STUDIES ON OPTIMAL OPERATION AND CONTROL OF BATCH DISTILLATION COLUMNS

by

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ABSTRACT

Batch distillation is an important industrial unit operation in which a liquid mixture is separated into two or more products of different compositions. Batch operation offers greater flexibility when compared to continuous distillation. A multicomponent mixture can be separated into a number of product fractions in a single batch column whereas, in continuous operation several columns are required. A batch column can also handle a wider range of feed compositions, relative volatilities and product specifications. The same column can therefore be used for a number of different separation tasks. However, this type of separation requires the input of relatively large amounts of energy.

During the last century there was a shift from batch to continuous operation. However, recently this trend has been reversed. Batch distillation is becoming increasingly important as a result of the expansion in the fine chemical and pharmaceutical industries characterized by small amounts of products with high added value and greater demands for flexibility and productivity. With this renewed interest, there is a need to develop a better understanding of how best to operate conventional batch columns. Alternatives to the traditional column configuration should also be considered.

In this thesis, a short cut formula for the estimation of required startup time is presented and found to yield good results.

In difficult separations, a novel cyclic operating policy, in which the condenser holdup is repeatedly filled and dumped, has been found to significantly reduce the operating time when compared to conventional operating policies. The optimal operating parameters are easy to calculate and no changes are needed in the process equipment of a regular column in order to run the column according to this new policy.

In a regular column, the feed is charged to the reboiler in the bottom and the product is withdrawn from the top. Alternatively, the feed can be charged to the top and the product withdrawn from the bottom. This latter configuration is denoted an inverted column. A regular and an inverted column have been compared in terms of minimum operating time and the inverted column is found to be preferable for certain separations.

It is generally found that conventional batch distillation is not well suited for difficult separations or for separations where the amount of light component in the feed charge is small. For such separations, inverted columns or cyclic operation may be preferred.

A reactor may be combined with a distillation column in order to increase productivity. The reaction will then act as a continuous feed to the batch column. This combined operation is, however, difficult to control due to strong interactions between the two units. This thesis addresses the dynamic behavior and controllability of an industrial case study. The conventional top-temperature control strategy is found to be less suitable for batch columns and therefore alternative control strategies are considered.

The thesis concludes with a discussion of optimal performance in the same industrial case study. On-line operation according to an optimal policy is demonstrated in conjunction with both a continuous and a real time control system.

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Contents

Chapter 1

Introduction and Literature Review

Research is to see what everybody else sees,
and to think what nobody else has thought.

Albert Szent-Gyorgyi

Abstract

This chapter sets out the historical background and importance of batch distillation and the motivation for the work in this thesis. It begins with a description of the normal operation of a conventional batch distillation column. A brief overview of the literature concerning modeling, simulation, optimization and control of batch distillation is then given in order to put the work in this thesis into a wider context. Much of this work has involved finding optimal operating parameters. Therefore, a general optimal control problem is defined and alternative solution methods are discussed. The formulation of this general problem applied to optimal reflux ratio and optimal recycle problems in batch distillation, as it is described in the literature, is presented. The chapter concludes with a brief description of the following chapters in this thesis.

1.1 Background

Distillation is both the oldest and most important method of producing chemically pure substances. It was probably first discovered by the Alexandrian Chemists in the first century A.D. and was mainly used to produce alcoholic beverages and essential oils (Forbes, 1970). The early apparatus consisted of a heated still or pot, one or more receivers and a spout or tube leading from the heated still to the receiver in which the vapors were condensed. This type of apparatus is the simplest form of *batch distillation* and is now often referred to as Rayleigh distillation after the author of the first theoretical work on this type of separation (Rayleigh, 1902). During the centuries the method of batch distillation was constantly improved. Better cooling avoided distillation losses and at the same time allowed for more rapid separation. Fractionation was obtained in a simple way by making the still-head larger. There were also some attempts at refluxing of distillate back to the still to obtain better fractionation. By 1860 the distillery industry generally began to demand for continuous distillation columns and this new mode of operation gradually became more common as the chemical industry grew. Today, distillation is still the most important separation method in the chemical and petrochemical industries. Distillation columns constitute a significant fraction of the capital investment in chemical plants and refineries. Their operating costs are often a major part of the total operating cost of many processes (Luyben, 1992). Distillation is also a major energy consumer in a broader perspective; in the USA about 7% of the total energy consumption is spent in distillation (Gmehling *et al.*, 1994). Batch distillation columns generally have a higher energy consumption than equivalent continuous columns. However, no real comparison has been found in the literature.

1.2 Motivation

Batch distillation is becoming increasingly important as a result of the expansion in the fine chemical and pharmaceutical industries characterized by small amounts of products with high added value. These speciality chemicals represent about 15% of worldwide chemicals production with approximately $150 billion produced annually (Sundaram and Evans, 1993b).

In the fine chemical industry, the amount of a specific product varies considerably from only a few kilos up to several hundred tons per year. Often, the same plant is used to manufacture a large number of products. Also the recipes of the different products may vary largely from one product to another. The process equipment in the plant must therefore be very flexible. Batch distillation has the advantage of being able to recover a number of products from a feed charge in a single column. A batch column can handle multicomponent mixtures, a wide range of feed compositions and degree of difficulty of separation in terms of a wide range of relative volatilities and product purities. In other words, batch distillation offers a maximum of flexibility to answer the random demand for chemical products.

In the pharmaceutical industry, each product must be accompanied by a list of raw-materials used (origin and specifications) and also of all the steps (reaction, separation, crystallization etc.) it has gone through during the manufacturing. This makes batch distillation more suited than continuous distillation for separation processes.

Batch columns are often linked closely with reactors (Lucet *et al.*, 1992). By removing one or more products from the reaction by distillation, much higher conversions will be achieved than otherwise possible. This is due to two factors; 1) the reaction is kept away from equilibrium by removal of one or more of the products and 2) the reaction temperature is increased since one or more of the light components are removed.

Many larger installations also feature batch columns. Material to be separated may be high in solids content, or it may contain tars or resins that would plug or foul a continuous column. Use of a batch unit can keep solids separated and permit convenient removal at the end of the process (Perry and Green, 1984).

Although distillation is one of the most intensively studied and better understood processes in the chemical industry, its batch version still represents an interesting field for academic and industrial research for a number of reasons:

1. Even for a simple binary mixture there are several alternative operations possible. With complex trade-offs as a result of the many degrees of freedom available, there is ample scope for optimization. Since the process is intrinsically dynamic, its optimization leads to an optimal control problem, for which problem formulation and numerical solution methods are not yet well established (Macchietto and Mujtaba, 1992). However, it is still possible to consider rather complex operating policies.

2. Industrial batch columns are often run as if they are merely a larger version of laboratory columns meaning that the control strategies are very simple. This is a general characteristic for small companies who are not able to support research and development in engineering as well as in the synthesis and production of their products (Lucet *et al.*, 1992). Therefore, there is a need for practical and reliable methods for development of control strategies for batch columns.

3. Batch distillation is a very old unit operation which has been run in more or less the same way for centuries. The demand for efficiency and productivity is continuously increasing. However, the optimal operation of columns is limited by the given operating policy. It is therefore important to develop new and improved policies in order to increase productivity.

1.3 Conventional batch distillation and its operation

A conventional batch distillation column consists of the following: a still or reboiler which is heated either by a heating jacket or by circulating its content through a heat exchanger; a column section with trays or filled with packing material; a condenser which condenses the vapor leaving the column section; a condenser reflux drum which collects the condensed vapor and one or more accumulator tanks where the distilled product is collected. Some of the distillate is returned to the column section as reflux. A schematic representation of the process is given in Figure 1.1. The order of appearance of the components in the distillate is determined by the phase equilibria characteristics of the mixture with the most volatile component first, then the second most volatile etc. The distillate composition will normally vary during the operation and usually a number of cuts or fractions are made. This is shown in Figure 1.2 for a binary mixture with A as the most volatile component. Some of the cuts are desired products while others are intermediate fractions which do not meet the product specifications. These off-cuts are either recycled to subsequent batches or processed separately. This type of operation is particularly useful when using an existing column with perhaps a less than optimal number of stages. At the end of the separation the content in the column section will drain down into the still and a residual bottom fraction may or may not be recovered as product.

Operation of a conventional batch distillation column can be conveniently described in three parts: 1) startup period, 2) production period and 3) shutdown period. The column is usually run under total reflux during the startup period until the unit is taken to a steady state or to a state when the distillate composition reaches the desired product

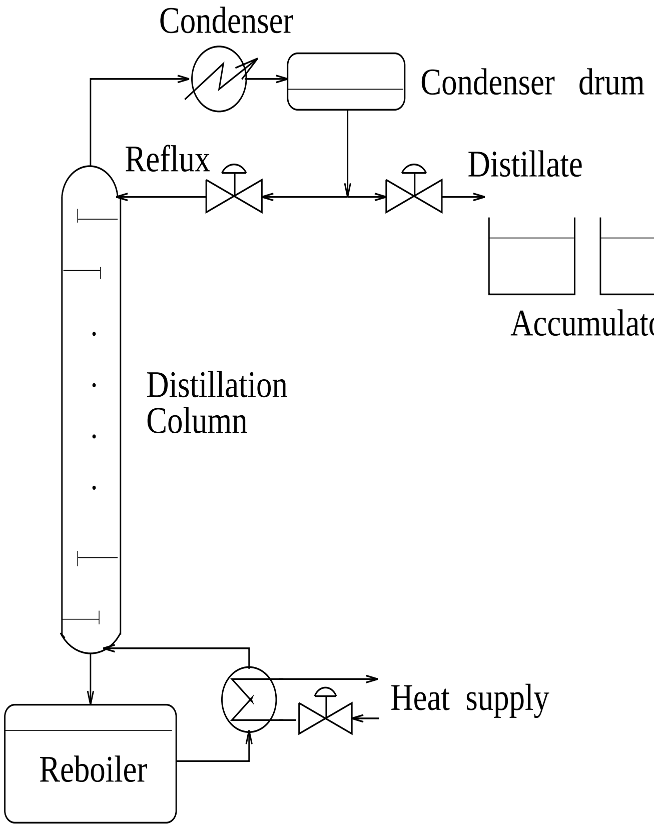


Figure 1.1: Conventional batch distillation column.

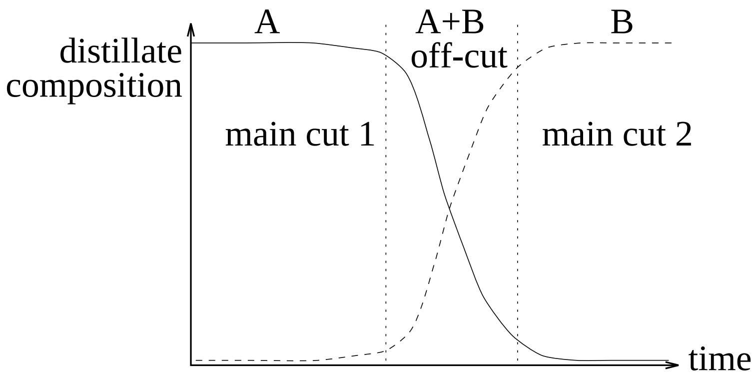


Figure 1.2: Distillate composition as a function of time.

purity. The production period is normally the most time consuming but for difficult separations such as for high purity or azeotropic separations, the startup time may also be significant. During the production period, the column is usually operated in one of the following ways (see Figure 1.3):

1. *Constant reflux ratio.* The reflux ratio is set at a predefined value at which it is maintained for the given cut. The instantaneous distillate composition is changing.

2. *Constant distillate composition.* When it is desired to maintain a constant distil-

late composition, the amount of reflux returned to the column must be constantly increased throughout the run.

3. *Optimal reflux ratio.* The reflux ratio is found based on some optimal criterion. Both the reflux ratio and the distillate composition will normally vary during the run. Operation according to an optimal distillate composition is equivalent.

The reflux ratio is defined as either the ratio between reflux and distillate flow (external reflux ratio), or as the ratio between reflux and vapor flow (internal reflux ratio).

The optimal reflux policy is generally found to be one of increasing reflux as the batch progresses. It has always been reported to lie between those from the constant reflux ratio and the constant distillate composition policies. The constant distillate composition policy is generally found to yield a better performance than the constant reflux policy.

1.4 Some basic definitions

The following basic definitions are used in later chapters:

Operating strategy: The decision regarding the number of product cuts and off-cuts together with a scheme for recycling off-cuts.

Operating policy: The decision on how to operate the column for a given operating strategy. Conventional operating policies are: 1) constant reflux ratio policy, 2) constant distillate composition policy and 3) optimal reflux ratio policy.

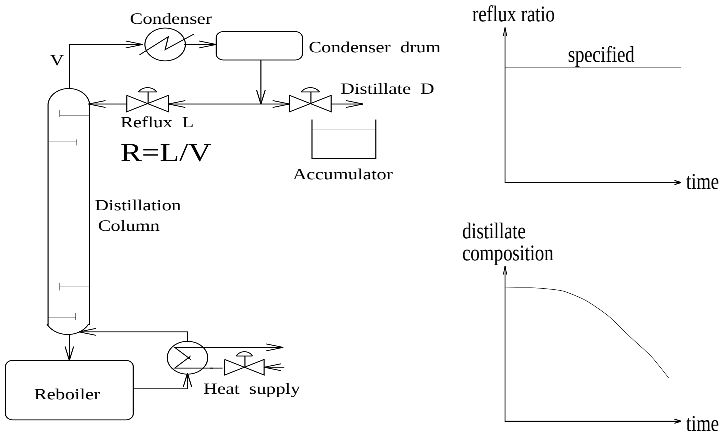
Column configuration: The arrangement of the column as to where the feed is added and where the product is withdrawn.

Control strategy: The decision on how to control the column for a given operating strategy and operating policy. This included the selection of type of measurements and controllers and also the location of the measurement points.

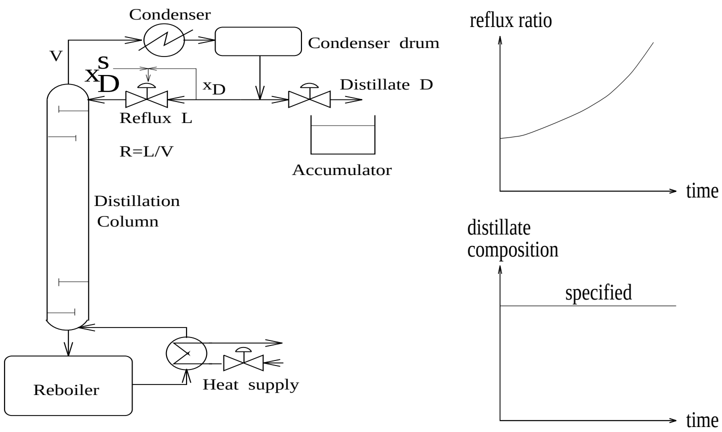
Control configuration: The desicion on how to pair the measured and manipulated variables. It is customary in continuous distillation to denote the control configuration on the basis of which controllers are used for composition control. An example is the LV-configuration where reflux flow L and boilup rate V are used to control the compositions in the top and bottom of the column. This notation is also used in this thesis.

Controllability: The definition of controllability used in this thesis is: The ability to achieve acceptable control performance irrespective of the controller. Controllability is an inherent property of the process and is therefore independent of controller tuning.

1. Constant reflux ratio policy



2. Constant distillate composition policy



3. Optimal reflux ratio policy

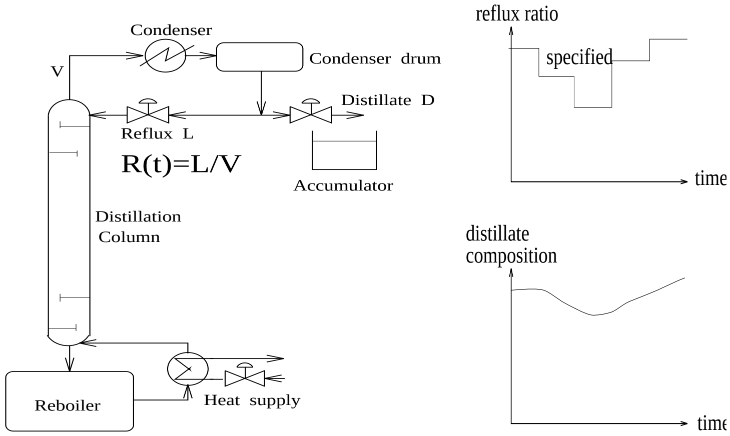


Figure 1.3: Conventional operating policies.

Table 1.1: Selected papers on modeling and simulation of batch distillation.

Authors	feed	reaction	plate hydraulics
Huckaba and Danly (1960)	binary	no	no
Meadows (1963)	multicomponent	no	no
Distefano (1968)	multicomponent	no	no
Domenech *et al.* (1974a)	multicomponent	no	no
Domenech *et al.* (1974b)	multicomponent	no	no
Egly *et al.* (1979)	multicomponent	yes	no
Boston *et al.* (1980)	multicomponent	no	no
Gallun and Holland (1982)	multicomponent	no	yes
Sadotomo and Miyahara (1983)	multicomponent	no	no
Cuille and Reklaitis (1986)	multicomponent	yes	no
Reuter *et al.* (1989)	multicomponent	yes	no
Galindez and Fredenslund (1988)	multicomponent	no	no
Ruiz (1988)	multicomponent	no	yes
Albet *et al.* (1991)	multicomponent	yes	no
Chiotti and Iribarren (1991)	binary	no	no
Diwekar and Madhavan (1991)	multicomponent	no	no
Albet *et al.* (1994)	multicomponent	yes	no
Diwekar (1994)	multicomponent	no	no

1.5 Modeling and simulation of batch distillation

Batch distillation is an inherently unsteady state process since material is being withdrawn and the reboiler holdup and compositions are continuously changing. A mathematical model of the process will therefore be dynamic and involve both differential and algebraic equations. A general model usually includes mass and energy balances, phase equilibria, plate hydraulics etc. The complexity of the models presented in the literature has increased in parallel with the development in computer power and speed. An overview of papers considering modeling and simulation of batch distillation is given in Table 1.1. (Other, and generally simpler models, are also presented in the papers considering optimization of batch distillation, Table 1.2.)

Common assumptions for batch distillation models are: 1) Negligible vapor holdup; 2) adiabatic plates; 3) perfect mixing and equilibrium on all trays; 4) constant pressure; 5) total condensation with no subcooling and 6) constant tray holdup. The most comprehensive model was presented by Ruiz (1988) and involved the geometry of the trays, heat transfer in the reboiler and column section etc. In contrast, Diwekar (1994) studied the trade-off between accuracy and efficiency for reduced order models based on a short-cut method.

With the increased computer speed and improved solutions methods for DAE (differential and algebraic equation) and ODE (ordinary differential equation) systems, rigorous simulation of batch distillation is now possible. Models of high complexity including physical properties from thermodynamic packages can be solved using, for example, commercial equation solvers like SPEEDUP (1993).

All simulations in this thesis have been performed using SPEEDUP (1993).

1.6 Optimization of batch distillation

The research on optimization of batch distillation has involved many aspects such as binary or multicomponent mixtures, columns with or without holdup, recycled off-cuts etc. Rigorous optimization of batch distillation presents serious theoretical and computational difficulties due to:

1. Time dependence of the operation with discontinuities
2. Modeling equations which involve differential algebraic equations (DAEs) whose dimensions strongly depends on the number of plates and components
3. The presence of several objectives when multicomponent multifraction distillation is considered
4. A mixture of discrete (e.g. sequence of product cuts and off-cuts, recycling of off-cuts) and continuous (e.g. reflux ratio, energy input) variables

In the literature before the last decade, the column performance was generally described by very simplified models due to limited computer resources. However, with the continuous increase in computational power the complexity of models has increased. In the more recent literature the emphasis has been more on solution methods than on the actual results of the optimization.

1.6.1 Discrete versus continuous variables

The separation of multicomponent mixtures in a batch column offers a large number of degrees of freedom. The number of product cuts and off-cuts and the possible recycling of off-cuts are typical *discrete* decision variables. Whether an off-cut should be produced or not is a yes/no decision and so is the decision as to if and how it is to be recycled. A specific instance of these decisions is often called an *operating strategy.* Other decision variables vary *continuously* during the batch. Although the main operating variables are the reflux ratio and energy input, the pressure and condenser drum holdup may also vary.

Even for binary separations there are a number of possible operating strategies (off-cut or no off-cut, recycle policy etc.). Also one or more operating variables may vary with time for each operating strategy. The complexity increases substantially with the number of components in the feed. Selecting the best operation thus presents a formidable challenge. The problem could in principle be posed as a large mixed integer nonlinear dynamic programming problem (MINDLP), but so far only small subsets of this problem have been studied (Macchietto and Mujtaba, 1992). The operating strategy is normally fixed a priori and in most papers only the reflux ratio is optimized. However, with the increasing development in the area of optimization theory, larger and larger problems are being considered. Indeed, during the last few years some authors have also considered optimal recycle policies.

1.6.2 The general optimal control problem

Since batch distillation is an intrinsically dynamic process, the equality constraints for the optimization problem are ordinary differential and algebraic equations (DAEs). This type of problem is generally referred to as an *optimal control problem* and is an element of the field of control theory which has experienced significant development since the 1950s.

The optimal control problem can generally be stated as:

$$\min_{\dot{x}(\cdot),x(\cdot),y(\cdot),u(\cdot),v,x(t_0),t_f} \Phi(\dot{x}(t_f), x(t_f), y(t_f), u(t_f), v, t_f)$$

subject to

$$g(\dot{x}(t), x(t), y(t), u(t), v, t_f, t) = 0$$

$$h(\dot{x}(t_f), x(t_f), y(t_f), u(t_f), v, t_f) \geq 0$$

where Φ is the objective function depending on the conditions at the final time t_f, g the equality constraints (DAEs) and h the inequality constraints at the end point. Interior point or path constraints may also exist. Upper and lower bounds may be defined for the control variables u, design variables v, initial states x_0 and final time t_f. The variable x denotes the state variables and y the algebraic variables. The optimization problem can also be reformulated to a maximization problem.

The optimal control problem is often referred to as an infinite dimensional problem because u is a function of time rather than a specific value at the optimum. This type of problem can be solved using Pontryagin's maximum principle. However, this solution method is usually time consuming. Therefore it is more efficient to employ nonlinear programming rather than the maximum principle to compute the optimal control. This method is preferred by more recent authors. The optimization problem can then be solved by numerical techniques that convert the problem into a nonlinear programming problem (NLP). Two general approaches have appeared in the chemical engineering literature in recent years. These are either based on

1. Control vector parameterization (e.g. Farhat *et al.*, 1991)

2. Full parameterization of both control and states variables; often referred to as the Collocation method (e.g. Logsdon and Biegler, 1989)

Control vector parameterization

By parameterizing the control vector, one can think of the true optimal control profile as being approximately represented by a simple basic function, for example, constant, linear, quadratic or exponential. The basic function is defined for a control interval and the number of control intervals is determined a priori. An example is given in Figure 1.4 where the optimal control profile is approximated by first a constant and then a linear function over the two control intervals. With this method, the control profile can be

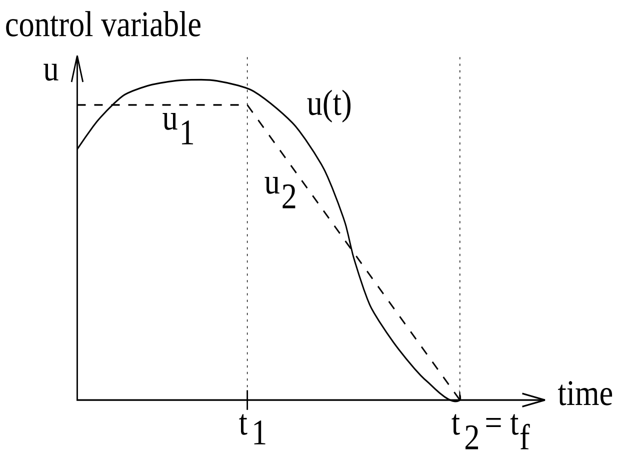


Figure 1.4: Example of control parameterization.

fully described by the number of control intervals, the control functions (constant value, linear etc.) and the duration of the intervals. These parameters are then added to the optimization problem to form a finite set of decision variables.

The solution method usually includes two levels. The first or inner level performs a simulation to solve all the equality constraints (DAEs) and calculates the inequality constraints and the objective function. The second or outer level performs the actual optimization which is a standard NLP problem, solvable by, for example, Sequential Quadratic Programming (SQP) (Vassiliadis, 1993). This approach is commonly known as the *feasible path approach*. The advantage is that the resulting optimization problems are in general small in terms of the number of variables. If the process is terminated at the outer optimization level before reaching a solution, the terminating point is feasible and may be acceptable as a practical, although sub-optimal, solution to the problem. However, since the feasible path approach requires a complete solution of the DAEs for each trial value of the decision variables, this may be computationally expensive. The simulation level may also fail since a feasible region may not exist for certain values of the decision variables.

Full parameterization, the Collocation method

The collocation method discretizes the control functions as well as the ordinary differential equations in the original DAE model, using collocation over finite elements. The result is a large system of algebraic equations, which, together with the objective function and the inequality constraints, forms a sizable NLP problem. For the same problem, however, the degrees of freedom for this large scale NLP is the same as for the small scale NLP in the control parameterization method. Since the DAEs and the optimization problem are solved simultaneously, this approach is called an *infeasible path approach*. The main advantage is that the repeated integration is avoided and the method should therefore be faster.

However, comprehensive comparisons between control vector parameterization and the collocation method have not been published, so it is not possible to make definite statements about their relative merits (Macchietto and Mujtaba, 1992).

