FEHE efficiency.

The reactor conversion is given by:

\[ X = \frac{\gamma(1) - \gamma(0)}{B} \]  \hspace{1cm} (3.8)

Equation 3.6 may be solved by a shooting technique: start with an initial guess for \( \gamma(0) \), integrate to find \( \gamma(1) \), check the boundary condition, and update \( \gamma(0) \) (for example, by the Newton method). This way, the problem is reduced to one algebraic equation with one state variable (reactor inlet temperature) for which we can apply the singularity theory of a single state variable (Golubitsky and Schaeffer, 1985).

---

**Figure 3.2. Conversion vs. dimensionless furnace duty, for different values of NTU.**

\( Da=5, \ \gamma=5, B=0.07, \ \theta_0=0.6, Q_c=0.15, M_c=M_h=0.75, M_h=M_c=0.2, Le=2000. \)

Continuous lines represent stable steady-state. Dashed lines represent unstable steady states. Bold lines are the loci of fold and Hopf bifurcations. Points C, DH, DZ, I and BL correspond to cusp, double Hopf, double zero, isola and boundary limit bifurcations.
The S-shaped curves (bifurcation diagrams) in Figure 3.2 represent the conversion vs. furnace duty (bifurcation variable), for a given set of values of $Da, B, Q_c, \theta_b, M_b, M_c, M_s, \text{and } Le$. Each curve is associated with a fixed value of $NTU$. The number of possible steady-states changes at fold bifurcation points. The upper and lower branches of the fold curve correspond to reaction extinction and ignition, respectively. The qualitative shape of the bifurcation diagram changes when the second parameter ($NTU$) is varied. At the cusp point (C) two fold points appear or disappear and the number of steady-states changes by two. The locus of cusp points is called hysteresis variety. Note that if the furnace duty is fixed to $Q_h=0.145$ (point I), the conversion does not depend on $NTU$. This corresponds to an isola bifurcation when $NTU$ (or $\varepsilon$) is taken as bifurcation parameter. When the $NTU$ parameter has very large values ($NTU \to \infty, \varepsilon \to 1$) the bifurcation parameter on the ignition and extinction branches reaches the asymptotic values $Q_h=0.15$ and $Q_h=0.08$ (not shown in Figure 3.2), respectively. They are called boundary-limit points (BL). For a first-order reaction, the heat-integrated PFR exhibits at most three steady-states, hence the double-limit variety (where the qualitative nature of the bifurcation diagram may also change) does not exist.

When multiple steady-states exist, the middle one is always unstable (dashed line). A steady-state may also become unstable at a Hopf bifurcation point. Here a branch of oscillating solution arises. When a second parameter is varied, the number of Hopf bifurcation points may change. At the double-Hopf (DH) bifurcation point two Hopf bifurcation points appear or disappear. At the double-zero (DZ) bifurcation point, one Hopf bifurcation point appears or disappears.

Details of the bifurcation points computation are presented in the Appendix.

**Steady-state behaviour**

In this study we considered two cases.

First, we used as bifurcation parameter:

$$Q = (1 - \varepsilon) \cdot \theta_b + (Q_h - \varepsilon Q_c)$$  \hspace{1cm} (3.9)

It represents the system energy input and captures the influence of feed temperature, furnace, and steam-generator duties on the steady-state. Note that $\theta_b, Q_h, \text{and } Q_c$ appear coupled as $Q$ in the steady-state equation 3.6 and they do not appear in the bifurcation equations A3.6 and A3.9.
A pitchfork bifurcation was identified. It corresponds to an isola bifurcation of the fold points locus and divides the $\varepsilon$–$B$ plane into two regions (Figure 3.3).

If the reaction is highly exothermic or the FEHE is very efficient (at the right of the pitchfork line), state multiplicity is possible irrespective of the value of the reaction activation energy. Otherwise (at the left of pitchfork line), state multiplicity is possible only for high or low activation energy.

**Figure 3.3. Pitchfork bifurcation**

At the right of the pitchfork bifurcation, multiple steady-states are possible irrespective of the value of activation energy.
Figure 3.4 presents a cross section of the hysteresis variety in \( (\gamma - B) \) plane, for different values of the FEHE efficiency. For fixed \( \varepsilon \), multiple steady-states are possible at the right of the hysteresis line. For fixed activation energy, an increase of either the FEHE efficiency or adiabatic temperature rise leads to multiplicity. The unicity region is larger for intermediate activation energies.

![Cross section of the hysteresis variety in \( (\gamma - B) \) plane for different values of FEHE efficiency.](image)

For fixed \( \varepsilon \), multiple steady-states are possible at the right of the hysteresis line.

Second, we considered the FEHE efficiency \( (\varepsilon) \) as bifurcation parameter. In this case, no other parameters may depend on it. Hence \( \Theta_b \), \( Q_h \) and \( Q_c \) may not be coupled as \( Q \), and their effect must be investigated separately.

The steady-state behaviour is more complicated. In addition to the hysteresis variety, one isola and two boundary-limit varieties exist. Figure 3.5 presents a typical phase diagram.
in the \((Q_c, Q_h)\) space. The hysteresis and boundary-limit varieties are described by the following equations:

\[
(1 - \epsilon^*) \cdot \theta_0 + Q_h - \epsilon^* Q_c = Q^* \tag{3.10}
\]

\[
Q_h = Q_c \tag{3.11}
\]

\[
Q_h = Q_c - B \tag{3.12}
\]

where \(\epsilon^*\) and \(Q^*\) correspond to the cusp bifurcation.

Figure 3.5. Phase diagram of heat-integrated PFR with FEHE efficiency as bifurcation parameter.

\(Da=5, \gamma=10, B=0.2, \theta_c=-0.4\). C - cusp, I - isola, BL - boundary limit.

The different types of bifurcation diagrams existing in regions I - VIII are presented in Figure 3.6.

The isola variety approaches asymptotically the BL₁ and BL₂ varieties. This observation together with the Eqs 3.10-3.12 can be used to sketch the phase diagram. The isola and hysteresis varieties intersect at a pitchfork bifurcation point.
These singularities divide the parameter space into eight regions where qualitatively different bifurcation diagrams exist (they are presented in Figure 3.6).

![Bifurcation Diagrams](image)

**Figure 3.6. Conversion vs. FEHE efficiency bifurcation diagrams.**
Diagrams I-VIII correspond to regions I-VIII in Figure 3.5.

In region I there is a unique steady-state. Crossing the hysteresis variety to region II, two fold points appear and state multiplicity is possible. When the BL₁ variety is crossed to region III, a low-conversion isolated branch appears and the multiplicity pattern becomes 1-3-1-3. The two branches coalesce when the isola is intersected and two fold points disappear. Hence, in region IV, a low conversion branch exists for all values of FEHE efficiency, while high conversion is possible only for high values of the ε parameter. When moving to regions V or VII, the high conversion branch disappears (at BL₂), or multiple states appear on the
low-conversion branch (at hysteresis), respectively. From region VII, the high conversion branch disappears at BL$_2$ (region VI) or the two branches coalesce at isola (region VIII).

Note that isolated branches exist between the BL$_1$ and BL$_2$ varieties.

**Dynamic behaviour**

Dynamic classification involves dividing the parameter space into regions where a different number of Hopf bifurcation points exists. Due to the large number of parameters, a complete classification is difficult. We consider the energy input as the bifurcation parameter and restrict our analysis to the influence of the dimensionless adiabatic temperature rise. We focus on high values of the FEHE efficiency, and thus high tube and shell residence times. We consider a catalytic reactor because its “wrong-way” behaviour is the cause of oscillations (and hence of Hopf bifurcations). This is reflected in the large value of $Le$.

The wrong-way behaviour of packed-bed reactors has been demonstrated by Mehta et al. (1981) who used the plug-flow pseudohomogeneous model. Pinjala et al. (1988) and Chen and Luss (1989) added more complexity. They showed that models including axial dispersion and/or heterogeneity (1) account for state multiplicity inside the reactor and (2) predict smaller and slower temperature peaks. The first matter is usually avoided in practice by the reactor design, however, and the second item does not change the qualitative transient behaviour. Besides, the heat-integrated PFR becomes a two-state-variables problem when heterogeneity or axial dispersion is included; thus, the classification methodology is not applicable. For these reasons, the use of the plug-flow pseudohomogeneous model is adequate for the purpose of this work.

A typical phase diagram is presented in Figure 3.7. The double-zero locus is tangential to the cusp locus. The double-Hopf intersects the double-zero at a bifurcation point of codimension 3. The $(e-B)$ space is divided into six regions where different types of bifurcation diagrams exist. The bifurcation diagrams of conversion vs. energy input are presented in Figure 3.8. In region I, one stable steady-state exists. After crossing the double-Hopf locus, two Hopf points appear. In-between, no stable steady-state exists and the system exhibits oscillatory behaviour. When the hysteresis variety is crossed, two fold points appear. Therefore, in region III there are multiple steady-states. Oscillatory behaviour is possible on both the ignited and extinguished branches. In region IV, one Hopf point disappears after crossing the double-zero locus. In region V, a second Hopf point appears on the ignited
branch. They both disappear when the double-Hopf variety is crossed; thus, in region VI steady-state multiplicity is possible but not the oscillatory behaviour.

Note that the phase diagram may change (the double zero and double Hopf loci do not intersect) when a third parameter is varied.

![Phase diagram of the heat-integrated PFR with energy input as bifurcation parameter.](image)

**Figure 3.7.** Phase diagram of the heat-integrated PFR with energy input as bifurcation parameter.

\( Da=5, \gamma=5, M_f=M_c=0.75, M_t=M_e=0.2, Le=2000. \)

C - Cusp. DH - Double Hopf. DZ - Double Zero.

The different types of bifurcation diagrams existing in regions I-VI are presented in Figure 3.8.

It is expected that the steady-state and dynamic behaviour of the heat-integrated PFR will change when a control system (for example, manipulating furnace duty to control reactor input temperature) is present. In addition, we expect more complicated phase diagrams when more detailed models are used for the furnace and steam-generator (for example with burn gas and cooling water temperatures as parameters, instead of furnace and steam-generator duties).
Chapter 3. Design of Heat-Integrated PFR

Design procedure for the heat-integrated PFR

In this section we propose a design methodology for the heat-integrated PFR. We are not concerned with detailed sizing of the equipment. We are interested in the values of FEHE efficiency, furnace, and cooler duties. These can be obtained by optimisation according to economic criteria, but certain restrictions arise when multiplicity, sensitivity, and stability are considered.

We propose the following design procedure for the heat-integrated PFR:

**Step 0.** Initial data: Feed flow rate and concentration; reactor inlet temperature; conversion; reactor type (catalytic or empty tube); reaction kinetics and thermodynamics.

Figure 3.8. Conversion vs. energy input bifurcation diagrams.
Diagrams I-VI correspond to regions I-VI in Figure 3.7. ◯ Fold; □ Hopf.
Step 1. Size the reactor based on adiabatic PFR equations.

Step 2. Choose the reference temperature. If $\bar{T}$ is chosen such that $Da = 5$, Figures 3.3 and 3.4 can be used during step 3. Calculate the dimensionless activation energy and adiabatic temperature rise.

Step 3. Select the FEHE, based on the following observations:

- When the heat-integrated PFR is designed for low conversion, the hysteresis variety is a conservative boundary against runaway (Balakotaiah et al., 1995, Patil et al., 1997). In this case, Figure 3.4 can be used to select FEHE efficiency so the $(\chi B)$ point is in the unicity region.
- When the system is designed for middle or high conversion, autothermal operation may be of interest. This is possible only when an ignited state exists. In this case, the $(\chi B)$ point must be in the multiplicity region of Figure 3.4. The extinction point gives the minimum energy input required.
- Close to the hysteresis variety the system is very sensitive to change of feed temperature, furnace, and cooler duties.
- The energy input is given by:
  \[ Q = (1 - \epsilon) \cdot \theta_t(0) - \epsilon \cdot B \cdot X \]  
  (3.13)

Step 4. Stability issues: The middle operating point is always unstable. The other operating points may be stable or unstable. Dynamic simulation may be used to assess the stability, but no information about the extent of the stability region will be obtained. We recommend computing the Hopf bifurcation points (which are on the boundary of the oscillatory behaviour region) in order to find possible instability near the operating point. Note that working at an unstable operating point is not by itself unattractive, as a temperature controller may stabilise the system. Loss of stability due to disturbances or design-parameter uncertainty is dangerous. If the dynamic behaviour is not satisfactory, the design must be changed. The following observations should be considered:

- Only for a small range of model parameters the whole ignited branch (high conversion) is stable (region VI in Figure 3.7). A domain of oscillatory behaviour bounded by Hopf and extinction points (regions III and IV) is very likely. In this case, increasing the energy input will stabilise the system, without significant change of conversion. If the heat of reaction is small, two Hopf points may bound the oscillation domain (region V). In this case decreasing the energy input may also stabilise the system. This solution may be
undesirable, as the operating point will move closer to the extinction point. Also, the change of the conversion will be more important.

- If the system is designed in the unicity region, oscillations are likely for highly exothermic reactions, especially at intermediate conversion (region II in Figure 3.5). In this case, decreasing the FEHE efficiency will ensure a stable and unique operating point.

- If no solution can be found, then some of the initial data have to be modified.

**Step 5.** Choose feed-temperature, furnace, and steam-generator duties. When the FEHE efficiency is constant, the bifurcation behaviour depends only on the energy input ($Q$) and does not depend on the distribution of the energy input between feed-temperature, furnace and steam-generator duties. Consequently, these variables can be linked to plant energy balance, site integration, combined heat and power production, or low equipment cost considerations. To prevent operational problems due to isolated branches when FEHE efficiency may change, operation in region I or II of Figure 3.5 should be preferred. As these regions are at the left of the BL$_1$ line, the following condition must be fulfilled:

$$0 < Q_e < (\theta_2 - \theta_0) \frac{e}{1-e} \cdot B \cdot X$$

(3.14)

Note that the ranges for furnace and steam-generator duties decrease when the FEHE efficiency increases.

We shall illustrate the proposed methodology by three different reaction systems. The first one, propylene chlorination, is highly exothermic with moderate activation energy. Smith (1970) gives kinetic parameters. With excess propylene, the reaction can be considered first order. The second system involves a mild exothermic reaction with high activation energy, toluene hydrodealkylation (Douglas, 1988). The third system is a catalytic reaction with intermediate heat effect and low activation energy. Table 1 contains the initial data and the results.

Both kinetic ($k_0$ and $E_a$) and thermodynamic data ($\Delta T_{ad}$) are known. Design specifications are expressed as feed temperature ($T_0$), reactor inlet temperature ($T_2$), and desired conversion ($X$). In the first two cases, we require autothermal operation (defined by the existence of an ignited state for some range of operating variables). In the last case, we require a unique, stable operating point.
# Design of Heat-Integrated PFR

## Table 3.1. Design results for three heat-integrated PFR systems

<table>
<thead>
<tr>
<th></th>
<th>Propylene chlorination</th>
<th>Benzene hydrodealkylation</th>
<th>Catalytic reaction</th>
</tr>
</thead>
<tbody>
<tr>
<td>$k_0$ (s$^{-1}$)</td>
<td>3.417 10$^4$</td>
<td>2.65 10$^{14}$</td>
<td>8.67 10$^3$</td>
</tr>
<tr>
<td>$E_a$ (J/mol K)</td>
<td>63070</td>
<td>282870</td>
<td>41430</td>
</tr>
<tr>
<td>$\Delta T_{ad}$ (K)</td>
<td>170</td>
<td>70</td>
<td>70</td>
</tr>
<tr>
<td>Reaction type</td>
<td>homogeneous</td>
<td>homogeneous</td>
<td>catalytic</td>
</tr>
<tr>
<td>$T_0$ (K)</td>
<td>300</td>
<td>300</td>
<td>300</td>
</tr>
<tr>
<td>$T_2$ (K)</td>
<td>573</td>
<td>910</td>
<td>700</td>
</tr>
<tr>
<td>$X$</td>
<td>0.995</td>
<td>0.75</td>
<td>0.53</td>
</tr>
<tr>
<td>Requirements:</td>
<td>autothermal operation</td>
<td>autothermal operation</td>
<td>unique, stable operating point</td>
</tr>
<tr>
<td>$t_0$</td>
<td>7.57</td>
<td>34.6</td>
<td>25</td>
</tr>
<tr>
<td>$\bar{T}$ (for which Da = 5)</td>
<td>700</td>
<td>972</td>
<td>1000</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>10.87</td>
<td>35</td>
<td>5</td>
</tr>
<tr>
<td>$B$</td>
<td>0.243</td>
<td>0.072</td>
<td>0.07</td>
</tr>
<tr>
<td>$\theta_0$</td>
<td>-0.57</td>
<td>-0.69</td>
<td>-0.7</td>
</tr>
<tr>
<td>$\theta_2$</td>
<td>-0.181</td>
<td>-0.062</td>
<td>-0.3</td>
</tr>
<tr>
<td>$\varepsilon^*$</td>
<td>0.21</td>
<td>0.288</td>
<td>0.78</td>
</tr>
<tr>
<td>$Q^*$</td>
<td>-0.192</td>
<td>-0.0609</td>
<td>-0.0947</td>
</tr>
</tbody>
</table>

**Multiplicity**
- multiple steady-states if $\varepsilon > 0.3, Q < -0.155$
- multiple steady-states if $\varepsilon > 0.35, Q < 0.058$
- state unicity if $\varepsilon < 0.7, Q > 0.116$

**Stability**
- No oscillatory behaviour is expected.
- Unstable operating point. Unicity does not insure stability.

**FEHE efficiency**
- $\varepsilon = 0.5$
- $\varepsilon = 0.6$
- $\varepsilon = 0.5$

**No isolated branches**
- $Q_c < 0.147$
- $Q_c < 0.547$
- $Q_c < 0.344$
First, we design the reactor using the adiabatic PFR equations. This gives the residence time ($t_0$). Now we can calculate the reference temperature (for which $Da=5$) and use it for dimensionless parameters. We mention that the dimensionless activation energy is high for the second reaction ($\gamma=35$) and low for the third one ($\gamma=5$). Also, the dimensionless temperature rise correctly indicates that the first reaction is highly exothermic ($B=0.243$). Consequently, the proposed choice of the reference temperature gave meaningful results.

To get state unicity (multiplicity) we might calculate the FEHE efficiency at the cusp bifurcation ($\epsilon^*$) and choose a lower (higher) value. Alternatively, we can simply use Figure 3.4 to place the $(B,\gamma)$ point in the desired region.

Next, we consider stability issues. For noncatalytic reactions (first system), the high conversion branch is stable and no oscillatory behaviour is expected. The second system must be operated on the middle branch, which is always unstable; hence, a stabilising controller must be installed. For catalytic reactions (third system), unicity does not ensure stability. Figure 3.7 suggests that FEHE efficiency must be further decreased to ensure stable operation (region I).

After the stability was taken into account, we may choose the FEHE efficiency and use Eq. 3.13 to calculate the system energy input.

Finally, we must divide the energy input between feed temperature, furnace, and steam-generator duties. Isolated branches for varying FEHE efficiency are undesirable. To avoid them, we use Eqs 3.10-3.12 to draw diagrams similar to Figure 3.4, and place the operating point in region I.

**Conclusions**

In this article, we analyzed the relation between the reaction characteristics, design and state multiplicity and stability of the heat-integrated PFR. We considered a first-order, irreversible, exothermic reaction.

First, the energy input was considered as bifurcation parameter. It captures the influence of the feed-temperature, furnace, and steam-generator duties on the state multiplicity and stability. The hysteresis, double-zero, and double-Hopf bifurcation points were used to classify the steady-state and dynamic behaviour of the system. The parameter space is divided into six regions, corresponding to different types of bifurcation diagrams. State multiplicity is likely for very exothermic reactions or high FEHE efficiency, irrespective
Chapter 3. 

Design of Heat-Integrated PFR

of the activation energy. The heat-integrated PFR may exhibit oscillatory behaviour for realistic values of model parameters.

Subramanian and Balakotaiah (1996) obtained similar phase diagrams for different adiabatic reactor models, when the Damkohler number was considered to be the bifurcation parameter. However, these systems do not exhibit oscillatory behaviour for practical values of model parameters.

Second, the FEHE efficiency was chosen as bifurcation parameter. The hysteresis, isola, and boundary-limit varieties were used to classify the steady-state behaviour. The parameter space is divided into eight regions where qualitative different bifurcation diagrams exist. State multiplicity and isolated branches are now possible.

A design procedure was proposed. We found that the selection of FEHE efficiency is the critical step for achieving the desired state multiplicity and to ensure stability. When feed temperature, furnace or steam-generator duties are the changing variables, multiplicity and stability are not affected by the partition of energy input between these variables. Consequently, the heat equipment may be designed based on plant energy balance, site integration, combined heat and power production or lowest equipment cost. Certain restrictions arise, however, when the change of FEHE efficiency is taken into account. The procedure was applied to three reaction systems with different kinetic and thermodynamic characteristics.

The approach we presented can be applied to other equipment arrangements as well as to more complex reaction systems. Also, more accurate models of the furnace, steam-generator, and catalytic reactor can be considered.

**Notation**

\[ A_T = \text{FEHE area, m}^2 \]
\[ B = \text{adiabatic temperature rise, dimensionless} \]
\[ = \frac{\Delta T_{ad}}{T} \]
\[ c_p = \text{specific heat, J/(kg K)} \]
\[ Da = \text{Damkohler number} \]
\[ = k(T)^{\nu_p} \]
\[ E_a = \text{activation energy, J/mol} \]

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Chapter 3. Design of Heat-Integrated PFR

- \( F \) = mass flow rate, kg/s
- \( K_T \) = FEHE heat transfer coefficient, W/(m\(^2\) K)
- \( k \) = reaction rate constant, s\(^{-1}\)
- \( L \) = length, m
- \( Le \) = Lewis number, dimensionless
- \( \frac{1-\varepsilon_f \cdot \rho_f \cdot c_{pf}}{\varepsilon_f \cdot \rho_f \cdot c_{pf}} \)
- \( M_k \) = residence time ratio (k = t, s, h, c)
- \( \frac{t_{ok}}{t_{or}} \)
- \( NTU \) = number of transfer units, dimensionless
- \( \frac{K_T A_T}{F c_p} \)
- \( q_k \) = duty (k = h, c), W
- \( Q_k \) = duty (k = h, c), dimensionless
- \( \frac{q_h}{F c_p T} \)
- \( Q \) = energy input, dimensionless
- \( (1-\varepsilon) \dot{\rho}_0 - \varepsilon Q_c + Q_h \)
- \( R \) = gas constant, J/(mol K)
- \( T \) = temperature, K
- \( \bar{T} \) = reference temperature, K
- \( \Delta T_{ad} \) = adiabatic temperature rise, K
- \( t \) = time, s
- \( t_0 \) = residence time, s
- \( \frac{V \rho}{F} \)
- \( u, v \) = eigenfunctions
- \( X \) = conversion, dimensionless
- \( z \) = axial coordinate, m
- \( V \) = reactor volume, m\(^3\)
Greek letters

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>γ</td>
<td>activation energy, dimensionless</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$= \frac{E_a}{RT}$</td>
<td></td>
</tr>
<tr>
<td>ε</td>
<td>FEHE efficiency, dimensionless</td>
<td></td>
</tr>
<tr>
<td>λ</td>
<td>eigenvalue</td>
<td></td>
</tr>
<tr>
<td>ρ</td>
<td>density, kg/m³</td>
<td></td>
</tr>
<tr>
<td>τ</td>
<td>time, dimensionless</td>
<td></td>
</tr>
<tr>
<td>θ</td>
<td>temperature, dimensionless</td>
<td></td>
</tr>
<tr>
<td>ξ</td>
<td>axial coordinate, dimensionless</td>
<td></td>
</tr>
</tbody>
</table>

Subscripts

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>t</td>
<td>FEHE tube side</td>
</tr>
<tr>
<td>s</td>
<td>FEHE shell side</td>
</tr>
<tr>
<td>h</td>
<td>furnace</td>
</tr>
<tr>
<td>r</td>
<td>reactor</td>
</tr>
<tr>
<td>c</td>
<td>steam-generator</td>
</tr>
<tr>
<td>sf</td>
<td>solid phase</td>
</tr>
<tr>
<td>ff</td>
<td>fluid phase</td>
</tr>
<tr>
<td>b</td>
<td>feasibility boundary</td>
</tr>
<tr>
<td>0</td>
<td>feed</td>
</tr>
<tr>
<td>1</td>
<td>furnace inlet</td>
</tr>
<tr>
<td>2</td>
<td>reactor inlet</td>
</tr>
<tr>
<td>3</td>
<td>reactor outlet</td>
</tr>
<tr>
<td>4</td>
<td>steam-generator outlet</td>
</tr>
</tbody>
</table>
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Appendix: Bifurcation points computation

The equations describing the dynamic behaviour of the heat-integrated PFR can be written in the following condensed form:

\[ C \frac{\partial y}{\partial \tau} = - \frac{\partial y}{\partial \xi} + f(y, \mu, p) \quad (A3.1a) \]

\[ Ay(\tau,0) + By(\tau,1) - c = 0 \]

\[ y(0,\xi) = y_0(\xi) \quad (A3.1b) \]

\[ y \] is the vector of model variables, \( C \) is a diagonal capacity matrix, \( f(y) \) is a non-linear function, \( \mu \) is the bifurcation parameter and \( p \) is the vector of remaining parameters. \( A \) and \( B \) are constant matrices and \( c \) is a constant vector.

Its steady-state solution is given by the boundary value problem:

\[ \frac{\partial \delta_{y_1}}{\partial \xi} = f(y, \mu, p) \quad (A3.2a) \]

\[ Ay_0(0) + By_1(1) - c = 0 \quad (A3.2b) \]

When Eq. A3.1 is linearized around steady-state and separation of variables is applied, the following eigenvalue problem is obtained:

\[ C(\lambda + \lambda_1)(u + iv) = - \frac{d(u + iv)}{d\xi} + f_y(u + iv) \quad (A3.3) \]

where:

\[ \delta_y = (y(\xi, \tau) - y_1(\xi)) = (\mu(\xi) + iv(\xi)) \cdot e^{(\lambda + \lambda_1)\tau} \quad (A3.4) \]

At fold points, one real eigenvalue crosses the imaginary axis. Substitution of \( \lambda_1 = \lambda_2 = 0 \) in Eq. A3.3 and identification of real and imaginary parts gives:
\[ \frac{du}{d\xi} = f_r \cdot u \]  

(A3.5a)

with the boundary condition:

\[ Au(0) + Bu(1) = 0 \]  

(A3.5b)

The eigenfunctions can be determined up to a multiplicative constant. Hence \( u(0) \) may be given an arbitrary value. We have two equations for two unknowns: the state variable \( y \) and the bifurcation parameter \( \mu \). Shooting technique is available as solution method.

For the heat-integrated PFR, the FEHE, furnace and steam-generator differential equations defining the fold points may be integrated analytically to give:

\[ \frac{du_r}{d\xi} = B \cdot Da \cdot \exp \left[ \frac{\phi \theta_r}{1 + \theta_r} \right] \left( \left( 1 - \frac{\theta_r - \theta_r(0)}{B} \right) \frac{\gamma}{(1 + \theta_r)^2} = \frac{1}{B} \right) \cdot u_r, \]  

(A3.6a)

\[ \epsilon \cdot u_r(1) - u_r(0) = 0 \]  

(A3.6b)

The fold bifurcation points of the heat-integrated PFR were found by solving the boundary value problem defined by Eqs 3.6 and A3.6.

The computation of the cusp points (hysteresis variety) is based on the following observation: the loci of the ignition and extinction points meet at the cusp point.

For the case of one algebraic equation (Eq. 3.6) with one state variable (\( y = \theta_r(0) \)), the defining condition for the isola variety is:

\[ f(y, \mu, p) = \frac{\partial f(y, \mu, p)}{\partial y} = \frac{\partial f(y, \mu, p)}{\partial \mu} = 0 \]  

(A3.7)

Subramaniam and Balakotaiah (1996) showed how to compute the isola variety in distributed parameter systems. However, we used a simpler approach: at the isola point, the fold locus has a local extreme. Note that all the steady solutions corresponding to varying NTU (or \( \epsilon \)) pass through point I in Figure 3.2. Consequently, this point satisfies Eq. A3.7 if NTU (or \( \epsilon \)) is considered as bifurcation parameter.

If the fold point is located at a feasibility boundary \( y_b \) (or \( \mu_b \)) we speak about a boundary-limit point, defined by:

\[ f(y, \mu, p) = \frac{\partial f(y, \mu, p)}{\partial y} = 0 \]  

(A3.8)

\[ y = y_b \quad \text{or} \quad \mu = \mu_b \]
Chapter 3. Design of Heat-Integrated PFR

State multiplicity is possible for non-zero values of the FEHE efficiency ($\varepsilon$). When this parameter approaches the limit $\varepsilon = 1$, the conversion at the ignition and extinction points goes toward $X = 0$ and $X = 1$, respectively. In these cases, the heat balance Eqs 3.6b and 3.8 reduce to Eqs. 3.11 and 3.12.

At Hopf bifurcation points, a pair of complex eigenvalues crosses the imaginary axis. Substitution of $\lambda_1=0$ in Eq. A3.3 and identification of real and imaginary parts gives:

\begin{align}
\frac{\partial u}{\partial \xi} &= f_x u + C\lambda_2 v \\
\frac{\partial v}{\partial \xi} &= f_y v - C\lambda_2 u 
\end{align}

(A3.9a)

with the boundary conditions:

\begin{align}
Au(0) + Bu(1) &= 0 \\
Av(0) + Bv(1) &= 0 
\end{align}

(A3.9b)

As the eigenfunctions are determined up to a multiplicative constant, initial values $u(0)$ and $v(0)$ may be fixed, so the eigenvalue $\lambda_2$ and the bifurcation parameter may be found using a shooting method.

The loci of Hopf and fold bifurcation points meet at a double zero and the locus of Hopf points has an extreme at a double Hopf bifurcation. Tracing the locus of Hopf points required the main computing effort. The local parametrization technique (Seydel and Hlavacek, 1987) with secant predictor worked well (only the temperatures $\theta_1(0)$, $\theta_3(0)$, $\theta_4(0)$, and $\theta_4(0)$ were admitted as local parameters). A significant reduction of computing time was achieved by using the Broyden method (instead of Newton) as corrector. The step length control strategy recommended by Seydel (1984) was used.
Chapter 4 Stability and Multiplicity Approach to the Design of Heat-Integrated Multibed PFR

Abstract

The non-linear behaviour of the heat-integrated multibed plug flow reactor, consisting of feed-effluent heat exchanger (FEHE), furnace, multibed adiabatic tubular reactor with intermediate cooling, and steam-generator is studied. A first-order, reversible, exothermic reaction is considered. The hysteresis, isola, boundary-limit, double-zero and double-Hopf varieties are calculated. They divide the parameter space into regions with different steady-state and dynamic behaviour. State multiplicity, isolated branches and oscillatory behaviour may occur for realistic values of model parameters. Implications on design are discussed.
Chapter 4. Design of Heat-Integrated Multibed PFR

Introduction

The multibed, adiabatic, tubular reactor with intermediate cooling is the usual option when the reaction is exothermic, but equilibrium limited. Significant energy saving can be achieved if the reactor effluent is used to preheat the feed. Apparently, there is no need of an additional heat source. However, there are several reasons to add supplementary units:

1. A heater (furnace) is always required for start-up. It may be placed upstream or downstream of the feed-effluent heat exchanger (FEHE). It is favourable to place the FEHE before the furnace, as it will work at lower temperatures. Frequently, a purge stream is available as fuel.

2. The size of the FEHE limits the energy recovery. Consequently, a cooler is required. It is advantageous to place a steam-generator before the FEHE, as it will allow heat recovery at high temperature.

3. In autothermal reactors, multiple steady-states may exist. The classic analysis of van Heerden (1958) shows that the middle steady-state is always unstable. Moreover, stability can be lost due to Hopf bifurcation. In such cases, stable operation is possible only if a control system is in place. If the furnace and steam-generator are included, their duties are available as manipulated variables in temperature control loops.

![Diagram of Heat-integrated multibed PFR](image)

**Figure 4.1. Heat-integrated multibed PFR**
The inclusion of furnace and steam-generator leads to the structure presented in Figure 4.1. This structure, with the reactor playing the role of a heat pump, is called heat-integrated multibed PFR. This site integration solution is more efficient than process-process heat integration, as proposed by Douglas (1988).

Typical initial design data are the condition of the feed stream; reactor type (empty tube or packed bed; and the required conversion. Reaction kinetics and thermodynamics are also known. During conceptual design, the designer is mainly interested in beds' volume, FEHE area, and furnace and steam-generator duties. An optimisation problem, according to some steady-state related economic criteria, can be formulated. The solution is expected to recommend a small reactor because this is an expensive unit. The difference between the heat provided in the furnace and the heat recovered in the steam-generator should be low, in order to achieve energy saving. Moreover, their absolute values should be related to the plant energy balance.

However, the operating conditions may be different. There is always uncertainty of the kinetic parameters. Feed flow rate or concentration may deviate from the design values. Disturbances in feed temperature, furnace and steam-generator duties are unavoidable. FEHE efficiency may change due to fouling. Therefore, the operating point may become unstable and / or sustained oscillations may occur. In addition, the system may exhibit high sensitivity, which makes control difficult. Consequently, the design must be assessed with regard to steady-state multiplicity, stability and sensitivity issues.

Several articles dedicated to the nonlinear behaviour of the heat-integrated reactors have been published. Lovo and Balakotaiah (1992) computed the uniqueness-multiplicity boundary of the tubular reactor with internal or external heat exchange and CSTR with external heat exchange. For limiting cases, they presented analytical expressions of the ignition, extinction, and cusp points. Subramanian and Balakotaiah (1996) classified the steady-state and dynamic behaviour of several distributed reactor models, including the CSTR with external heat exchange and the tubular reactor with internal heat exchange. Silverstein and Shinnar (1982) evaluated the stability of the heat-integrated PFR using the frequency response of individual equipment components.

Although singularity theory (Balakotaiah and Luss, 1982, 1984, Golubitsky and Schaeffer, 1985) and bifurcation theory (Iooss and Joseph, 1981, Guckenheimer and Holmes, 1983, Scott, 1993) proved their power during the analysis of nonlinear systems, few
applications to design are reported. Russo and Bequette (1995) studied the influence of design parameters on the multiplicity behaviour of the jacketed exothermic CSTR. Heiszwolf and Fortuin (1997) presented an application-oriented design procedure for unique and stable operation of first-order reaction systems in a CSTR. Morud and Skogestad (1998) discussed the dynamics of an industrial multibed ammonia reactor, where positive feedback due to heat integration led to oscillatory behaviour. They showed that instability occurs at a Hopf bifurcation point. Khinast et al. (1998) analysed the continuously stirred decanting reactor. They computed loci of codimension-2 singular points, which divide the space of the design parameters into regions with different steady-state and dynamic bifurcation diagrams. This way, desirable regions of operation and potential stability or operability issues were identified.

In a recent article (Bildea and Dimian, 1998) the heat-integrated adiabatic PFR where a first-order, irreversible, exothermic reaction takes place, was investigated. Its nonlinear behaviour was studied. A design procedure was proposed to ensure a desired multiplicity pattern and a stable point of operation, and to avoid high sensitivity. The procedure was applied to three reaction systems with different kinetic and thermodynamic characteristics.

This work analyses a more complex system. We consider a first-order, reversible, exothermic reaction, and adiabatic operation of two-bed tubular reactor. We present the model equations and the types of bifurcation of interest. Afterwards, we classify the steady-state and dynamic behaviour by computing the hysteresis, isola, boundary-limit, double-zero and double-Hopf varieties. State multiplicity, isolated branches and oscillatory behaviour are possible for realistic values of model parameters. Subsequently, we discuss how the results may be used to avoid operational problems.

**Model equations**

The dynamic behaviour of the heat-integrated PFR is described by plug-flow, pseudo-homogeneous models. The dimensionless equations are given below:

**FEHE (tube and shell-side):**

\[
M_t \frac{\partial \theta_t}{\partial \tau} = - \frac{\partial \theta_t}{\partial \xi} + NTU \cdot (\theta_s - \theta_t)
\]

\[
M_s \frac{\partial \theta_s}{\partial \tau} = \frac{\partial \theta_s}{\partial \xi} - NTU \cdot (\theta_s - \theta_t)
\]

(4.1a)
Chapter 4. Design of Heat-Integrated Multibed PFR

Furnace:

\[ M_a \frac{\partial \theta_a}{\partial \tau} = - \frac{\partial \theta_a}{\partial \xi} + Q_a \]  

(4.1b)

First bed:

\[ \frac{\partial X_1}{\partial \tau} = - \frac{\partial X_1}{\partial \xi} + Da \cdot r(X_1, \theta_1) \]  

(4.1c)

\[ Le \frac{\partial \theta_1}{\partial \tau} = - \frac{\partial \theta_1}{\partial \xi} + B \cdot r(X_1, \theta_1) \]

Intermediate cooler:

\[ M_{cl} \frac{\partial \theta_{cl1}}{\partial \tau} = - \frac{\partial \theta_{cl1}}{\partial \xi} - Q_{cl1} \]  

(4.1d)

\[ M_{cl} \frac{\partial X_{cl1}}{\partial \tau} = - \frac{\partial X_{cl1}}{\partial \xi} \]

Second bed:

\[ \frac{\partial X_2}{\partial \tau} = - \frac{\partial X_2}{\partial \xi} + \alpha \cdot Da \cdot r(X_2, \theta_2) \]  

(4.1e)

\[ Le \frac{\partial \theta_2}{\partial \tau} = - \frac{\partial \theta_2}{\partial \xi} + \alpha \cdot B \cdot r(X_2, \theta_2) \]

Steam-generator:

\[ M_{cl} \frac{\partial \theta_{cl2}}{\partial \tau} = - \frac{\partial \theta_{cl2}}{\partial \xi} - Q_{cl2} \]  

(4.1f)

with appropriate initial and boundary conditions.