In this thesis, the optimization program DAEOPT developed at Imperial College in London (Vassiliadis, 1993) has been used. The program is based on control vector parameterization and the optimization is solved by Sequential Quadratic Programming (SQP). At present, there are no commercial programs available for dynamic optimization.

1.6.3 Reflux ratio optimization

In most of the available literature on optimization of batch distillation only the reflux ratio is considered as an optimization variable. The operating strategy, e.g. the number of product cuts and off-cuts, is specified a priori. The *optimal control problem* is in the literature usually formulated in one of the following ways:

1. The *minimum time problem*: The optimal policy is that which produces a required quantity of distillate of specified purity in the shortest possible time. The optimization problem can be stated as

$$\min_{R(t)} t_f$$

$$\begin{aligned} x_A &\geq x_A^{spec} \\ H_A &\geq H_A^{spec} \end{aligned}$$

which in words is to find the optimal reflux ratio as a function of time $R(t)$ which minimizes the total operating time t_f subject to the constraints that the composition and amount of distillate must meet their specifications (x_A^{spec} and H_A^{spec} respectively).

2. The *maximum distillate problem*: The optimal policy is that which produces the maximum amount of distillate of specified purity in a fixed duration of time. The optimization problem can be stated as

$$\max_{R(t)} H_A$$

$$\begin{aligned} x_A &\geq x_A^{spec} \\ t_f &= t_f^{spec} \end{aligned}$$

equivalent to finding the optimal reflux ratio as a function of time $R(t)$ which maximizes the amount of distillate H_A subject to the constraint that the composition of distillate must meet its specification x_A^{spec} with the total operating time given a priori $t_f = t_f^{spec}$.

3. The *maximum profit problem*: It is also possible to determine the optimal operating policy by optimization of some economic criterion. The optimization problem can then be stated as

$$\max_{R(t)} P = \max_{R(t)} \frac{C_A H_A - C_F H_F - \int_0^{t_f} Q_R(t)dt}{t_f}$$

$$
\begin{aligned}
x_A &\geq x_A^{spec} \\
H_A &\geq H_A^{spec}
\end{aligned}
$$

or finding the optimal reflux ratio as a function of time $R(t)$ which maximizes the hourly profitability P where C_A and C_F are the prices of the distillate product and the feed respectively and $\int_0^{t_f} Q_R(t)dt$ is the total energy consumption. Possible constraints are that the composition and amount of distillate must meet their specifications (x_A^{spec} and H_A^{spec} respectively).

Other types of constraints, e.g. recovery of a component, conversion of a reactant etc. are also possible and have been considered in the literature.

The three control objectives differ in the way the optimal performance of the column is defined. In the first objective, it is considered to be optimal to use as little time as possible for the production of a fixed quantity, for example, a given amount of material of a set purity required by a downstream process. Also storage may be limited thereby restricting the amount produced. The time frame might be fixed, for example to the duration of one shift. The control objective will then be to produce as much as possible within this time frame; in other words a maximum quantity of distillate. Normally less production time will give a higher profit. The main reason why objective three, maximum profit, is rarely used is because this requires additional cost parameters which can be difficult to estimate.

Table 1.2 sets out an overview of papers on optimization of batch distillation in terms of optimal reflux ratio for a given operating strategy. It also includes the control problem solved and the solution method used. Generally, in the earlier literature the optimization problem was solved using Pontryagin's maximization principle. In the more recent studies Non Linear Programming has been preferred.

Earlier studies considered very simple dynamic models. With the continuous increase in computational power the complexity of both the model and the optimization problem has increased.

Only a few papers consider other operating variables than the reflux ratio. However, Hansen and Jørgensen (1986) and Christensen and Jørgensen (1987) also considered an optimal boilup rate.

The results from the optimization studies generally showed an increased performance of the column using an optimal reflux policy, compared to the conventional constant reflux ratio and constant distillate composition policies. The improvement in either time saving or productivity ranged from marginal up to about 20% depending on the mixtures investigated. The increase in profit was reported to be up to 40%.

1.6.4 Recycle optimization

When a mixture is distilled in a batch column it is not necessary that each charge is separated into a distillate and a residue which simultaneously meet the imposed specifications. Any off-spec material may be reprocessed in one of the following ways:

1. The off-cuts are mixed with fresh feed in the next batch

Table 1.2: Selected papers on optimization of batch distillation.

Problem solved, Authors	holdup	off-cuts	feed	solution
MINIMUM TIME:				
Coward (1967a)	no	no	binary	max. principle
Coward (1967b)	no	no	binary	max. principle
Robinson (1969)	yes	no	key comp.	max. principle
Robinson (1970)	yes	no	key comp.	max. principle
Robinson (1971a)	no	yes	binary	max. principle
Robinson (1971b)	yes	no	binary	max. principle
Mayur *et al.* (1970)	no	yes	binary	max. principle
Mayur and Jackson (1971)	no	no	multicomp.	max. principle
Egly *et al.* (1979)	yes	no	multicomp.	max. principle
Hansen and Jørgensen (1986)	no	no	binary	max. principle
Christensen and Jørgensen (1987)	no	yes	binary	max. principle
Mujtaba and Macchietto (1988b)	yes	yes	binary	NLP
Macchietto *et al.* (1989)	yes	yes	binary	NLP
Mujtaba and Macchietto (1991)	yes	no	binary	NLP
Mujtaba and Macchietto (1992b)	yes	yes	multicomp.	NLP
MAXIMUM DISTILLATE:				
Converse and Gross (1963)	no	no	binary	max. principle
Converse and Huber (1965)	yes	no	binary	max. principle
Keith and Brunet (1971)	no	no	binary	max. principle
Diwekar *et al.* (1987)	no	no	multicomp.	max. principle
Mujtaba and Macchietto (1988a)	yes	no	multicomp.	NLP
Farhat *et al.* (1990)	no	no	multicomp.	NLP
Farhat *et al.* (1991a)	yes	no	multicomp.	NLP
Farhat *et al.* (1991b)	no	no	multicomp.	NLP
MAXIMUM PROFIT:				
Kerkhof and Vissers (1978)	no	no	binary	max. principle
Diwekar *et al.* (1989)	no	no	multicomp.	NLP
Logsdon *et al.* (1990)	no	no	multicomp.	NLP
Mujtaba and Macchietto (1992a)	yes	yes	multicomp.	NLP
Mujtaba and Macchietto (1993)	yes	yes	multicomp.	NLP

2. A number of off-cuts are stored and subsequently reprocessed separately

3. Each off-cut is stored and reprocessed separately (all the first off-cuts together, all the second off-cuts together etc.)

4. The off-cuts are charged to the column at an appropriate tray and time during the course of the next batch (Luyben, 1988)

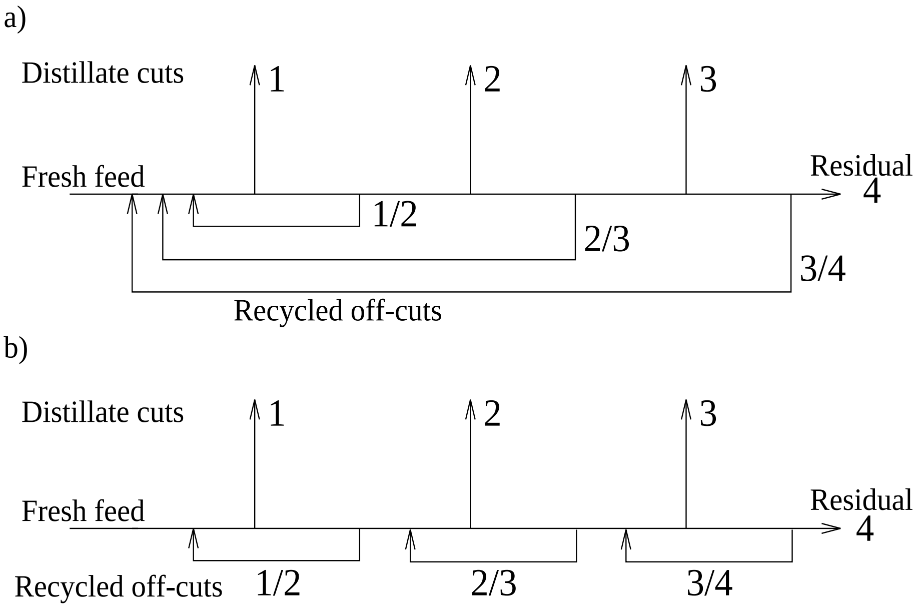


Figure 1.5: Recycling of off-cuts: a) to the next feed or b) to the reboiler at an appropriate time during the next batch.

5. Each off-cut is charged to the reboiler at an appropriate time during the course of the next batch, e.g. each cut is recycled back to the point where it is to be taken off as product (Mujtaba and Macchietto, 1992b)

The first and last alternative are schematically represented in Figure 1.5 for a four component mixture with three distillate product cuts, three off-cuts and one residue product cut. The numbers are for the components in order of decreasing volatility. Reprocessing the off-cuts will increase recovery of the products but at the expense of a reduced processing capacity or a longer operating time giving an interesting trade-off. The simplest procedure is to mix the off-cuts with fresh feed in the next batch. Here no extra storage tanks are needed and complex charging procedures are avoided. In the multicomponent case this means mixing components which have already been separated and this is clearly not optimal. However, optimal recycling of off-cuts at different times during the batch may be difficult to achieve in practice due to disturbances and changing conditions. In a binary separation, the composition of the off-cut will not be much different from that of the feed (Luyben, 1988). The mixing with fresh feed is therefore justified.

When the off-cuts are recycled to the next batch, a cyclic operation is established which will be quasi-steady state, i.e. subsequent batches will follow the same trajectories and produce the same product cuts and off-cuts. Luyben (1988) indicates that in practice such a quasi steady state mode will be achieved after approximately three or four cycles.

It should be noted that there are two common policies regarding the *size* of the consecutive batches of fresh feed (Chiotti, 1993):

1. The reboiler is full for every cycle such that successive admissions of fresh feed are smaller as the size of the off-cuts increase. The size of the feed charge to be processed (fresh and recycled) remains constant.

Table 1.3: Selected papers on recycle optimization of batch distillation.

Authors	feed
Mayur *et al.* (1970)	binary
Christensen and Jørgensen (1987)	binary
Luyben (1988)	ternary
Mujtaba and Macchietto (1988b)	binary
Mujtaba (1989)	multicomponent
Quintero-Marmol and Luyben (1990)	ternary
Macchietto and Mujtaba (1992)	multicomponent
Mujtaba and Macchietto (1992b)	multicomponent
Chiotti *et al.* (1993)	multicomponent

2. The amount of fresh feed is constant such that the total feed charge (fresh and recycled) increases with the size of the off-cuts.

An overview of papers that have considered optimization of recycled off-cuts is given in Table 1.3. The optimal recycle policy for the binary case was studied by Mayur *et al.* (1970), Christensen and Jørgensen (1987) and Mujtaba and Macchietto (1988b). Christensen and Jørgensen developed a measure for the degree of difficulty of separation in order to establish the advantages associated with recycling the off-cut. This measure was extended to the multicomponent case by Mujtaba (1989). It was generally found that recycling of off-cuts was advantageous for difficult separations with significant time savings. Luyben (1988) considered ternary mixtures where the off-cuts were recycled back to the feed of the next batch. The results were based on repeated simulations. Quintero-Marmol and Luyben (1990) considered six different off-cut handling schemes and found that, in general, mixing had a detrimental effect on the rate of products obtained. Again, the results were based on repeated simulations. Mujtaba and Macchietto (1992b) developed a strategy to calculate the optimal amount and composition of the off-cuts for multicomponent mixtures to yield a minimum batch cycle time for the quasi-steady state. Simultaneously, they also found the optimal reflux ratios. The off-cuts were assumed to be stored separately and recycled to the reboiler in sequential order. The overall problem was decomposed into a sequence of pseudo-binary minimum time problems. Macchietto and Mujtaba (1992) considered the same recycle policy but with an outer maximum profit optimization loop. The same recycle policy was also considered by Chiotti *et al.* (1993) but their results were based on steady state calculations only.

1.7 Control of batch distillation

The off-line optimization of batch distillation has received much attention over the years both in terms of reflux profiles and recycle polices (see Tables 1.2 and 1.3). These optimal results can in turn be used as set points in on-line operation. However, so far controllability

Table 1.4: Selected papers on control of batch distillation.

Authors	type of study
Egly *et al.* (1979)	optimization and non-linear control
Luecke *et al.* (1985)	Dynamic Matrix Control (DMC)
Yoon (1986)	Dynamic Matrix Control (PhD.-thesis)
Macchietto *et al.* (1989)	on-line control using optimal profiles
Reuter *et al.* (1989)	modeling and PI control
Quintero-Marmol (1990)	inferential control (PhD.-thesis)
Quintero-Marmol and Luyben (1991)	extended Luenberger observer
Bosley and Edgar (1992)	Nonlinear Model Predictive Control (NMPC)
Quintero-Marmol and Luyben (1992)	inferential model-based control

of batch columns using these optimal parameters has not been studied. Also design of controllers and control strategies to track the set points has received very little attention.

A summary of papers considering closed loop control of batch distillation is given in Table 1.4. Egly *et al.* (1979) considered optimization and control of an esterification process using a non-linear control system based on temperature measurements at different locations in the column. They also discussed how to select the measurement points. Luecke *et al.* (1985) used Dynamic Matrix Control (DMC) to operate an integrated farm energy system in which a packed batch column was used. The process was approximated as first order with dead time. Macchietto *et al.* (1989) used off-line optimal control policies together with supervisory control techniques for conducting binary batch distillation campaigns on-line. Reuter *et al.* (1988) incorporated the simulation of PI-controllers in their model of a batch column with reaction only in the reboiler. They stated that their model could be used for the investigation of control structure with the aid of Relative Gain Array analysis (RGA) but no details were given. Quintero-Marmol and Luyben (1991) explored the use of observers to predict compositions in multicomponent batch distillation through the use of temperature measurements. A design methodology was developed that addressed the issues of number and location of temperature sensors, the placement of the closed-loop poles of the observer and the sensitivity of initial condition estimates. Bosley and Edgar (1992) considered implementation of optimal profiles using Nonlinear Model Predictive Control (NMPC). They reported varying dynamic gains and response times from operation with a variable reflux ratio. Quintero-Marmol and Luyben (1992) presented results of a study of two types of model-based inferential control systems for multicomponent batch distillation. They showed that both steady-state and dynamic estimators provided good estimates of the distillate compositions using only one temperature measurement.

1.8 Alternative operating policies for batch distillation

The demand for efficiency and productivity of batch distillation is continuously increasing. However, the optimal operation of columns is limited by the given operating policy and the process equipment. So far only the reflux ratio and the recycle policy have been considered in the literature as ways of increasing productivity. Other factors should therefore be considered. Examples are:

1. Varying condenser holdup (as an additional optimization parameter)
2. Varying operating pressure (as an additional optimization parameter)
3. Varying boilup rate (as an additional optimization parameter), for example, reducing it as the reboiler is gradually depleted
4. Starting the batch with product from the previous batch in the condenser and/or column section to reduce the startup time
5. Concentrating the product in several steps when the required separation can not be achieved in the given column
6. Inverting the column by charging the feed to the condenser drum and withdraw product from the bottom
7. Adding an entrainer to improve the separation, for example, to break an azeotrope
8. Using steam distillation, or generally continuously adding a volatile component to effectively reduce the pressure

Most of these items are of course well known, but they have usually not been considered as optimization variables. The two last items are also well known from continuous distillation. The addition of a volatile component to the batch charge will effectively reduce the pressure and thereby lower the boiling point temperature. This makes it possible to use a lower-value energy source and to avoid possible thermal decomposition. One special case is where the heat is direct supply of energy in terms of steam as in steam distillation. Another possible strategy, when the top product is sent to waste-water treatment, is to use pure water as reflux similar to an absorption column.

Several authors have suggested the use of an inverted column, where the feed is charged to the condenser. However, the inverted column is not truly the inverse of a regular batch column, since this would require the batch, which is stored in the condenser, to be in vapor phase which is very unlikely.

When the desired purity is not obtainable in the column, the batch may be processed in several steps each increasing the purity of the product. A well known example of this is the separation of ethanol from water by evaporation (Rayleigh distillation).

The production period is normally the most time consuming compared to the startup and shutdown periods. However, for difficult separations such as for high purity or azeotropic separations, the startup time may also be significant. Alternative ways of

Table 1.5: Text books which include a discussion of batch distillation.

Authors	Title
Billet (1973)	Industrielle Destillation (in German)
Coulson and Richardson (1978)	Chemical Engineering
Ellerbe (1979)	Handbook of Separation Techniques for Chemical Engineers
Ellerbe (1980)	Separation Techniques I. Liquid-Liquid Systems
Holland (1981)	Fundamentals of Multicomponent Distillation
Holland and Liapis (1983)	Computer Methods for Solving Dynamic Separation Problems
Perry and Green (1984)	Perry's Chemical Engineering Handbook
Rose (1985)	Distillation Design in Practice
Domenech and Pibouleau (1989)	Process Modeling and Control in Chemical Engineering
Luyben (1992)	Practical Distillation Control

reducing the length of this period, such as starting the batch with light product in the condenser, should therefore be considered.

So far, the reflux ratio has been the only operating variable considered for optimization. However, both the boilup rate, column pressure and condenser holdup are operating variables which should be considered in order to optimize the operation.

Most of the factors mentioned above are considered in this thesis.

1.9 Text books on batch distillation

No text books are available which consider batch distillation alone. However, most books on distillation or separation processes contain a chapter on batch distillation. Some examples are given in Table 1.5. Coulson and Richardson (1978) and Perry and Green (1984) describe the basic principles of batch distillation in a straightforward manner. They also include some dynamic models. Holland (1981) and Holland and Liapis (1983) mainly discuss the dynamic equations for batch distillation and alternative mathematical solution methods. Billet (1973) and Rose (1985) concentrate on design of batch columns. Domenech and Pibouleau (1989) present different mathematical models and also discuss optimal operating policies for batch distillation. Luyben (1992) discusses control of batch columns based on the papers by Quintero-Marmol and Luyben (1991, 1992).

1.10 Thesis overview

This thesis consists of 5 separate papers on optimal operation and control of batch distillation columns which may be read independently. Literature references are also given in each chapter. The thesis is divided into two parts:

Part I: Optimal operation of batch distillation

Chapter 2 studies the optimal *startup* of batch columns. Normally the production period is the most time consuming but in some separations the startup time may be up to 50% of the total operating time. The paper discusses the relative significance of the startup time compared to the total operating time for different separations. The startup time is found to have a limited influence on the total operating time unless it is very different from the optimal value. Neglecting the initial filling of the column section and condenser holdup is a normal assumption in the literature. It is established that for optimization studies, this is in most cases acceptable. A short cut formula for the estimation of required startup time is presented and found to yield good results. In the last part of the paper alternative changes in the startup procedure are considered. These are: using partial backmixing equipment; finite reflux with some reflux directly to the reboiler and starting the batch with light product in the condenser drum initially.

Chapter 3 presents a novel *cyclic operating policy* for batch distillation which may be an alternative to conventional policies for some separations. The cyclic operating policy involves active use of a varying condenser holdup with periodic filling and dumping. The cyclic policy is presented in detail and a comparison is made to the conventional policies (constant distillate, constant reflux ratio and optimal reflux ratio operation) with respect to total operating time. The cyclic policy has been noted earlier because it may be easier to operate and control. Surprisingly, it is found that for difficult separations, the cyclic policy may significantly reduce the operating time compared to conventional operating strategies. A simplified cyclic policy is presented which is almost as good as the optimal cyclic policy but easier to calculate and implement. The effect of condenser holdup on the minimum operating time is discussed.

Chapter 4 presents a comparison between a regular and an *inverted column* in terms of minimum operating time for given separations. In the inverted column, the feed is charged to the condenser drum and the products are withdrawn from the reboiler. However, the inverted column is not the true inverse of a regular batch column, since this would require the batch, which is stored in the condenser, to be in the vapor phase which is very unlikely. In the paper, examples are presented which show that using an inverted column may be preferable to a regular column for some separations when the objective is to minimize the operating time.

Part II: Optimal operation and control of reactive batch distillation

Chapter 5 is devoted to the study of *controllability and control strategies for reactive batch distillation* in which a reaction takes place in the reboiler. A batch reactor may be combined directly with a distillation column by distilling off the light product in order to increase the reactor temperature or to improve the product yield of an equilibrium reaction. In this paper, an industrial esterification reaction is considered. It is found that, at any time, the same amount of light product should be removed as the amount being formed by the reaction. The controllability of the process is found to depend strongly on operating conditions and on the time during the batch. Several control strategies are presented and found to yield good performance.

Chapter 6 continues the discussion of reactive batch distillation using the same industrial process case study as in Chapter 5. In this chapter the *optimal operation* of the process is considered. Both reflux ratio and boilup rate are considered as optimization variables. The importance of proper feedback *control* of reactive batch distillation is discussed. Simple control strategies are presented and the effect of disturbances is considered for both a continuous controller and a real time control system.

Final conclusions and suggestions for future work are presented in Chapter 7.

Presentations

Preliminary versions of some of the papers in this thesis have been presented at international chemical engineering conferences:

- American Institute of Chemical Engineers (AIChE) 1991 Annual Meeting, Los Angeles, November 1991. (Parts of Chapter 5.)
- NATO Advanced Study Institute on Batch Processing Systems Engineering, Antalya, Turkey, May-June 1992. (Parts of Chapter 5.)
- 1993 European Symposium of Computer Aided Process Engineering-3 (ESCAPE-3), Graz, Austria, July 1993. (Chapter 6.)
- 1994 International Symposium on Process Systems Engineering (PSE-94), Kyongju, Korea, May-June 1994. (Chapter 3.)

The paper from ESCAPE-3 (Chapter 6) has been published in a supplement to Computers & Chemical Engineering (Vol. 18, 1994). It has also been selected for publication in a special issue of the same journal. The paper from NATO ASI 1992 (Parts of Chapter 5) has been accepted for publication in Batch Processing Systems Engineering: Current Status and Future Directions, G.V. Reklaitis *et al.* eds., Spring-Verlag, Berlin (1994). Chapter 5 has been accepted for publication in Journal of Process Control, special issue on Batch Processing (1994).

In addition to the work presented in this thesis, the author has also participated in industrial research projects governing modeling, optimization and control of batch distillation during the course of study. Reports from these studies are, however, confidential and therefore not included in this thesis.

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Chapter 2

Optimal Startup Procedures for Batch Distillation

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Abstract

Operation of a conventional batch distillation column can be conveniently described in three parts: 1) startup period, 2) production period and 3) shutdown period. For standard separation processes, the production period is the most time consuming. However, for difficult separations, such as for high purity or azeotropic separations, the startup time may also be significant. In this paper, we present results for optimal operation of a number of different separations taking the startup time into consideration. Alternative ways of reducing the duration of the startup period are discussed. The main conclusions are:

1. It is not necessary to include the initial filling of trays and condenser drum in optimization studies.
2. The exact value of the startup time is of limited significance unless it is very different from the optimal value.
3. A short cut formula can provide an estimation for the required startup time based on operating parameters which are normally known a priori.

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2.1 Introduction

During the last decades, batch distillation has become increasingly important as a result of the expansion in the fine chemical and pharmaceutical industries characterized by small amounts of products with high added value. Also the need to increase the recovery of useful resources from wast products has forced a reappraisal of this unit.

Operation of a conventional batch distillation column can be conveniently described in three parts: 1) startup period (normally under total reflux), 2) production period and 3) shutdown period. For standard separation processes the production period is the most time consuming. Although, for difficult separations, such as for high purity or azeotropic separations, the startup time may also be significant as shown in this paper.

The importance of the startup period relative to the total separation has only been studied by few authors. Nad and Spiegel (1987) presented experimental results for the separation of 3 components into two main cuts, two off-cuts and a residue where the startup period was more than 1/4 of the total operating time. Holland and Liapis (1983) presented simulation results for the separation of a five component mixture where the startup time was more than 50% of the total operating time. Luyben (1971) presented simulation results for a number of cases where the startup time varied from negligible up to 50% of the total operating time.

Consequently, to optimize the process, the startup period may have to be considered as part of the complete batch distillation cycle. However, the startup time has not been included in many previous studies. This reduces the validity of these studies to cases where the production period is dominant. To the best of our knowledge, no studies of optimal startup procedures have been presented in the literature so far.

The purpose of this paper is to discuss various aspects of startup procedures for batch distillation as well as issues related to simulation and optimization of batch columns taking startup into consideration. The paper is divided into four parts:

Part I. An equilibrium time is defined based on the approach to equilibrium under total reflux. The influence of the initial conditions specified for the mathematical model used in optimization studies is discussed (trays initially filled or not etc.). We discuss the effect of type of mixture and column characteristics on the equilibrium time (parameter values for relative volatility, number of trays, feed composition etc.). The influence of the definition of equilibrium time is considered. Finally, we present typical dynamic responses under total reflux.

Part II. We discuss under which conditions the optimal startup time is significant compared to the optimal total operating time. The influence of startup time on the operating time is discussed.

Part III. We present a short cut formula for the estimation of equilibrium and startup time.

Part VI. We conclude by discussing alternative ways of reducing the startup time.

We will in each part give a brief literature review of previous work.

2.2 Dynamic model

The dynamic model used in this paper for a binary mixture is valid under the following assumptions:

A1 Staged batch distillation column with trays numbered from the top and down

A2 Perfect mixing and equilibrium on all trays

A3 Negligible vapor holdup

A4 Constant stage pressures and tray efficiencies

A5 Constant vapor flows (the energy balance is neglected)

A6 Total condensation with no subcooling in the condenser

A7 Constant relative volatility

A8 Constant molar condenser drum holdup (perfectly controlled)

A9 Constant molar liquid holdup on all trays (liquid dynamics are neglected)

The column is given in Figure 2.1. With these assumptions the following differential and algebraic equations result:

reboiler:

$$dH_B/dt = L_N - V_B \tag{2.1}$$

$$d(H_B x_B)/dt = L_N x_N - V_B y_B \tag{2.2}$$

column tray, j=1,N:

$$0 = L_{j-1} + V_{j+1} - L_j - V_j \tag{2.3}$$

$$H_j dx_j/dt = L_{j-1} x_{j-1} + V_{j+1} y_{j+1} - L_j x_j - V_j y_j \tag{2.4}$$

$$V_j = V_{j+1} \tag{2.5}$$

condenser and condenser drum:

$$0 = V_1 - L - D \tag{2.6}$$

$$H_C dx_D/dt = V_1 y_1 - L x_D - D x_D \tag{2.7}$$

$$R = L/V_1 \tag{2.8}$$

accumulator:

$$dH_A/dt = D \tag{2.9}$$

$$d(H_A x_A)/dt = D x_D \tag{2.10}$$

equilibrium, j=1,N and B:

$$y_j = \frac{\alpha x_j}{1 + (\alpha - 1) x_j} \tag{2.11}$$

Note that the reflux ratio R used here is the <u>internal</u> reflux ratio $R = R_{internal} = L/V_1$, not the usual external ratio $R_{external} = L/D$. (This is better solved numerically, since $R_{internal} \in [0, 1]$ whereas $R_{external} \in [0, \infty >$.) It is assumed that the vapor flow V_B can be manipulated directly. SPEEDUP (1993) is used for the simulations.

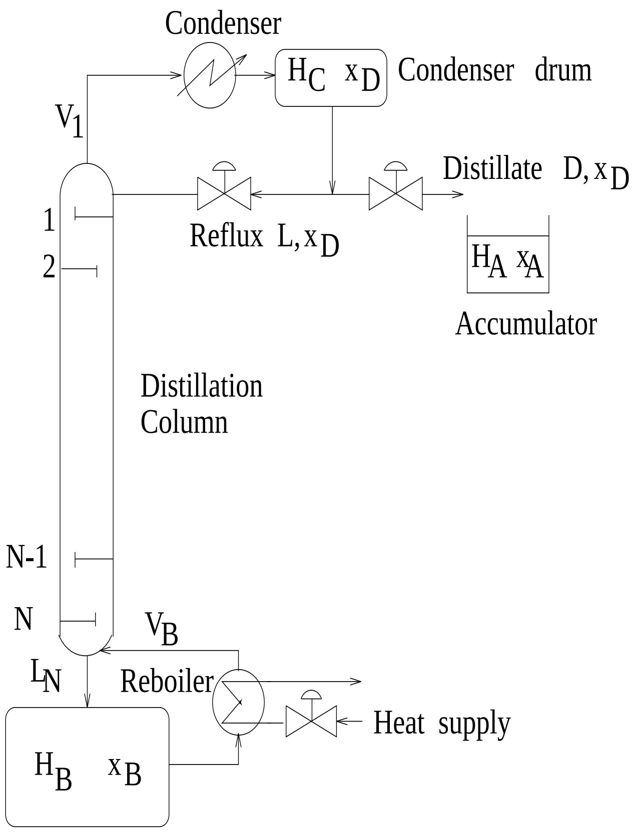

Figure 2.1: Batch distillation column.

2.3 PART I. Startup of a batch column

In the first part of this paper we will discuss the normal startup of a batch column with emphasis on the duration of the startup period given by a defined equilibrium time. We will consider the effect of various operating parameter values on the equilibrium time, e.g. how the equilibrium time varies with increasing holdups, feed compositions, relative volatility etc. We will also compare different initial conditions, or different assumptions as to how the column is filled up initially, in terms of dynamic behavior and required equilibrium time. These initial conditions are needed in the mathematical solution of the dynamic model in simulation and optimization. Different definitions of equilibrium time are discussed. We will conclude this part by presenting typical dynamic responses for startup under total reflux.

2.3.1 Normal startup of a batch column

The startup of a batch column, packed or tray, can generally be described by the following sequence:

1. The liquid feed is initially charged to the reboiler. Heat is introduced to bring the material to its boiling point temperature.

2. Boiling starts and vapor travels up through the column and some of the vapor condenses inside the column section.

3. The condenser starts to operate as the vapor reaches the top tray and the condenser drum starts to fill up.

4. The reflux valve is opened when the liquid fills the condenser drum. Normally, operation at total reflux begins. However, some product may also be collected at this point with a given reflux flow instead.

5. The liquid begins to flow into the top section of the column and the liquid holdup in the top starts to increase.

6. The column section will be filled up from the top and down by the returned liquid until the lowest part of the column is filled and the liquid reaches the reboiler.

7. The column is now run under total reflux operation until the unit is taken to a steady state or to a state where the distillate composition reaches the desired product purity.

The first step, heating the feed, can be in the order of hours depending on the heat input to the reboiler. Typically, about 1 hr is needed for this step in a normal laboratory column. During step 2, the column section is heated up by the vapor traveling upwards in the column. Some of the vapor will condense in the column section. The filling of the column and condenser by vapor and liquid, steps 2-6, is in the order of minutes. The last step is normally the most time consuming. Typically, a total reflux time of up to 15 hr may be needed for difficult separations. For extreme separations such as isotope separations, even days may be needed.

This startup procedure is the same for packed and tray columns. However, in the rest of this paper we will limit the discussion to tray columns.

The simulation of the startup period as described above is complicated. A very rigorous model which includes the geometry of the trays in the column, the heat transfer in the reboiler and column section etc., is needed. Ruiz (1988) used a dynamic model presented by Ruiz *et al.* (1988), (originally developed by Gani *et al.* (1986)), for the simulation of four different cases from the earlier literature (Boston *et al.* (1980), Domenech *et al.* (1981), Holland and Liapis (1983) and Nad and Spiegel (1987)) where two of the cases where compared with experiments (Domenech *et al.* (1981) and Nad and Spiegel (1987)). Their model included variable holdups, variable internal flow rates, tray geometry and hydraulics. It also includes the possibility of taking the heat transfer in the column section into consideration. Such a model is however, not suitable for optimization studies since its solution is very time consuming. In optimization of a dynamic process, using e.g. the feasible path approach, the dynamic model must be solved for each trial value(s) of the optimization variable(s) and must therefore be reasonably simple (Vassiliadis, 1993).

A normal assumption in the literature is that the feed is present initially at its boiling point, i.e. step 1 is negligible. This means that the initial heating is "decoupled" from the rest of the process. However, Albet *et al.* (1994) included the heating of the initial charge in their simulation of the startup of a batch reactive column. In their simulation, the heating time was about 25% of the total operating time.

Steps 2-6 are in simulations normally approximated by assuming that the trays are filled up according to some filling procedure. We will in the following consider different filling procedures and comment on the possible differences in the duration of the startup period.

2.3.2 Definition of equilibrium time

For an infinite startup time, the column would eventually reach a new equilibrium state (steady state). To describe how fast the column reaches this new state we introduce the equilibrium time t_{eq}^{γ}:

Equilibrium time: t_{eq}^{γ} is the time when the distillate composition has reached a fraction γ of its steady state value under total reflux relative to the initial feed composition

$$\frac{x_D(\mathbf{t}_{\mathbf{eq}}^{\gamma}) - x_F}{x_D^{ss} - x_F} = \gamma$$

or equivalently

$$x_D(\mathbf{t}_{\mathbf{eq}}^{\gamma}) = x_F + \gamma(x_D^{ss} - x_F) \equiv x_D^{\gamma} \tag{2.12}$$

In this paper we will select $\gamma = 0.98$, that is the equilibrium time $t_{eq}^{98\%}$ is when the distillate composition x_D has reached 98% of its steady state value x_D^{ss} relative to the feed composition x_F.

Other definitions have been used in the literature but most of these are difficult to apply generally as they mostly depend on absolute and not relative values. We will at the end of this section discuss possible differences in the conclusions depending on the actual *definition* of the equilibrium time.

2.3.3 Examples studied

We have computed the equilibrium time given by Eq. 2.12 for a number of examples under varying feed composition x_F, column tray holdup H_j and condenser drum holdup H_C. We have also studied the effect of relative volatility α and number of trays N. The examples studied are given in Table 2.1. Note that Examples 7, 21, 24 and 27 are identical. Also included is the steady state distillate composition, x_D^{ss} and the distillate composition at 98% approach to equilibrium, $x_D^{98\%}$. The last three columns of the table give the equilibrium time $t_{eq}^{98\%}$ for different filling procedures INIT-1, INIT-2 and INIT-3. These results are discussed in more detail in the next two sections.

2.3.4 Alternative simulation procedures for the initial filling

In the beginning of this section we described the different steps in the startup of a batch column. These steps are in simulations normally approximated by assuming that the trays and the condenser are initially filled up according to some filling procedures.

Table 2.1: Steady state distillate composition x_D^{ss}, distillate composition at 98% approach to equilibrium $x_D^{98\%}$ and equilibrium time $t_{eq}^{98\%}$ for filling procedures INIT-1, INIT-2 and INIT-3. (The amount of initial charge is $H_F = 10.0\ kmol$ in all cases.)

Example	N	α	x_F	V $kmol/hr$	H_C $kmol$	H_j $kmol$	x_D^{ss}	$x_D^{98\%}$	INIT-1 $t_{eq}^{98\%}$ hr	INIT-2 $t_{eq}^{98\%}$ hr	INIT-3 $t_{eq}^{98\%}$ hr
1	10	2.0	0.05	10	0.1	**0.001**	0.9884	0.9696	0.275	0.275	0.275
2						**0.01**	0.9868	0.9680	0.402	0.402	0.402
3						**0.05**	0.9736	0.9552	1.062	1.064	1.064
4						**0.1**	0.9343	0.9167	1.874	1.879	1.879
5			0.10			**0.001**	0.9951	0.9772	0.155	0.154	0.155
6						**0.01**	0.9948	0.9769	0.214	0.214	0.214
7						**0.05**	0.9926	0.9747	0.506	0.507	0.507
8						**0.1**	0.9874	0.9696	0.924	0.929	0.929
9			0.50			**0.001**	0.9995	0.9895	0.087	0.086	0.086
10						**0.01**	0.9995	0.9895	0.100	0.099	0.099
11						**0.05**	0.9995	0.9895	0.179	0.178	0.176
12						**0.1**	0.9994	0.9894	0.289	0.289	0.286
13			0.80			**0.001**	0.9999	0.9959	0.081	0.079	0.079
14						**0.01**	0.9999	0.9959	0.091	0.088	0.087
15						**0.05**	0.9999	0.9959	0.150	0.147	0.142
16						**0.1**	0.9999	0.9959	0.236	0.231	0.223
17	10	2.0	0.10	10	**0.05**	0.01	0.9951	0.9771	0.137	0.137	0.137
18					**0.1**		0.9948	0.9769	0.214	0.214	0.214
19					**0.5**		0.9902	0.9724	0.960	0.962	0.962
20					**0.05**	0.05	0.9931	0.9753	0.421	0.422	0.422
21					**0.1**		0.9926	0.9747	0.506	0.507	0.507
22					**0.5**		0.9827	0.9651	1.429	1.433	1.433
23	10	**1.5**	0.10	10	0.1	0.05	0.8729	0.8574	1.213	1.214	1.214
24		**2.0**					0.9926	0.9747	0.506	0.507	0.507
25		**3.0**					0.9999	0.9819	0.199	0.200	0.200
26	**5**	2.0	0.10	10	0.1	0.1	0.8393	0.8245	0.644	0.644	0.644
27	**10**					0.05	0.9926	0.9747	0.506	0.507	0.507
28	**15**					0.033	0.9997	0.9817	0.376	0.377	0.377
29	**20**					0.025	1.0000	0.9820	0.311	0.313	0.313
30	**25**					0.02	1.0000	0.9820	0.274	0.275	0.275
31	20	4.0	0.005	10	0.025	0.005	0.9995	0.9796	0.429	0.429	0.429
32	10	3.0	0.05	10	0.1	0.01	0.9998	0.9808	0.179	0.179	0.179
33	10	2.0	0.5	10	0.1	0.01	0.9995	0.9895	0.100	0.099	0.099

Description of procedures

In this paper, we consider the following filling procedures:

INIT-0 The feed charge is initially distributed in the condenser drum, on the trays and in the reboiler. The condenser and tray holdups are constant and the initial compositions are equal to the total reflux compositions (steps 2-7 negligible, employed by Distefano (1968)).

INIT-1 The feed charge is initially distributed in the condenser drum, on the trays and in the reboiler. The condenser and tray holdups are constant and all compositions are initially equal to the feed composition (for instance Mujtaba and Macchietto (1991) and Logsdon and Biegler (1992)).

INIT-2 The column section and condenser drum are initially empty and all the feed is in the reboiler. The vapor from the reboiler is condensed and stored in the overhead equipment until liquid first fills the condenser drum and then the column trays. (This is equivalent to operating the column without reflux, i.e., with only one theoretical rectification stage.) As soon as the condenser is filled up the trays are filled from the top and down. After a tray has been filled, its holdup is assumed constant (suggested by Luyben (1971) and applied by González-Velasco *et al.* (1987)).

INIT-3 Same as INIT-2 but it is assumed that some of the vapor (here 20%) is condensed on the trays before reaching the condenser drum. The trays are then filled up from the top and down (for example Sadotomo and Miyahara (1983)).

It should be noted that these procedures are all different mathematical descriptions, only with varying assumptions, of the *same* filling process. It should also be stressed that the *steady state profiles* (or x_j's at $t = \infty$) for all procedures are identical.

Alternative INIT-0 neglects the startup period altogether. We will in this section consider procedures INIT-1, INIT-2 and INIT-3 and discuss the differences in the duration of the startup period using the different filling procedures. We will show that there is almost no difference and the simplest procedure INIT-1 is therefore recommended.

Variations of the same procedures have also been compared by Abdul Aziz *et al.* (1992) with the equilibrium time defined as the time when the distillate composition has reached $x_D = 0.95$. They studied the effect of holdups and number of trays on two different ternary mixtures. Furthermore, empirical correlations between holdups and equilibrium time were presented.

For startup of the column using INIT-2 and INIT-3, assumptions $A8$ and $A9$ of the dynamic model in section 2.2 is replaced by:

A8 Molar condenser drum holdup H_C not constant during filling, perfectly controlled otherwise

A9 Molar tray holdups H_j not constant

A10 Linear tray hydraulics, $\Delta L_j = \Delta M_j / \tau_h$

and the equations for the column section, Eq. 2.3-2.5, and for the condenser, Eq. 2.6-2.8, are replaced by:

column tray, j=1,N:

$$\begin{aligned}
dH_j/dt &= L_{j-1} + V_{j+1} - L_j - V_j && (2.13)\\
d(H_j x_j)/dt &= L_{j-1}x_{j-1} + V_{j+1}y_{j+1} - L_j x_j - V_j y_j && (2.14)\\
V_j &= V_{j+1} \quad \text{(INIT-2)} && (2.15)\\
V_j &= \begin{cases} 0 & \text{if } H_j < 0.2 \cdot H_j^{spec} \quad \text{(INIT-3)} \\ V_{j+1} & \text{otherwise} \end{cases} && (2.15)\\
L_j &= \begin{cases} 0 & \text{if } H_j < H_j^{spec} \\ L_j^o + \frac{H_j - H_j^o}{\tau_h} & \text{otherwise} \end{cases} && (2.16)
\end{aligned}$$

condenser and condenser drum:

$$\begin{aligned}
dH_c/dt &= V_1 - L - D && (2.17)\\
d(H_C x_D)/dt &= V_1 y_1 - L x_D - D x_D && (2.18)\\
L &= \begin{cases} 0 & \text{if } H_C < H_C^{spec} \\ R \cdot V_1 & \text{otherwise} \end{cases} && (2.19)
\end{aligned}$$

The initialization variables for the different procedures are given in Table 2.2.

Table 2.2: Initial values for filling procedures INIT-1, INIT-2, and INIT-3 (—: no composition specified since the holdup is zero).

	INIT-1	INIT-2,3
accumulator	— $H_A^o = 0$	— $H_A^o = 0$
condenser	$x_D^o = x_F$ $H_C^o = H_C^{spec}$	— $H_C^o = 0$
trays	$x_j^o = x_F$ $H_j^o = H_j^{spec}$	— $H_j^o = 0$
reboiler	$x_B^o = x_F$ $H_B^o = H_F$ $-\sum^N H_j^o - H_C^o$	$x_B^o = x_F$ $H_B^o = H_F$

Results for comparison of filling procedures

The initial tray holdups H_j, liquid flows L_j and tray compositions x_j for example 7 ($x_F = 0.1$ and $H_j = 0.05$ $kmol$) are plotted in Figure 2.2 for the three procedures. The initial peaks in compositions for INIT-3 are due to composition changes caused by the initial filling of 20% of the tray holdups.

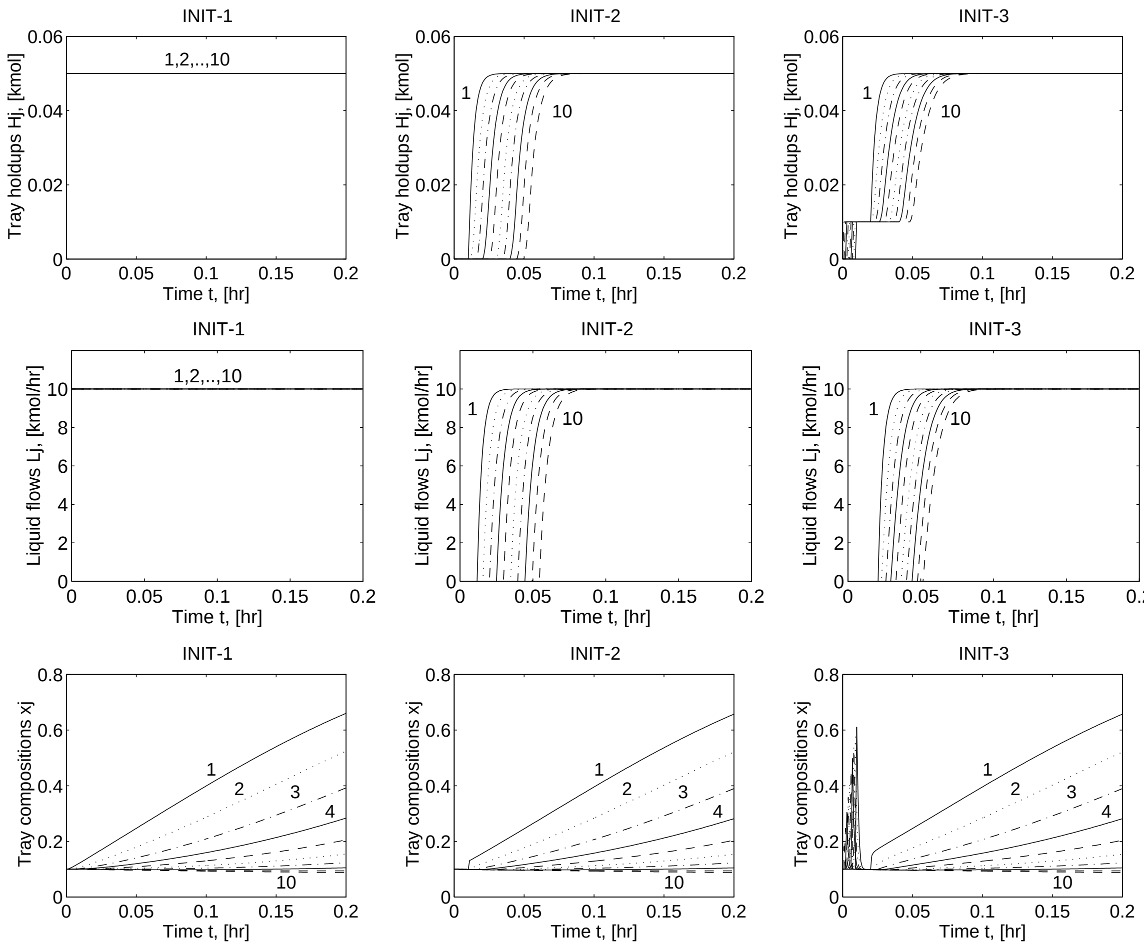

Figure 2.2: Initial tray holdups H_j, liquid flows L_j and tray compositions x_j for filling procedure INIT-1 (left), INIT-2 (middle) and INIT-3 (right) (example 7, $t_{eq} = 0.58\ hr$).

The difference between the responses for the three procedures is quite significant. Nevertheless, from Table 2.1 we see that the equilibrium times for the different filling procedures are almost identical for all the examples. For mixtures rich in light component (large x_F) together with large tray holdups there is some difference; in the extreme case about 5% (example 16: $x_F = 0.8$ and $H_j = 0.1\ kmol$ where $t_{eq}^{98\%} = 0.236\ hr$ for INIT-1 and $t_{eq}^{98\%} = 0.223\ hr$ for INIT-3). Note that when the amount of initial filling is increased in INIT-3, the equilibrium time will be somewhat reduced.

It should be noted that including the filling of the trays (INIT-2 or INIT-3) in optimizations is time consuming due to the very small integration steps needed initially. Also, with the optimization program used in this study (Vassiliadis, 1993), it was found that the calculations of sensitivities (needed to find the new search directions in optimization) may in some cases be infeasible for INIT-2 and INIT-3.

The filling procedure normally used in the literature is INIT-1 (assuming feed composition on all trays and in the condenser initially). Based on the results in this paper, it can be concluded that *procedure INIT-1 is indeed in most cases accurate enough when the* *objective is to describe the duration of the startup period.* However, it must be emphasized that the filling procedures are purely mathematical formulations. They do not define the filling of the column in real practice. In the rest of this paper we will use INIT-1 for initialization.

2.3.5 Effect of operating parameters on equilibrium time

We now discuss in more detail the effect of operating parameters on the equilibrium time. Some of the results from Table 2.1 are plotted in Figure 2.3 where we show the effect on the equilibrium time, $t_{eq}^{98\%}$, of changes in internal tray holdup ($\sum^N H_j/H_F \cdot 100\%$), condenser drum holdup ($H_C/H_F \cdot 100\%$), feed composition, relative volatility and number of trays (tray efficiency). Note that in the last case with varying number of trays N (bottom plot), the total column holdup $\sum^N H_j$ is constant. This is equivalent to assuming that the tray efficiency has increased, e.g. by changing the packing material in a packed column.

Results

From Figure 2.3 we find that the equilibrium time $t_{eq}^{98\%}$ is:

- slightly more than proportionally increasing with increasing tray holdup H_j
- slightly more than proportionally increasing with increasing condenser holdup H_C
- decreasing with increasing feed composition x_F (light component)
- decreasing with increasing relative volatility α
- decreasing with increasing number of trays N (with total tray holdup constant)

The proportional increase in equilibrium time with increasing column holdup H_j and condenser holdup H_C is expected since this directly increases the residence time. The reason why it increases slightly more than proportionally when the holdup is increased is due to the fact that the composition in the reboiler changes which results in a more difficult separation. (Mujtaba and Macchietto (1991) have some further discussion on holdup effects).

The equilibrium time increases with decreasing feed composition, relative volatility and number of trays. This is as expected since the separation then becomes more difficult. Note in particular the sharp increase in equilibrium time as the feed composition approaches zero. The smallest value of x_F given in Figure 2.3 is $x_F = 0.05$ (Example 3) with an equilibrium time of 1.1 hr. However, if x_F is reduced to 0.005 (not given in Table 2.3) then the equilibrium time increases from 1.1 hr to 3.6 hr. However, note that the distillate composition is significantly reduced, $x_D^{ss} = 0.31$.

Note that with our dynamic model, the equilibrium time will decrease with V_B; if the boilup rate V_B is decreased with a factor of 10 the equilibrium time will be increased by the same factor. This is a result of the assumptions in our model were the heat capacities in the column wall and internals are neglected and the tray efficiency and initial tray holdup is assumed independent of liquid and vapor flow.