For a first-order, reversible reaction, the dimensionless reaction rate is given by:

\[ r(X, \theta) = e^{\phi (l+\theta)} \cdot (1 - X) \cdot \left( 1 - \phi \cdot e^{\phi (l+\theta)} \cdot \frac{X}{1 - X} \right) \]  

(4.2)

The model variables are dimensionless temperatures in the shell and tube-side of the FEHE (\( \theta_a, \theta_i \)), furnace (\( \theta_0 \)), reactor (\( \theta_1, \theta_2 \)), intermediate cooler (\( \theta_{cl1} \)), and steam-generator (\( \theta_{cl2} \)), and conversion (\( X \)). The parameters represent reactor residence time (\( Da \)), FEHE area (\( \varepsilon \)), activation energy (\( \gamma \)), adiabatic temperature rise (\( B \)), furnace, cooler and steam-generator duties (\( Q_a, Q_{cl1}, Q_{cl2} \)), feed temperature (\( \theta_0 \)), residence time ratios (\( \alpha, M_a, M_i, M_b, M_{cl1}, M_{cl2} \)), pre-exponential factors ratio (\( \phi \)), heat capacity (\( \delta \)) and heat capacity ratio (\( Le \)).

To study the steady-state and dynamic behaviour with varying feed temperature, only one dimensionless parameter must depend on it; hence, the reference temperature \( \bar{T} = T_0 \) is not
a good choice. For this reason, we follow Heiszwolf and Fortuin (1997) and arbitrarily set \( \gamma = 25 \). Then we calculate the corresponding temperature and use it as reference. In this way, changes of furnace, cooler or steam-generator duty, feed temperature and concentration, and FEHE efficiency are reflected by independent changes of the model parameters: \( Q_h, Q_{c1}, Q_{c2}, \theta_h, B, \) and \( \varepsilon \), respectively. Chen and Luss (1989) used a similar procedure setting \( Da = 1 \).

The steady-state model is obtained by dropping the time derivatives in Eq. 4.1. After analytical integration of the FEHE, furnace, cooler and steam-generator equations, the following boundary value problem is obtained:

\[
\begin{align*}
\frac{\partial X_1}{\partial \xi} &= Da \cdot r(X_1, \theta_1) \\
\frac{\partial \theta_1}{\partial \xi} &= B \cdot r(X_1, \theta_1) \\
\frac{\partial X_2}{\partial \xi} &= \alpha \cdot Da \cdot r(X_2, \theta_2) \\
\frac{\partial \theta_2}{\partial \xi} &= \alpha \cdot B \cdot r(X_2, \theta_2) \\
X_1(0) &= 0 \\
X_2(0) &= X_1(1) \\
\theta_2(0) &= \theta_1(1) - Q_{c1} \\
g(\theta_1(0), p^*) &= (1 - \varepsilon)\theta_0 + (Q_h - \varepsilon Q_{c2}) + \varepsilon \cdot \theta_2(1) - \theta_1(0) = 0
\end{align*}
\]

where:

\[
\varepsilon = \frac{NTU}{1 + NTU}
\]

is the FEHE efficiency

\[
p^* = (\theta_0, Q_h, Q_{c1}, Q_{c2}, \varepsilon, B, \gamma, \phi, \delta, Da, \alpha)
\]

is the vector of model parameters.

The methodology we will use to classify the steady-state and dynamic behaviour may be applied to systems that are described by a single intrinsic variable. This implies that, at least theoretically, it is possible to reduce the model equations to one equation with one variable. To demonstrate the validity of this assumption, we outline one possible approach to the solution of model equations: start with an initial guess for \( \theta_1(0) \), integrate Eqs. 4.3a-c to find \( \theta_2(1) \), check the boundary condition (Eq. 4.3d) and update \( \theta_1(0) \).
**Bifurcation Points**

In this section, we will introduce the types of bifurcation occurring in the heat-integrated multibed PFR. The reader is referred to the cited literature on bifurcation and singularity theories for a mathematically rigorous presentation. Details on the bifurcation points computation are given in the Appendix.

The S-shaped curves (bifurcation diagrams) in Figure 4.2 represent the conversion vs. furnace duty (bifurcation variable), for a given set of values of model parameters. Each curve is associated with a fixed value of NTU. The number of possible steady-states may change at *fold bifurcation points*. The upper and lower branches of the fold curve correspond to reaction *extinction* and *ignition*, respectively.

The middle steady-state is always unstable (dashed-line). A steady-state may also become unstable at *Hopf bifurcation point*. Here a branch of oscillating solution arises.

![Figure 4.2. Conversion vs. furnace duty for different values of the FEHE efficiency.](image)

Figure 4.2. Conversion vs. furnace duty for different values of the FEHE efficiency. 
\(\gamma=25, \alpha=0.293, \beta=0.50, D\theta=0.00361, \alpha=3.54, \phi=10^3, Q_{cl}=0.214, Q_{el}=0.286, \theta_i=-0.325, Le=2000\). Continuous lines represent stable steady-states. Dashed lines represent unstable steady-states. Bold lines are the loci of fold and Hopf bifurcations. Points C, DH, DZ, I and BL correspond to cusp, double-Hopf, double-zero, isola and boundary-limit bifurcations. When multiple steady-states exist, the middle one is always unstable (dashed line).
When the FEHE efficiency is fixed, the feed temperature, furnace and steam-generator duties may be coupled in one parameter, which will be called net energy input:

\[ Q = (1 - \varepsilon) \cdot \theta_h + Q_h - \varepsilon Q_c \]  

(4.5)

Note that \( \theta_h, Q_h \) and \( Q_c \) appear coupled as \( Q \) in the steady-state equation (Eq. 4.3) and they do not appear in the equations describing the bifurcation points (Eqs A4.5 and A4.7).

The qualitative shape of the bifurcation diagram changes when the second parameter \( (\varepsilon) \) is varied. At the cusp point (C) two fold points appear or disappear and the number of steady-states changes by two. The locus of cusp points is called hysteresis variety.

The bifurcation diagram may also change when isola variety is crossed (point I in Figure 4.2). Here, two separate solution branches coalesce. The existence of the isola variety depends on the choice of the bifurcation parameter. For the heat-integrated multibed PFR, the isola variety exists when the FEHE efficiency \( (\varepsilon) \) is the bifurcation parameter.

When the \( \varepsilon \) parameter has very large values \( (\varepsilon \rightarrow 1) \) the bifurcation parameter on the ignition and extinction branches reaches the asymptotic values \( Q_h=0.5 \) and \( Q_h=0.28 \) (not shown in Figure 4.2), respectively. They are called boundary-limit points (BL).

For a first-order reaction, the heat-integrated multibed PFR exhibits at most three steady-states, hence the double-limit variety (where the qualitative nature of the bifurcation diagram may also change) does not exist.

The number of Hopf bifurcation points may also change. At the double-Hopf (DH) bifurcation point two Hopf bifurcation points appear or disappear. At double-zero (DZ) bifurcation point, a Hopf bifurcation point appears or disappears.

**Steady-state behaviour**

First, we consider the net energy input as bifurcation parameter. It captures the influence of feed temperature, furnace and steam-generator duties on the steady-state. In this case the existence of multiple steady-states depends on the (fixed) value of FEHE efficiency, but is independent on how the net energy input is distributed between feed temperature, furnace and steam-generator duties.

For the multibed, adiabatic reactor where an exothermic, reversible reaction takes place, one can choose the thermal policy (beds inlet and outlet temperatures) in order to get a reactor with minimum volume. However, computation of the fold bifurcation points revealed
that, when state multiplicity exists, the required energy input is dangerously close to the extinction value (Figure 4.3).

![Figure 4.3. Extinction, ignition and required energy input for optimum reactor design](image)

Therefore, the FEHE efficiency should be chosen in the unicity region, which is bounded by the hysteresis variety. We remark that extinction may not be a problem when the thermal policy is sub-optimal. In this case the FEHE efficiency should be chosen so that the operating point is far enough from the cusp point, so high sensitivity is not a concern.

The steady-state behaviour is more complex when we consider the FEHE efficiency as bifurcation parameter. In this case, no other parameters are allowed to depend on it. Hence, \( \theta_b, Q_s \) and \( Q_{c2} \) can not be coupled in a single parameter \( Q \), and their effect must be investigated separately.

Figure 4.4 presents a typical phase diagram in the \( (Q_{c2}, Q_s) \) space. In addition to the hysteresis variety, one isola and two boundary-limit varieties exist. The isola variety approaches asymptotically the BL\(_1\) and BL\(_2\) varieties. The isola and hysteresis varieties intersect at a pitchfork bifurcation point.
These singularities divide the parameter space into eight regions where qualitative different bifurcation diagrams exist (they are presented in Figure 4.5). In region I there is a unique steady-state. Crossing the hysteresis variety to region II, two fold points appear and state multiplicity is possible. When the BL$_1$ variety is crossed to region III, a low-conversion isolated branch appears and the multiplicity pattern becomes 1-3-1-3. The two branches coalesce when the isola is intersected and two fold points disappear. Hence, in region IV, a low conversion branch exists for all values of FEHE efficiency, while high conversion is possible only for high values of the $\varepsilon$ parameter. When moving to regions V or VII, the high conversion branch disappears (at BL$_2$), or multiple states appear on the low-conversion branch (at hysteresis), respectively. From region VII, the high conversion branch disappears at BL$_2$ (region VI) or the two branches coalesce at isola (region VIII).
Dynamic behaviour

Dynamic classification involves dividing the parameter space into regions where a different number of Hopf bifurcation points exists. Due to the large number of parameters, a complete classification is difficult. We consider the energy input as the bifurcation parameter and restrict our analysis to the influence of the FEHE efficiency and dimensionless adiabatic temperature rise. We consider a catalytic reactor because its “wrong-way” behaviour (Mehta et al., 1981) is the cause of oscillations (and hence of Hopf bifurcations). This is reflected in the large value of the $Le$ parameter.
A typical phase diagram is presented in Figure 4.6. The cusp, double-zero and double-Hopf varieties divide the ($\varepsilon$–$B$) space into regions where different types of bifurcation diagrams exist. The bifurcation diagrams of conversion vs. energy input are presented in Figure 4.7. In region I, one stable steady-state exists. After crossing the double-Hopf locus to region II, two Hopf points appear. In-between, no stable steady-state exists and the system exhibits oscillatory behaviour. When the hysteresis variety is crossed, two fold points appear. Therefore, in region III there are multiple steady-states. Oscillatory behaviour is possible on both the ignited and extinguished branches. In region IV, after crossing the double-zero locus, one Hopf point disappears and oscillatory behaviour is possible only on the ignited branch.

![Phase diagram of the heat-integrated multibed PFR with energy input as bifurcation parameter.](image)

Figure 4.6. Phase diagram of the heat-integrated multibed PFR with energy input as bifurcation parameter.

$\gamma = 25, B = 0.293, \delta = 50.50, Da = 0.00361, \alpha = 3.54, \varphi = 10^4, Q_{c1} = 0.214, Le = 2000$.

The different types of bifurcation diagrams existing in regions I-IV are presented in Figure 4.7.
Figure 4.7. Conversion vs. energy input bifurcation diagrams
The diagrams I-IV correspond to regions I-IV in Figure 4.6.
○ - fold bifurcation; ■ - Hopf bifurcation

Design considerations

During the conceptual design, we are mainly interested in the reactor thermal policy and the values of FEHE efficiency, furnace and steam-generator duties.

According to economic criteria, optimisation will give close to minimum reactor volume (an expensive unit) and high FEHE efficiency (for energy savings). However, the operating point will be in the multiplicity region, near the extinction point. When the net energy input accidentally decreases, the system will be driven in the unicity region, where only the extinguished state is possible. In this case, a special procedure will be required to restart the reaction. For these reasons, when the reactor had been designed close to its optimum, operation in the multiplicity region is not recommended. Because the hysteresis variety bounds the unicity region, FEHE efficiency should be less than the value corresponding to the cusp bifurcation. We remark that the system exhibits high sensitivity if the operating point is close to the hysteresis. This may be undesirable. Also, note that state unicity does not ensure stability.
One can increase the energy saving by reactor overdesign (sub-optimal thermal policy). In this case the system exhibits input multiplicity, i.e. the same conversion may be obtained on the middle or on the upper branch. In the first case, the net energy input is lower, but the operating point is always unstable. In the second case, the operating point may be stable or unstable. Dynamic simulation can be used to assess the stability, but no information about the extent of the stability region will be obtained. We recommend the computation of the Hopf bifurcation points (which are on the boundary of the oscillatory region) to find possible instability near the operating point. Note that working at an unstable operating point is possible, as a temperature controller may stabilise the system. More dangerous is the loss of stability due to disturbances or design parameter uncertainty.

When FEHE efficiency is constant, the number of steady-states and their stability depends only on the net energy input ($Q$) and does not depend on how this is distributed between feed temperature, furnace and steam-generator duties. Consequently, these variables can be linked to plant energy balance, site integration, combined heat and power production or low equipment cost considerations. To prevent operational problems due to isolated branches when FEHE efficiency may change, operation in region I of Figure 4.4 should be preferred.

**Conclusions**

Nonlinear behaviour of a multibed heat-integrated PFR was studied, for a first-order, reversible reaction. Hysteresis, isola, boundary-limit, double-Hopf and double-zero varieties were used to classify the steady-state and dynamic behaviour of the system. State multiplicity, isolated branches and oscillatory behaviour are possible for realistic values of the model parameters. The phase diagrams are similar with those obtained for different adiabatic reactor models, as reported by Subramanian and Balakotaiah (1996) and Bildea and Dimian (1998).

There is a strong relationship between the design of individual pieces of equipment and the non-linear behaviour of the whole system. The results of significance are:

1. If the reactor is designed for a minimum volume, the selection of FEHE efficiency is constrained by the requirement of state unicity. This design rule ensures against reaction extinction due to thermal disturbances. The rejection of disturbances becomes difficult before state multiplicity appears (near the cusp bifurcation point). Consequently, optimal reactor design is robust only coupled with a proper FEHE efficiency.
2. When energy saving is of interest, a large FEHE must be used to achieve autothermal operation (defined by the existence of an ignited state). However, the reactor thermal policy should be sub-optimal, resulting in reactor overdesign.

3. The same conversion may be obtained for two different values of the net energy input (feed enthalpy, furnace and steam-generator duties). When multiple steady-states exist, the operating point corresponding to low net energy input is always unstable. Even when only one steady-state exists, its stability is not guaranteed. Hence, stabilising control may be necessary. Alternatively, stability can be achieved by changing the design such that the operating point corresponds to high net energy input.

Bifurcation and singularity theories may be applied to similar problems, where the design is constrained by state multiplicity and instability. In this way, desirable regions of operation are identified and potential stability or operability problems are avoided.

**Notation**

\( A_T \) = FEHE area, \( m^2 \)

\( B \) = adiabatic temperature rise, dimensionless

\[ = \frac{\Delta T_{\text{ad}}}{T} \]

\( c_p \) = specific heat, \( J/(kg \, K) \)

\( Da \) = Damkohler number, dimensionless

\[ = k(T)^{\frac{V_p}{F}} \]

\( E_a \) = activation energy, \( J/mol \)

\( F \) = mass flow rate, \( kg/s \)

\( K_T \) = FEHE heat transfer coefficient, \( W/(m^2 \, K) \)

\( k_i \) = pre-exponential factor (i=1,-1), \( s^{-1} \)

\( L \) = length, \( m \)

\( Le \) = Lewis number, dimensionless

\[ = 1 + \frac{1 - \varepsilon_{\text{fl}}}{\varepsilon_{\text{fl}}} \cdot \frac{\rho_d \cdot c_p_{\text{fl}}}{\rho_{\text{fl}} \cdot c_{p,\text{fl}}} \]

\( M_k \) = residence time ratio (k=t, s, c1, c2)

\[ = \frac{t_{\text{res}}}{t_{01}} \]
\[ NTU = \text{number of transfer units, dimensionless} \]
\[ = \frac{K_i A_r}{F \rho_c} \]

\[ q_k = \text{duty (k = h, c1, c2), W} \]
\[ Q_k = \text{duty (k = h, c1, c2), dimensionless} \]
\[ = \frac{q_k}{F \rho_c \bar{T}} \]

\[ Q = \text{energy input, dimensionless} \]
\[ = (1-\varepsilon) \theta - \varepsilon Q_c + Q_h \]

\[ R = \text{gas constant, J/(mol K)} \]
\[ T = \text{temperature, K} \]
\[ \bar{T} = \text{reference temperature, K} \]
\[ \Delta T_{ad} = \text{adiabatic temperature rise, K} \]
\[ r = \text{reaction rate, mol/(m}^3\text{s)} \]
\[ t = \text{time, s} \]
\[ t_{0i} = \text{residence time (i=1,2), s} \]
\[ = \frac{V_i \rho}{F} \]

\[ u, v = \text{eigenfunctions} \]
\[ X = \text{conversion, dimensionless} \]
\[ z = \text{axial coordinate, m} \]
\[ V_i = \text{reactor volume (i=1,2), m}^3 \]

**Greek letters**
\[ \alpha = \text{residence time ratio, dimensionless} \]
\[ = \frac{t_{0i}}{t_{02}} \]
\[ \gamma = \text{activation energy, dimensionless} \]
\[ = \frac{E_{al}}{RT} \]
\[ \varepsilon = \text{FEHE efficiency, dimensionless} \]
\[ \varphi = \text{pre-exponential factors ratio, dimensionless} \]
Chapter 4. Design of Heat-Integrated Multibed PFR

\[
\frac{k_1}{k_{-1}} = \lambda
\]

\[
\lambda = \text{eigenvalue}
\]

\[
\rho = \text{density, kg/m}^3
\]

\[
\tau = \text{time, dimensionless}
\]

\[
\frac{\tau}{\tau_0} = \theta
\]

\[
\theta = \text{temperature, dimensionless}
\]

\[
\xi = \frac{z}{L}
\]

\[
u, \psi = \text{eigenfunctions}
\]

**Subscripts**

- c1 - intermediate cooler
- c2 - steam-generator
- h - furnace
- s - FEHE shell side
- sf - solid phase
- ff - fluid phase
- t - FEHE tube side
- 0 - feed
- 1 - first reactor bed
- 2 - second reactor bed
- b - feasibility boundary

**Literature cited**


Chapter 4. Design of Heat-Integrated Multibed PFR


Subramanian, S. and V. Balakotaiah, “Classification of Steady-State and Dynamic Behaviour
Chapter 4. Design of Heat-Integrated Multibed PFR


Appendix. Bifurcation points computation

To derive the defining equation of the fold points, we consider the condensed form of the mathematical model:

\[ C \frac{\partial y}{\partial \tau} = - \frac{\partial y}{\partial \xi} + f(y, \mu, p) \]  \hspace{1cm} (A4.1a)

\[ Ay(\tau,0) + By(\tau,1) - c = 0 \]

\[ y(0,\xi) = y_0(\xi) \]  \hspace{1cm} (A4.1b)

where \( y \) is the vector of model variables; \( C \) is a diagonal capacity matrix; \( f(y) \) is a non-linear function; \( \mu \) is the bifurcation parameter; and \( p \) is the vector of remaining parameters. \( A \) and \( B \) are constant matrices and \( c \) is a constant vector.