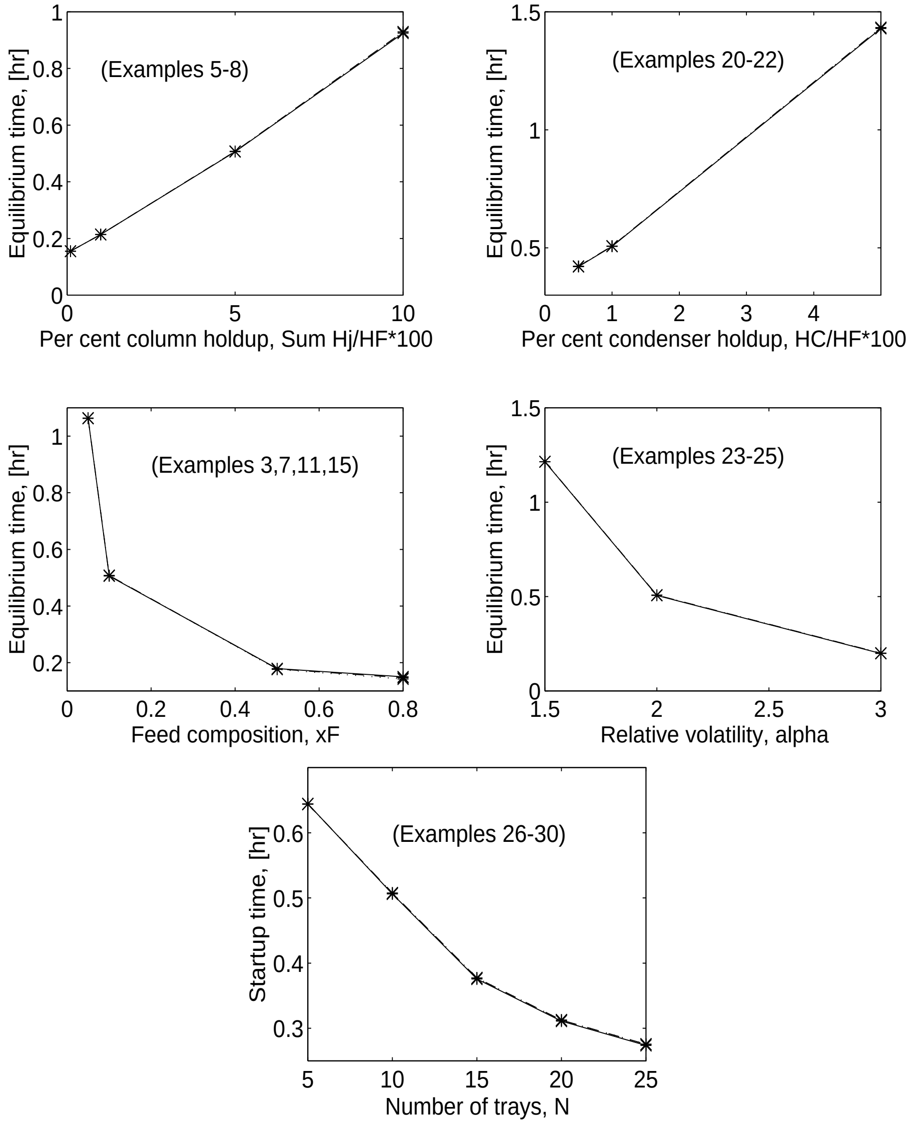

Figure 2.3: Equilibrium time $t_{eq}^{98\%}$ for filling procedures INIT-1 (—), INIT-2 (- -) and INIT-3 (-·-) under varying conditions.

Our results are mostly in agreement with those found by Abdul Aziz *et al.* (1992) who used a definition of equilibrium time, $x_D(t_{eq}) = 0.95$. However, they found a different dependency on the number of trays N since they used a constant tray holdup H_j for all trays such that the total internal column holdup $\sum^N H_j$ changed with N (equivalent to an increase in the height of the column).

In conclusion we have found that *the equilibrium time will be long for difficult separations (low relative volatility, few trays), for large holdups and when only a small amount of light component is present in the feed.* In part II of this paper we will present optimal startup times for different cases taking the whole operating time (startup and production) into account.

2.3.6 Dependency on the definition of equilibrium time

We have in this paper defined the equilibrium time to be the time when the distillation composition has reached 98% of its steady state value at total reflux relative to the feed composition:

I $x_D(\mathbf{t}_{\mathbf{eq}}^{\mathbf{98\%}}) = x_F + 0.98(x_D^{ss} - x_F)$ (this paper, Eq. 2.12)

Other definitions used in the literature are:

II $x_D(\mathbf{t}_{\mathbf{eq}}) = x_D^{ss} - 0.1(x_D^{ss} - \alpha x_B^{ss})$ (Jackson and Pigford, 1956)

III $dx_D(\mathbf{t}_{\mathbf{eq}})/dt < 0.01\ hr^{-1}$ (Luyben, 1971)

IV $x_D(\mathbf{t}_{\mathbf{eq}}) = x_D^{ss} - 10^{-4}$ (González-Velasco *et al.*, 1987)

V $x_D(\mathbf{t}_{\mathbf{eq}}) = 0.95$ (Abdul Aziz *et al.*, 1992 and González-Velasco *et al.*, 1987)

Definition II is similar to Definition I and depends on the approach to equilibrium using relative values. Definitions III and IV both involve the approach to equilibrium in some sense but using absolute instead of relative values. The results for different separations will therefore not be directly comparable for any of them. Definition V involves a given concentration and will not even be attainable for low purity separations.

The required equilibrium time for the different definitions are given in Table 2.3 for three examples. It can be seen that the equilibrium time varies considerably with its definition as expected. Definitions II and III give a varying approach to equilibrium depending on the given example. Definition IV is very strict; giving a long equilibrium time which takes the column very close to steady state. Definition V is found to give an infinite equilibrium time for example 4 since $x_D^{ss} = 0.93 < 0.95$.

Table 2.3: Equilibrium time for different definitions.

example	x_D^{ss}	I $t_{eq}^{98\%}$, hr	II t_{eq}, hr	III t_{eq}, hr	IV t_{eq}, hr	V t_{eq}, hr
4	0.9343	1.874	0.842	2.512	5.162	∞
7	0.9926	0.506	0.311	0.844	1.191	0.398
11	0.9995	0.179	0.098	0.325	0.404	0.103

It must be emphasized that we are not stating that our definition is the right one, merely that *relative* values should be used. The factor 0.98 was arbitrarily selected and should probably be increased (e.g. to 0.995) for difficult separations with high purity demands and relaxed (e.g. to 0.95) for easier ones with lower purity. One possibility to reduce the dependency on such a factor is to consider logarithmic compositions instead, e.g. $X = \ln[x/(1-x)]$. This has not been considered in this paper.

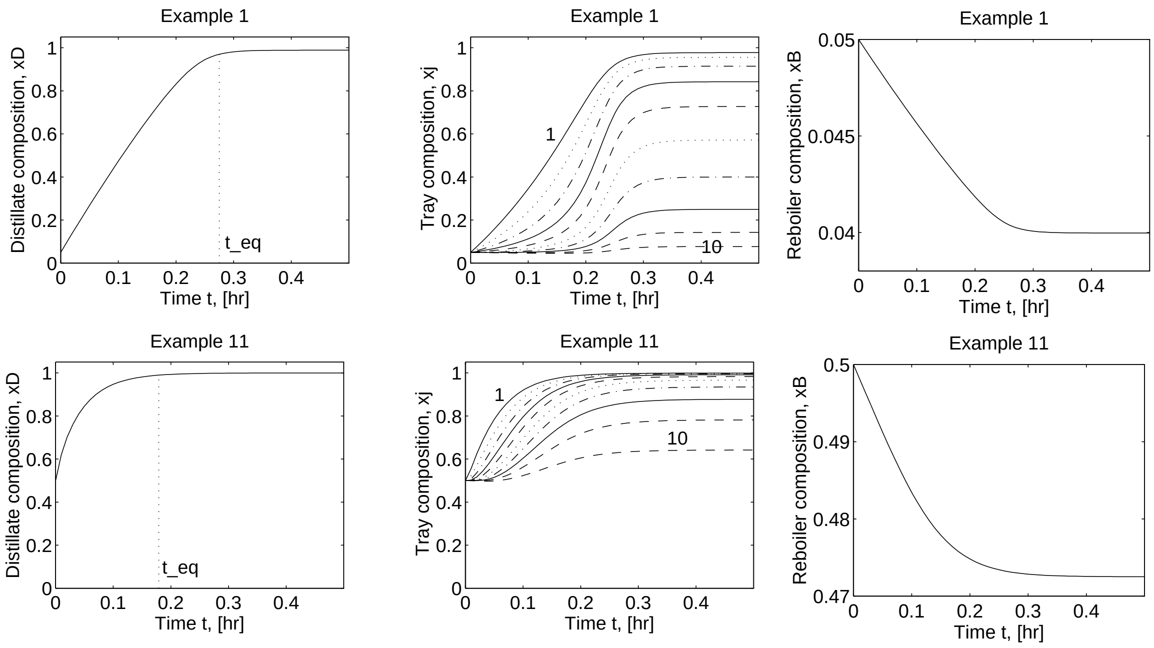

Figure 2.4: Dynamic responses for examples 1 and 11 (INIT-1 for initialization) (Note that the y-axis is different for the reboiler composition x_B).

2.3.7 Dynamic responses for startup

In the previous sections we have discussed the equilibrium time for a number of examples. In all the cases, the equilibrium time was defined according to Eq. 2.12, that is depending on the approach to steady state in the top of the column. An obvious question will then be: Is the approach to equilibrium different for the rest of the column and how does the dynamic response depend on the given separation?

Consider Figure 2.4 where the approach to equilibrium is given for the distillate composition x_D, the tray compositions x_j and the reboiler composition x_B for example 1 and 11. Example 1 has a very low feed composition and tray holdup ($x_F = 0.05$ and $H_j = 0.001\ kmol$) and yields $t_{eq}^{98\%} = 0.275\ hr$. Example 11 has both a larger composition and a larger holdup ($x_F = 0.5$ and $H_j = 0.05\ kmol$) and yields $t_{eq}^{98\%} = 0.179\ hr$. Note that all the responses start from the feed composition x_F since INIT-1 is used for initialization.

The responses have a different shape in the two examples; initially x_D increases almost linearly in example 1 but exponentially in example 11. The tray compositions also have a different response and are almost S-shaped for some trays. This difference is independent of the filling procedure.

For both examples, the reboiler composition is only slightly reduced from that of the feed. This is due to the gradual depletion of light component from the reboiler. If the reboiler holdup had been very large compared to the holdup in the column section and condenser, the reboiler composition would have been unchanged (this is assumed in the derivation of a short cut formula for required equilibrium time in Section 2.5.2).

For both examples, equilibrium is reached first in the top of the column. This is best seen by plotting the normalized compositions; see Figure 2.5. The compositions are

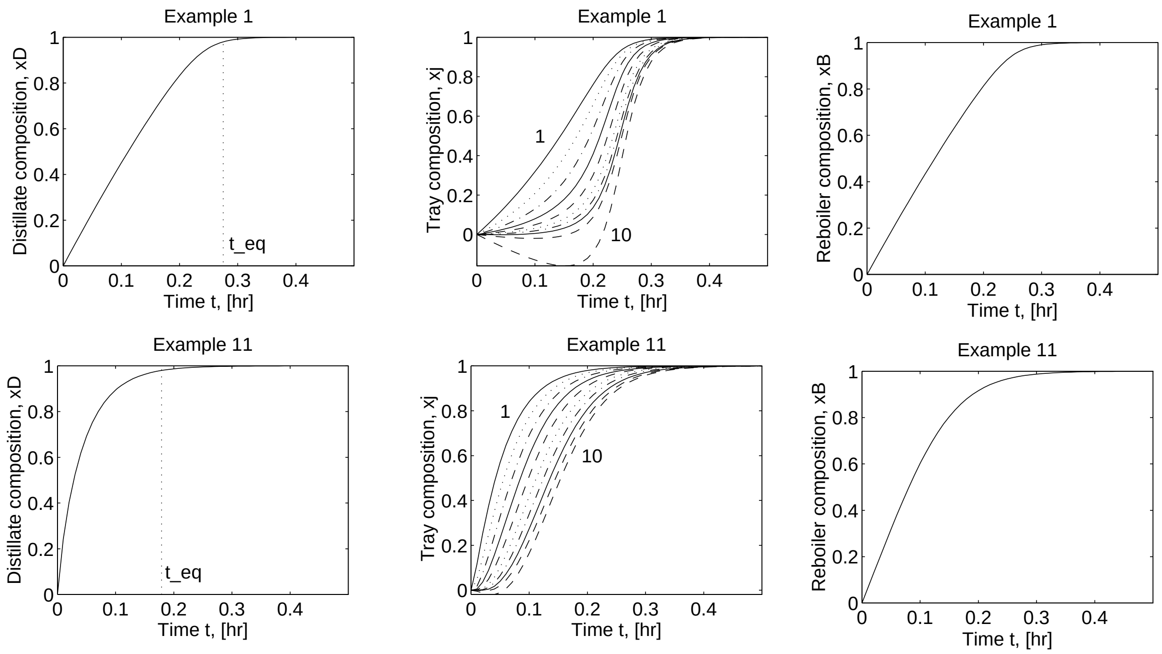

Figure 2.5: Normalized dynamic responses for examples 1 and 11.

normalized according to:

$$x^{norm}(t) = \frac{x(t) - x^o}{x^{ss} - x^o} \tag{2.20}$$

From Figure 2.5 it can be seen clearly that the responses vary with location in the column and with the feed mixture. In particular, the responses in the bottom of the column are slower than those in the top. (In our later development of a short cut formula in Section 2.5.2 we assume that all the trays have the same first order response which is clearly not the case.) The responses can not be approximated by any simple function, e.g. first order, second order etc.

Note that the normalized responses for the lower trays in the column go below zero (e.g. Figure 2.5, top middle plot). In other words, there is an inverse response (opposite direction of final change) for these trays. This effect is due to the assumption of initially filled trays (INIT-1) and is not observed if the filling of the trays is taken into account (INIT-2 or INIT-3) and would therefore probably not occur in practice.

2.3.8 Summary of Part I

In this section we have defined the equilibrium time to be the time to reach 98% approach to equilibrium in the top of the column relative to the feed composition. We have found that the equilibrium time will be long for separations with low relative volatility, few trays, large holdups and when only a small amount of light component is present in the feed. The approach to equilibrium is found to be quicker in the top of the column than in the bottom. We have also found that it is not necessary to include the initial filling of the trays and the condenser in optimizations studies where only the duration of the equilibrium time is of importance and not the exact responses.

2.4 PART II. Significance of startup time for optimal operation

As mentioned in the introduction, the startup time for a given separation may be long and in some cases more than 50 % of the total operating time. In this part we will discuss under which conditions the startup time is significant compared to the total operating time. To the best of our knowledge, no such comparison has been made in the literature so far.

We will consider a binary mixture separated into two or three fractions (see Figure 2.6):

Two fractions. After the initial startup period under total reflux ($t_{startup}$), there is a production period (time t_1) where a light product with mole fraction x_A and amount H_A is accumulated.

Three fractions. After the first production period (t_1), there is a second production period (time t_2) where an intermediate off-cut is produced (with composition x_{off} and amount H_{off}) and collected in a second accumulator tank.

In both cases, the heavy product is the residual material in the reboiler with mole fraction x_R and amount H_R. Note that this residual also includes the holdup in the condenser and the column section which is assumed to be drained to the reboiler at the end of the batch. (If the condenser holdup was large, one would probably want to drain this to the accumulator instead).

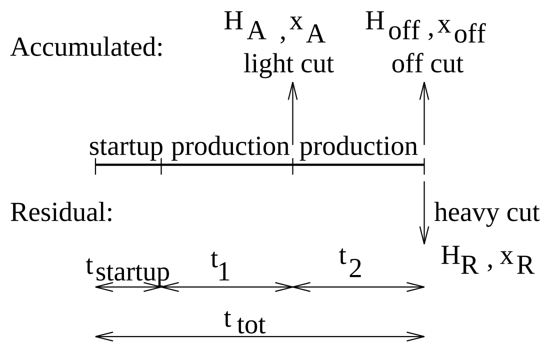

Figure 2.6: Binary separation into two main cuts and one off-cut.

In the following we will consider an operating policy with constant reflux ratios R_1 and R_2 in each of the two production periods. For a given separation the optimal startup time $t^{opt}_{startup}$, the times to reach the specifications for the product cuts, t_1 and t_2, and the corresponding constant reflux ratios R_1 and R_2 can be found by minimizing the total operating time t_{tot}:

$$\min_{t^{opt}_{startup}, t_1, R_1, t_2, R_2} t_{tot} = t^{opt}_{startup} + t_1 + t_2 \tag{2.21}$$

In addition, there are constraints on the composition and recovery percentage of light product at t_1 and heavy product at t_2:

$$x_A(t_1) \geq x_A^{spec} \tag{2.22}$$
$$100\% \cdot \frac{x_A H_A}{x_F H_F}(t_1) \geq \% \text{ light recovery} \tag{2.23}$$
$$x_R(t_2) \leq x_R^{spec} \tag{2.24}$$
$$100\% \cdot \frac{(1-x_R)\,H_R}{(1-x_F)\,H_F}(t_2) \geq \% \text{ heavy recovery} \tag{2.25}$$

Also, there may be upper and lower bounds on times and reflux ratios. However, all the constraints are not necessarily active at the optimal solution (inequalities instead of equalities). Note that if we had not allowed for the off-cut, only two constraints could be independently specified due to the total mass and component balances:

$$H_A + H_R(+H_{off}) = H_F \tag{2.26}$$
$$H_A x_A + x_R H_R(+x_{off} H_{off}) = x_F H_F \tag{2.27}$$

The dynamic model is given by Eq. 2.1-2.11 and the initial compositions are assumed to be equal to the feed composition (INIT-1). The column is run under total reflux during startup ($R_{startup} = 1.0$).The optimization program DAEOPT (Vassiliadis, 1993) is used with an optimization accuracy of 10^{-4}. The purpose of this study is to investigate the relative importance of the startup time for a given separation. We do therefore not consider what is done with the off-cut after the separation, e.g. recycling back to the next feed charge or separate reprocessing.

2.4.1 Optimal results

Optimal results for several cases are given in Table 2.4. The specifications are chosen so as to illustrate the influence of startup time for different types of separations (varying purity and recovery specifications). Cases C1 and C2 correspond to example 31 in Table 2.1, cases C3 and C4 to example 32 and cases C5, C6 and C7 to example 33. For all cases the total holdup in the column section $\sum^N H_j$ is 1% of the initial charge. The condenser drum holdup H_C is assumed to be 0.25% of the initial charge for cases C1 and C2 and 1% for the others. A time t_2 equal to zero means the constraints are met without the production of an off-cut (C2, C3 and C6). Note that for some cases (C2, C3, C6 and C7), not all the constraints are active (for example C2 where $x_R = 0.0025 < 0.01 = x_R^{spec}$). This will in general be the case when an off-cut is not produced (C2, C3 and C6) since then only two constraints can be independently specified.

The approach to equilibrium given in Table 2.4 is defined as:

$$\% \text{ approach to equilibrium} = \frac{x_D\left(t_{startup}^{opt}\right) - x_F}{x_D^{ss} - x_F} \cdot 100\% \tag{2.28}$$

Let us now consider the results in more detail. For case C1, the startup time is almost 70% of the total operating time. In this case only a very small amount of light component

Table 2.4: Optimal results (minimum operating time) for different separations for a piecewise constant reflux policy with one startup period and one or two production periods ($H_F = 10\ kmol$ and $V_B = 10.0\ kmol/hr$ for all cases) (*: on the lower bound, −: not specified).

	C1 (Ex 31)	C2 (Ex 31)	C3 (Ex 32)	C4 (Ex 32)	C5 (Ex 33)	C6 (Ex 33)	C7 (Ex 33)
Cases:							
N	20	20	10	10	10	10	10
α	4.0	4.0	3.0	3.0	2.0	2.0	2.0
H_j, $kmol$	0.005	0.005	0.01	0.01	0.01	0.01	0.01
H_C, $kmol$	0.025	0.025	0.1	0.1	0.1	0.1	0.1
x_F	0.005	0.005	0.05	0.05	0.5	0.5	0.5
Specifications:							
x_A^{spec}	0.5	0.9	0.99	0.95	0.95	0.99	0.9
% light recovery	90	50	75	-	75	90	-
x_R^{spec}	0.0001	0.01	0.05	0.01	0.05	0.1	0.01
% heavy recovery	90	-	-	90	75	-	90
Results:							
x_A	0.500	0.900	0.990	0.950	0.950	0.990	0.955
% light recovery	90.00	50.00	75.00	9.88	75.00	90.00	83.04
x_R	0.00010	0.0025	0.013	0.010	0.050	0.092	0.010
% heavy recovery	90.00	99.97	99.96	90.00	75.00	99.09	90.00
x_{off}	0.0043	-	-	0.277	0.500	-	0.725
$t_{startup}^{opt}$, hr	0.886	0.556	0.183	0.162	0.031	0.077	0.035
t_1, hr	0.009	0.003	2.285	0.0008	1.652	4.674	2.178
R_1, hr	0.01*	0.01*	0.983	0.342	0.761	0.903	0.800
t_2, hr	0.387	0	0	0.699	0.860	0	1.963
R_2, hr	0.754	-	-	0.806	0.755	-	0.944
t_{tot}^{opt}, hr	1.282	0.559	2.468	0.862	2.543	4.751	4.175
$t_{startup}^{opt}/t_{tot}^{opt}\cdot 100\%$	**69.1 %**	**99.5 %**	**7.4 %**	**18.8 %**	**1.2 %**	**1.6 %**	**0.8 %**
$x_D(t_{eq}^{opt})$	0.9984	0.9927	0.9841	0.9609	0.8246	0.9725	0.8450
x_D^{ss}	0.9995	0.9995	0.9998	0.9998	0.9995	0.9995	0.9995
approach to eq.	**99.9 %**	**99.3 %**	**98.3 %**	**95.9 %**	**65.0 %**	**68.7 %**	**69.0 %**

(impurity) is to be recovered from the feed charge leaving a heavy product of high purity. (It should be noted that the startup time is significantly reduced if there is no purity constraint on the light product).

For case C2, the startup time is 99.5% of the total operating time. The optimal solution is in fact a cyclic operating policy with one cycle (see Chapter 3 of this thesis) where a condenser drum holdup equal to the desired amount of light product is used and the column is run under total reflux until the desired product is obtained.

For case C5, which is an easy equimolar separation with lower product purities, the startup time is only 1 % of the total operating time. The results depend of course on the specifications for the product compositions and the percentage of recovery of the components. If a high percentage of recovery with a high purity is desired, the startup time will be longer than if the specifications are less strict. This is illustrated by case C3 and C4 which only differ in the specifications for the products.

We see from Table 2.4 that the approach to equilibrium at the end of the startup time varies considerable between the cases (99.9 % for case C1 to 65.0 % for case C5). Thus, the approach to equilibrium during startup will, as the startup time, depend on the mixture and the specifications.

In summary, the startup time will be significant compared to the total operating time for difficult separations (high purities and recoveries) and when the combined column and condenser holdup is large. The last conclusion regarding holdup follows from the fact that the equilibrium time increases proportionally with holdup (see Figure 2.3 and Part III in this paper) whereas the total operating time increases much less than proportionally (see Chapter 3 of this thesis).

2.4.2 Optimal results with pre-specified startup time

In the investigation of equilibrium time in Part I we used the time for 98% approach to equilibrium (Eq. 2.12). This definition clearly does not correspond to the optimal startup time. For difficult separations the column is run closer to steady state than for easier separations as seen in the previous section. The question is now *how sensitive* the results are to the exact value of the startup time. To this effect, we select the startup time equal to the equilibrium time $t_{eq}^{98\%}$ (Eq. 2.12) and compute the corresponding optimal total operating time $t_{tot}^{98\%}$. (The results are found by first running a simulation under total reflux to find the steady state distillate composition x_D^{ss} and thereby the startup time $t_{eq}^{98\%}$. In the new optimizations, the startup time is pre-specified to this value and $t_{tot}^{98\%}$ is found.) These values are compared with the previous optimal values in Table 2.5.

Table 2.5: Optimal results (minimum operating time $t_{tot}^{98\%}$) for different cases when the startup time is pre-specified, $t_{startup} = t_{eq}^{98\%}$.

	C1 (Ex 31)	C2 (Ex 31)	C3 (Ex 32)	C4 (Ex 32)	C5 (Ex 33)	C6 (Ex 33)	C7 (Ex 33)
$t_{eq}^{98\%}$, hr	0.429	0.429	0.179	0.179	0.100	0.100	0.100
$t_{tot}^{98\%}$, hr	1.287	0.909	2.469	0.863	2.589	4.760	4.218
$t_{eq}^{98\%}/t_{startup}^{opt}$	0.51	0.81	0.98	1.10	3.23	1.30	2.86
$t_{tot}^{98\%}/t_{tot}^{opt}$	1.004	1.626	1.000	1.001	1.018	1.002	1.010

Interestingly, although the startup time is quite different ($t_{eq}^{98\%}/t_{startup}^{opt} \neq 1$), the increase in total operating time as expressed by $t_{tot}^{98\%}/t_{tot}^{opt}$ is very small for all the cases studied (except for case C2 where a cyclic operating policy is more optimal than the reflux policy studied here).

Total operating time as a function of startup time

For a given prespecified startup time one may compute the optimal total operating time. This relationship is shown graphically in Figure 2.7 for cases C3, C5 and C7. We see that

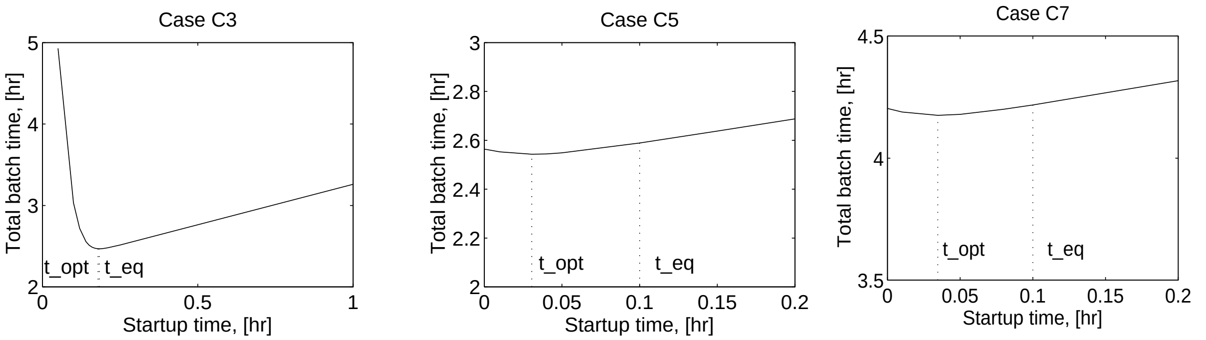

Figure 2.7: Total operating time t_{tot} as a function of startup time $t_{startup}$ for cases C3, C5 and C7 (Note that the scaling is different).

for all cases, the total operating time increases moderately when the operating time is longer than optimal. However, the optimum is very flat for cases C5 and C7 but not for case C3. For case C3 there is a very large penalty in terms of longer operating time when the startup time is less than the optimal value. Two factors contribute to this:

1. The startup time and approach to equilibrium is more important for cases where the tightest specification is for the light product (case C3).

2. The startup time and approach to equilibrium is less important when an off-cut is produced since then there are more degrees of freedom (cases C5 and C7).

2.4.3 Summary of Part II

In this section we have compared the optimal startup time with the optimal total operating time for several cases. It was found that the startup time is a considerable part of the total operating time for difficult separations with high purity and recovery specifications. This is as expected. However, the actual duration of the startup time is of limited significance unless it is very different from the optimal value. The correct startup time is most important for separations with high purity constraints for the lightest product since this is the component taken off first.

2.5 PART III. Estimation of equilibrium and startup time

In the previous section we found that the optimal startup time varies considerable with the type of separation. We found that selecting the startup time equal to the 98% equilibrium time (Eq. 2.12) gives a close to optimal total operating time and this value can therefore be used as an estimate for the required startup time. However, to calculate this value, a detailed dynamic model is needed. In this part we will therefore present a short cut formula for a-priori estimation of the required equilibrium time. This formula only depends on parameters that are usually known or can be easily estimated (number of trays, relative volatility, feed composition, holdups and flows).

We will start by giving a brief overview of previous work. Then a short cut formula for the time constant $\tau_{est} = t_{eq}^{63\%}$ is presented. The derivation of the formula is given in Appendix. We compare estimate values with those found from simulations and we will show that the short cut formula presented here can be used to obtain a good estimate for the equilibrium time and thereby the startup time for a given separation.

2.5.1 Previous work

Some of the early work on batch distillation focused on estimating equilibrium time, for example, Huffman and Urey (1937), Cohen (1940), Berg and James (1948), Jackson and Pigford (1956) and Sellers and Augood (1956). The work prior to 1961 has been reviewed by Archer and Rothfus (1961).

Huffman and Urey (1937) studied the equilibrium time of a fractionating column for the separation of oxygen isotopes and developed an equation for the estimation of equilibrium time for these systems (characterized by a very small difference in volatility, $\alpha \approx 1.0$). The derivation was based on the assumption that "the time needed to achieve the steady state is equal to the quantity of the more volatile component needed to fill the column divided by the rate of flow of the component into the column from the reboiler". Isotope systems were also studied by Cohen (1940) and Sellers and Augood (1956). In their papers, the rate of approach to equilibrium was estimated from an analytical solution of the differential equations describing the system. In the case of isotope separations, equilibrium is found to be approximated only after many days of total reflux operation.

Berg and James (1948) rederived and modified the theoretical equations from Cohen (1940) in order to permit the determination of startup times for columns commonly encountered in industrial practice. They derived equations which allow the prediction of the time involved in reaching a given reflux composition. However, we found their equations to yield totally misleading estimates with some equilibrium times up to 1000 times longer than those from simulations. Therefore, no further discussion on their approach is given in this paper.

2.5.2 Short cut formula for the estimation of equilibrium time

In this part of the paper, we will compare simulated equilibrium times with equilibrium time constants estimated from a model presented by Jackson and Pigford (1956). The equilibrium time constant is defined as the time needed to reach 63% (0.63=1-exp(-1)) of the steady state condition:

$$\tau = t_{eq}^{63\%} \tag{2.29}$$

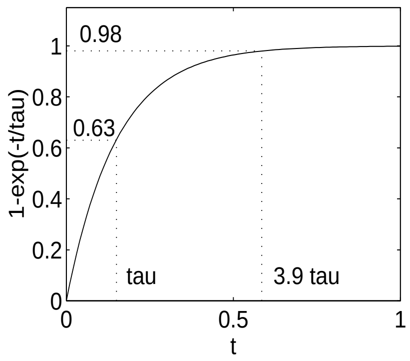

Figure 2.8: First order response with time constant τ.

Jackson and Pigford (1956) derived the following expression for τ_{est} based on the assumption given by Huffman and Urey (1937) ("the time needed to achieve..."):

$$\tau_{est} = \left(\frac{NH_j + H_C}{L}\right)\left(\frac{\bar{x}^{ss} - x_F}{y_F - x_F}\right) \tag{2.30}$$

Jackson and Pigford (1956) derived this expression from a qualitative starting point, but we present in Appendix a detailed derivation following that of Skogestad and Morari (1987) for continuous columns. The main assumptions for deriving this formula are:

A10 All trays have the same response (clearly a poor assumption as can be seen from Figure 2.5).

A11 The reboiler holdup is much larger than the holdup in the column section and condenser.

The assumption of identical responses yields:

$$\Delta x_j(t) = \Delta x_j(1 - k(t))$$

where

$$\begin{aligned} \Delta x_j(t) &= x_F - x_j(t) \\ \Delta x_j &= x_F - x_j^{ss} \end{aligned}$$

By using material balances (see Appendix) we get a first order response where $k(t)$ is given by

$$k(t) = \exp(-t/\tau_{est}) \tag{2.31}$$

and illustrated in Figure 2.8. The *estimated* time to reach 98% approach to equilibrium, $t_{est}^{98\%}$, can then be calculated from:

$$1 - \exp\left(-t_{est}^{98\%}/\tau_{est}\right) = 0.98 \tag{2.32}$$

and we find that

$$t_{est}^{98\%} = 3.9\tau_{est} \tag{2.33}$$

Below we shall compare $t_{est}^{98\%} = 3.9\tau_{est}$ with the simulated value $t_{eq}^{98\%}$ (more precisely we will compare τ_{est} with $t_{eq}^{98\%}/3.9$).

The average steady state composition in the column and the condenser, $\bar{x}^{ss}$ in Eq. 2.30, is calculated from one of the following three equations (see Appendix):

1. Exact relationship for case with infinite reboiler (no change in reboiler composition)

$$\bar{x}_1^{ss} = \frac{\sum^N H_j \frac{\alpha^{N+1-j} x_F}{1+(\alpha^{N+1-j}-1)x_F} + H_c \frac{\alpha^{N+1} x_F}{1+(\alpha^{N+1}-1)x_F}}{N H_j + H_C} \tag{2.34}$$

2. Simplified expression by Jackson and Pigford (1956)

$$\bar{x}_2^{ss} = \frac{1}{2} \frac{\ln \left(\frac{1+\left(\alpha^{N+1}-1\right)x_F}{1+(\alpha-1)x_F} \right) \left(\frac{1+\left(\alpha^{N+2}-1\right)x_F}{1+(\alpha^2-1)x_F} \right)}{N \ln \alpha} \tag{2.35}$$

3. Short cut formula by Skogestad and Morari (1987)

$$\bar{x}_3^{ss} = \left(1 + \frac{1}{\frac{N \ln \alpha}{\ln \frac{1-x_F}{x_F}} - 1} \right)^{-1} \tag{2.36}$$

One advantage with the last two expressions, and in particular the last one $\bar{x}_3^{ss}$, is that the calculations involved are much simpler than for $\bar{x}_1^{ss}$.

In the following we will compare the estimated time constant τ_{est} with $t_{eq}^{98\%}/3.9$ obtained from simulations. The estimated time constant is given for the three average steady state compositions $\bar{x}^{ss}$ (Eq. 2.34-2.36) and is denoted τ_{est1}, τ_{est2} and τ_{est3}.

Comparison with simulations

The average compositions $\bar{x}_1^{ss}$, $\bar{x}_2^{ss}$ and $\bar{x}_3^{ss}$ and the corresponding estimated time constants, τ_{est1}, τ_{est2} and τ_{est3} are given in Table 2.6. The corresponding values from dynamic simulations, $\bar{x}_{sim}^{ss}$, $t_{eq}^{98\%}$ and $t_{eq}^{98\%}/3.9$ are also given. The examples are from Table 2.1. Note that identical examples have been omitted (Example 7=21=24=27). The time constants and the equilibrium time are compared in Figure 2.9.

It is found that the average column and condenser compositions $\bar{x}_1^{ss}$, $\bar{x}_2^{ss}$ and $\bar{x}_3^{ss}$ deviate from those found from simulations $\bar{x}_{sim}^{ss}$ and as expected $\bar{x}_1^{ss}$ shows the smallest deviation. (The difference between $\bar{x}_1^{ss}$ and the simulated value $\bar{x}_{sim}^{ss}$ is due to the assumption of infinite reboiler holdup.) The mean deviation between $\bar{x}_1^{ss}$ and the simulated value $\bar{x}_{sim}^{ss}$ is 0.071 whereas the deviation for $\bar{x}_2^{ss}$ or $\bar{x}_3^{ss}$ is 0.122 and 0.115 respectively. Nevertheless, since $\bar{x}_3^{ss}$ is much easier to compute, it may be preferred.

The time constants τ_{est1}, τ_{est2} and τ_{est3} differ as expected since the average compositions $\bar{x}_1^{ss}$, $\bar{x}_2^{ss}$ and $\bar{x}_3^{ss}$ are different. For all the examples in Table 2.6, the estimated time constant τ_{est} is much larger than the time constant $t_{eq}^{98\%}/3.9$ found from simulations (see Figure 2.9, left plot). The main reason for this deviation is that $t_{eq}^{98\%}$ is defined at the top

Table 2.6: Average compositions $\bar{x}_1^{ss}$, $\bar{x}_2^{ss}$ and $\bar{x}_3^{ss}$, time constants τ_{est1}, τ_{est2} and τ_{est3} and $\bar{x}_{sim}^{ss}$, $t_{eq}^{98\%}$ and $t_{eq}^{98\%}/3.9$ from simulations. (The amount of feed is $H_F = 10.0\ kmol$ in all cases).

Ex.	$\bar{x}_1^{ss}$	$\bar{x}_2^{ss}$	$\bar{x}_3^{ss}$	$\bar{x}_{sim}^{ss}$	τ_{est1} hr	τ_{est2} hr	τ_{est3} hr	$t_{eq}^{98\%}$ hr	$t_{eq}^{98\%}/3.9$ hr
1	0.957	0.705	0.575	0.952	0.221	0.159	0.128	0.275	0.072
2	0.804	0.705	0.575	0.777	0.333	0.290	0.232	0.402	0.105
3	0.679	0.705	0.575	0.556	0.835	0.869	0.697	1.062	0.276
4	0.651	0.705	0.575	0.399	1.461	1.593	1.277	1.874	0.487
5	0.970	0.793	0.683	0.968	0.117	0.093	0.078	0.155	0.040
6	0.854	0.793	0.683	0.843	0.184	0.169	0.143	0.214	0.056
7	0.760	0.793	0.683	0.704	0.484	0.508	0.428	0.506	0.132
8	0.739	0.793	0.683	0.612	0.859	0.931	0.784	0.924	0.240
9	0.993	0.955	1.000	0.993	0.033	0.030	0.033	0.087	0.023
10	0.962	0.955	1.000	0.960	0.055	0.055	0.060	0.100	0.026
11	0.936	0.955	1.000	0.931	0.157	0.164	0.180	0.179	0.047
12	0.931	0.955	1.000	0.919	0.284	0.300	0.330	0.289	0.075
13	0.998	0.987	1.000	0.998	0.025	0.023	0.025	0.081	0.021
14	0.988	0.987	1.000	0.988	0.042	0.042	0.045	0.091	0.024
15	0.981	0.987	1.000	0.979	0.122	0.126	0.135	0.150	0.039
16	0.979	0.987	1.000	0.976	0.222	0.232	0.248	0.236	0.061
17	0.807	0.793	0.683	0.797	0.130	0.127	0.107	0.137	0.036
18	0.854	0.793	0.683	0.843	0.184	0.169	0.143	0.214	0.056
19	0.949	0.793	0.683	0.923	0.622	0.508	0.428	0.960	0.250
20	0.739	0.793	0.683	0.686	0.429	0.466	0.392	0.421	0.109
22	0.854	0.793	0.683	0.757	0.922	0.846	0.713	1.429	0.372
23	0.573	0.582	0.458	0.514	0.662	0.675	0.501	1.213	0.315
25	0.862	0.905	0.800	0.818	0.305	0.322	0.280	0.199	0.052
26	0.543	0.613	0.366	0.485	0.325	0.376	0.195	0.644	0.167
28	0.840	0.861	0.789	0.795	0.543	0.558	0.505	0.376	0.098
29	0.880	0.896	0.842	0.843	0.572	0.584	0.544	0.311	0.081
30	0.904	0.917	0.873	0.872	0.590	0.599	0.567	0.274	0.071
Deviation	0.071	0.122	0.115						

of the column which responds much faster than the trays inside the column (recall Figure 2.5) which is used for calculating the average composition. In addition, the assumption of identical responses on all stages is clearly incorrect.

As a final remark we note with some surprise that the estimated time constant τ_{est} in most cases is quite close to the equilibrium time $t_{eq}^{98\%}$ (and not $t_{eq}^{98\%}/3.9$) (see Figure 2.9, right plot). In the following we shall compare the estimated time constant τ_{est} with the equilibrium time $t_{eq}^{98\%}$ and the optimal startup time $t_{startup}^{opt}$.

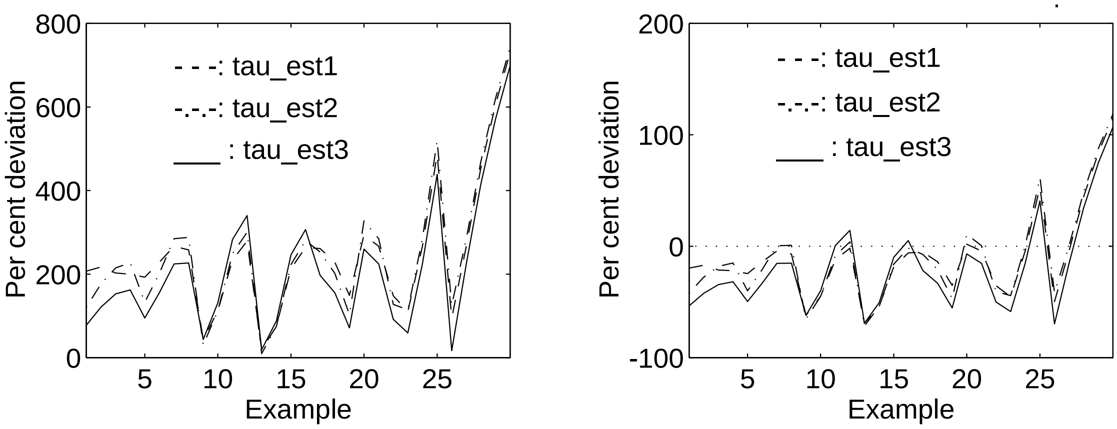

Figure 2.9: Deviation of estimated equilibrium time constant τ_{est} from a) simulated time constant $((\tau_{est} - t_{eq}^{98\%}/3.9)/t_{eq}^{98\%}/3.9)$ and b) simulated 98% equilibrium time $((\tau_{est} - t_{eq}^{98\%})/t_{eq}^{98\%})$.

Table 2.7: Optimal startup time $t_{startup}^{opt}$, 98% equilibrium time $t_{eq}^{98\%}$ and estimated time constants τ_{est1}, τ_{est2} and τ_{est3}.

Case	$t_{startup}^{opt}$ hr	$t_{eq}^{98\%}$ hr	τ_{est1} hr	τ_{est2} hr	τ_{est3} hr
C1	0.886	0.450	0.733	0.746	0.684
C2	0.556	0.450	0.733	0.746	0.684
C3	0.183	0.179	0.194	0.187	0.158
C4	0.162	0.179	0.194	0.187	0.158
C5	0.031	0.100	0.055	0.055	0.060
C6	0.077	0.100	0.055	0.055	0.060
C7	0.035	0.100	0.055	0.055	0.060

Comparison with optimal startup time

From a practical point of view it is more interesting to compare the estimated equilibrium time constant τ_{est} with the optimal startup times found in Part II (see Table 2.4 and 2.5). In Table 2.7 the optimal startup time, $t_{startup}^{opt}$, and the simulated time to reach 98% approach to equilibrium, $t_{eq}^{98\%}$, are given together with the estimated time constants calculated from Eq. 2.30 for cases C1 to C7 in Part II.

We see that the estimated time constants are quite close to the optimal startup times. Also keep in mind the results from Part II where we found that the startup time had a limited influence on the total operating time unless it was very different from the optimal value. The estimated time constant τ_{est} therefore provides a good estimate for the startup time and we recommend to use:

$$t_{startup} \approx \tau_{est} \tag{2.37}$$

2.5.3 Summary of Part III

In this part we have presented a short cut formula for the estimation of required startup time. Three different versions of the formula have been investigating depending on the estimation of the average composition in the column and condenser. We have found that the three estimated time constants τ_{est1}, τ_{est2} and τ_{est3} all provide good estimates for the startup time for several cases. The average column and condenser composition $\bar{x}^{ss}$ calculated from the three different versions, all deviate from the simulated value. However, this deviation is of little importance compared to the assumptions made in the development of the formula. We therefore recommend to use the simplest equation, Eq. 2.36 ($\bar{x}_3^{ss}$). The following equations should therefore be used for the estimation of startup time:

$$t_{startup} \approx \tau_{est} = \left(\frac{NH_j + H_C}{L}\right)\left(\frac{\bar{x}^{ss} - x_F}{y_F - x_F}\right) \tag{2.38}$$

$$\bar{x}^{ss} = \bar{x}_3^{ss} = \left(1 + \frac{1}{\frac{N \ln \alpha}{\ln \frac{1-x_F}{x_F}} - 1}\right)^{-1} \tag{2.39}$$

where $t_{startup}$ is the *estimated startup time* and $\bar{x}^{ss}$ the average steady state composition in the column and condenser calculated from the short cut formula by Skogestad and Morari (1987).

2.6 PART IV. Improvements in the startup procedure

Obviously a quick composition change has to take place during the startup period in order to reach steady state or a prescribed reflux composition as soon as possible. Changes in the startup procedure and/or the equipment characteristics may be necessary in order to reduce the duration of this period. The following suggested improvements will be considered in the last part of this paper:

1. Partial backmixing equipment
2. Finite reflux with some reflux directly to the reboiler
3. Light material in the condenser drum initially

2.6.1 Previous work

Partial backmixing equipment. González-Velasco *et al.* (1987) proposed to modify the overhead equipment of the batch distillation unit in order to more closely approach a plug flow behavior within the condenser. They demonstrated that this behavior, instead of the normal complete mixing, reduces the inertia to composition changes and permits a more rapid startup of the unit. One of the modifications, partial backmixing equipment, is illustrated in Figure 2.10. The partial backmixing was run as follows: When drum 1

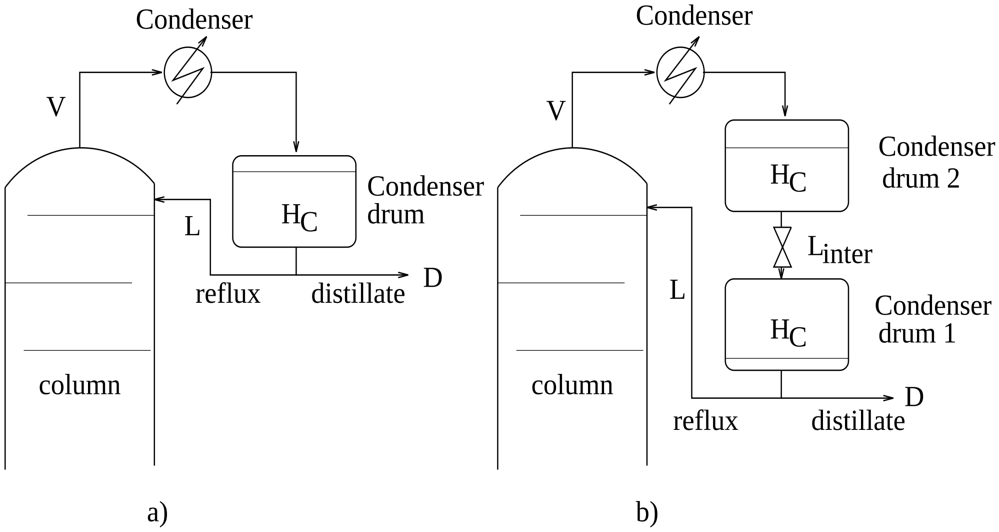

Figure 2.10: a) Usual equipment with backmixing in the condenser; and b) partial backmixing equipment with two condenser drums.

had been initially filled up during startup, the valve was closed and total reflux operation begun. The reflux was provided from drum 1 and the condensed vapor stored in drum 2. As soon as drum 1 was empty, drum 2 was completely full; at this time the valve was opened and liquid transferred from drum 2 to 1 during a period of time which was considered to be negligible. This cycle was repeated until steady state or a prescribed reflux concentration was reached. The authors reported time savings in the startup time of up to 25%.

Finite reflux with some reflux directly to the reboiler. González-Velasco *et al.* (1987) also proposed to return a portion of the condensed vapors as reflux to the column and the rest to the reboiler during the startup period. However, they did not considered the proposal in detail. The same procedure was also suggested by Domenech and Pibouleau (1989), who claimed that the sudden change in distillate composition at the beginning of the production period was thereby avoided and Albet *et al.* (1994) who stated that this procedure gives more flexibility to reach the desired distillate composition. A schematic representation of this configuration is given in Figure 2.11. Intuitively, this sounds more like a deterioration than an improvement as we will indeed show in the following.

Light component in the condenser drum initially. A third possible improvement in the startup procedure is to start the batch with either light product from the previous batch or pure light component in the condenser initially. This way, the reflux will already be at its prescribed value and steady state will be reached quicker. This mode of operation is particularly easy to implement if the light component is water. Luyben (1988) proposed to use the first off-cut from a batch to fill up the condenser drum prior to startup of the next batch. However, no results were given. One drawback with this method is that light component which has already been separated is returned to the column and re-processed. The time saved during startup must therefore compensate for a longer production time if this procedure is to be beneficial.

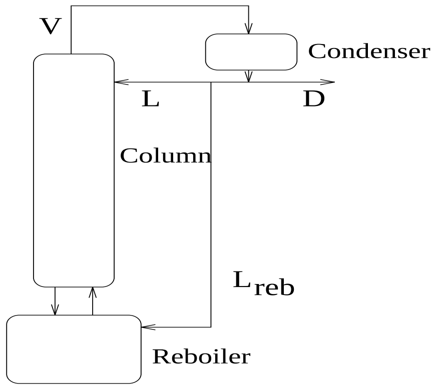

Figure 2.11: Batch distillation column with some reflux to the reboiler.

2.6.2 Partial backmixing equipment

In this section we will compare the equilibrium startup time, defined as the time for 98% approach to equilibrium (Eq. 2.12), for a number of cases using one condenser drum (conventional equipment) and two condenser drums (partial backmixing equipment). The following changes are made in the dynamic model to describe the case with two condenser drums (Eq. 2.6-2.8 replaced by):

condenser and condenser drums:

$$dH_{C2}/dt = V_1 - L_{inter} \tag{2.40}$$

$$dH_{C2}x_{C2}/dt = V_1 y_1 - L_{inter} x_{C2} \tag{2.41}$$

$$dH_{C1}/dt = L_{inter} - L - D \tag{2.42}$$

$$dH_{C1}x_D/dt = L_{inter} x_{C2} - Lx_D - Dx_D \tag{2.43}$$

$$R = L/V_1 \tag{2.44}$$

$$L_{inter} = \begin{cases} L_{inter}^{max} & \text{during dumping} \\ 0 & \text{otherwise} \end{cases} \tag{2.45}$$

The tray holdups are assumed constant and with an initial composition equal to the feed composition (INIT-1). The bottom condenser drum is assumed initially filled ($H_{C1}^o = H_C$) whereas the top drum is empty ($H_{C2}^o = 0$). The time to dump the content from the top drum to the bottom drum is assumed almost negligible (L_{inter}^{max} very large).

Results

The results are given in Table 2.8 for different feed compositions and condenser holdups. The condenser drum holdup H_C is 0.5, 2.5, 5 and 10% of the initial charge respectively. Note that the total drum holdup for the case with two condenser drums is equal to the holdup in the single drum case ($H_{C1} + H_{C2} = H_C$). The total internal column holdup $\sum^N H_j$ is assumed to be 5% of the initial charge.

The difference in equilibrium time is small when $x_F = 0.1$, or when there is a low amount of light component in the feed. However, using two drums will actually slightly increase the equilibrium time for low x_F and large H_C. The largest time saving (12%) is

Table 2.8: Steady state distillate composition x_D^{ss}, equilibrium time $t_{eq}^{98\%}$ and % time saving for one and two condenser drums for different feed composition x_F and condenser drum holdup H_C. (Parameter values: $N = 10$, $\alpha = 2.0$, $V_B = 10.0\ kmol/hr$, $H_j = 0.05\ kmol$, $H_F = 10\ kmol$, INIT-1 for initialization).

Parameters x_F	H_C $kmol$	distillate composition x_D^{ss}	Equilibrium time one drum $t_{eq}^{98\%}$, hr	two drums $t_{eq}^{98\%}$, hr	% time saving
0.1	0.05	0.9931	0.421	0.418	0.7%
	0.25	0.9905	0.794	0.790	0.5%
	0.5	0.9827	1.429	1.428	0.07%
	1.0	0.8566	3.230	3.263	-1.0%
0.5	0.05	0.9995	0.144	0.143	0.7%
	0.25	0.9994	0.295	0.280	5.1%
	0.5	0.9994	0.502	0.458	8.8%
	1.0	0.9993	0.927	0.814	12.2%

for the case with the largest feed composition and the largest condenser drum ($x_F = 0.5$, $H_C = H_{C1} + H_{C2} = 1.0\ kmol$, $t_{eq}^{98\%} = 0.93\ hr$ for one drum and $t_{eq}^{98\%} = 0.81\ hr$ for two drums). González-Velasco *et al.* (1987) reported time savings from 2 to 25% using two drums instead of one. However, they only studied cases where the condenser holdup was 5 or 10% of the initial charge and the feed composition $x_F = 0.5$ to 0.6.