Its steady-state solution is given by the boundary value problem:

\[ \frac{\partial y_s}{\partial \xi} = f(y_s, \mu, p) \]  \hspace{1cm} (A4.2a)

\[ Ay_s(0) + By_s(1) - c = 0 \]  \hspace{1cm} (A4.2b)

When equation (A4.1) is linearized around steady-state and separation of variables is applied, the following eigenvalue problem is obtained:

\[ C(\lambda + \lambda \xi i)(u + iv) = d(u + iv) \frac{d^2}{d\xi^2} + f_y \cdot (u + iv) \]  \hspace{1cm} (A4.3)

where:

\[ \delta_y = (y(\xi,\tau) - y_s(\xi)) = (u(\xi) + iv(\xi)) \cdot e^{(\lambda_1+\lambda_2)\tau} \]  \hspace{1cm} (A4.4)

At fold points, one real eigenvalue crosses the imaginary axis. Substitution of \( \lambda_1=\lambda_2=0 \) in Eq. A4.3 and identification of real and imaginary parts gives:

\[ \frac{du}{d\xi} = f_y \cdot u \]  \hspace{1cm} (A4.5a)

with the boundary condition:
Chapter 4. Design of Heat-Integrated Multibed PFR

\[ Au(0) + Bu(1) = 0 \quad (A4.5b) \]

The eigenfunctions can be determined up to a multiplicative constant. Hence \( u(0) \) may be given an arbitrary value. We have two equations for two unknowns: the state variable \( y \) and the bifurcation parameter \( \mu \). Shooting technique is available as solution method.

At Hopf bifurcation points, a pair of complex eigenvalues crosses the imaginary axis. Two additional conditions must be satisfied:

\[ - \frac{d\lambda_1}{d\mu} \neq 0 \quad (A4.6a) \]

- the real parts of the other eigenvalues are negative. \( (A4.6b) \)

Substitution of \( \lambda_1 = 0 \) in Eq. A4.3 and identification of real and imaginary parts gives:

\[ \frac{\partial u}{\partial \xi} = f_yu + C\lambda_2 v \]
\[ \frac{\partial v}{\partial \xi} = f_yv - C\lambda_2 u \quad (A4.7a) \]

with the boundary conditions:

\[ Au(0) + Bu(1) = 0 \]
\[ Av(0) + Bv(1) = 0 \quad (A4.7b) \]

As the eigenfunctions are determined up to a multiplicative constant, initial values \( u(0) \) and \( v(0) \) may be fixed, so the eigenvalue \( \lambda_2 \) and the bifurcation parameter may be found using a shooting method.

The fold and Hopf points have codimension-1 because they fix the value of one parameter.

For the case of one algebraic equation (Eq. 4.3d) with one state variable \( (y = \theta_r(0)) \), the defining condition for the cusp variety is:

\[ g(y, \mu, p) = \frac{\partial g(y, \mu, p)}{\partial y} = \frac{\partial^2 g(y, \mu, p)}{\partial y^2} = 0 \quad (A4.8) \]

A direct method for computing the cusp variety in distributed parameter systems is presented by Witmer et al. (1986). However, we used a simpler approach based on the following observation: the loci of the ignition and extinction points meet at the cusp point.

The defining condition for the isola variety is:

\[ g(y, \mu, p) = \frac{\partial g(y, \mu, p)}{\partial y} = \frac{\partial g(y, \mu, p)}{\partial \mu} = 0 \quad (A4.9) \]
Point I in Figure 4.2 belongs to the locus of fold points; hence the first equality in Eq. A4.9 is satisfied. Moreover, it also satisfies the second equality in Eq. A4.9 because it is common to all steady-state solutions corresponding to varying \( \varepsilon \), hence, point I represents an isola bifurcation when \( \varepsilon \) is considered as bifurcation parameter. We note that it can be identified as an extreme of the fold locus.

If a fold point is located at a feasibility boundary (\( \varepsilon = 1 \)), we speak about a boundary-limit point (BL), defined by:

\[
\frac{\partial}{\partial y} g(y, \mu, p) = 0 \quad (A4.10a)
\]

\[
y = y_0 \quad \text{or} \quad \mu = \mu_0 \quad (A4.10b)
\]

At double-Hopf bifurcation, the transversality condition defined by Eq. A4.6a is broken. It can be identified as an extreme on the locus of Hopf points.

When double-zero variety is crossed, the condition A4.6b is no longer satisfied. The double-zero point can be identified as the intersection between the loci of Hopf and fold bifurcation points.

Cusp, isola, boundary-limit, double-Hopf and double-zero points fix the value of two parameters; hence, they are codimension-2 varieties.

When codimension-2 bifurcation points were calculated by continuation of codimension-1 varieties, tracing the locus of Hopf points required the main computing effort. The local parametrization technique (Seydel and Hlavacek, 1987) with secant predictor worked well (only the temperatures \( \theta_1(0), \theta_2(0), \theta_3(0), \theta_4(0), \theta_5(0) \) and \( \theta_6(0) \) were admitted as local parameters). A significant reduction of computing time was achieved by using the Broyden method (instead of Newton) as corrector. The step length control strategy recommended by Seydel (1984) was used.
Chapter 5 Interaction between Design and Control of Heat-Integrated PFR

Abstract

The interaction between design and control of a heat-integrated plug-flow reactor (PFR) is analysed. Four design alternatives and three control structures are considered. Linear controllability analysis shows that designs with small steam-generator and large feed-effluent heat exchanger (FEHE) cannot be stabilised. At steady-state, control structures using furnace duty as manipulated variable can reject disturbances in feed flow rate, steam-generator duty and FEHE fouling. However, fast control cannot be achieved. Moreover, there is no incentive to add a bypass around the FEHE. The nonlinear behaviour of the controlled system is analyzed using bifurcation theory. The points in the design parameter space are classified according to their position relative to the cusp and Hopf-and-pitchfork varieties. The range of design parameters for which the system cannot be stabilised is detected. The results of linear and nonlinear analysis are confirmed by dynamic simulation.
**Introduction**

In industrial processes where an exothermic reaction takes place, heat exchange between the effluent of an adiabatic reactor and the feed stream may be used for energy saving. There are several reasons to include additional units in the system:

- A heater (furnace) is required for startup. Moreover, positive feedback due to heat integration may lead to state multiplicity or instability. In this case, furnace duty can be the manipulated variable in a temperature control loop, in order to achieve stable operation.

- The heat excess must be removed in a cooler (steam-generator). Placing the steam-generator before the FEHE allows heat recovery at higher temperature and is therefore preferable in view of exergetic considerations.

- Rapid quench of the reactor effluent may be necessary to minimize coking and to avoid the fouling of heat-exchange units.

The resulting structure, presented in Figure 5.1, will be called heat-integrated PFR.

![Figure 5.1. Heat-integrated plug-flow reactor.](image)

The feed is preheated by the reactor effluent in the feed-effluent heat exchanger (FEHE). Start-up, control or plant energy balance may require additional units: furnace, quench, steam-generator.

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Although this structure is attractive from the viewpoint of energy saving, state multiplicity, isolated solution branches and oscillatory behaviour are possible. Hence, control problems are expected. A design procedure was proposed to ensure a desired multiplicity pattern and a stable point of operation, and to avoid high sensitivity (Bildea and Dimian, 1998).

Silverstein and Shinnar (1982) evaluated the stability of a very similar system, using the frequency response of the individual equipment components. They considered the multiplicity region and concentrated on the intermediate, open-loop unstable operating point. A control loop, maintaining the furnace exit temperature by adjusting the fuel rate to the furnace, could stabilise the system. The authors pointed out that industrial furnaces are normally sluggish and a bypass around FEHE could provide a manipulated variable to achieve quick control.

Tyreus and Luyben (1993) analyzed a reactor / preheater process. Reactor dynamics (dead-time and inverse response) was captured by a transfer function containing a gain, a positive zero, dead-time and two first-order lags. The dynamics of the FEHE was neglected. The coupled system was unstable for loop gain greater than unity. Bypass around the FEHE was used to stabilise the system. The authors pointed out that, contrary to conventional wisdom, the addition of integral action in the control loop improved closed loop stability.

Luyben et al. (1998) considered the control of a heat-integrated PFR, without steam-generator. Using nonlinear dynamic simulation, they showed that systems with a large furnace (and low heat integration) are easier to control.

This work studies the interaction between design and control of a heat-integrated PFR. In the next section, the nonlinear, dynamic model is presented. It is shown that two design parameters have to be set during conceptual design: FEHE efficiency and steam-generator duty. Four different design alternatives, for which controllability problems are expected, are investigated. Three different control structures are considered. Linear controllability analysis shows that systems with small steam-generator and large FEHE are difficult to stabilise. Bypass around the FEHE can not be used to reject disturbances and there is no incentive to control both FEHE and furnace outlet temperatures. Next, the nonlinear behaviour of the controlled system is analyzed by bifurcation theory. The range of design parameters for which the system can not be stabilised is detected. The operating points are classified according to
their position relative to different bifurcation varieties. Doing this, the meaning of “small” or “large” units becomes clear. The results are confirmed by nonlinear dynamic simulation.

**Design considerations and model equations**

During conceptual design, the reactor volume, FEHE efficiency, furnace and steam-generator duties are of interest. Moreover, detailed equipment sizing is not necessary.

Feed flow rate \( (F) \), reactor inlet temperature \( (T_2) \) and the conversion \( (X) \) are typical initial data. Consequently, the reactor may be designed if reaction kinetics and thermodynamics (adiabatic temperature rise, \( \Delta T_{ad} \)) are known.

The first design decision refers to FEHE efficiency \( (\varepsilon) \). The size of FEHE determines the multiplicity pattern, the stability of the operating point and the sensitivity to energy disturbances. Bildea and Dimian (1998) discuss this issue in detail.

When the FEHE efficiency is known, the net energy requirement \( (q) \) can be calculated:

\[
q = (1-\varepsilon) \cdot Fc_p T_0 + (q_b - \varepsilon \cdot q_c) = (1-\varepsilon) \cdot Fc_p T_2 - \varepsilon \cdot Fc_p \Delta T_{ad} X
\]

(5.1)

However, it must be split between feed enthalpy \( (T_0) \), furnace and steam-generator duties \( (q_b, q_c) \). Considering fixed feed temperature, the second decision concerns the steam-generator duty. Note that the furnace duty can be calculated from the overall energy balance.

In this article, we consider a case where moderate conversion is required. Table 5.1 presents the kinetic, thermodynamic and design parameters. They correspond to the toluene hydrodealkylation (Douglas, 1988). First-order kinetics can be assumed because of the large hydrogen excess. For this reaction, the boundary between unicity and multiplicity regions is located at a rather low value of FEHE efficiency \( (\varepsilon^* = 0.32) \). Four design alternatives are presented in Table 5.2.

Design 1 has a small FEHE \( (A_{st}=130 \text{ m}^2, \varepsilon=0.28) \). The operating point is in the unicity region, but close to the unicity-multiplicity boundary. Design 2 has a large FEHE \( (A_{st}=1500 \text{ m}^2, \varepsilon=0.745) \). Hence, the operating point is in the multiplicity region. In Designs 1A and 2A, a large quantity of energy is used to generate steam, and a large amount of energy is provided in the furnace. Conversely, less steam is generated in Designs 1B and 2B, a smaller furnace being necessary. Note that energy recovered in the FEHE increases from Design 1A to Design 2B.
Table 5.1. Kinetic, thermodynamic and design parameters

<table>
<thead>
<tr>
<th>Feed</th>
<th>Feed flowrate, ( F ) (kg/s)</th>
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</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>temperature, ( T_0 ) (°C)</td>
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<tr>
<td></td>
<td>pressure, ( p ) (atm)</td>
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</tr>
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<td></td>
<td>reactant mole fraction, ( y )</td>
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</tr>
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<td></td>
<td>specific heat, ( c_p ) (J/(kg K))</td>
<td>4000</td>
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<tr>
<td>Reactor</td>
<td>reaction enthalpy, ( \Delta H_r ) (J/mol)</td>
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<td></td>
<td>pre-exponential factor, ( k_0 ) (s(^{-1}))</td>
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<tr>
<td></td>
<td>activation energy, ( E_a ) (J/(mol K))</td>
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</tr>
<tr>
<td></td>
<td>conversion, ( X )</td>
<td>0.75</td>
</tr>
<tr>
<td></td>
<td>inlet temperature, ( T_2 ) (°C)</td>
<td>637</td>
</tr>
<tr>
<td>FEHE</td>
<td>heat transfer coefficient, ( K_{st} ) (W/(m(^2) K))</td>
<td>100.0</td>
</tr>
<tr>
<td></td>
<td>bypass, ( 1-\alpha )</td>
<td>0.2</td>
</tr>
<tr>
<td>Furnace</td>
<td>tube diameter, ( d_w ) (m)</td>
<td>0.05</td>
</tr>
<tr>
<td></td>
<td>tube wall thickness, ( \delta_w ) (m)</td>
<td>0.005</td>
</tr>
<tr>
<td></td>
<td>tube density, ( \rho_w ) (kg/m)</td>
<td>7800</td>
</tr>
<tr>
<td></td>
<td>tube specific heat, ( c_{p,w} ) (J/(kg K))</td>
<td>2000</td>
</tr>
<tr>
<td></td>
<td>heat transfer coefficient, ( K_{wf} ) (W/(m(^2)K))</td>
<td>200</td>
</tr>
<tr>
<td></td>
<td>tube temperature, ( T_w ) (°C)</td>
<td>800</td>
</tr>
</tbody>
</table>

Table 5.2. Design alternatives of the HDA heat-integrated reactor.

<table>
<thead>
<tr>
<th></th>
<th>Design 1A</th>
<th>Design 1B</th>
<th>Design 2A</th>
<th>Design 2B</th>
</tr>
</thead>
<tbody>
<tr>
<td>Furnace</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>duty, ( q_b /10^3 ) (W)</td>
<td>16436</td>
<td>13969</td>
<td>12665</td>
<td>6011.2</td>
</tr>
<tr>
<td>area, ( A_{wf} ) (m(^2))</td>
<td>223.9</td>
<td>205.3</td>
<td>194.48</td>
<td>121.9</td>
</tr>
<tr>
<td>Steam-generator</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>duty, ( q_c /10^3 ) (W)</td>
<td>10576.3</td>
<td>1047</td>
<td>10577.5</td>
<td>1048.96</td>
</tr>
<tr>
<td>( T_5 ) (°C)</td>
<td>297</td>
<td>588</td>
<td>297</td>
<td>588</td>
</tr>
<tr>
<td>FEHE</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>efficiency, ( \epsilon )</td>
<td>0.28</td>
<td>0.28</td>
<td>0.745</td>
<td>0.745</td>
</tr>
<tr>
<td>area, ( A_{st} ) (m(^2))</td>
<td>130.0</td>
<td>130</td>
<td>1500</td>
<td>1500</td>
</tr>
<tr>
<td>hot side outlet temperature, ( T_1 ) (°C)</td>
<td>104.7</td>
<td>186</td>
<td>228.8</td>
<td>445.5</td>
</tr>
<tr>
<td>cold side outlet temperature ( T_6 ) (°C)</td>
<td>223.85</td>
<td>435.6</td>
<td>102.7</td>
<td>181.9</td>
</tr>
<tr>
<td>duty / (10^3 ) (W)</td>
<td>2390.8</td>
<td>4992</td>
<td>6362</td>
<td>13295.8</td>
</tr>
</tbody>
</table>

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The assumptions made to develop the dynamic model are:

1. Reactor dynamics may be described by a plug-flow, pseudo-homogeneous model.
2. A detailed model of the steam-generator is not necessary, since we regard its duty as a disturbance.
3. For FEHE, an efficiency model is adequate because of fast dynamics.
4. A fast fuel flow rate – flue-gas temperature control loop is used to set the furnace duty at the prescribed value (Shinskey, 1988). Consequently, the heat transfer from burner to tubes (hot-side) is not included in the model. Moreover, temperature variation along the tubes is neglected and the cold-side heat-transfer coefficient is assumed to be constant. These assumptions allow an approximate furnace design (number of tubes, diameter, length, and thickness) and development of a model describing the essential dynamic behaviour.

The model equations are presented below:

**FEHE:**

\[
T_i = (1 - \varepsilon) \cdot T_0 + \varepsilon \cdot T_s
\]  

(5.2a)

**Furnace** (fluid, and tubes wall)

\[
\frac{\partial T_h}{\partial t} = -w_h \cdot \frac{\partial T_h}{\partial z} + \frac{4}{d_w} \cdot \frac{K_{wt}}{\rho \cdot c_p} \cdot (T_w - T_h)
\]  

(5.2b)

\[
\frac{\partial T_w}{\partial t} = \frac{1}{\rho \cdot c_{pw} \cdot V} \left( q_b \cdot \pi \cdot d_w \cdot K_{wt} \cdot \int_0^1 (T_w - T_h) dz \right)
\]  

(5.2c)

**Reactor**

\[
\frac{\partial C}{\partial t} = -w_r \cdot \frac{\partial C}{\partial z} - r(C, T_r)
\]  

(5.2d)

\[
\frac{\partial T_r}{\partial t} = -w_r \cdot \frac{\partial T_r}{\partial z} + \frac{(-\Delta H_r)}{\rho \cdot c_p} \cdot r(C, T_r)
\]  

(5.2e)

**Steam-generator**

\[
T_s = T_4 - \frac{q_c}{F \cdot c_p}
\]  

(5.2f)

Additionally, first-order lags of 30 seconds were considered for temperature measurements, FEHE bypass flow rate, and furnace duty.
Controllability analysis

The main control objective is to keep the reaction conversion and selectivity within acceptable limits. Their control demands composition analysers that are expensive, require maintenance and have unfavourable dynamics. However, both variables depend on the reaction temperature. Hence, we consider control structures that include the reactor inlet temperature as the main controlled variable.

Our analysis will evaluate stability and disturbance rejection properties of different design alternatives. We consider the furnace duty and bypass around FEHE as potential manipulated variables, and feed flow rate, steam-generator duty and FEHE heat transfer coefficient as disturbances.

Three different control structures are investigated (Table 5.3). First (CS 1), furnace duty is used to control the reactor inlet temperature. Secondly (CS 2), FEHE bypass and furnace inlet temperature are the manipulated and controlled variables, respectively. The setpoint of this loop can be used, in a cascade manner, to control reactor inlet temperature. Both furnace duty - reactor inlet temperature and FEHE bypass - furnace inlet temperature loops are used in the third control structure.

Table 5.3. Control structures

<table>
<thead>
<tr>
<th>Outputs</th>
<th>CS 1</th>
<th>CS 2</th>
<th>CS 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>$y_1$</td>
<td>$T_2$</td>
<td>$T_2$</td>
<td>$T_2$</td>
</tr>
<tr>
<td>$y_2$</td>
<td>-</td>
<td>-</td>
<td>$T_1$</td>
</tr>
<tr>
<td>Inputs</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$u_1$</td>
<td>$q_h$</td>
<td>$\alpha$</td>
<td>$q_h$</td>
</tr>
<tr>
<td>$u_2$</td>
<td>-</td>
<td>-</td>
<td>$\alpha$</td>
</tr>
<tr>
<td>Disturbances</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$d_1$</td>
<td>$F$</td>
<td>$F$</td>
<td>$F$</td>
</tr>
<tr>
<td>$d_2$</td>
<td>$q_c$</td>
<td>$q_c$</td>
<td>$q_c$</td>
</tr>
<tr>
<td>$d_3$</td>
<td>$K_{st}$</td>
<td>$K_{st}$</td>
<td>$K_{st}$</td>
</tr>
</tbody>
</table>
Linear analysis

The axial coordinate in Eqs. 5.2 was discretized by finite differences and the model was linearized around steady-state. The following state-space representation was obtained:

\[ \frac{dx(t)}{dt} = A \cdot x(t) + B \cdot u(t) + B_d \cdot d(t) \] (5.3a)
\[ y(t) = C \cdot x(t) + D \cdot u(t) + D_d \cdot d(t) \] (5.3b)
\[ e(t) = r(t) - y(t) \] (5.3c)

where \( x, u, d, y, e \) and \( r \) are the vectors of the state variables, manipulated inputs, disturbances, controlled outputs, control errors and setpoints, respectively. \( A, B, B_d, C, D \) and \( D_d \) are matrices of appropriate dimensions.

In order to obtain meaningful controllability results, the inputs, disturbances and outputs were scaled. In terms of scaled variables, the control objective is to keep the control error \( |e(t)| < 1 \), using manipulated inputs \( |u(t)| < 1 \), when disturbances \( |d(t)| < 1 \) affect the process. The scaling factors are 1 K for temperatures and 25% of the nominal value for other variables.

In terms of transfer functions, the linear model of the process is given by:

\[ y(s) = G(s) \cdot u(s) + G_d(s) \cdot d(s) \] (5.4)

where:

\[ G(s) = C \cdot (sI - A)^{-1} \cdot B + D \] (5.5a)
\[ G_d(s) = C \cdot (sI - A)^{-1} \cdot B_d + D_d \] (5.5b)

Skogestad and Postlewaite (1996) review the linear controllability indices. Linear models were used only for controllability analysis. The full nonlinear model was used to evaluate the performance of the control system.

Stability

Computation of the eigenvalues of the \( A \) matrix shows that the operating points are stable for Designs 1A and 1B, but unstable for Designs 2A and 2B.

Figure 5.2 presents a part of the root-locus plot for Design 2A and CS1 (similar results are obtained with CS2). Two eigenvalues located near the imaginary axis are of interest. They are denoted by \( \lambda_+ \) and \( \lambda_- \), according to their sign if the controller gain is zero.
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Figure 5.2. Root locus plot.
There is a limited range of controller gain that ensures stability.

When the controller gain is increased, $\lambda_+$ is shifted towards the left half plane and crosses the imaginary axes. Hence, there is a minimum controller gain for stability. In the same time, $\lambda$ moves to the right. When $\lambda_+$ and $\lambda$ meet, a pair of complex conjugate eigenvalues emerges and moves to the right half plane. Hence, there is also a maximum controller gain for stability.

For Design 2B, $\lambda_+$ and $\lambda$ meet in the right half plane, near the imaginary axis. Hence, for all controller gains, there is at least one eigenvalue located in the right half plane. Consequently, linear analysis predicts that Design 2B can not be stabilised by a P-controller.

For the heat-integrated PFR, integral controller action has the usual, destabilising effect. This is in contrast with the catalytic reactor / preheater process (Tyreus and Luyben, 1993), where integral action improved closed loop stability.

**Interactions**

For CS3 we compute the Relative Gain Array in order to evaluate the input-output pairing under decentralised control:

$$\Lambda = G \odot (G^{-1})^T \quad (5.6)$$
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The diagonal RGA elements for designs 1A, 1B, 2A and 2B are 4.274, 4.289, -0.954 and -0.964, respectively. Decentralised integral controllability requires positive values for stable systems, but negative values for unstable systems with one RHP pole. This condition is fulfilled in all cases.

Close-to-one diagonal elements denote little interaction. Small values in the RGA matrix also indicate that model uncertainty is not a problem.

The RGA_number, defined as:

\[ RGA\_number = \| I - \Lambda(j\omega) \|_{\text{sum}} \]  

has small value and falls to zero for high frequency (Figure 5.3), showing that good control performance is possible.

![Figure 5.3. CS3: RGA_number.](image)

- Design 1A; ■ - Design 1B; ▲ - Design 2A; × - Design 2B

RGA_number has small values and drops to zero at high frequency.

**Disturbance rejection**

To analyse the disturbance rejection properties, we calculate the Closed Loop Disturbance Gain (CLDG), defined by:

\[ \tilde{G}_d = \tilde{G} \cdot G^{-1} \cdot G_d \]  

where \( \tilde{G} \) is a matrix consisting of diagonal elements of \( G \).
Its elements, \( \tilde{g}_{ik} \), give the apparent gain of the \( k \)th disturbance on the \( i \)th output under decentralised control. For SISO systems, disturbance gain and CLDG are equivalent. The necessary condition to avoid inputs saturation is:

\[
|g_u| > |\tilde{g}_{ik}|, \quad \forall k
\]  

(5.9)

Figure 5.4 presents loop and disturbance gains when furnace duty is the manipulated variable (CS1). At low frequency, condition 5.9 is fulfilled for Design 1A and 2A. Hence, disturbances can be rejected. For Design 1B and 2B, it is necessary to slightly increase the maximum allowed control action, in order to avoid input saturation. It all cases, fast disturbances can not be rejected because condition 5.9 is not satisfied at high frequencies. Feed flow rate is the worst disturbance. Large steam-generator duty (Design 1A and 2A), is the second demanding disturbance. Note that fast change of FEHE heat transfer coefficient is not expected, so we are only concerned with low frequency response.

![Figure 5.4. CS1: Frequency-dependent (rad/s) loop and disturbance gains.](image)

- \( * \): \( g \), \( \ast \), \( \triangle \), \( \times \), \( g_{ik} \), \( k=1,2,3 \).
- Slow disturbances can be rejected, but fast control can not be achieved.
Diagrams in Figure 5.5 present the loop and disturbance gains when bypass around FEHE is the manipulated variable (CS2). Changes of the feed flow rate can not be handled. Disturbances in steam-generator duty can be rejected only if the steam-generator duty is small (Design 1B, 2B). Only Designs 2A and 2B can cope with FEHE fouling. On the whole, Design 2B (large FEHE, small steam-generator) performs the best.

![Figure 5.5](image)

**Figure 5.5. CS2: Frequency-dependent (rad/s) loop and disturbance gains.**

- *g*: ■, ▲, ×, g_{dk}, k=1,2,3.
  - Feed flow rate disturbances can not be rejected.
  - Designs 1A and 2A can not handle change of steam-generator duty.
  - Only Design 2A can cope with FEHE fouling.

In CS3, a second controller is added to CS1. It regulates the furnace inlet temperature using a bypass around FEHE. RGA elements show that, compared to CS1, the gain of main loop (furnace duty - reactor inlet temperature) is lower for Design 1A and 1B, and is practically unchanged for Designs 2A and 2B.

The change in the effect of the $k^{th}$ disturbances on $i^{th}$ output, caused by decentralised control, is given by the relative disturbance gain (RDG) defined as $\beta_{ik} = \tilde{g}_{ik}/g_{ik}$. Figure 5.6 presents the RDG of the feed flow rate disturbance. Interactions increase the gain from
disturbance to reactor inlet temperature; Hence, there is little incentive to add the second
control loop. Moreover, except Design 2B, bypass around FEHE is not powerful enough to
control furnace inlet temperature when the system is affected by disturbances.

![Graphs showing frequency-dependent (rad/s) RDG of feed flow rate disturbance.]

**Figure 5.6. CS3: Frequency-dependent (rad/s) RDG of feed flow rate disturbance.**

- **1A** - \( \beta_{21} \), **1B** - \( \beta_{22} \). Interactions increase the gain from disturbance to reactor inlet
temperature.

**Nonlinear analysis**

The control of the heat-integrated reactor in the toluene hydrodealkylation (HDA)
plant has been considered by Luyben et al. (1998). They pointed out that is difficult to
stabilise systems with large FEHE and small furnace. In this section, we identify the region in
the design parameter space leading to designs that can not be stabilised. To achieve this goal,
we use elements of the bifurcation theory (Guckenheimer and Holmes, 1983). The model
equations can be written in the following dimensionless form:
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Furnace:

\[ H_i \cdot M_h \frac{\partial \theta_h}{\partial \tau} = -\frac{\partial \theta_h}{\partial \xi} + Da \cdot H_i \cdot (\theta_u - \theta_h) \]

(5.10a)

\[ H_i \cdot M_u \frac{\partial \theta_u}{\partial \tau} = Da \cdot H_b - Da \cdot H_i \cdot \int_0^1 (\theta_u - \theta_h) \cdot d\xi \]

(5.10b)

Reactor:

\[ \frac{\partial X}{\partial \tau} = -\frac{\partial X}{\partial \xi} + Da \cdot \exp \left( \frac{\gamma \cdot \theta_r}{1 + \theta_r} \right) \cdot (1 - X) \]

(5.10c)

\[ Le \frac{\partial \theta_r}{\partial \tau} = -\frac{\partial \theta_r}{\partial \xi} + B \cdot Da \cdot \exp \left( \frac{\gamma \cdot \theta_r}{1 + \theta_r} \right) \cdot (1 - X) \]

(5.10d)

Temperature sensor:

\[ M_s \cdot \frac{d \theta_s}{d \tau} = Da \cdot (\theta_h(1) - \theta_s) \]

(5.10e)

P-controller (SP=0) and duty valve:

\[ M_v \cdot \frac{d H_b}{d \tau} = Da \cdot \left( H_b^- - K_c \cdot \theta_s - H_b \right) \]

(5.10f)

Boundary conditions:

\[ \theta_b(0) = (1 - \varepsilon) \cdot \theta_0 + \varepsilon \cdot (\theta_r(1) - Da \cdot H_c) \]

(5.10g)

\[ \theta_s(0) = \theta_h(1) \]

(5.10h)

Figure 5.7 presents, qualitatively, the conversion vs. controller gain and Damkohler number for two sets (FEHE efficiency, steam-generator duty). The first diagram corresponds to design 2A (large FEHE, large steam-generator). Model equations are used to compute the controller bias (the value of the manipulated variable when the controlled variable equals its set point). This is denoted as the nominal case and is represented by the dark lines. The system exhibits three steady-states for small controller gain. The middle state is of interest, but is unstable. Increasing the controller gain, the branches become closer. They coalesce for a critical value of the controller gain. This is a pitchfork bifurcation point. For higher values of the controller gain, only one steady-state is possible. When the controller gain is further increased, the system loses stability due to a Hopf bifurcation. Hence, the pitchfork and Hopf points bound the range of controller gain that ensures state unicity and stability.

The extent of the unicity and stability region decreases when the FEHE efficiency increases or the steam-generator duty decreases. For a critical value of FEHE efficiency,
Pitchfork and Hopf bifurcations occur at the same value of the controller gain. Incidentally, the Design 2B is very close to this limit. This situation is presented in the second diagram of Figure 5.7. If FEHE efficiency is further increased, the design can no longer be stabilised.

Figure 5.7. Conversion vs. controller gain and Damkohler number.
Pitchfork and Hopf bifurcation points bound the range of controller gain that ensures state unicity and stability. Middle steady-state can not be stabilised for large FEHE efficiency and small steam-generator duty.

The correct value for the controller bias can not be calculated when design parameters are uncertain. This is denoted as the perturbed case and is presented in Figure 5.7 by the light lines. The Hopf point is one limit of the stability region. Depending on the model parameter uncertainty, the other limit is the fold point located on the lower or the upper solution branch. Note that the region of state unicity and stability is larger. However, zero control error can not by achieved by proportional control.

In order to classify the steady-state and dynamic behaviour of the controlled system, we choose the controller gain as bifurcation parameter. The Hopf and fold points are 1-codimensional, which means that the value of one parameter is fixed (Guckenheimer and
Holmes, 1983). In the general case, a pitchfork point has codimension 3. However, the assumption that the parameters used for bias calculation are exactly known introduces $Z_2$-symmetry. Consequently, the pitchfork point becomes also 1-codimensional (Golubitsky and Schaeffer, 1985).

Consider a fixed, large FEHE efficiency. Then, there is a particular value of the steam-generator duty for which the pitchfork and Hopf bifurcation occur at the same value of the controller gain, like in the second diagram of Figure 5.7. This point has codimension 2. We call it “Hopf-and-pitchfork”. Computing its locus in the space of the design parameters (FEHE efficiency, steam-generator duty), we trace the boundary between the designs can be stabilised, and designs that can not. Similarly, computing the locus of the cusp variety (Guckenheimer and Holmes, 1983), we trace the boundary between state unicity and multiplicity.

The results are presented in Figure 5.8. The cusp and Hopf-and-pitchfork varieties divide the design parameter space into three regions. In region I, the unique steady-state is stable for low controller gain, but unstable for high controller gain. State multiplicity appears when the cusp variety is crossed to region II, but it is possible to find a controller gain that gives a unique stable steady-state. When the Hopf locus is crossed to region III, it is impossible to get both state unicity and stability.

Figure 5.8. Phase diagram in the space of design parameters.
Cusp and Hopf-and-pitchfork varieties divide the parameters space into regions with different conversion vs. controller gain bifurcation diagrams.
Dynamic simulation

We verify first that design 2B can not be stabilised. Figure 9 presents the results of dynamic simulation, when CS1 is used. Initially, the system is at steady-state. After 2000 s., the feed flow rate is changed by 0.1%, for 100 s. In the first simulation, the controller gain is lower than the gain at the pitchfork point. For this reason, there are multiple steady-states and the operating point is unstable. Hence, the disturbance drives the system to the extinguished state. In the second simulation, the controller gain is slightly increased past the pitchfork point. Consequently, only one steady-state exists. However, it is unstable and surrounded by a limit cycle.

![Dynamic simulation results](image)

**Figure 5.9. Dynamic simulation results.**

For large FEHE and small steam-generator duty, it is impossible to get both state unicity and stability

A question that naturally arises is: “The results of linear controllability analysis are applicable to such nonlinear systems?” To answer this question, we apply feed flow rate and steam-generator duty disturbances to Designs 1A, 1B and 2A. CS1 with Tyreus - Luyben controller settings (Table 5.4) is used. The results are presented in Figure 5.10.
In all cases, the manipulated variable is strong enough to reject disturbances. However, disturbance rejection is slow and the control error is initially large. For Designs 1A and 1B, feed flow rate is the worst disturbance and input saturation occurs. For design 2A, the disturbances considered are almost equally difficult. These results agree with linear controllability analysis.

**Figure 5.10. Dynamic simulation**

Results of linear controllability analysis are confirmed.
Conclusions

In this article, we analyzed the interaction between design and control of a heat-integrated PFR. The following conclusions are drawn:

- The controllability of the heat-integrated PFR is determined by two design decisions: FEHE efficiency and steam-generator duty.
- Systems with small steam-generator and large FEHE can not be stabilized. The Hopf-and-pitchfork variety bounds the range of design parameters for which the system can be stabilized.
- At steady-state, control structures using furnace duty as manipulated variable can reject disturbances in feed flow rate, steam-generator duty and FEHE fouling. However, fast control can not be achieved. There is no incentive to add a bypass around FEHE because it enhances the effect of disturbances.
- Designs with small FEHE are also difficult to control if the operating point is close to the cusp variety.
- The results of linear and nonlinear analysis are confirmed by dynamic simulation.

The approach presented here may be extended to similar problems, where state multiplicity and instability limits the range of controllable designs.

Notation

\begin{align*}
A &= \text{area, m}^2 \\
c_p &= \text{specific heat, J/(kg K)} \\
C &= \text{concentration, mol/m}^3 \\
Da &= \text{Damkohler number, dimensionless} \\
&= \frac{V_r \cdot \rho \cdot k(T)}{F} \\
d &= \text{diameter, m} \\
F &= \text{mass flowrate, kg/s} \\
\Delta H_r &= \text{heat of reaction, J/mol} \\
H_h &= \text{furnace duty, dimensionless} \\
&= \frac{q_h}{\rho \cdot c_p \cdot \bar{T} \cdot V_r \cdot k(T)} \\
H_c &= \text{steam-generator duty, dimensionless}
\end{align*}
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\[
Q_c = \frac{Q_c}{\rho \cdot c_p \cdot \bar{T} \cdot V_r \cdot k(\bar{T})}
\]

\[H_t = \text{furnace heat transfer area, dimensionless}\]

\[
= \frac{K_{nt} \cdot A_{nt}}{\rho \cdot c_p \cdot V_r \cdot k(\bar{T})}
\]

\[H_f = \text{FEHE heat transfer area, dimensionless}\]

\[
= \frac{K_{nt} \cdot A_{nt}}{\rho \cdot c_p \cdot V_r \cdot k(\bar{T})}
\]

\[K = \text{heat transfer coefficient, W/(m}^2 \text{K)}\]

\[Le = \text{Lewis number, dimensionless (Le}=1\text{)}\]

\[m = \text{mass, kg}\]

\[M_w = \text{furnace, mass of the tube walls, dimensionless}\]

\[
= \frac{\rho \cdot c_{pw} \cdot \delta_w \cdot k(\bar{T})}{K_{nt}}
\]

\[M_h = \text{furnace, inertia due to gas holdup, dimensionless}\]

\[
= \frac{d_w \cdot \rho \cdot c_p \cdot k(\bar{T})}{4 \cdot K_{nt}}
\]

\[NTU = \text{number of transfer units, dimensionless,}\]

\[
= \frac{K_{nt} \cdot A_{nt}}{F \cdot c_p}
\]

\[q = \text{duty, W}\]

\[t = \text{time, s}\]

\[i = \text{reference time, s}\]

\[
= \frac{V_r \cdot \rho}{F}
\]

\[T = \text{temperature, K}\]

\[\bar{T} = \text{reference temperature (}\bar{T}=T_2\text{), K}\]

\[V = \text{volume, m}^3\]

\[w = \text{velocity, m/s}\]

\[z = \text{axial coordinate, m}\]
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Greek letters

\( l-\alpha \) = bypass around FEHE
\( \rho \) = mass density, \( \text{kg/m}^3 \)
\( \xi \) = thickness of furnace tube walls, \( \text{m} \)
\( \varepsilon \) = FEHE efficiency, dimensionless

\[ \varepsilon = \frac{1 - e^{-\kappa \gamma (l-\alpha)}}{1 - \alpha \cdot e^{-\kappa \gamma (l-\alpha)}} \]

Subscripts

\( c \) = steam-generator
\( f \) = furnace, fluid phase
\( h \) = furnace
\( r \) = reactor
\( s \) = FEHE, shell-side
\( t \) = FEHE, tube-side
\( w \) = furnace, tubes wall

Literature cited


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Chapter 6 Singularity Theory Approach to Ideal Binary Distillation

Abstract

Computation of codimension-2 singular points and parameter sets was used to classify the steady state behaviour of ideal, constant molar overflow, binary distillation. Eleven different bifurcation diagrams are possible. Multiple steady states are likely to appear for practical situations when the relative volatility is large, the components have very different molar mass, or the feed concentration is small.
Introduction

The possibility of state multiplicity and instability in ideal distillation was simultaneously recognised by researchers at University of Trondheim and Technical University of Denmark. Jacobsen and Skogestad (1990, 1991) pointed out the following sources of multiplicity: 1) the nonlinear transformation between mass and molar flow rates, and 2) interaction between flows and composition due to energy balance. Nielsen (1990) presents experimental purity vs. volume reflux rate diagrams, that prove the existence of multiple steady states. Kienle et al. (1995) and Køggersbol et. al (1996) found experimentally multiple steady states in the methanol-propanol separation, the first item above being the source of multiplicity. They computed the locus of limit points (codimension-1 singularities) and showed how the interaction with feasibility boundaries leads to different bifurcation diagrams. Similar results were obtained for equilibrium model with constant molar overflow and for detailed model including energy balance and mass transfer resistance. However, they considered only one column design and only one mixture. Moreover, they did not provide a complete classification of the steady state behaviour.