The distillate composition for operation with one and two condenser drums is given in Figure 2.12 for the case with $x_F = 0.5$ and $H_C = 0.5\ kmol$. The distillate composition is stepwise constant for the case with two condenser drums. Also, for this case steady state is reached quicker when two drums are used.

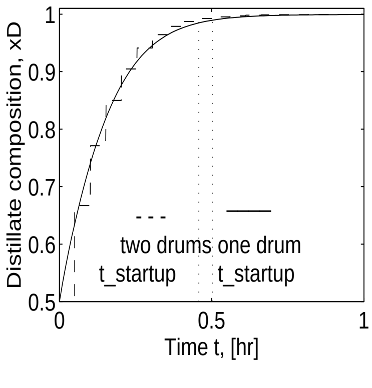

Figure 2.12: Distillate composition x_D for one and two condenser drums when $x_F = 0.5$ and $H_C = 0.5\ kmol$ (Parameter values: $N = 10$, $\alpha = 2.0$, $V_B = 10.0\ kmol/hr$, $H_j = 0.05\ kmol$, $H_F = 10\ kmol$, INIT-1 for initialization).

It can therefore be concluded that *the partial backmixing equipment is advantageous*

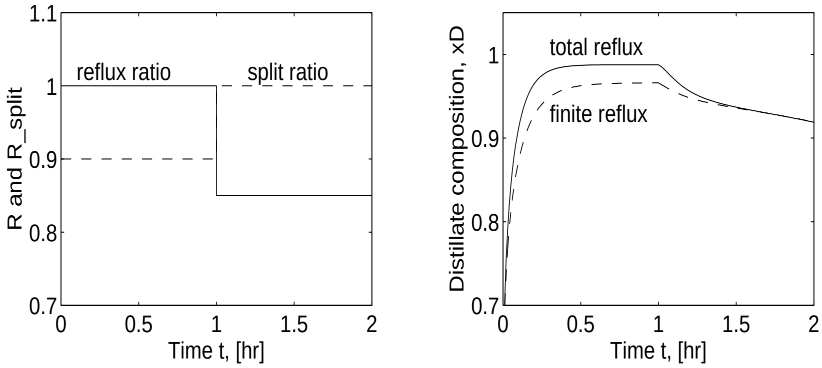

Figure 2.13: Reflux ratio R, split ratio R_{split} and distillate composition x_D as a function of time for the cases with total reflux and finite reflux. (Parameter values: $N = 10$, $\alpha = 2.0$, $V_B = 10.0\ kmol/hr$, $H_j = 0.05\ kmol$, $H_F = 10\ kmol$, $H_C = 10^{-6}\ kmol$, $x_F = 0.5$, R=1.0 when $t < 1\ hr$ and 0.85 otherwise and $R_{split} = 0.9$ when $t < 1\ hr$ and 1.0 otherwise, INIT-1 for initialization).

if the feed composition x_F is large and the condenser holdup H_C has to be large too for some reason.

2.6.3 Finite reflux with some reflux directly to the reboiler

González-Velasco *et al.* (1987) discussed the possibility of running the column with a finite reflux ratio during the startup period, i.e. returning a portion of the condensed vapors as reflux and the rest to the reboiler. However, this method leads to a steady state with a lower distillate composition as indeed pointed out by González-Velasco *et al.* (1987). The advantage of this procedure is, according to Domenech and Pibouleau (1987), that the sudden change in distillate composition at the beginning of the production period is avoided. This is illustrated in Figure 2.13 where the distillate composition is given as a function of time for the conventional case with total reflux and the case with finite reflux during the startup period. The startup time is 1 hr in both cases. For the case with finite reflux, 10% of the total reflux flow is returned to the reboiler during the startup period. The main equations are given below:

$$V = L + L_{reb} + D \tag{2.46}$$

$$R = \frac{L}{L+D} \tag{2.47}$$

$$R_{split} = \frac{L}{L+L_{reb}} \tag{2.48}$$

From the plot is can be seen that the steady state distillate composition is indeed lower for the finite reflux case. But the change in composition from $t = 1\ hr$, the beginning of the production period, is less than for the total reflux case. However, by reducing the reflux ratio for the conventional case stepwise at the beginning of the production period, the sudden change in distillate composition can easily be avoided. In the example the condenser holdup is assumed negligible, but the same results are obtained with a finite condenser holdup. Therefore, *there are no benefits in terms of time savings from using finite reflux to the reboiler instead of total reflux during the startup period.*

2.6.4 Material in the condenser drum initially

If the material initially charged to the condenser drum has a higher purity than the feed, the startup period may be reduced. An obvious disadvantage is that material which has already been separated is recycled back to the process and has to be re-separated. The time saved during startup must therefore compensate for a longer separation time if this procedure is to be beneficial.

We here consider the following alternative startup procedures:

P1: Normal startup where the condenser is initially filled with feed, $x_D^o = x_F$ (INIT-1).

P2: The condenser is initially filled with light product from the previous batch, $x_D^o = x_A^{spec}$.

P3: The condenser is initially filled with pure light component, $x_D^o = 1.0$ (This may be reasonable if the light component is water).

The tray holdups are assumed negligible (to avoid numerical problems in optimization) and the feed is initially charged to the reboiler (except for the part in the condenser in $P1$).

The optimal separation of a given mixture according to the four procedures can be found in terms of minimum operating time. Here, we consider a piecewise constant reflux ratio policy with one startup and one production period:

$$\min_{t^i_{startup}, t^i_{prod}, R^i_{prod}} \quad t^i_{tot} = t^i_{startup} + t^i_{prod} \tag{2.49}$$

where i is the given procedure $P1$, $P2$ or $P3$. The constraints are given by

$$H_A \geq H_A^{spec} + H_C^o \tag{2.50}$$

$$x_A \geq \frac{H_A^{spec} x_A^{spec} + H_C^o x_D^o}{H_A^{spec} + H_C^o} \tag{2.51}$$

where H_C^o is the amount of added material for procedures $P2$ and $P3$. ($H_C^o = 0$ for procedure $P1$.) In words, the problem is to find the optimal time periods $t_{startup}$ and t_{prod} and the optimal constant reflux ratio R_{prod} which minimizes the total operating time t_{tot} subject to constraints on the composition and amount of accumulated product, x_A and H_A. (Equivalently we could have specified the compositions x_A and x_R.) Note that the startup time is also optimized. For procedures $P2$ and $P3$, the amount of light product added initially must also be removed during the course of operation if these procedures are to be beneficial (Eq. 2.50). For procedure $P3$ this means that the purity specification is increased (Eq. 2.51). This will ensure the same purity and recovery of the heavy product (but will give a more difficult separation).

Table 2.9: Optimal results for a constant reflux ratio policy with one startup and one production period for procedure P1 (normal startup), P2 (light product in condenser initially) and P3 (pure light component in condenser initially). (Parameter values: $N = 10$, $\alpha = 2.0$, $V_B\ kmol/hr$, $H_F = 10\ kmol$, $H_j = 1e-6\ kmol$, INIT-1 for initialization).

x_F	H_C $kmol$	x_A^{spec}	H_A^{spec} $kmol$	% light rec.	t_{tot}^{P1} hr	t_{tot}^{P2} hr	t_{tot}^{P3} hr
0.2	0.1	0.95	1.895	90	**4.85**	4.88	5.00
0.1	0.1	0.95	0.789	75	**2.85**	2.92	3.09
0.08	0.1	0.95	0.632	75	**3.35**	3.43	3.76
0.07	0.1	0.95	0.553	75	**3.83**	3.90	4.43
0.06	0.1	0.95	0.474	75	4.77	**4.75**	5.79
0.05	0.1	0.95	0.395	75	7.36	**6.74**	10.03
0.01	0.01	0.90	0.056	50	2.83	**2.76**	4.48

The dynamic model is given by Eq. 2.1-2.11. During startup the column is run under total reflux ($R_{startup} = 1.0$). The reflux ratio R_{prod} is constant during the production period. The optimization program DAEOPT (Vassiliadis, 1993) is used with an optimization accuracy of 10^{-4}.

Results

The optimal results for some examples are given in Table 2.9. It is found that filling the condenser drum with light product from the previous batch ($P2$), may be beneficial for mixtures with a very low contents of light component. However, the time saved with procedure $P2$ relative to the normal procedure $P1$ is small. Filling the condenser with pure light component ($P3$) yields in all cases a longer operating time than the normal procedure ($P1$). However, note that the reflux ratio was assumed constant during the production period and a varying reflux ratio might yield different results. Also the effect of holdup was not considered.

The startup times for procedures $P2$ and $P3$ are negligible for all the examples, i.e. product removal can be started immediately. Thus although the startup time is reduced in procedures $P2$ and $P3$, this advantage is lost when the total operating time is considered.

2.6.5 Summary of Part VI

In this last section we have investigated alternative changes in the startup procedure and the equipment characteristics suggested to reduced the startup time. Using two condenser drums in series, here called partial backmixing equipment, is found to be advantageous when the feed is rich in light component (x_F large) and the condenser holdup H_C is large. Also, filling the condenser drum with light product from the previous batch may be beneficial for some mixtures. However, using finite reflux, with some reflux to the reboiler during the startup period, reduces the achievable purity in the column and should therefore be avoided.

2.7 Conclusions

In this paper we have discussed various aspects of startup of a batch distillation column. A summary of the main results are given below.

- It is not necessary to include the filling of trays and condenser drum in optimization studies.
- When defining a measure for the necessary startup time for a batch column, relative and not absolute values should be used.
- The exact value of the startup time is of limited significance unless it is very different from the optimal value.
- A short cut formula can be used to provide an estimate for the required startup time.
- Using two condenser drums in series can reduce the startup time for some separations.

In this paper we have used a simplified model of the column and also the separations studied were not too difficult. In practice, the startup time will be significant for separations with very low relative volatilities, where the feed is low in one component and where the purity specifications for the product is very high (typically ppm-level of the undesired components). In addition, these separations often involve azeotropes. The validity of the short cut formula and the possible improvements in the startup procedure should therefore also be investigated for this type of mixtures.

The short cut formula is based on the assumption of equal first order responses in the column which is shown to be a poor assumption. Possible improvements should be considered.

Notation

D	distillate flow	$kmol/hr$
H_A	liquid holdup in accumulator	$kmol$
H_C	liquid holdup in condenser drum	$kmol$
H_F	amount of initial liquid feed charge	$kmol$
H_j	liquid holdup on tray j	$kmol$
H_j^o	initial liquid holdup on tray j	$kmol$
H_{off}	amount of off-cut	$kmol$
H_R	amount of residual in column, reboiler and condenser	$kmol$
L	reflux flow	$kmol/hr$
L_{inter}	liquid flow between first and second condenser drum (backmixing equipment)	$kmol/hr$
L_j	liquid flow from tray j	$kmol/h$
L_j^o	initial liquid flow from tray j	$kmol/hr$
L_{reb}	reflux flow to the reboiler (finite reflux case)	$kmol/hr$
N	number of trays in the column section (excl. reboiler and condenser)	
R	internal reflux ratio $=L/V_1$ (or $= L/(L+D)$ for finite reflux case)	
R_{split}	split ratio between reflux to column section and reboiler (finite reflux case)	
t	time	hr
t_{tot}	total operating time	hr
$t_{startup}$	startup time	hr
$t_{eq}^{98\%}$	time to reach 98% of steady state as defined by Eq. 2.12	hr
V_j	vapor flow from tray j	$kmol/hr$
x_A	mole fraction of light component in accumulated product	
x_B	mole fraction of light component in reboiler	
x_D	mole fraction of light component in distillate	
$x_D^{98\%}$	mole fraction of light component in distillate at 98% approach to equilibrium	
x_F	mole fraction of light component in initial feed charge	
x_j	mole fraction of light component in liquid leaving tray j	
x_{off}	mole fraction of light component in off-cut	
x_R	mole fraction of light component in residual in column and reboiler	
$\bar{x}$	average mole fraction of light component in the column section and condenser	
y_B	mole fraction of vapor leaving the reboiler	
y_F	mole fraction of vapor in equilibrium with liquid feed	
y_j	mole fraction of light component in vapor leaving tray j	
Greek letters		
α	relative volatility	
γ	fraction of approach to equilibrium in Eq. 2.12	
Δ	deviation from initial state	
τ_h	hydraulic time constant $= 2/3 \cdot H_j^o/L_j^o$	hr^{-1}
τ_{est}	estimated time constant from short cut formula	hr
Scripts		
j	tray number	
$norm$	normalized	
o	initial conditions	
opt	optimal value	
$spec$	specified value	
ss	steady state	

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Appendix. Short cut formula

The derivation of the short cut formula for the estimation of equilibrium time used in section 2.5, is based on the same 9 assumptions as for the dynamic model in section 2.2, A1-A9, and in addition:

A10 All the trays and the condenser have the same dynamic response (a poor assumption as shown in Figure 2.5)

A11 Large reboiler holdup $H_B \gg H_j, H_C$

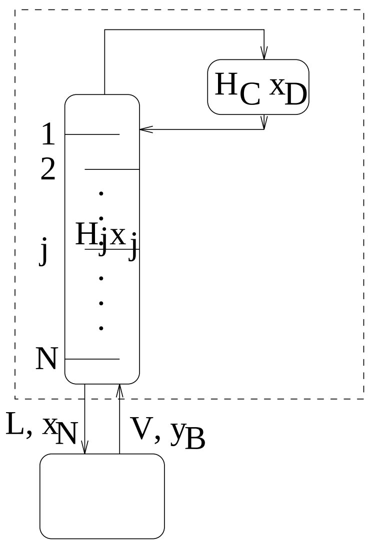

Figure 2.14: Batch distillation column under total reflux.

A12 Total reflux during startup, $D = 0$, $L = V$ and $R = 1$

A13 Initial composition x_F on all trays and in condenser (INIT-1)

An overall material balance for the column section (see Figure 2.14):

$$\frac{d\left(\sum^N H_j x_j + H_C x_D\right)}{dt} = V y_B - L x_N \tag{2.52}$$

Subtracting the final steady state under total reflux

$$0 = V y_B^{ss} - L x_N^{ss} \tag{2.53}$$

gives

$$\frac{d\left(\sum^N H_j x_j + H_C x_D\right)}{dt} = V(y_B - y_B^{ss}) - L(x_N - x_N^{ss}) \tag{2.54}$$

Since the reboiler is large compared to the holdup on the trays (A11), the reboiler vapor composition can be assumed constant $y_B \approx y_B^{ss} \approx y_F$ (see Figure 2.4 for cases where this does not hold). At $t = 0$, $x_N = x_N^0 = x_F$. Also $x_N^{ss} = y_B^{ss} \approx y_F$ giving

$$\frac{d\left(\sum^N H_j \Delta x_j + H_C \Delta x_D\right)}{dt} = -L\Delta x_N \tag{2.55}$$

where $\Delta x_N = x_F - y_F$. All the trays and the condenser are assumed to have the same dynamic response (A10)

$$\Delta x_j(t) = \Delta x_j \cdot (1 - k(t)) \tag{2.56}$$

where $\Delta x_j = x_F - x_j^{ss}$. This gives

$$\frac{d\left(\sum^N H_j \Delta x_j k(t) + H_C \Delta x_D k(t)\right)}{dt} = -L\Delta x_N k(t) \tag{2.57}$$

$$\frac{dk(t)}{k(t)} = \frac{1}{-\frac{\sum^N H_j \Delta x_j + H_C \Delta x_D}{L \Delta x_N}} dt \tag{2.58}$$

The solution to Eq. 2.58 is a first order equation

$$k(t) = \exp(-t/\tau_{est}) \tag{2.59}$$

where the time constant for the process is

$$\begin{aligned}
\tau_{est} &= \frac{\sum^N H_j \Delta x_j + H_C \Delta x_D}{L \Delta x_N} \qquad (2.60)\\
&= \frac{\sum^N H_j \left(x_F - x_j^{ss}\right) + H_C \left(x_F - x_D^{ss}\right)}{L \left(x_F - y_F\right)}\\
&= \frac{\sum^N H_j x_F - \sum^N H_j x_j^{ss} + H_C x_F - H_C x_D^{ss}}{L \left(x_F - y_F\right)}\\
&= \frac{\left(N H_j + H_C\right) x_F - \left(N H_j + H_C\right) \bar{x}^{ss}}{L \left(x_F - y_F\right)}
\end{aligned}$$

where

$$\bar{x}^{ss} = \frac{\sum^N H_j x_j^{ss} + H_c x_D^{ss}}{N H_j + H_C} \quad \text{(weighted average)} \tag{2.61}$$

and we get

$$\tau_{est} = \left(\frac{N H_j + H_C}{L}\right) \left(\frac{\bar{x}^{ss} - x_F}{y_F - x_F}\right) \tag{2.62}$$

where $\bar{x}_{ss}$ is the average composition in the column section and condenser. This gives the same results as the "intuitive" derivation of Jackson and Pigford (1956) (at least for $H_C = 0$):

$$\tau_{jackson} = \frac{\text{amount of light component needed to fill the column}}{\text{rate of flow of light component from reboiler to column}} \tag{2.63}$$

$$\tau_{jackson} = \frac{H_j \sum^N \left(x_j^{ss} - x_j^o\right)}{L \left(y_F - x_F\right)} \tag{2.64}$$

$$\tau_{jackson} = \left(\frac{N H_j}{L}\right) \left(\frac{\bar{x}^{ss} - x_F}{y_F - x_F}\right) \tag{2.65}$$

The average steady state composition $\bar{x}^{ss}$ needed in Eq. 2.62 can be found either from (in all cases we assume $x_B^{ss} = x_F$, A11):

1. The relationship between liquid and vapor composition when a constant relative volatility is assumed and the fact that $x_j^{ss} = y_{j+1}^{ss}$ at steady state giving:

$$y_B^{ss} = \frac{\alpha x_B^{ss}}{1 + (\alpha - 1) x_B^{ss}} \quad \text{(reboiler)} \tag{2.66}$$

$$x_j^{ss} = \frac{\alpha^{N+1-j} x_B^{ss}}{1 + \left(\alpha^{N+1-j} - 1\right) x_B^{ss}} \quad \text{(column section)} \tag{2.67}$$

$$x_D^{ss} = \frac{\alpha^{N+1} x_B^{ss}}{1 + (\alpha^{N+1} - 1)\, x_B^{ss}} \quad \text{(condenser)} \tag{2.68}$$

$$\bar{x}_1^{ss} = \frac{\sum^N H_j x_j^{ss} + H_c x_D^{ss}}{N H_j + H_C} \quad \text{(weighted average)} \tag{2.69}$$

$$\mathbf{\bar{x}_1^{ss}} = \frac{\sum^N H_j \frac{\alpha^{N+1-j} x_B^{ss}}{1+(\alpha^{N+1-j}-1)x_B^{ss}} + H_c \frac{\alpha^{N+1} x_B^{ss}}{1+(\alpha^{N+1}-1)x_B^{ss}}}{N H_j + H_C} \tag{2.70}$$

The average composition is weighted according to column and condenser drum holdup. Eq. 2.67-2.68 are found from Eq. 2.66 and $x_j^{ss} = y_{j+1}^{ss}$. Eq. 2.69-2.70 gives the average steady state composition in the column and condenser, $\bar{x}_1^{ss}$, based on the composition in the reboiler x_B^{ss}.

2. Jackson and Pigford (1956) approximated the average column composition by taking the arithmetic average of the sum of the compositions:

$$\mathbf{\bar{x}_2^{ss}} = \frac{1}{2} \frac{\ln \left(\frac{1+\left(\alpha^{N+1}-1\right)x_B^{ss}}{1+(\alpha-1)x_B^{ss}} \right) \left(\frac{1+\left(\alpha^{N+2}-1\right)x_B^{ss}}{1+(\alpha^2-1)x_B^{ss}} \right)}{N \ln \alpha} \tag{2.71}$$

Note that with this approximation the compositions are not weighted according to holdup.

3. Alternatively the average steady state composition can be calculated as given by Skogestad and Morari (1987) (assuming constant relative volatility and that the composition profile can be approximated by straight lines over 3 intervals):

$$\bar{x} = \left(1 + \frac{\ln \frac{1-x_B}{x_B}}{\ln \frac{x_D}{1-x_D}} \right)^{-1} \tag{2.72}$$

The separation factor S is defined by

$$S \equiv \frac{\frac{x_D}{1-x_D}}{\frac{x_B}{1-x_B}} \tag{2.73}$$

and with total reflux we have $S = \alpha^N$. The average composition in the column section then becomes

$$\bar{x} = \left(1 + \frac{\ln \frac{1-x_B}{x_B}}{N \ln \alpha - \ln \frac{1-x_B}{x_B}} \right)^{-1} \tag{2.74}$$

$$\mathbf{\bar{x}_3^{ss}} = \left(1 + \frac{1}{\frac{N \ln \alpha}{\ln \frac{1-x_B^{ss}}{x_B^{ss}}} - 1} \right)^{-1} \tag{2.75}$$

With this approximation the compositions are not weighted according to holdup. Note that this equation is originally developed for a binary mixture in a continuous column but is here applied to a batch column.

Chapter 3

A Cyclic Operating Policy for Batch Distillation

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with emphasis on the cyclic operating policy"

Abstract

This paper sets out a cyclic operation policy for batch distillation with repeated filling and dumping of the condenser drum. It is compared to conventional policies (constant distillate, constant reflux ratio and optimal reflux ratio operation) with respect to minimum operating time. The cyclic policy has been noted earlier because it may be easier to operate and control. Surprisingly, we also find that the cyclic policy may significantly reduce the operating time compared to the conventional operating policies for difficult separations. A simplified cyclic policy is presented which is almost as good as the optimal cyclic policy but much easier to calculate. The effect of constant condenser holdup on operating time is discussed. The paper concludes with a brief discussion of the practical implementation of the cyclic policy.

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3.1 Introduction

Batch distillation is used in the chemical industry in the production of small amounts of products with high added value and for processes where flexibility is needed, for example, when there are large variations in feed composition or when production demand is varying. With an increasing demand for product quality and production flexibility, the ability to run columns as efficiently, or as optimally, as possible is of great economic importance.

The optimization of batch distillation has been studied in the past (for example: Diwekar *et al.* (1987), Farhat *et al.* (1991), Logsdon and Biegler (1992), Mujtaba and Macchietto (1988, 1992)), but mostly the optimal reflux ratio, and to a lesser extent the optimal reboiler heat duty, has been considered as optimization variables. However, there are other factors that may be much more important for the optimal operation of a batch column, such as:

1. Varying the condenser holdup (e.g., cyclic operation with filling and dumping)
2. Starting the batch with product from the previous batch in the condenser and/or column section
3. Recycling and mixing of off-cuts from different batches
4. Concentrating the product in several steps
5. Inverting the column by charging the feed to the condenser drum and withdrawing product from the reboiler
6. Varying the operating pressure
7. Using steam distillation, or generally to continuously add a volatile component to effectively reduce the pressure
8. Adding an entrainer to improve the separation, for example to break an azeotrope

We will in this paper discuss one of these factors, - *the varying condenser holdup.* Other factors are discussed elsewhere in this thesis.

Given a batch column with a fixed reboiler heat duty, the amount of reflux determines the degree of separation and therefore the product rate. In batch distillation, the reflux rate is normally controlled according to one of the following three conventional operating policies (Perry and Green, 1984):

1. Changing the reflux to keep a constant product composition (constant distillate composition policy)
2. Maintaining a constant ratio of reflux flow to distillate flow (constant reflux ratio policy)
3. Changing the reflux ratio according to some optimization criterion (optimal reflux ratio policy)

A fourth alternative usually not considered is

4. Alternating between total reflux and low reflux (cyclic policy)

The first three alternatives all assume a constant condenser holdup whereas the fourth uses *the condenser holdup as a degree of freedom.* A generalization of the cyclic policy is to let the reflux ratio and the condenser holdup vary freely with time and find their optimal profiles by optimization.

In this paper we focus on the cyclic operating policy. It has several advantages compared to the conventional schemes. It utilizes the maximum attainable separation in the column. There is a minimal need for control and also it is less sensitive to disturbances and therefore safer to operate. For small laboratory columns accurate measurement of flows may be difficult and the cyclic operation is therefore well suited for these columns since no flow measurements are needed. A disadvantage, however, is that the composition of the feed charge must be known quite exactly.

We will first give a description of the cyclic policy and compare it with the conventional policies in terms of minimum operating time for a given example. We will then present a simplified cyclic policy which is easier to calculate and implement than the optimal cyclic policy. This simplified policy is compared with the reflux ratio policies for a number of examples with varying operating conditions. The effect of constant condenser holdup on operating time is discussed. We conclude by discussing how the cyclic policy can be implemented in practice.

3.2 The cyclic operating policy

In this paper we consider the cyclic operation of a batch column in which the *external* reflux and distillate flows are cycled. This should not be confused with the controlled cycling of continuous distillation columns in which the internal flows are cycled (e.g. Cannon (1961) or Baron *et al.* (1980)). The *cyclic operation* is characterized by repeating three periods of operation: "filling up", "total reflux" and "dumping". The three periods are illustrated in Figure 3.1. During the first period the condenser is filled up and the filling time is determined by the boilup rate. During the second period the column is run under total reflux and the maximum attainable separation in the given column is achieved. During this period the light component accumulates in the condenser drum until the column reaches equilibrium or steady state. During the last period the condenser is dumped and the product is withdrawn. The dumping time is determined by the maximum distillate flow out of the condenser. This three steps procedure is repeated until the specifications are satisfied for either the product in the accumulator or for the residue in the reboiler and column. The number of cycles is largely determined by the feed composition but also by the product specifications. The cycle times for this policy will be in the order of minutes and hours.