This work analyses the multiplicity of states in binary distillation by rigorous application of the singularity theory (Balakotaiah and Luss, 1984; Golubitsky and Schaeffer, 1985). We consider the mass reflux flow rate ($L_w$) as bifurcation parameter and compute codimension-2 varieties that divide the feed composition ($z_F$) – boilup ($V$) parameter space into regions where different types of bifurcation diagrams exist. Finally, we investigate the effect of physical and design parameters on the location and extent of the multiplicity regions.

State multiplicity in ideal, binary distillation

Steady state multiplicity does exist in distillation. Let consider the separation of a binary mixture with ideal vapour-liquid equilibrium and assume constant molar overflows. The reflux is specified on mass basis, while the boilup flow rate is specified on molar basis (the $L_w, V$ configuration). The model equations are well known (Seader and Henley, 1998). We note that the tray molar balance equation can be written in dimensionless form by dividing with the feed flow rate ($F$). For this reason, we will use symbols as $L$ (molar reflux), $V$ (molar boilup), $D$ (distillate flowrate), $B$ (bottom flowrate) for dimensionless variables and symbols as $\bar{F}, \bar{L}, \bar{V}, \bar{D}, \bar{B}$ for dimensional variables. Concentration notation will refer to the light component.
Two feasibility boundaries exist for distillation. At total reflux, the distillate flow rate is zero and only a bottom product is obtained (this is different from the total reflux used in Fenske equation, which applies for a closed system). Similarly, at total reboil, there is no bottom product but a distillate.

![Diagram of distillation process and state multiplicity](image)

**Figure 6.1. State multiplicity in ideal, binary distillation.**
Continuos line represents distillate composition vs. molar reflux. Dashed line represents the relationship between molar reflux and distillate composition, for fixed mass reflux. There are three different feasible steady states. The middle state is unstable. $\alpha=3.55$; $M_1 / M_2 = 0.533$; $z_F=0.5$; $V=2$; $L_w=50$; $N=8$; $N_F=4$;

Figure 6.1 explains the occurrence of state multiplicity. The continuos line represents the dependence of distillate composition versus molar reflux, obtained from tray-by-tray material balance and equilibrium equations. The dashed line represents the relationship between molar reflux and distillate composition, for fixed mass reflux. The two curves may intersect at three points, corresponding to three different steady states that are possible for the same operating parameters (mass reflux and boilup). The McCabe-Thiele diagrams of the three operating points are also displayed.
We remark that the middle steady state is unstable. Consider a small, positive deviation of the distillate composition. The molar reflux becomes higher than the one necessary to achieve the actual purity; hence the distillate composition will increase further, until the upper operating point is reached.

**Solution method**

The methodology we will use to classify the steady state behaviour may be applied to systems that are described by a single intrinsic variable. This implies that, at least theoretically, it is possible to reduce the model equations to one equation with one variable. To demonstrate the validity of this assumption, we outline one possible approach to the solution of model equations.

We start with an initial guess for the distillate composition, $x_D$. The dimensionless molar reflux is given by:

$$L = \frac{L_w}{1+(M_1/M_2 - 1) \cdot x_0}$$  \hspace{1cm} (6.1)

$M_1/M_2$ is the molar mass ratio, $L_w = \bar{L}_w/(\bar{F} \cdot M_2)$ and $L = \bar{L}/\bar{F}$ are the dimensionless mass and molar reflux, respectively.

Application of the equilibrium and operating equations gives the composition of the bottom product:

$$y_1 = x_D$$

$$x_1 = e(y_1)$$

$$\text{do} \quad k = 2 \ldots N$$

$$y_k = o(x_{k-1})$$

$$x_k = e(y_k)$$

$$\text{end do}$$

$x_k$ and $y_k$ are the composition of the liquid and vapour leaving the $k^{th}$ tray, and $e(.)$ and $o(.)$ represent equilibrium and operating equations, respectively.

Finally, the global mole balance defined by Eq. 6.3 is checked, and $x_D$ is updated.

$$f(x_D) = F \cdot z_F -(V-L) \cdot x_D -(F - V + L) \cdot x_N = 0$$  \hspace{1cm} (6.3)

We remark that this approach is independent of the vapour-liquid equilibrium model used. Moreover, no fundamental limitations arise by including the energy balance. Although not straightforward, the reduction to one-equation model is theoretically possible.
Singularity theory states that the qualitative features of the bifurcation diagram may change only when the parameter set crosses the *hysteresis* (H), *isola* or *double limit* varieties. Only the hysteresis variety exists for ideal, constant molar overflow, binary distillation with mass reflux as bifurcation parameter. When the hysteresis variety is crossed, the number of possible steady states changes by two, as two limit points appear or disappear.

When feasibility boundaries exist, the bifurcation diagram may also change at special sets of parameters (Balakotaiah and Luss, 1984). In this work we considered:

- The *boundary-limit set*: a limit point exists at a feasibility boundary. There are two such sets, corresponding to one limit point at the total reboil (BL₁) or total reflux (BL₂) boundaries, respectively.

- The *cross-and-limit set*: the position of one limit point relative to one solution located at the feasibility boundary changes. There are two such sets, corresponding to total reboil (CL₁) or total reflux (CL₂) and one limit point occurring for the same value of the bifurcation parameter.

- The *double-cross set* (DC): the relative position of two solutions located at the feasibility boundary changes. There is one double-cross set, corresponding to total reboil and total reflux occurring for the same value of the bifurcation parameter.

The above-mentioned varieties are 2-codimensional, i.e. they fix the value of two parameters. Balakotaiah and Luss (1984) present their defining equations. The derivatives involved in the definition of the singular points can be easily computed by successively applying the chain rule for differentiation.

For the first stage,

\[
\frac{\partial y_1}{\partial x_d} = 1 \quad (6.4)
\]

\(x_1\) is computed from \(y_1\) using the equilibrium equation; hence:

\[
\frac{\partial x_1}{\partial x_d} = \frac{\partial y_1}{\partial y_1} \cdot \frac{\partial y_1}{\partial x_d} = \frac{\partial y_1}{\partial x_d} \quad (6.5)
\]

\(y_2\) is computed from \(x_1\) using the operating relation; hence:

\[
\frac{\partial y_2}{\partial x_d} = \frac{\partial y_2}{\partial x_1} \cdot \frac{\partial x_1}{\partial x_d} = \frac{\partial y_2}{\partial x_1} \quad (6.6)
\]

We continue this way, until we reach the bottom stage.

Differentiation of Eq. 6.3 gives Eq. 6.7, where all the terms are available.
\[
\frac{\partial f}{\partial x_D} = -(V - L) + (x_D - x_N) \frac{\partial L}{\partial x_D} - (F - V + L) \frac{\partial x_N}{\partial x_D}
\] (6.7)

Extension to higher order derivatives is straightforward.

**Results**

Due to the great number of parameters involved, the complete classification of the steady state behaviour is difficult. It is useful to divide the set of parameters into three groups:

1) Operation parameters: mass reflux (Lw), boilup (V), and feed composition (zf);
2) Physical parameters: relative volatility (\(\alpha\)), and molar mass ratio (\(M_1/M_2\));
3) Design parameters: number of trays (N), and feed tray (NF).

A codimension-2 variety determines the state variable and two parameters. One of them is the bifurcation parameter (Lw). We choose the boilup (V) as the second parameter. The location of a codimension-2 point (Lw, V) changes when a third parameter (zf) is varied. This dependence can be traced and plotted in the (V-zf) plane, for fixed values of the \(\alpha\), \(M_1/M_2\), N and NF parameters. We used a continuation method based on local parameterisation (Seydel and Hlavacek, 1987).

The codimension-2 varieties dividing the (zf-V) parameter space are presented in Figure 6.2.

![Typical phase diagram for ideal, constant molar overflow, binary distillation.](image)

**Figure 6.2. Typical phase diagram for ideal, constant molar overflow, binary distillation.**

\(\alpha=1.1; M_1/M_2 =0.9; N=8; NF=4.\) H - hysteresis; BL - boundary limit; DC - double cross; CL - cross-and-limit. Bold line represents the unicity-multiplicity boundary. Dashed line corresponds to occurrence of two unfeasible limit points. Different types of bifurcation diagrams existing in regions I - V are presented in Figure 6.3.
Figure 6.3 presents the bifurcation diagrams existing in different regions of Figure 6.2. In order to illustrate clearly the steady state behaviour, we chose extreme parameter values (a small column is used to separate a low relative volatility mixture). We will discuss latter the influence of more realistic operation and physical parameters.

The hysteresis variety is intersected tangentially by the two boundary-limit sets at codimension-3 singular points \((P_1 \text{ and } P_2)\). In region Ia, there is one steady state. Crossing the hysteresis variety to region Ib or Ic, two limit points appear. However, because they are located outside the feasibility region, state unicity is preserved. Crossing the BL\(_1\) set from region Ib to region IV, one limit point passes the total reboil boundary and enters the feasibility region; hence, the multiplicity pattern in region IV is 2-1. Similarly, when the BL\(_2\) set is crossed from region Ic to III, one limit point enters the feasibility region through the total reflux boundary; hence, the multiplicity pattern in region III is 1-2. Consequently, the multiplicity boundary (bold line in Figure 6.2) is given by a segment of BL\(_1\) down to \(P_1\), a segment of H between \(P_1\) and \(P_2\), and a segment of BL\(_2\) beyond \(P_2\). When going from region IV or III to region V, the BL\(_2\) and BL\(_1\) sets are crossed, respectively. In each case, one limit
point goes out of the feasibility region (through the total reflux and the total reboil boundaries, respectively); hence, in region V, we have again state unicity.

Regions III and IV are further divided by the double-cross set into parts with different relative location of the total reboil and total reflux conditions. We denote by L, M, and U the lower, middle and upper branches of the bifurcation diagram, respectively (regardless their feasibility). Then, in regions IIIa and IIIb the multiplicity patterns are L-LM and M-LM, respectively. Similarly, the multiplicity patterns in regions IVa and IVb are MU-U and MU-M, respectively.

Two feasible limit points appear when the hysteresis variety is crossed from region Ia to region II. Region II is bounded by the H, BL₁ and BL₂ sets and divided by two cross-and-limit sets and the double cross set. The 1-3-1 multiplicity pattern (IIa) becomes 2-3-1 (IIb), 1-3-2 (IIc) or 2-3-2 (IId), as CL₁ and CL₂ are crossed and the relative positions of one limit point and one feasibility boundary change. When the DC set is passed to region He, the relative location of feasibility boundaries changes; consequently, the multiplicity pattern becomes 2-1-2.

Figure 6.4 shows phase diagrams obtained for values of the α, M₁/M₂, N and N_f parameters that are more realistic than those used in Figure 6.2. Although all the varieties and regions discussed previously are present, we displayed, for clarity, only the unicity-multiplicity boundary.

Diagram A shows the influence of relative volatility, for a mixture with small molar mass ratio. When α has practical values, the BL₁ variety moves very close to z_F=1. The BL₂ variety is located at very small feed concentration and does not intersect the hysteresis for positive values of the boilup. Hence, the boundary between the unicity and multiplicity regions is the hysteresis variety. (This conclusion is also valid for other practical situations). There is a region of small z_F that can lead to multiple steady states even at low boilup. For high z_F, practical boilup values may lead to multiple steady states if the components to be separated have high relative volatility. We also note that, at small boilup, increasing α slightly shifts the unicity region towards lower feed concentration.

Diagram B presents the influence of the molar mass ratio, for a usual value of α. The size of the unicity region is larger when the components have similar molar mass. The effect of M₁/M₂ is more important at high feed concentration. When M₁/M₂ is close to the unity, multiplicity is probable to occur for low feed concentration or high internal flow rates. The
number of trays has a small effect (Diagram C), while the location of the feed tray does not affect appreciable the multiplicity boundary.

Figure 6.4. Influence of the model parameters on the extent and location of the multiplicity region.
Multiplicity region is larger for high relative volatility, low molar mass ratio or feed close to the column bottom.
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In diagrams A-C the BL₁ and BL₂ varieties are located for very high or low feed concentration, respectively. One exception is the low relative volatility case (Diagram D). In this situation, moving the feed location to the column bottom enlarges the multiplicity region.

In conclusion, computation of codimension-2 singular points and parameter sets was used to classify the steady state behaviour of ideal, constant molar overflow, binary distillation. Eleven different bifurcation diagrams are possible. Multiple steady states are likely to appear for practical situations when the relative volatility is large, the components have very different molar mass or feed concentration is small.

Literature cited


Chapter 7 Nonlinear behaviour of reactor-separator-recycle systems: Implications for integrating conceptual design and plantwide control

Abstract

This article studies the nonlinear behaviour of reactor-separator-recycle systems. The dimensionless mole-balance equations are parameterized by the plant Damköhler number (Da) and the separation specifications. Reactant accumulation does not occur if $Da > Da^{ct}$. $Da^{ct}$ corresponds to a bifurcation point of the mole-balance equations and depends on separation performance. For one reactant recycle and high purity separation, $Da^{ct} = 1$. When two reactants are recycled, multiple steady states are possible. In this case, $Da^{ct}$ depends also on the control structure. Close to $Da^{ct}$, control structures must change the reaction conditions in order to reject effectively the disturbances. Reviewing several plantwide control studies that reported bad controllability, we found that the designs considered are close to the critical value of the plant Damköhler number.
Introduction

During the traditional design of chemical processes, many alternatives are generated. Early in the project development, economic criteria are used to discard unattractive designs. Later, when the flowsheet structure is established and the units are sized, controllability issues become more important. Often, the designer finds that the alternatives developed do not have the required operability properties. Hence, designs that have been rejected because of lower economic performance are re-evaluated. Because this approach may be time and effort consuming, the need of integrating process design and control is well recognized. Yet, a procedure to generate controllable design alternatives is not available.

Chemical plants are nonlinear systems. Today simulation software makes possible the development of detailed, nonlinear dynamic models. Nevertheless, for the purpose of controllability analysis (Skogestad and Postlethwaite, 1996), linear models are (frequently successfully) employed. This happens because the mathematical tools needed to analyze linear differential equations are well developed, while the theory of nonlinear differential equations is still an active field of research. However, many of the control problems originate from a bad design. Dynamic modeling requires equipment sizing, which is not available during conceptual design. Consequently, the designer relies upon steady state models. However, he still uses linear tools, although analysis of nonlinear algebraic equations is possible.

The nonlinearity of chemical processes can manifest as parametric sensitivity, state multiplicity, instability, or oscillatory behaviour (Seider and Brengel, 1991). These features can be predicted by singularity (Golubitsky and Schaeffer, 1985) and bifurcation (Guckenheim and Holmes, 1983) theories, which have been extensively applied in the field of chemical reaction engineering. A classic example is the first-order, exothermic reaction in CSTR, axial dispersion reactor, or catalytic particle. Generally, bifurcation diagrams are used to represent the dependence of the system’s state vs. one distinguished operating variable (bifurcation parameter). Then, bifurcation varieties are computed. They divide the space of the remaining parameters into regions corresponding to qualitative-different bifurcation diagram. From the viewpoint of design and control, designs close to bifurcation varieties generally should be avoided. The reason is that the uncertainty of the design parameters and the disturbances affecting the process may shift the operating point to a region where the
qualitative behavior is different from the expected one. Then, undesirable phenomena may occur: loss of stability, reaction ignition or extinction, reverse sign of the gain in control loops, etc. Moreover, close to bifurcation points, high sensitivity to disturbances is expected.

Most of the previous studies considered the stand-alone reactor. Although the effect of recycling a fraction of the reactor effluent has been occasionally studied (Pareja and Reilly, 1969, Recke and Jorgensen, 1999), the systems considered are not good examples for chemical reactors in recycle plants. Typically, the reactor effluent is processed by the separation section. Hence, the composition and temperature of the recycle stream are different from the reactor effluent. Moreover, often temperature controllers keep constant reaction temperature, or, for adiabatic reactors, constant temperature of the reactor feed.

This article addresses the nonlinear behaviour of certain reactor-separator-recycle systems. Figure 7.1 presents the flowsheet considered. In a real plant, several feed, product, or recycle streams may exist. Hence, each general streams of Figure 7.1 (Plant feed; Reactor feed; Reactor effluent / Separation feed; Recycle; Products) represents an arbitrary number of real streams.

Figure 7.1. General structure of Reactor-Seperator-Recycle systems.
Throughout this work, the molar flow rate of the streams enumerated above are denoted by \( F, R, S, Y, \) and \( P \), respectively. Symbols as \( F_A, S_B \), are used for the components flow rates, where the subscript refers to chemical species. Dimensionless flow rates are obtained by division to the feed flow rate of a reference reactant \( (F_A) \). They are represented by lower-case symbols (for example, \( r = R/F_A, s_B = S_B/F_A \)). Greek letters are used for flow rate ratios (for example, \( \alpha_{YS} = Y_A/S_A \cdot \varphi_{RF} = R/F \)). \( c_{ik} \) and \( z_{ik} \) signify concentration and mole fraction, respectively, of the \( i^{th} \) component in the \( k^{th} \) stream.

The behaviour of reactor-separator-recycle systems is relevant for integrating conceptual design and plantwide control because, early during conceptual design, one reaches a stage when the recycle structure of the flowsheet is established. Then, the reactor is the first unit to be considered in detail because the chemical species present in the reactor effluent determine the separation section. Hence, reactor modeling, sizing, and control are considered before separation is addressed. In this work, we will treat the separation as a black box, and consider a kinetic model of the chemical reactor.

This article is organized as follows. In the next section, we study a first order reaction in a CSTR-separator-recycle system. The dimensionless mole-balance equations are parameterized by the plant Damköhler number \( (Da) \) and the separation specifications. It is demonstrated that reactant accumulation does not occur if \( Da > Da^{ct} \), where the critical value \( Da^{ct} \) depends on separation performance. For \( Da = Da^{ct} \), a transcritical bifurcation of the mole balance equations takes place. For high purity of product and recycle streams, \( Da^{ct} = 1 \). These conclusions also apply to the PFR, first-order consecutive, and \( n^{th} \)-order reactions. Further, we compare two different control structures and show that their relative performance depends on the design. Close to \( Da^{ct} \) (small reactor or slow kinetics), it is necessary to change the reaction conditions when disturbances affect the process. In the last section, we discuss two different control structures for a second order reaction. In these cases, the critical value of the plant Damköhler number corresponds to a fold bifurcation of the mole balance equations. Reviewing several plantwide control studies that reported bad controllability, we found that the designs considered are close to the critical value of the plant Damköhler number.

**Plant Damköhler number**

In this section it will be shown that there is a minimum-volume constraint for a chemical reactor placed in a recycle plant.
We consider the first-order $A \rightarrow B$ reaction, taking place in a CSTR. Mole balance equations can be solved for consistent specifications. One possible choice is: flow rates of A and B in the feed stream ($F_A$, $F_B$); reactor volume ($V$); separation performance, for example as recoveries ($\alpha_{YS} = Y_A / S_A$ and $\beta_{PS} = P_B / S_B$). It must be pointed that often the reactant is completely recovered in the separation section ($\alpha_{YS} = 1$).

The reactor feed is chosen as tear stream and the reaction conversion ($x_A = 1 - S_A / R_A$) is introduced as auxiliary variable. Then, the following dimensionless balance equations can be easily derived:

$$ r_A = f_A + \alpha_{YS} \cdot r_A \cdot (1 - x_A) \quad (7.1) $$
$$ r_B = f_B + (1 - \beta_{PS}) \cdot \left( r_B + r_A \cdot x_A \right) \quad (7.2) $$

If the product B is not present in the feed stream ($f_B = 0$) and is completely recovered in the separation section ($\beta_{PS} = 1$), then:

$$ r_A = \frac{R_A}{F_A} = \frac{1}{1 - \alpha_{YS} \cdot (1 - x_A)} \quad (7.3) $$
$$ r_B = \frac{R_B}{F_A} = 0 \quad (7.4) $$
$$ z_{AS} = 1 - x_A \quad (7.5) $$

To find the conversion, the CSTR equation:

$$ R_A - S_A = k \cdot V \cdot c_{AS} \quad (7.6) $$
is written in the following dimensionless form:

$$ g(x_A, Da, \alpha_{YS}) = Da - \frac{x_A}{(1 - x_A) \cdot (1 - \alpha_{YS} \cdot (1 - x_A))} = 0 \quad (7.7) $$

$$ Da = \frac{k \cdot V}{F_A \cdot V_{mA}} $$
is the plant Damköhler number, which includes the reactor volume, the reaction kinetics, the reactant feed flow rate, and its molar volume at reactor temperature and pressure. Eq. 7.7 assumes that the molar volume does not depend on composition. This is true for gas-phase reactions. For liquid-phase reactions, an additional parameter, $v_B = V_{µB} / V_{µA}$, should be introduced in the balance equations. If A and B are isomers and have similar densities, then $v_B \approx 1$. 

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Eq. 7.7 has two Damköhler-dependent solutions. They are presented in Figure 7.2a, for different values of the \( \alpha_{Y,S} \) parameter. For stand-alone CSTR (\( \alpha_{Y,S} = 0 \)) or incomplete reactant recycle (\( \alpha_{Y,S} < 1 \)), the feasible (\( 0 < x_A < 1 \)) and unfeasible (\( x_A < 0 \)) solution branches do not intersect. When all the reactant is recycled (\( \alpha_{Y,S}=1 \)), one solution (\( x_A = 0 \)) involves infinite recycle. The other solution has physical significance (\( 0 < x_A < 1 \)) if, and only if, \( Da > 1 \). Moreover, the two branches intersect at the point \( T \) having the coordinates \( (x_A, Da) = (0, 1) \).

Figure 7.2. First-order reaction in CSTR-Separator-Recycle: conversion, yield and reactor outlet/separation inlet flow rate
In the framework of the bifurcation theory, the point \( T \) represents a transcritical bifurcation of the model equations. The defining condition for the transcritical bifurcation is (Guckenheimer and Holmes, 1983):

\[
\frac{dg(x_A, Da, \alpha_{Y,S})}{dx_A} = \frac{1 - \alpha_{Y,S} + \alpha_{Y,S} \cdot x_A^2}{(1-x_A)^2 \cdot (1-\alpha_{Y,S} + \alpha_{Y,S} \cdot x_A)^2} = 0 \tag{7.8}
\]

The only feasible case \((0 < \alpha_{Y,S} \leq 1)\) when the real root

\[
x_A^\alpha = \pm \sqrt{\frac{\alpha_{Y,S} - 1}{\alpha_{Y,S}}} \tag{7.9}
\]

exists is \( \alpha_{Y,S} = 1 \). Substitution in Eq. 7.7 gives:

\[
\lim_{\alpha_{Y,S} \to 1} \lim_{x_A \to x^2} g(x_A, Da, \alpha_{Y,S}) = Da - 1 = 0 \tag{7.10}
\]

Bifurcation theory states that an exchange of stability takes place at the transcritical bifurcation point. If the reactor volume is small or the reaction rate is slow, the reactant fed in the process can not be entirely transformed into products. Because the reactant can not leave the plant, it accumulates, and the recycle becomes infinite. Hence, for \( Da < 1 \), the trivial solution \((x_A = 0, \infty, infinite recycle)\) is stable (solid line in Figure 7.2a). Increasing the Damkohler number \( Da > 1 \), the stable, feasible solution appears, while the infinite-recycle solution loses stability.

Figure 7.2b presents the yield, defined as \( p_B = P_B/F_A \), of the CSTR – separator – recycle system, for different values of the reactant recycle ratio \( \alpha_{Y,S} \). When the entire amount of unreacted \( A \) is recycled, the yield equals the maximum value, \( p_B = 1 \). For the same feed, the yield of the stand-alone CSTR is the lowest.

In recycle systems, the yield enhancement is achieved through separation and increased flow rates. Figure 7.2c presents the flow rate of the stream entering the separation system. High values of the recycle ratio, small reactor volume or slow reaction rate lead to high flow rates. If the operating point is close to \( Da = 1 \), then the flow rates exhibit extremely high sensitivity to change of reactant feed flow rate, reaction rate, or reactor volume, and severe control problems are expected.

The set of specifications \((\alpha_{Y,S} < 1, \beta_{F,S} = 1)\) may represent a separation section that, after the A-B mixture is split into components, purges a fraction of A. This has a stabilising
effect. It should be remarked that systems where the purge flow rate is *fixed* are qualitatively identical with systems where the entire amount of reactant is recycled.

Often, the separation section is operated in such a way that the composition of the outlet streams are kept to required values \( z_{A,Y} < 1 \) and \( z_{B,P} < 1 \). Hence, a small quantity of reactant leaves the plant in the product stream, while some product is recycled. Such systems behave as systems with pure separation products \((\alpha_{Y,S} = 1, \beta_{P,S} = 1)\). To demonstrate this statement, we follow the procedure outlined above, using \( z_{A,Y} \) and \( z_{B,P} \) as specifications for the separation section. After some algebraic manipulations and cancellation of the trivial solution \( x_A = 0 \), the following equation is obtained:

\[
x_A = \frac{D_a \cdot z_{A,Y} - z_{B,P}}{D_a \cdot z_{A,Y} + z_{A,Y} - 1}
\]

(7.11)

The solution of Eq. 7.11 is feasible \((0 < x_A < 1)\) if, and only if:

\[
D_a > D_a' = \frac{z_{B,P}}{z_{A,Y}}
\]

(7.12)

The conclusion of this section is that, if the reactant is not allowed to leave the plant due to the recycle or composition control of the product stream, then the internal flow rates are finite if, and only if, the plant Damkohler number exceeds a critical value. For complete reactant and product recovery in the separation section, the critical value of the Damkohler number is \( D_a' = 1 \). This result also applies for n-th order reactions, with the plant Damkohler number defined as:

\[
D_a = k \cdot \frac{V}{F_A \cdot V_{\mu_A}} \left( \frac{1}{V_{x_A}} \right)^{n-1}
\]

(7.13)

**Interaction between design and control**

The set of specifications used in the previous section \((F_A, V, z_{A,Y}, z_{B,P})\) can be viewed as a plantwide control structure (Figure 7.3a): the reactor volume is kept constant; the separation section is dual-composition controlled; and the plant throughput is set by the reactant feed. For this control structure, the feed disturbances affect the flow rate and composition of the reactor outlet. Hence, manipulated variables internal to separation section are used to reject the disturbances.

The CSTR-separator-recycle was previously analysed in several plantwide controllability studies. Luyben (1994) pointed out that the conventional control structure
exhibits high sensitivity to feed disturbances. He proposed (Figure 7.3b) to keep the reactor outlet on flow control and to modify the plant throughput by changing the reaction conditions (reactor volume or temperature). This control structure is equivalent to the following set of design specifications: $F_A$, $S$, $z_{A,Y}$, $z_{B,P}$. When disturbances affect the process, the load of the separation section changes only due to composition modification. Consequently, disturbances are rejected mainly by changing the reaction conditions.

Figure 7.3. CSTR-Separator-Recycle: controls structures.
In order to compare the conventional and Luyben's control structures, we perform a steady state sensitivity analysis for different values of the design parameters. The equations are presented, for a more general case, in the Appendix.

Let \( F_A^* \) and \( V^* \) represent the nominal feed flow rate and reaction volume, respectively. Figure 7.4 presents, for the conventional control structure, the flow rate to separation vs. reactant feed flow rate, for different values of the nominal Damkohler number \( (Da^* = k \cdot V^*/(F_A^* \cdot V_{inA})) \) and recycle stream purity \( (z_{A,Y}) \). For Luyben's control structure, reactor volume \( (V \neq V^*) \) vs. reactant feed flow rate is presented. In both cases, small changes mean that small effort is needed to reject disturbances.

**Figure 7.4. First-order reaction in CSTR-Separator-Recycle: sensitivity analysis for different control structures**
Results presented in Figure 7.4 demonstrate that systems with small reactor or slow reaction rate \((Da^* < 2)\) are better controlled by the Luyben’s structure. However, systems with large reactor or fast reaction rate \((Da > 3)\) are better controlled by the conventional control structure. To explain this, we recall that close to \(Da = 1\) the system is sensitive to \(Da\). For the conventional control structure, the plant \(Da\) number represents a disturbance (because it contains \(F_A\)). Because low sensitivity is required, designs with \(Da\) large perform better. For the Luyben control structure, the plant \(Da\) number is a manipulated variable (through the reaction volume \(V\)). Consequently, disturbances can be rejected with small effort if \(Da\) is small. It should be also remarked that high purity separation \((z_{A,Y} = 1)\) improves the performance of the conventional control structure. This is contrary to the Luyben’s structure. This observation is explained by Eq. 7.12: increasing \(z_{A,Y}\) shifts \(Da^{*}\) toward smaller values; then, the difference \(Da - Da^{*}\) becomes larger, with a beneficial effect for the conventional structure.

It is worthy to mention that the design analysed by Luyben (1994) had \(Da\)=1.8. Hence, poor performance of the conventional control structure is expected. Larsson et al. (1999) pointed out that there is an economic penalty for fixing the reactor effluent and the conventional control structure works very well if the reaction volume is set to a larger value.

A closer look at the dimensionless mole-balance equations suggests a third control structure (Figure 7.3c). It is obvious that as long as the dimensionless model parameters do not change, all flow rates are proportional to feed. Moreover, stream compositions are constant. Hence, production change can be achieved by changing the feed and modifying the reaction conditions \((V\) or \(k)\) to keep constant Damkohler number. This control structure (Figure 7.3c) is similar to the alternatives proposed by Wu and Yu (1996). They called it “balanced control structure” because the disturbances are rejected changing both the reactor and the separation conditions.

**The Plug-Flow Reactor**

Considering a high-purity separation, the mole balance equations of the PFR-separator-recycle system can be reduced to:

\[
Da = \frac{k \cdot V}{V_{mA} \cdot F_A} \cdot \frac{1}{1 - \alpha_{yA} \cdot (1-x_A)} = \ln \frac{1}{1-x_A}
\]

(7.14)
For given $Da$ and $\alpha_{Y,S}$, Eq. 7.14 is solved numerically to find the corresponding conversion. Then, flow rate and composition of various streams in the system can be calculated. Figure 7.5 presents the conversion, flow rate to separation, and production rate vs. plant Damkohler number. The behaviour of the PFR coupled with separation and recycle is similar to the behaviour of the CSTR. For complete reactant recycle, internal flow rates are finite if, and only if, the plant Damkohler number exceeds a critical value. For high purity separation, the critical value of the Damkohler number is $Da^{cr}=1$. Close to the critical value of the Damkohler number, high sensitivity to disturbances is expected.

Figure 7.5. First-order reaction in PFR-Separator-Recycle: conversion, yield and reactor outlet / separation inlet flow rate
Consecutive reactions

In this section, we consider the following first-order, consecutive reactions:

\[
\frac{k_1}{k_2} \quad A \rightarrow B \rightarrow C
\]

B is the main product. \(k_1\) and \(k_2\) are the reaction constants. The flowsheet specifications are: flow rates of A and B in the feed stream \((F_A, F_B=0)\); reactor volume \((V)\); separation performance \((\alpha_{YS} = 1, \beta_{pS} = 1, \gamma_{pS} = 1)\).

Continuous Stirred Tank Reactor

The dimensionless reaction extents, \(\xi_{v,1} = \xi / F_A0\), are given by:

\[
\xi_{v,1} = Da \cdot \frac{(1 - \xi_{v,1})}{1 - \alpha_{YS} \cdot \xi_{v,1}}
\]

with the solutions:

\[
\xi_{v,1} = \frac{Da + 1 + \sqrt{(Da + 1)^2 - 4 \cdot Da \cdot \alpha_{YS}}}{2 \cdot \alpha_{YS}}
\]

and:

\[
\xi_{v,2} = \frac{Da \cdot k_2 \cdot \frac{1 - \alpha_{YS}}{k_1 \cdot (1 - \alpha_{YS} \cdot \xi_{v,1}) \cdot \xi_{v,1}}}{1 + \frac{1 - \alpha_{YS}}{1 - \alpha_{YS} \cdot \xi_{v,1}}}
\]

Then, the following expressions can be derived:

reactor outlet flow rate: 
\[
s = \frac{1 - \alpha_{YS} \cdot \xi_{v,1}}{1 - \alpha_{YS}}
\]

reactor outlet composition: 
\[
z_{AS} = \frac{1 - \xi_{v,1}}{1 - \alpha_{YS} \cdot \xi_{v,2}}, \quad z_{BS} = \frac{(\xi_{v,1} - \xi_{v,2}) \cdot (1 - \alpha_{YS})}{1 - \alpha_{YS} \cdot \xi_{v,2}}
\]

production rate: 
\[
p_B = \xi_{v,1} - \xi_{v,2}
\]

reaction selectivity: 
\[
\sigma_{BA} = \frac{\xi_{v,1} - \xi_{v,2}}{1 - (1 - \alpha_{YS}) \cdot s_A}
\]

Results in Figure 7.6 demonstrate that the reactant recycle increases the production rate and improves selectivity, especially close to \(Da = 1\). However, the stream processed by
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the separation section is more diluted in the B product and has higher flow rate (similar to Figure 7.2).

Figure 7.6. Consecutive, first-order reactions in CSTR-separator-recycle: yield, selectivity and main product concentration at reactor outlet / separation inlet.

Consecutive reactions in PFR

To solve the mole balance equations, the reactor feed is chosen as tear stream. Further, the mole balance equations of the reactant A can be reduced to:
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\[ Da = r_A \cdot \ln \frac{\alpha_{YS} \cdot r_A}{r_A - 1} \]  \hspace{1cm} (7.22)

Eq. 7.22 is solved numerically, and the following variables are computed:

reactor outlet component flow rates:

\[
s_A = r_A \cdot e^{r_A}, \quad s_B = \frac{r_A}{1 - k_2/k_1} \left( e^{r_A} - e^{r_A} \right), \quad s_C = r_A - (s_A + s_B) \]  \hspace{1cm} (7.23)

production:  \[ p_B = s_B \]  \hspace{1cm} (7.24)

selectivity:  \[ \sigma_{B/A} = \frac{p_B}{1 - (1 - \alpha_{YS}) \cdot s_A} \]  \hspace{1cm} (7.25)

Figure 7.7. Consecutive, first-order reactions in PFR-separator-recycle: yield, selectivity and main product concentration at reactor outlet / separation inlet.
The dependence of production rate, selectivity and product concentration at reactor outlet vs. plant Damkohler number is presented in Figure 7.7. When placed in a recycle system, the PFR behaves very similar to the CSTR. However, higher product concentration at the reactor outlet can be obtained.

**Second order reaction in CSTR**

In this section we will analyse a process where the second-order reaction $A + B \rightarrow C$ takes place in a solvent $S$. The process consists of a temperature-controlled CSTR and a separation section. The process may be represented by the general reactor-separator-recycle flowsheet in Figure 7.1. It should be remarked that the second reactant (and the solvent) not only changes the reaction kinetics, but also adds more dimensionality to the problem.

We will consider two control structures, presented in Figure 7.8. They follow naturally when the degrees of freedom and consistent specifications of the mass balance equations are considered. Thus, for simultaneous, independent specification of the A and B feed flow rates ($F_A$, $F_B$), the mass balance equations can not be solved. The reason is that, in recycle systems, the ratio of reactants in feed must reflect the reaction stoichiometry. Hence, control structures in which both reactants are on flow control are unfeasible. Moreover, due to inherent measurement errors, ratio control can not avoid the imbalance.

In control structure CS1, the A feed is flow-controlled (FC1). This stream is used to achieve production changes. The reactor effluent controls the reactor volume (LC1). We assume that in the separation section there are some locations reflecting species inventory. In Figure 7.8, they are denoted by $A$, $B$, $C$, $S$, respectively. The recycle of A and the product stream control species inventory (LC2 and LC4). The recycles of B and S are on flow control (FC2 and FC3). Reactant B is fed in the separation section in order to keep constant inventory (LC3). Solvent make-up may be necessary. This control structure corresponds to the following set of flowsheet specifications: feed ($F_A$, $F_C=0$, $F_S=0$); reactor volume ($V$); separation as recoveries ($\alpha_{Y,S}=1$, $\beta_{Y,S}=1$, $\gamma_{Y,S}=1$, $\sigma_{Y,S}=1$); component recycles ($Y_B$, $Y_S$).
To solve the mole-balance equations, the conversion of A is introduced as auxiliary variable. The flow rate of A, B, C and S at the reactor inlet are $F_A \cdot \frac{1-x_A}{x_A}$, $Y_B$, 0 and $Y_S$, respectively. Then, the volumetric flow rate is given by:

$$S^{\text{vol}} = F_A \cdot V_{\text{in},A} \left( \frac{1-x_A}{x_A} + v_B \cdot (y_B - 1) + v_S \cdot y_S + v_C \right)$$  \hspace{1cm} (7.26)$$

where $v_k = \frac{V_{\text{in},k}}{V_{\text{in},A}}$ is the dimensionless molar volume of the $k$ component.
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The reactant concentrations:

\[ c_{AS} = \frac{1-x_A}{x_A} \frac{1-x_A}{1-x_A + v_B \cdot (y_B - 1) + v_s \cdot y_s + v_c} \]  \hspace{1cm} (7.27)

\[ c_{BS} = \frac{1-x_A}{y_B - 1} \frac{1-x_A}{1-x_A + v_B \cdot (y_B - 1) + v_s \cdot y_s + v_c} \]  \hspace{1cm} (7.28)

are introduced in the CSTR's equation. The following dimensionless form is obtained

\[ g(x_A, Da, y_B, v_B, v_s \cdot y_s, v_c) = Da \left( \frac{1-x_A}{x_A} \frac{1-x_A}{1-x_A + v_B \cdot (y_B - 1) + v_s \cdot y_s + v_c} \right)^2 \]  \hspace{1cm} (7.29)

where \( Da = \frac{k \cdot V}{F_A \cdot V_{\mu A}} \cdot \frac{1}{V_{\mu A}} \) is the plant Damkohler number for a second-order reaction.