The *cyclic operating policy* for batch distillation columns has only been mentioned by a few authors in the literature. Barb and Holland (1967) presented a dynamic and a steady state model for the total reflux period as well as dynamic models for the dumping period

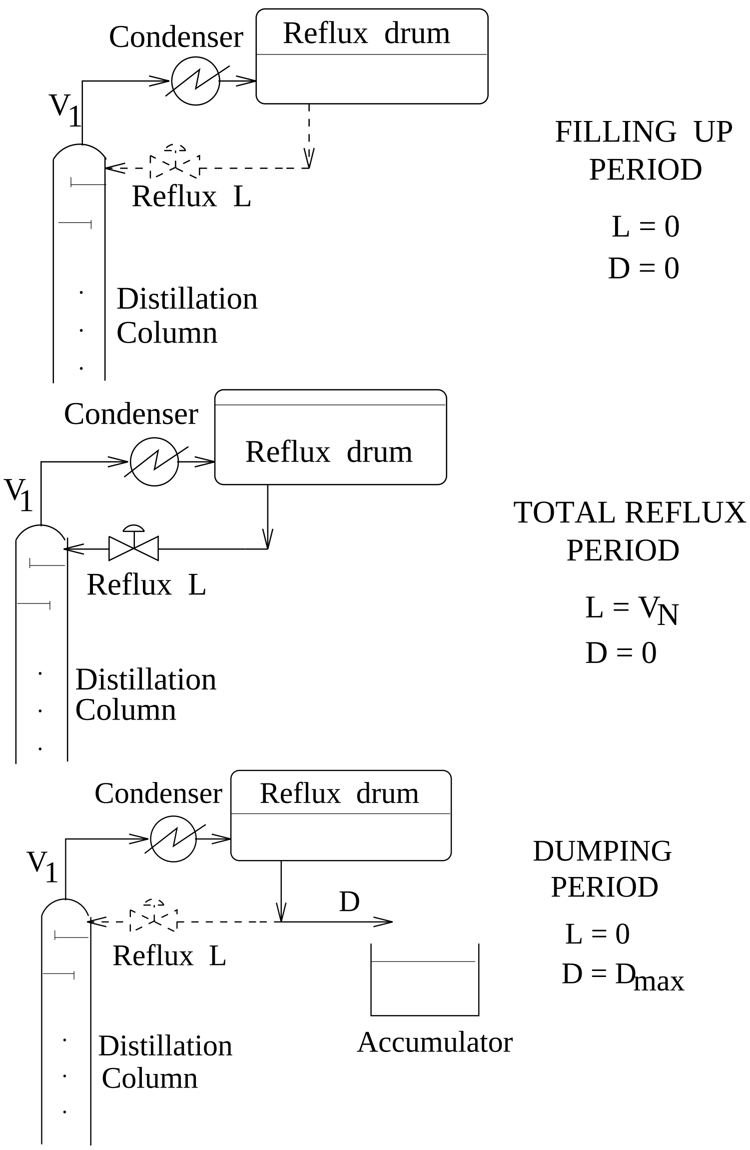

Figure 3.1: The three characteristic periods in the cyclic operation of a batch distillation column.

of the cyclic operation and for the conventional operation with constant or varying reflux ratio. The results were compared with experiments. However, only the models' ability to describe the course of the distillation in terms of distillate composition as a function of accumulated product was considered. The results by Barb and Holland (1967) were also presented in the book by Holland and Liapis (1983). Block (1967) discussed control of batch distillation columns. He proposed alternating between total reflux and no reflux (cyclic operation) as a mean for controlling the reflux rate but without giving any details about this operating scheme. However, he reported that one of the main difficulties is how to establish proper intervals of total and zero reflux. Gonzalez-Velasco *et al.* (1987) found that the cyclic operating policy (in their paper called *periodic batch distillation*) produces a higher sharpness of separation between components than the conventional constant reflux ratio policy. Their results were based on calculations of the steady state conditions corresponding to the end of the total reflux period for separations with 22 operating cycles and with varying relative volatilities between three components. No comparison was made between the two policies in terms of time consumption. Nowicki and Gorak (1988) presented a steady state model for the determination of the end point of the total reflux period for cyclic operation. They considered an example where a high purity separation was performed but used only one cycle to accomplish the separation. They reported that the energy consumption for this one cycle was only slightly higher than for the equivalent constant reflux ratio operation.

The cyclic operating policy has, to the best of our knowledge, not been studied as an alternative to the conventional operating policies with respect to optimal operation. The purpose of this paper is to compare the cyclic operating policy with the conventional operating policies in terms of minimum operating time. We will present guidelines for:

1. When the cyclic policy is preferred over the conventional operating policies,
2. How to determine the end point of the total reflux period and also
3. How to select the number of cycles for a given separation.

3.3 Comparison of operating policies

In this section we compare the optimal operation of the various operating policies. By optimal operation we generally mean the policy that maximizes profit for a given separation. For many practical cases maximizing the profit will be equivalent to minimizing the operating time t_f with the heat input (boilup rate) at its maximum. This is the problem considered in this paper. We solve the *minimum time problem* given constraints on the final accumulated light product. The optimization problem can be formulated as:

$$\min_{oper.param.} t_f \tag{3.1}$$

where t_f is the total operating time and subject to the following end point constraints for the accumulated product:

$$H_A(t_f) \geq H_A^{spec} \quad , \quad x_A(t_f) \geq x_A^{spec} \tag{3.2}$$

In words, the objective is to find the optimal operating parameter(s) which minimizes the total operating time t_f subject to constraints on the product composition x_A^{spec} and amount H_A^{spec}. In this paper, the optimization problem has been solved in the following way for the four policies:

Constant distillate composition. The optimal set point x_D^s for a composition controller which minimizes the operating time is found. The distillate composition is kept at the specified set point value x_D^s using a PI-controller with the reflux ratio R as manipulated variable. (The use of the distillate composition directly as an optimization variable would result in a high index problem which can not be solved by the given optimization software and which in any case can not be realized in practice).

Constant reflux ratio. The optimal constant reflux ratio R is found which minimizes the total operating time.

Optimal reflux ratio. The optimal reflux ratio as a function of time $R(t)$ is found which minimizes the total operating time. (This problem is equivalent to finding the optimal $x_D(t)$.) In this paper we use a slightly suboptimal policy where R is either piecewise constant or piecewise continuously linear over certain intervals.

Cyclic policy. For a given number of cycles, N_{cyc}, we find for each cycle k the optimal condenser holdup $H_{C,k}$ and the total reflux time $t_{totalreflux,k}$ which minimizes the operating time (see Figure 3.2). The filling and dumping times will be given by the condenser holdup and the reboiler heat duty (here given in terms of the boilup rate V) and distillate flow respectively:

$$t_{fill,k} = \frac{H_{C,k}}{V_{max}} \tag{3.3}$$

$$t_{dump,k} = \frac{H_{C,k}}{D_{max}} \tag{3.4}$$

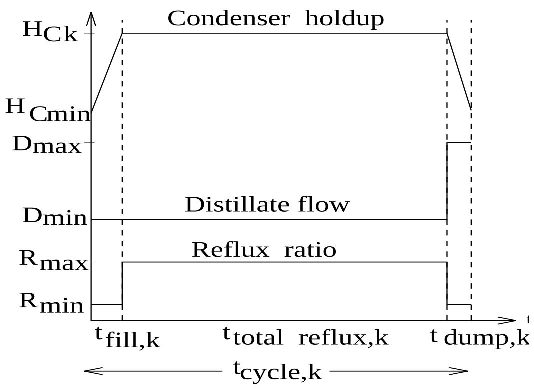

Figure 3.2: Reflux ratio, distillate flow and condenser holdup for one cycle of a cyclic operation.

3.3.1 Numerical example

As an example we consider the separation of a binary mixture with constant relative volatility $\alpha = 1.5$ in a column consisting of a reboiler, 10 theoretical trays, a condenser and an accumulator. The data used for the optimizations are given in Table 3.1. (This example is denoted Example 7 in the discussion of the simplified cyclic policy later in this paper.) Before the separation starts the feed is added to the reboiler. It is assumed that some of the feed is initially distributed on the trays in the column (i.e. $H_N^o = H_F - NH_j^o$) with a composition equal to that of the feed ($x_j^o = x_F$). (See Chapter 2 of this thesis for a discussion of initial profiles in simulation and optimization.) The initial holdups in the accumulator and in the condenser are negligible. The vapor flow (which is indirectly given by the reboiler heat duty) is in this paper assumed constant at its maximum value

Table 3.1: Process data used for comparison of operating policies.

Relative volatility:	$\alpha = 1.5$
Number of trays:	$N = 10$
Feed composition:	$x_F = 0.25$
Feed charge:	$H_F = 10$ kmol
Initial tray holdups:	$H_j^o = 0.01$ kmol
Amount of product:	$H_A^{spec} = 1.875$ kmol (67.5% recovery)
Product composition:	$x_A^{spec} = 0.9$
Bounds on variables,	
Condenser holdup:	$[H_C^{min}, H_C^{max}] = [10^{-6}, 1.875]$ kmol
Distillate flow:	$[D^{min}, D^{max}] = [0.0, 25.0]$ kmol/hr
Vapor flow:	$[V_B^{min}, V_B^{max}] = [10^{-6}, 10.0]$ kmol/hr
Reflux ratio:	$[R^{min}, R^{max}] = [0.01, 1.0]$
Controller parameters,	
constant distillate policy:	$K_p = 5.0$ and $\tau_I = 0.05$ hr

except during the dumping period in the cyclic policy when it is reduced. This is done for numerical reasons but the boilup rate would in practice be constant during the whole cycle. The maximum vapor and distillate flows are given by the column design and are in this paper chosen to be $V_B^{max} = 10$ kmol/hr and $D^{max} = 25$ kmol/hr. The minimum reflux ratio is set to a small value for numerical reasons but would in practice be zero. It should be noted that the reflux ratio is given as the internal reflux ratio since this is better solved numerically:

$$R = \frac{L}{L + D} \tag{3.5}$$

For the conventional policies the condenser holdup is assumed to be negligible ($H_C = H_C^{min}$) since this will normally minimize the operating time (Mujtaba and Macchietto, 1991). The effect of a finite condenser holdup is discussed later in this paper. For the constant distillate composition policy, the parameters for the PI-controller are given in Table 3.1.

The assumptions for the mathematical model used in this study are: (1) staged distillation column; (2) perfect mixing and equilibrium between vapor and liquid on all stages; (3) negligible vapor holdup; (4) equal molar vapor flows; (5) linear tray hydraulics; (6) total condensation with no subcooling in the condenser and (7) constant relative volatility. The model for the linear tray hydraulics has the form $L_j = L_j^o + (H_j - H_j^o)/\tau$. L_j^o and L_j are the initial and current instant liquid flow rate from the trays and H_j^o and H_j the initial and current molar holdup on the trays. The hydraulic time constant τ is given by $\tau = 2/3 \cdot H_j/L_j$.

3.3.2 Results

The optimal results for the four policies were obtained using the optimization program DAEOPT developed at Imperial College, London (Vassiliadis, 1993). The optimization accuracy was 10^{-3} for the cyclic policy and 10^{-4} for the others. It should be noted that for optimizations with more than one interval or cycle, several local minima may exist which makes the problem difficult to solve numerically.

For the optimal reflux ratio policy both a continuous linear and a piecewise constant policy is presented. The optimal results for the cyclic policy are presented for 1, 2, 3 and 4 cycles. The case with 5 cycles did not converge. The optimization for the cyclic operation is however, very time consuming. The solution for 4 cycles took about 17 CPU hours on a Sparc 10. This is because the number of optimization variables using DAEOPT is $13N_{cyc}$ where N_{cyc} is the number of cycles, whereas the actual number of optimization variables is only $2N_{cyc}$ (total reflux period and condenser holdup). A total of $9N_{cyc}$ variables are pre-specified in DAEOPT (vapor flow, reflux ratio and distillate flow for each cycle consisting of filling, total reflux and dumping periods respectively). Of the remaining $4N_{cyc}$ variables, $2N_{cyc}$ of these (the values of $t_{fill,k}$ and $t_{dump,k}$) are found by optimization, even though their optimal values are given by Eq. (3.3-3.4). Therefore "only" $4N_{cyc}$ (periods and condenser holdup) are actually optimized, but the sensitivities for all the $13N_{cyc}$ variables must be calculated for each intermediate solution of the optimization problem thus making the solution very time consuming.

The optimal results for the four policies are presented in Table 3.2 and in Figure 3.3.

Conventional policies

The constant distillate composition policy (case 1) is found to be by far the most time consuming policy for this example with $t_f = 9.53$ hr. The constant reflux policy (case 2a) yields $t_f = 5.73$ hr. Normally the constant reflux policy is operated with an initial period with total reflux. An initial total reflux period of 0.05 hr reduces the total operating time to 5.60 hr (case 2b).

For the optimal reflux policy we consider two constant intervals (case 3a), two continuously linear intervals (case 3b) and ten constant intervals (case 3c). When comparing these, we see as expected that the operating time decreases with increasing number of intervals (compare the constant interval cases 2a, 3a and c with $t_f = 5.73$ hr, 5.53 hr and 5.26 hr, respectively).

Note that for the case with two constant intervals (case 3a), letting the reflux ratio vary freely does not yield an initial total reflux solution. However, when the number of degrees of freedom is increased by allowing more intervals, the optimal initial conditions are found to be total reflux (case 3b and c). Case 1 will also have an initial period with total reflux until the specified distillate composition is reached.

As can be seen, using only two linear intervals for the optimal reflux policy (case 3b with t_f =5.32 hr) is almost as good as using 10 piecewise constant intervals (case 3c with t_f =5.26 hr) and the optimization problem is much easier in the first case (5 versus 20 optimization variables). This indicates that our solution is close to the true optimal reflux policy.

Table 3.2: Optimal solutions to the minimum time problem. For cases with more than 1 interval, t_i is the time for each interval (i.e. $t_f = \sum t_i$). The $R(t)$-values are the corresponding reflux ratios for each interval. For the cyclic policy, $t_{cycle,k} = t_{fill,k} + t_{totalreflux,k} + t_{dump,k}$. (*: on the upper or lower bound). (Data from Table 3.1.)

Case	Operating policy	Optimal t_i [hr]	Optimal variable	operating time t_f, [hr]
1	Constant dist. composition 1 interval	9.5268	x_D^s=0.90	**9.529**
2a	Constant reflux ratio, 1 interval	5.7345	R =0.9673	**5.734**
2b	Case 2a with initial total reflux	0.0529-5.5471	R = 0.9999*-0.9662	5.600
3a	Optimal reflux ratio, 2 constant intervals	3.1382-2.3955	$R(t)$ = 0.9613-0.9724	5.534
3b	Optimal reflux ratio, 2 linear intervals	0.1420-5.1804	$R(t)$ = 0.9999*-0.9522-0.9767	5.322
3c	Optimal reflux ratio, 10 constant intervals	0.0626-0.8938-0.9574-0.8925-0.8479-0.3047-0.4996-0.0971-0.7009-0.0025*	$R(t)$ = 0.9999*-0.9530-0.9582-0.9631-0.9675-0.9702-0.9720-0.9728-0.9753-0.9237	**5.259**
4a	Cyclic policy 1 cycle	$t_{cycle,1}$=7.0896	$H_{c,1}$=1.875*	7.090
4b	Cyclic policy 2 cycles	$t_{cycle,1}$=2.2354 $t_{cycle,2}$=1.8403	$H_{c,1}$=1.1642 $H_{c,2}$=0.7103	4.076
4c	Cyclic policy 3 cycles	$t_{cycle,1}$=2.3560 $t_{cycle,2}$=0.6880 $t_{cycle,3}$=0.9784	$H_{c,1}$=1.2261 $H_{c,2}$=0.2910 $H_{c,3}$=0.3574	**4.022**
4d	Cyclic policy 4 cycles	$t_{cycle,1}$=1.2365 $t_{cycle,2}$=0.1155 $t_{cycle,3}$=0.8111 $t_{cycle,4}$=1.8682	$H_{c,1}$=0.6896 $H_{c,2}$=0.0500* $H_{c,3}$=0.4096 $H_{c,4}$=0.7255	4.031

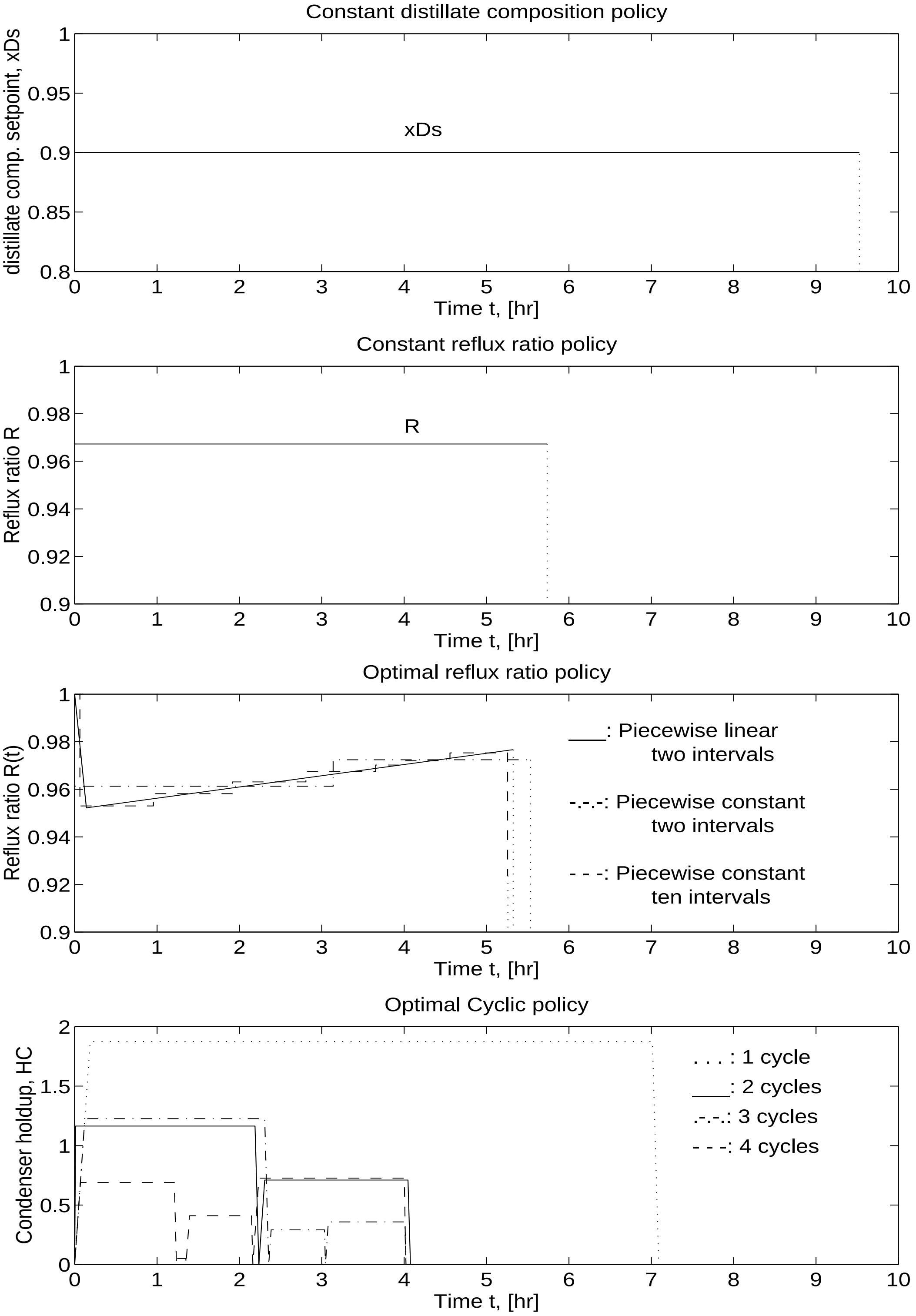

Figure 3.3: Optimal solutions to the minimum time problem. (Data from Table 3.1.)

The optimal cyclic policy

Next consider the cyclic policy. For case 4a, with only one cycle, the total operating time is t_f =7.09 hr. However, with more than one cycle (cases 4b, c and d) it is much less time consuming than any of the other policies. Specifically, the best reflux ratio policy (case 3c with t_f=5.26 hr) is about 30% more time consuming than the best cyclic policy with 3 cycles (case 4c with t_f=4.02 hr). Note that for the cyclic policy, the contribution of the filling and dumping times to the total operating time is constant and independent of the number of cycles. For our example,

$$\sum t_{fill} + t_{dump} = \frac{H_A^{spec}}{V_B^{max}} + \frac{H_A^{spec}}{D_{max}} = 0.1875 + 0.075 = 0.2625\,hr$$

Thus, in the optimal case the filling and dumping time is about 6.5% of the total operating time. Note that we have used only one condenser drum. By implementing two drums in parallel (such that the filling of the second is started at the same time as the dumping of the first one) the dumping time will be zero for all but the last cycle and the total operating time thereby reduced.

The optimal cyclic policy with 3 cycles (case 4c with t_f=4.02 hr) seems to be slightly better than with 4 cycles (case 4d with t_f=4.03 hr), although this may be due to the optimization accuracy. However, it is expected that there is a minimum somewhere since, for an infinite number of cycles, the cyclic policy would approach the optimal reflux policy (t_f=5.26 hr) but with an additional penalty for filling and dumping (0.2625 hr).

The condenser holdup $H_{C,k}$ in each period varies and from the results no conclusions as to the best condenser holdup profile (e.g. smaller holdups for later cycles) can be made. The amount and composition of the *accumulated* product for the case with 4 cycles (case 4d) is given in Table 3.3. The composition is high for the first cycle but then decreases with time until the specification is reached at the end of the last cycle.

Table 3.3: Composition x_A and amount H_A of accumulated product for the cyclic policy with 4 cycles (case 4d). (Data from Table 3.1.)

Cycle	t, [hr]	x_A	H_A, [kmol]
1	1.237	0.926	0.670
2	1.352	0.924	0.740
3	2.163	0.918	1.149
4	4.031	**0.900**	**1.875**

It is interesting to compare the sharpness of separation in the column for the different operating policies. Figure 3.4 gives the distillate composition as a function of accumulated product for case 2a and 4d compares to the total reflux case (maximum separation). It can be seen that for the cyclic policy, the distillate composition drops at the end of each cycle. This is because the vapor flow is reduced for numerical reasons and this would not occur in practice. The compositions for the cyclic policy and the reflux policy are

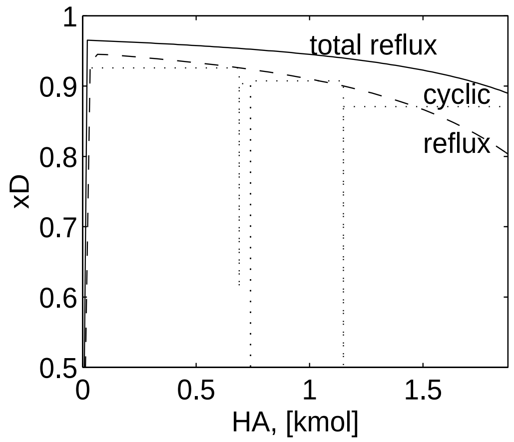

Figure 3.4: Distillate composition x_D as a function of amount of accumulated product H_A for total reflux, constant reflux ratio (case 2a) and cyclic policy with 4 cycles (case 4d). (Data from Table 3.1.)

close. The difference between the two cases is surprisingly small when one knows that the difference in operating time is 30% (5.73 hr and 4.03 hr). The results thus show that the cyclic policy is able to achieve the desired separation in a shorter time.

3.4 Simplified cyclic policy

It was found above that the cyclic policy is less time consuming and therefore better than the conventional policies. The optimization of this policy is however, very time consuming. To simplify the calculations for the cyclic policy we therefore consider a simplified cyclic policy with only one optimization parameter:

1. Select the number of cycles N_{cyc} and use the same condenser holdup for each cycle:

$$H_{C,k} = H_A^{spec}/N_{cyc} \tag{3.6}$$

2. A cycle is stopped when a given approach to equilibrium has been reached in the column. At equilibrium with total reflux, the liquid and vapor between two trays have the same composition, $x_j = y_{j+1}$. In this paper, the approach to equilibrium is defined in terms of the difference ϵ in liquid and vapor composition between tray 1 and 2 (see Figure 3.5):

$$|x_1 - y_2| \leq \epsilon \tag{3.7}$$

   The total reflux period is thus stopped when Eq. 3.7 is satisfied. The same value for ϵ is used for each cycle k and it is adjusted to meet the product composition specification. (The specification on the amount is always satisfied due to the way the condenser holdup is defined). Note that the cycle times will vary.

Instead of optimizing the condenser holdup and the total reflux time ($2N_{cyc}$ optimization variables) we now have only one variable ϵ which is selected such that the composition

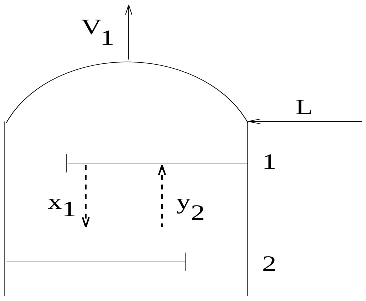

Figure 3.5: Steady state in the top of the column when $x_1 = y_2$.