Eq. 7.29 has two \( Da \)-dependent solution branches:

- \( Da < 0 \): the conversion is negative and has no physical significance.
- \( Da^{\alpha} > Da > 0 \): the solutions are complex.
- \( Da > Da^{\alpha} \): two meaningful solutions exist. They are presented in Figure 7.9, for different values of the model parameters.

The critical value of the Damköhler number corresponds to a fold bifurcation of the model equations. The defining condition for the fold bifurcation is (Guckenheimer and Holmes, 1983):

\[ \frac{\partial g(x_A, Da, y_B, v_B, v_s \cdot y_s, v_c)}{\partial x_A} = 0 \]  \hspace{1cm} (7.30)

Solution of Eqs. 7.29 and 7.30 gives the location of fold points:

\[ Da^{\alpha} = 4 \cdot \frac{v_B \cdot (y_B - 1) + v_s \cdot y_s + v_c}{y_B - 1} \]  \hspace{1cm} (7.31)

\[ x_A^{\alpha} = \frac{1}{1 + v_B \cdot (y_B - 1) + v_s \cdot y_s + v_c} \]  \hspace{1cm} (7.32)

Generally, large \( B \) and small \( S \) recycles enlarge the region where the feasible solutions exist.
Bifurcation theory states that one stable and one unstable steady state emerge from the fold point. The unusual behaviour on the lower branch (larger reactor gives lower conversion) suggests that this steady state is unstable.

It is remarkable that near the fold bifurcation point, the system is very sensitive to disturbances. Consequently, from a control point of view, designs with $Da$ close to $Da^{cr}$ should be avoided.

Figure 7.9. Second-order reaction in CSTR-Separator-Recycle and CS1: multiple solutions of the mole-balance equations.
In the second control structure of Figure 7.8 (CS2), the A feed is flow-controlled (FC1). This stream is used to achieve production changes. The reactor effluent is on flow control (FC2). The recycles of A and B control species inventory in the separation section (LC2 and LC3). The recycle of S is on flow control (FC3). Reactant B is fed in order to keep constant reactor volume (LC1). This control structure corresponds to the following set of flowsheet specifications: feed ($F_A, F_C=0, F_S=0$); reactor volume ($V$); separation as recoveries ($\alpha_{Y,S}=1, \beta_{Y,S}=1, \gamma_{Y,S}=1, \sigma_{Y,S}=1$); solvent recycle ($y_S$); reactor effluent volumetric flow rate ($s^{vol}$).

The following mole-balance equation can be derived:

$$g(x_A, Da, s^{vol}, v_B, y_S, v_C) = Da - \frac{{\left( x_A s^{vol} \right)^2}}{1 - x_A v_B} \cdot \left( \frac{1 - x_A + v_C + v_S y_S}{x_A} \right)$$

(7.33)

Eq. 7.33 has two solutions $0 < x_A < 1$ for $Da > Da^{cr}$. The critical value of the plant Damkohler number corresponds to a fold bifurcation, and is given by:

$$Da^{cr} = 4 \cdot v_B \left( \frac{s^{vol}}{s^{vol} - v_C - v_S y_S} \right)^2$$

(7.34)

$$x^{cr} = \frac{2}{2 + s^{vol} - v_C - v_S y_S}$$

(7.35)

Figure 7.10 presents the physical-significant solutions of Eq. 7.33, for different values of the model parameters.

The analysis presented here is based on a black box model of the separation units. Nevertheless, the results apply when more detail is considered. Luyben and Luyben (1997) studied the controllability of the second-order reaction – separator – recycle system, with rigorous dynamic models for the separation section (distillation columns). For CS1 and a design having $29.7 = Da > Da^{cr} = 10.66$, they could achieve large production changes. However, with CS2 they could not achieve production changes larger than 2%. This is not a surprise, because the design considered had $Da = 29.7 > 28.4 = Da^{cr}$.  

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Figure 7.10. Second-order reaction in CSTR-Separator-Recycle and CS2: multiple solutions of the mole-balance equations.
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Conclusions

When placed in recycle plants, chemical reactors behave different from the stand-alone reactors:

1. Production rate reflects the amount of reactants fed in the plant. There is no way to change the plant throughput without changing the reactants feed. Consequently, in plant-wide control structures, reactant feeds must change (directly or indirectly) when production change is required. For stand-alone reactors, it is enough to modify the reaction conditions in order to change the production rate.

2. When a reactant is recycled, the reactor must transform the whole amount fed in the process. Hence, in order to prevent accumulation and infinite recycle, large reactor volume or fast kinetics are required. The minimum-volume constraint depends on the control structure. It can be expressed in terms of plant Damköhler number and separation specifications. For one reactant recycle, $Da^a$ correspond to a transcritical bifurcation of the mole-balance equations. When two reactants are recycled, multiple steady states may exist, and $Da^a$ correspond to a fold bifurcation of the mole-balance equations. Near $Da^a$, the system exhibits high sensitivity to disturbances. Hence, control structures must use the reaction conditions as manipulated variables, in order to reject effectively the disturbances. There are no restrictions on the size of a stand-alone reactor.

3. The feed ratio of reactants must reflect the reaction stoichiometry. Consequently, only one reactant feed may be on flow control. The other reactants must be fed in such a way to control their inventory in the process. For stand-alone reactors, the ratio of reactants in the feed may have any value.

The results of this study reveals the important role played by the chemical reactor in the (plantwide) controllability of recycle plants.

Literature cited


Appendix. Solution of mole-balance equations for first-order reaction in CSTR-separator-recycle system, for two control structures.

We consider that the reaction takes place in a solvent S, which is recovered in the separation section and recycled. Because the solvent does not leave the plant, its flow rate is constant throughout the recycle loop \( F_S \). The molar volumes of A and B species are assumed to be equal. The dimensionless parameters \( v_S = V_{k,S} / V_{k,A} \) corresponds to the molar volume of the solvent. The product stream is pure \( (\alpha_{Y,S} = 1) \), but some amount of B is recycled \( (\alpha_{A,Y} < 1) \).

Conventional control structure, dual composition control

The parameters of the mole-balance equations are: flow rates of A and B in the feed stream \( (F_A, F_B = 0) \); reactor volume \( (V) \); separation performance, \( (z_{B,P} = 1 \) and \( z_{A,Y} \)); solvent flowrate \( (F_S) \). The mole-balance equations give:

\[
x_A = \frac{Da \cdot z_{A,Y} - 1}{z_{A,Y} \cdot (Da + f_S \cdot v_S) + (1 - z_{A,Y}) \cdot (f_S - 1)}
\]

\[
z_{BS} = \frac{(Da - 1) \cdot z_{A,Y} + f_S \cdot (1 - z_{A,Y}) \cdot (Da - 1 + v_S)}{Da \cdot z_{A,Y} + f_S \cdot (Da - 1 + v_S)}
\]

\[
s = \frac{Da \cdot z_{A,Y} + f_S \cdot (Da + v_S - 1)}{Da \cdot z_{A,Y} - 1}
\]
Luyben control structure, dual composition control

The parameters of the mole-balance equations are: flow rates of A and B in the feed stream \(F_A, F_B = 0\); flowrate to separation \((S)\); separation performance, \((z_{B,P} = 1 \text{ and } z_{A,Y})\); solvent flowrate \((F_S)\). The mole-balance equations give:

\[
\begin{align*}
  x_A &= \frac{1}{s \cdot z_{A,Y} + (1 - s)} \quad \text{(A7.4)} \\
  z_{B,S} &= (1 - z_{A,Y}) + \frac{z_{A,Y}}{s} \quad \text{(A7.5)} \\
  \frac{k \cdot V}{F_A \cdot V_{pA}} &= \frac{s + s \cdot (v_s - 1)}{(s-1) \cdot z_{A,Y} - s} \quad \text{(A7.6)}
\end{align*}
\]
Chapter 8. Conclusions and Suggestions for Further Research

The goal of this thesis was to investigate the integration between design and control of chemical plants. In contrast with the traditional methodology, where first several design alternatives are developed and afterwards their controllability is analysed, a method to generate only easily controllable designs was sought.

Due to the complexity of the problem, a systemic approach emerged. More precisely, the activity of conceptual design is viewed as selecting and assembling building blocks (called Basic Flowsheet Structures, BFS) in such a way that a system with required functionality is obtained. When this point of view is adopted, the main objectives of the plantwide control are achieved by coordination of the (local) BFS control. This way, distinction is made between local and plantwide control. Consequently, research of integration between design and control should proceed along two lines:

1. **Design of controllable BFS.** There is a lot of industrial experience regarding the design of unit operations having good controllability properties. However, in many cases, the interaction between some units is so strong that they must be considered as one system (BFS). Heat-integrated distillation and heat-integrated reactors are examples that have been analysed in this thesis. Other systems, as thermally coupled distillation systems, reactive distillation, azeotropic distillation with solvent recycle, are possible subjects for further research.

2. **Couple the BFS in a controllable flowsheet.** In this thesis, it is shown that the main task of the plantwide control is to maintain the mass balance of the plant, by avoiding accumulation / depletion of all chemical species involved in the process. Because the chemical reactor is the place where species are formed / consumed, it plays an important role in the plantwide controllability. Hence, study of reactor-separator-recycle systems can provide valuable insights into design of controllable flowsheet. In this thesis, the steady-state behaviour of several isothermal reactor-separator-recycle systems has been analysed, and implications on control have been discussed.
Although necessary, good steady-state controllability is not sufficient. Therefore, the dynamic aspects should also be introduced. This raises the subject of development of dynamic models to be used in the early stages of conceptual design.

Further research can be also directed towards reaction systems with a more complex stoichiometry, where more than one reactor might be necessary. Moreover, including the heat effects (no-isothermal reactors) may add more complexity.

The author of this thesis considers that the interaction between design and control can be revealed only by mathematical models that take into account the nonlinearities existing in every chemical process. This does not exclude the use of linear models, but requires the linearisation to be performed around several operating points. The use of nonlinear analysis during design can be summarised as follows: divide the space of the design parameters into regions with different steady-state and dynamic bifurcation diagrams, and identify desirable regions of operation and potential stability or operability issues. Avoid operation near bifurcation varieties.

However, there might be situations when high-performance requires operation near bifurcation points. In this cases, because of unavoidable disturbances and design parameter uncertainties, the control system must ensure that possible bifurcations are supercritical, that is they do not lead to a catastrophic change of system’s behaviour. This is an active field of mathematical research.

The classification methodology presented in this thesis can be applied to one state-variable problems. Although many chemical systems fall in this category, this is a hypothesis that should always be checked. Systems with more than one state-variable may exhibit very complex steady-state and dynamic behaviour whose complete classification, unfortunately, has not been achieved.

Finally, the following steps are recommended to integrate design and control:

1. Non-linear analysis: choose / identify the design variables; formulate controllability criteria; classify the operating points according to controllability criteria; identify feasible / unfeasible regions.
2. Generate design alternatives, using any available methodology.
3. Perform linear controllability analysis for the design alternatives
4. Select a suitable design
Summary

This thesis brings the following contributions to the field of integration between design and control:

1. Systemic, two-level approach to integrated design and control of chemical processes (Chapter 1). This involves: i) design of controllable basic flowsheet structures (BFS), the building blocks of chemical plants, and ii) coupling the BFS in a controllable flowsheet. Taking into account the main task of plantwide control, which is to maintain the plant’s mass balance, the integration of design and control can be achieved at a fundamental level of the hierarchical conceptual design procedure. The reactor and separation are assumed to be controllable, this assumption being an explicit task for their design. Then, plantwide control structures can be proposed and their performance can be assessed based on steady state sensitivity analysis or on more advanced tools. Toluene hydrodealkylation plant is used as a case study.

2. Guidelines for controllable design of a heat-integrated prefractionator / side-stream column configuration for ternary separation. In Chapter 2, several design alternatives are presented and discussed. Multi-input multi-output (MIMO) controllability analysis is performed based on linear models. It is demonstrated that the forward heat-integration arrangement with a small prefractionator has the best dynamic performances.

3. Nonlinear approach to problems where state multiplicity and instability limits the range of controllable designs. Chapters 3 and 4 study the steady-state and dynamic behaviour of heat-integrated Plug-Flow Reactors. First-order, irreversible and reversible, exothermic reactions, and adiabatic reactor operation are considered. The steady-state and dynamic behaviour is classified by computing the hysteresis, isola, boundary-limit, double-zero and double-Hopf varieties, which divide the space of design parameters in regions with different steady-state and dynamic bifurcation diagrams. State multiplicity, isolated solution branches and oscillatory behaviour are possible for realistic values of model parameters. The influence of reaction kinetics and thermodynamics and feed-effluent heat-exchanger (FEHE) efficiency on the extent of multiplicity region is studied. Subsequently, it is discussed how the results can be used to avoid operational problems and a design methodology is proposed. Further, Chapter 5 studies the interaction between design and
control of a heat-integrated PFR. Four different design alternatives, for which controllability problems are expected, are investigated. Three different control structures are considered. Linear controllability analysis shows that systems with small steam-generator and large FEHE are difficult to stabilise. Bypass around the FEHE can not be used to reject disturbances and there is no incentive to control both FEHE and furnace outlet temperatures. The nonlinear behaviour of the controlled system is analysed by bifurcation theory. The range of design parameters for which the system can not be stabilised is computed, by classifying the operating points according to their position relative to different bifurcation varieties. This way, the meaning of "large" and "small" units becomes clear. The results are confirmed by nonlinear dynamic simulation.

4. Steady-state classification of ideal, binary distillation by rigorous application of the singularity theory (Chapter 6). The mass reflux flow rate is considered as bifurcation parameter. Codimension-2 varieties, dividing the feed composition – boilup parameter space into regions with different types of bifurcation diagrams, are computed. In this way, all possible bifurcation diagrams are identified. The effect of physical and design parameters on the location and extent of the multiplicity regions is investigated.

5. Introduction of plant Damköhler number to study the nonlinear behaviour of the reactor separator-recycle systems (Chapter 7). The plant Damköhler number ($Da$) and the separation specifications are parameters of the dimensionless mole-balance equations. It is demonstrated that reactant accumulation does not occur and operation is possible if $Da > Da^\text{cr}$. The critical value $Da^\text{cr}$ corresponds to a bifurcation point of the mole-balance equations and depends on separation performance. For one reactant recycle and high purity separation, $Da^\text{cr} = 1$. When two reactants are recycled, multiple steady states are possible. In this case, $Da^\text{cr}$ depends also on the control structure. Close to $Da^\text{cr}$, control structures must change the reaction conditions in order to reject effectively the disturbances.

Chapter 8 presents authors comments on the research needed in the field of integration between design and control.
Samenvatting

Dit proefschrift gaat op verschillende manieren in op de integratie van procesontwerp en procesregeling:

1. Systematische aanpak van integratie van de regeling in het ontwerp van chemische processen (Hoofdstuk 1). Dit betreft: I) ontwerp van regelbare basic flowsheet structures (BFS) als bouwstenen van chemische fabrieken en ii) het samenstellen van een regelbaar fabrieksontwerp vanuit BFS'ën. Waar uitgegaan wordt van het instandhouden van de massabalans als primaire taak van de plantwide regeling, kan de integratie bereikt worden op het fundamentele niveau van de hiërarchische procedure voor conceptual design. Het regelbaar maken van de reactor en de scheiding is een expliciet onderdeel in hun ontwerpfase. Vervolgens worden plantwide regelstructuren ontworpen en getest op hun prestaties met behulp van steady-state gevoeligheidsanalyses of meer geavanceerde methoden. Als case study functioneert hier een Tolueen Hydroalkylatiefabriek.

2. Voorschriften om tot eenregelbaar ontwerp van een warmte-geïntegreerd, ternair scheidingsysteem van een prefractionator met zijstromen. In Hoofdstuk 2 wordt een aantal ontwerpalternatieven gepresenteerd en besproken. Multi-input, multi-output (MIMO) regelbaarheidsanalyse wordt uitgevoerd met behulp van lineaire modellen. Aangetoond wordt, dat voorwaardelijke warmte-integratie en een kleine prefractionator tot de beste resultaten leidt.

Samenvatting

Operationele problemen, en wordt er een nieuwe ontwerpmethodologie op basis hiervan voorgesteld. Verder gaat Hoofdstuk 5 in op de interactie tussen ontwerp en regeling van een warmte-geïntegreerde PFR. Vier verschillende ontwerpalternatieven, waarvoor regelbaarheidsproblemen verwacht worden, staan ter discussie, alsmede drie verschillende regelstructuren. Lineaire regelbaarheidsanalyse toont aan, dat systemen met een kleine stoomgenerator en een grote FEHE moeilijk te stabiliseren zijn. Een bypass rond de FEHE kan niet worden toegepast ter onderdrukking van verstoringen, noch bestaat er aanleiding om de uitgangstemperaturen van beide de FEHE en het fornis te regelen. Van het geregelde systeem wordt het niet-lineaire gedrag geanalyseerd met behulp van de bifurcatietheorie. De waarden van de ontwerpparameters, waarvoor geen stabilisatie bereikt kan worden, worden uitgerekend door de werkpunten naar hun positie ten opzichte van de verschillende bifurcatiepunten te classificeren. Op deze wijze wordt de betekenis van "grote" en "kleine" units duidelijk. Tenslotte blijken de resultaten te worden bevestigd met behulp van niet-lineaire, dynamische simulaties.


5. Introductie van het fabrieks-Damkohlergetal ter bestudering van het gedrag van reactor-separator-recycle-systemen (Hoofdstuk 7). Dit Da-getal en de specificaties van de scheidings zijn parameters in de dimensieloze molbalansen. Aangetoond wordt, dat accumulatie van reactant niet optreedt en het systeem operationeel kan zijn als $Da > Da^c$. De kritische waarde $Da^c$ is afgeleid van een bifurcatiepunt in de vergelijkingen voor de molbalansen en hangt af van de prestaties van het scheidingsysteem. Indien één reactant gerecycled wordt en hoge scheidingszuiverheid geldt dat $Da^c=1$. Bij recycling van twee reactanten gaan multipliciteiten voorkomen. In dat geval hangt $Da^c$ ook af van de regelstructuur. Het blijkt, dat dichtbij $Da^c$ de regeling reactiecondities wijzigt om effectief storingen te onderdrukken.

Hoofdstuk 8 bevat opmerkingen van de auteur van het proefschrift betreffende verder onderzoek dat nodig is op het gebied van integratie van ontwerp en regeling.

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