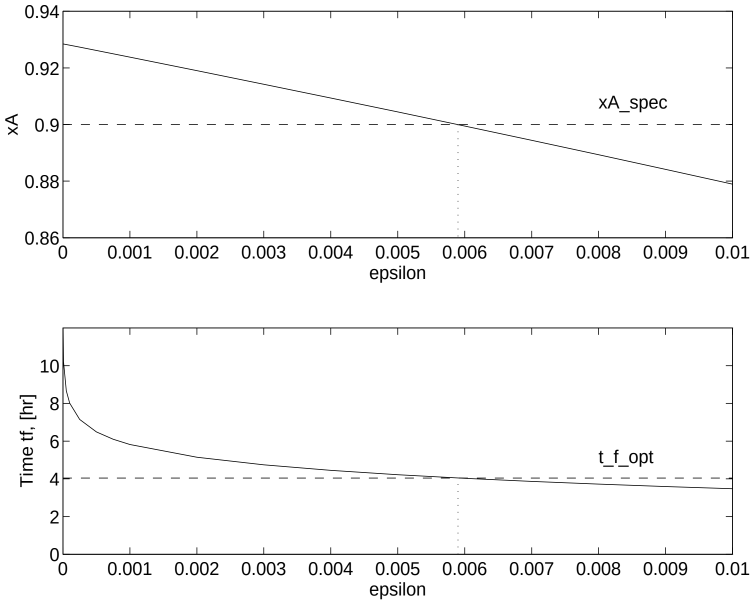

Figure 3.6: Accumulated product composition x_A and total operating time t_f as a function of ϵ for the simplified cyclic policy with 3 equal cycles. (Data from Table 3.1.)

specification is met. The procedure is demonstrated in Figure 3.6 where the final operating time and the composition of the accumulated product is presented as a function of ϵ for 3 equal cycles with the same ϵ for all cycles. The relationship between ϵ and x_A is almost linear and only a few trial values for ϵ are needed to find the value where $x_A = x_A^{spec}$. A simple batch distillation simulation program can therefore be used to find the right ϵ in the simplified policy instead of a large and complicated optimization program which is needed for the optimal cyclic policy.

Simplified versus optimal cyclic policy

Optimization of the simplified policy is fast and has few convergence problems. Importantly, we have found for the example in the previous section, that the optimal operating time from this simplified approach is only marginally larger than the operating time found by the very time consuming optimization procedure. A comparison is given in Table 3.4 for 1, 2, 3 and 4 cycles and in Figure 3.7 for 4 cycles. These results demonstrate that

Table 3.4: Minimum operating time t_f for the optimal and the simplified cyclic policy for different number of cycles N_{cyc}. (Data from Table 3.1.)

N_{cyc}	optimal cyclic policy t_f, [hr]	simplified cyclic policy t_f, [hr]
1	7.09	7.02
2	4.08	4.17
3	4.02	4.05
4	4.03	4.04

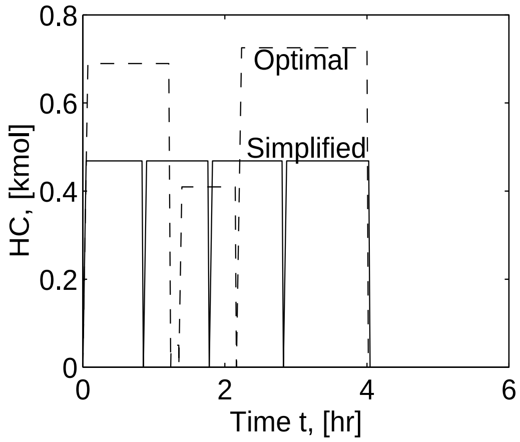

Figure 3.7: Condenser holdup H_C as a function of time t for the optimal and the simplified cyclic policy for 4 cycles. (Data from Table 3.1.)

varying the condenser holdup for each cycle as given by the optimal results is not very important for this example.

The same comparison between the optimal and the simplified cyclic policy has also been made for other examples with the same conclusion as for the example shown here.

3.5 Comparison of policies for varying conditions

We have shown above that the cyclic policy is less time consuming than any of the conventional operating policies for a given example. An obvious question is therefore: Is this always the case and if not *when is the cyclic policy to be preferred?* In this section we will try to answer this question with reference to a measure for the degree of difficulty of separation q. We will compare the simplified cyclic operating policy with the constant and optimal reflux ratio policies for varying values of q, that is for varying operating conditions. We will also give guidelines for how to select the number of cycles N_{cyc} in the simplified cyclic policy.

3.5.1 A measure for the degree of difficulty of separation

A measure for the difficulty of a given separation in a distillation column is the ratio between the minimum number of trays N_{min} required to achieve a specified separation under total reflux (i.e. the minimum value of N) and the actual number of trays N in the given column:

$$q = \frac{N_{min}}{N} \tag{3.8}$$

If $N \gg N_{min}$ (q small) the separation is considered easy but if $N \approx N_{min}$ ($q \approx 1$) it will be difficult. Fenske's equation (e.g. Perry and Green, 1984) can be used to calculate N_{min} for a binary separation in a continuous column:

$$N_{min} = \frac{\log\left(\frac{x_D}{1-x_D}\right)\left(\frac{1-x_B}{x_B}\right)}{\log\alpha} \tag{3.9}$$

For continuous distillation, x_D and x_B will be constant. However, for batch distillation, one or both compositions will vary with time according to the operating policy chosen. For the constant overhead policy, x_D will be constant and only x_B will vary with time. For the other operating policies both x_D and x_B will vary.

One alternative is therefore to use an average number of N_{min} for batch distillation where N_{min} is averaged over time:

$$\overline{N_{min}} = \frac{\int_{t_0}^{t_f} N_{min}(t)\,dt}{(t_f - t_0)} \tag{3.10}$$

where

$$N_{min}(t) = \frac{\log\left(\frac{x_D(t)}{1-x_D(t)}\right)\left(\frac{1-x_B(t)}{x_B(t)}\right)}{\log\alpha} \tag{3.11}$$

By assuming that x_D is constant as for the constant overhead policy and that x_B is a linear function of time, Eq. 3.10 can be rewritten to an integral which can be solved numerically a priori:

$$\overline{N_{min}} = \frac{\int_{x_{Bf}}^{x_{B0}} N_{min}(x_B)\,dx_B}{x_{B0} - x_{Bf}} \tag{3.12}$$

where

$$N_{min}(x_B) = \frac{\log\left(\frac{x_D^s}{1-x_D^s}\right)\left(\frac{1-x_B}{x_B}\right)}{\log\alpha} \tag{3.13}$$

and q is given by

$$q = \frac{\overline{N_{min}}}{N} \tag{3.14}$$

Equation (3.12-3.14) can by solved numerically and thereby provide a measure of the degree of difficulty of separation a priori. Note that Eq. 3.14 is independent of the amount of feed charged. Christensen and Jørgensen (1987), who first introduced this measure, used it in the study of waste cut recycling.

3.5.2 Results for comparison of cyclic and reflux policies

In the following we compare the simplified cyclic policy with the constant and optimal reflux policies. As already described in the simplified cyclic policy, we use the same condenser holdup $H_{C,k}$ and approach to equilibrium ϵ in each cycle. For the constant reflux policy the batch is started directly without an initial total reflux period. For the optimal reflux policy, a piecewise linear profile over two intervals is used with total reflux initially, $R(t_0) = 1.0$, in all cases.

The 32 examples studied are given in Table 3.5. We have varied the number of theoretical stages in the column N (5, 10, 15 or 20), the relative volatility α (1.25, 1.5 or 2), the feed composition x_F (0.15-0.85) and the product specifications for the distillate and the residual in the column, x_A^{spec} (0.9, 0.95 or 0.975) and x_R^{spec} (0.05 or 0.1). All other operating parameters are the same as the ones used in Section 3.3.1 (Table 3.1). The specification for the accumulated distillate holdup H_A^{spec} can be calculate from a total mass balance:

$$H_F x_F = H_A^{spec} x_A^{spec} + H_R x_R^{spec} \tag{3.15}$$

$$H_F = H_A^{spec} + H_R \tag{3.16}$$

The same assumptions for the dynamic model as in section 3.3.1 were used. Note that the example studied in the previous section is example 7 with $q = 0.85$. The results are presented in Table 3.6. The time saving is defined as:

$$\text{time saved } (\%) = \frac{t_{reflux} - t_{cyclic}}{t_{reflux}} \cdot 100\% \tag{3.17}$$

We have tried to correlate the time saving with the cyclic policy relative to the reflux policies with various parameters. The best single variables are found to be:

1. The degree of difficulty of separation, q
2. The feed composition x_F

In particular, we find that these two variables taken together give a good correlation with the time saved.

Time saving as a function of q

The time saved by using the simplified cyclic policy compared to the constant and optimal reflux policies is given as a function of q in Figure 3.8. From the plot is can be seen that the cyclic policy is better than the constant reflux ratio policy for most cases when the measure q is larger than about 0.7 and better than the optimal reflux policy when q is larger than about 0.8. However, the correlation is not unique. Still, it can be concluded that the time saved increases with increasing q value, that is, when the separation becomes more difficult. This is as expected because of the improved separation attainable under total reflux for the cyclic policy. Also the time saving is less compared to the optimal reflux policy than to the constant reflux policy which is obvious due to the larger number of degrees of freedom.

Table 3.5: Operating conditions for examples used in comparing the simplified cyclic policy and the constant and optimal reflux ratio policies (†: Example 7 is used in the study of the optimal cyclic policy in Section 3.3.2).

	Parameters			Specifications			Diff. of sep.
Example	α	N	x_F	x_A^{spec}	x_R^{spec}	H_A^{spec}	q
1	1.50	15	0.15	0.975	0.05	1.0811	0.91
2	1.25	15	0.45	0.900	0.10	4.3750	0.91
3	2.00	5	0.55	0.950	0.10	5.2941	0.90
4	1.50	10	0.80	0.975	0.10	8.0000	0.88
5	1.50	10	0.60	0.950	0.05	6.1111	0.85
6	1.25	20	0.45	0.950	0.10	4.1176	0.85
7†	1.50	10	0.25	0.900	0.10	1.8750	0.85
8	1.50	10	0.35	0.900	0.05	3.5294	0.83
9	1.25	15	0.65	0.900	0.05	7.0588	0.83
10	1.50	15	0.30	0.975	0.05	2.7027	0.82
11	2.00	5	0.40	0.900	0.10	3.7500	0.81
12	1.25	20	0.55	0.950	0.10	5.2941	0.80
13	1.25	15	0.65	0.900	0.10	6.8750	0.78
14	2.00	5	0.85	0.950	0.05	8.8889	0.78
15	1.25	20	0.40	0.900	0.05	4.1176	0.76
16	2.00	10	0.20	0.975	0.05	1.6216	0.74
17	2.00	10	0.15	0.975	0.10	0.5714	0.74
18	1.50	15	0.30	0.950	0.05	2.7778	0.71
19	1.50	10	0.50	0.900	0.10	5.0000	0.70
20	2.00	10	0.35	0.975	0.05	3.2432	0.68
21	1.50	15	0.30	0.950	0.10	2.3529	0.67
22	2.00	5	0.75	0.900	0.05	8.2353	0.65
23	1.50	15	0.45	0.950	0.05	4.4444	0.64
24	2.00	10	0.40	0.975	0.10	3.4286	0.63
25	2.00	10	0.25	0.950	0.05	2.2222	0.62
26	2.00	10	0.55	0.975	0.05	5.4054	0.61
27	2.00	10	0.30	0.950	0.10	2.3529	0.57
28	2.00	5	0.85	0.900	0.10	9.3750	0.56
29	1.50	15	0.65	0.950	0.10	6.4706	0.54
30	1.50	15	0.50	0.900	0.05	5.2941	0.51
31	2.00	15	0.35	0.975	0.10	2.8571	0.45
32	2.00	15	0.25	0.950	0.10	1.7647	0.41

Table 3.6: Results for the simplified cyclic policy, the constant and optimal reflux ratio policies for varying operating conditions or q values (†: Example 7 is used in the study of the optimal cyclic policy in section 3.3.2) ($*$: upper bound).

Exa-mple	q	Simplified cyclic policy N_{cyc}	$\epsilon \cdot 10^{-3}$	t_f hr	Const. reflux policy R_{opt}	t_f hr	% time saving	Opt. reflux policy t_f hr	% time saving
1	0.91	4	0.929	4.660	0.991	12.303	62.1	9.347	50.1
2	0.91	18	1.873	16.506	0.987	32.675	49.5	27.911	40.9
3	0.90	22	5.814	5.726	0.939	8.693	34.1	7.045	18.7
4	0.88	50$*$	3.530	10.527	0.938	12.858	18.1	8.608	-22.3
5	0.85	40	4.290	10.748	0.966	18.055	40.5	11.568	7.1
6	0.85	9	2.259	14.247	0.981	21.746	34.4	17.895	20.4
7†	0.85	4	6.538	4.045	0.967	5.734	29.5	5.322	24.0
8	0.83	11	5.945	7.706	0.970	11.813	34.8	9.284	17.0
9	0.83	50$*$	3.783	17.249	0.972	24.779	30.4	16.843	-2.4
10	0.82	4	2.480	6.861	0.975	10.662	35.7	7.760	11.6
11	0.81	8	15.740	3.476	0.904	3.895	10.8	3.430	-1.4
12	0.80	8	3.085	14.709	0.972	18.778	22.0	14.622	-0.6
13	0.78	50$*$	5.878	13.338	0.955	15.340	13.1	11.894	-12.1
14	0.78	50$*$	14.900	4.637	0.804	4.529	-2.4	2.766	-67.6
15	0.76	25	4.054	13.870	0.978	18.851	26.4	14.298	3.0
16	0.74	1	5.662	2.272	0.942	2.808	19.1	2.208	-2.9
17	0.74	1	9.167	0.771	0.937	0.900	14.4	0.730	-5.5
18	0.71	2	5.778	5.573	0.961	7.148	22.0	5.272	-5.7
19	0.70	5	13.286	5.946	0.919	6.140	3.2	5.084	-16.9
20	0.68	1	5.543	3.431	0.924	4.279	19.8	2.762	-24.2
21	0.67	1	8.629	3.618	0.940	3.889	7.0	3.405	-6.2
22	0.65	50$*$	30.050	3.966	0.775	3.653	-8.6	2.487	-59.5
23	0.64	2	5.915	6.732	0.948	8.491	20.7	5.518	-22.0
24	0.63	1	9.245	3.010	0.875	2.743	-9.7	2.144	-40.4
25	0.62	1	14.385	2.284	0.920	2.776	17.7	2.101	-8.7
26	0.61	50$*$	10.411	4.786	0.900	5.404	11.4	4.031	-18.7
27	0.57	1	20.135	1.919	0.869	1.796	-6.8	1.559	-23.1
28	0.56	50$*$	37.297	2.201	0.544	2.056	-7.1	1.512	-45.6
29	0.54	50$*$	13.347	6.172	0.892	5.975	-3.3	4.255	-45.0
30	0.51	50$*$	15.60	6.194	0.919	6.518	5.0	4.601	-34.6
31	0.45	1	12.25	2.558	0.860	2.035	-25.7	1.630	-56.9
32	0.41	1	24.11	1.440	0.861	1.267	-13.7	1.178	-22.3

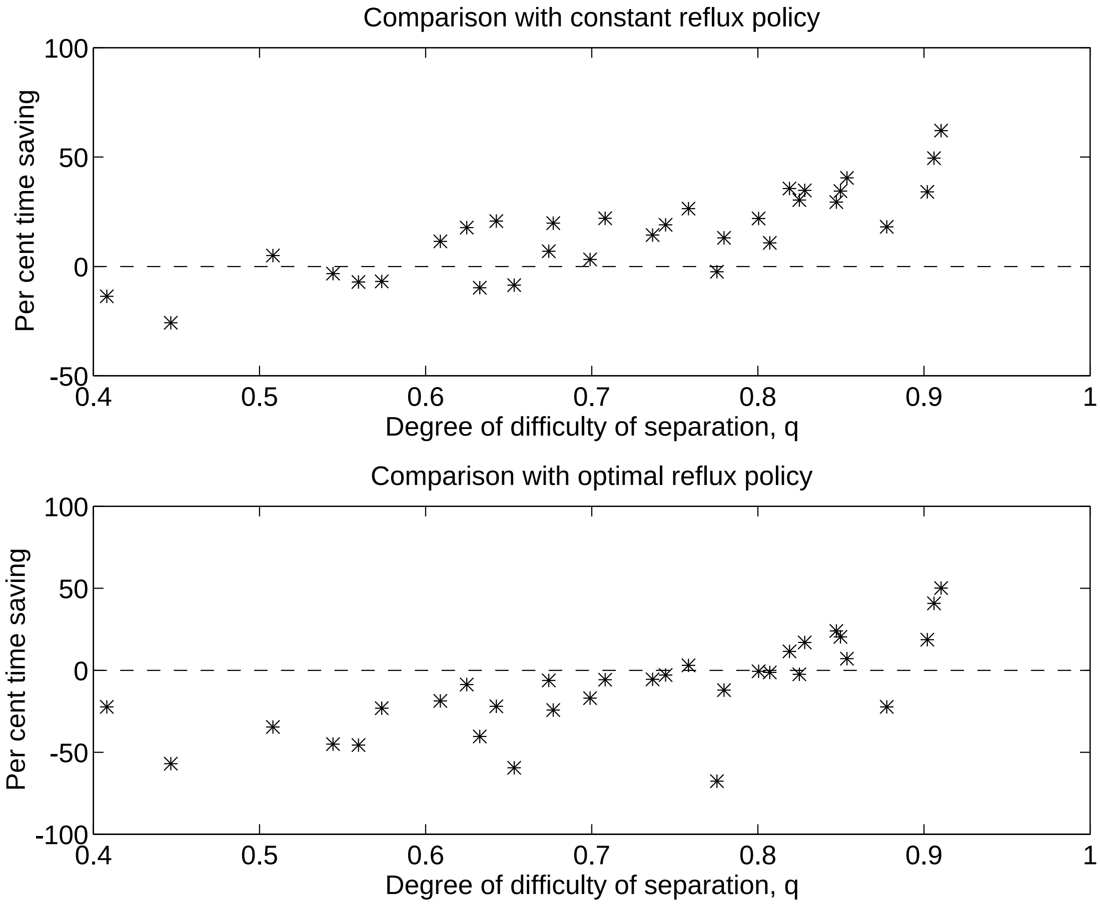

Figure 3.8: Per cent time saved as a function of the measure q by using the simplified cyclic policy compared to the constant and optimal reflux policies. (Data from Table 3.6.)

Time saving as a function of x_F

The time saved using the cyclic policy relative to the optimal reflux ratio policy is plotted in Figure 3.9. The correlation is far from unique, but the optimal reflux ratio is seen to be preferred for most mole fractions x_F larger than about 0.6.

Time saving as a function of q and x_F

The results in Figures 3.8-3.9 are combined in Figure 3.10 where we see that the combined effect of difficulty of separation, q, and feed composition, x_F, explains most of the time saving. From this plot it can be concluded that the cyclic policy is preferred over the optimal reflux policy for cases where q is high and x_F is small. In the opposite case, low q and high x_F, the optimal reflux policy is preferred.

3.5.3 Number of cycles in the cyclic policy

From Table 3.6 it can be seen that N_{cyc} varies considerably from only 1 cycle for some separations up to the maximum of 50 for others. We have tried to correlate the optimal

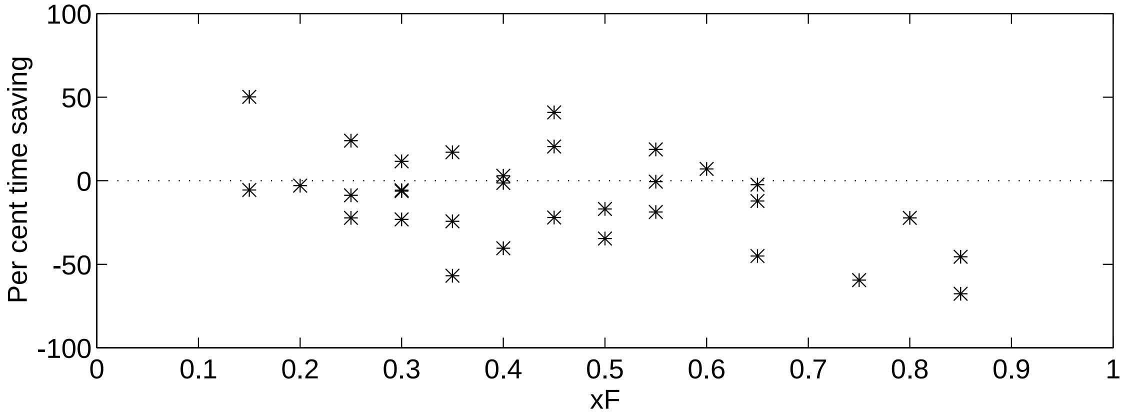

Figure 3.9: Per cent time saved as a function of x_F by using the simplified cyclic policy compared to the optimal reflux policy. (Data from Table 3.6.)

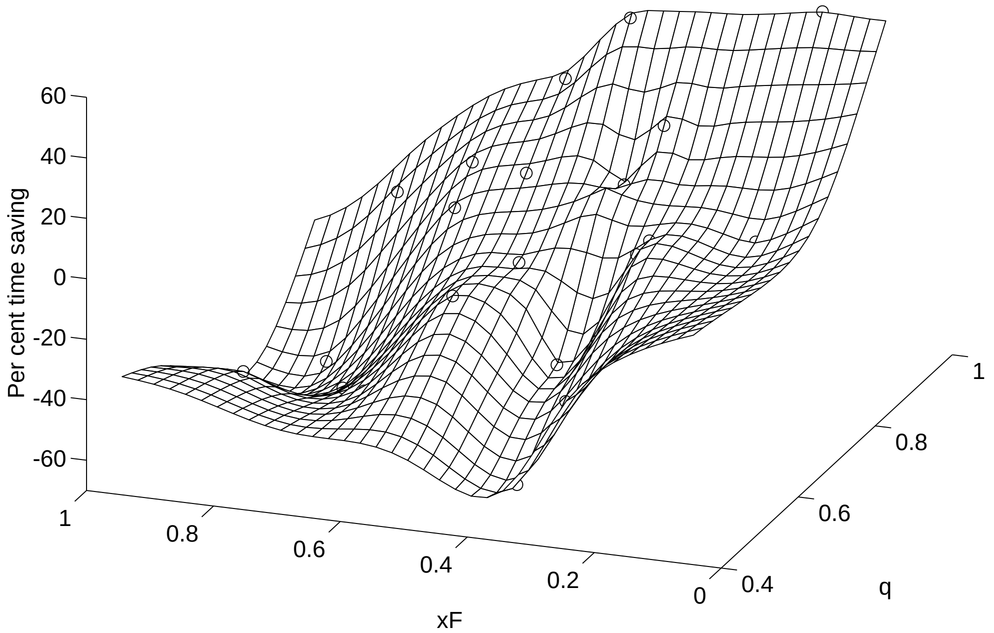

Figure 3.10: Per cent time saved as a function of q and x_F by using the simplified cyclic policy compared to the constant and optimal reflux policies. (Data from Table 3.6.)

N_{cyc} with various variables. The best correlation seems to be the feed composition x_F. From the plot in Figure 3.11 it can be seen that for high feed compositions $x_F > 0.6$, it is optimal to use many cycles (that is, the optimal reflux policy is preferred) whereas when only a small amount of light component is present, $x_F < 0.3$, it is optimal to use few cycles (typically 5 or less).

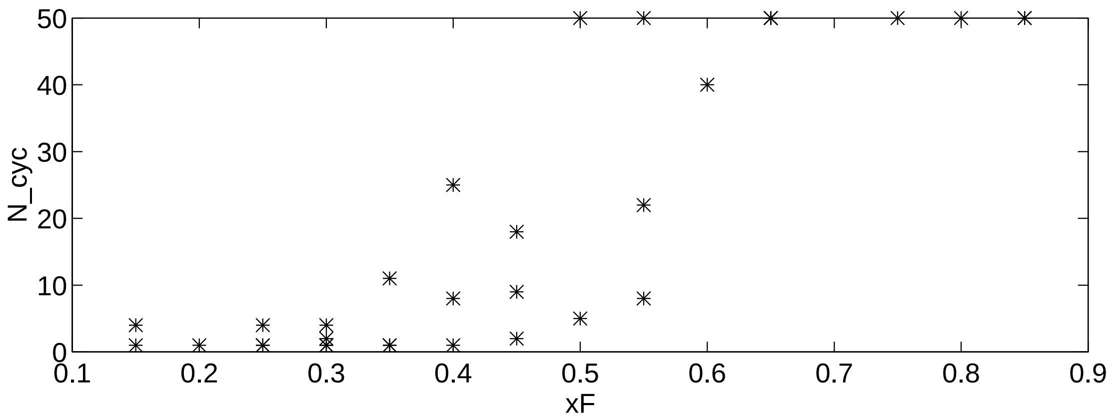

Figure 3.11: Number of cycles as a function of x_F for the simplified cyclic policy. (Data from Table 3.6.)

Obtaining the optimal number of cycles N_{cyc}. The optimal number of cycles N_{cyc} for each separation in Table 3.6 is found by running the simplified cyclic policy for different number of cycles until the minimum in terms of time consumption is found. The upper bound was in this study 50 cycles. An example of values for ϵ and total operating time as a function of number of cycles is given for Example 4 in Figure 3.12. For this example, the total operating time t_f decreases continuously when the number of cycles N_{cyc} is increased. The optimal number of cycles for this separation is larger than 50 cycles which was the upper limit in this study. However, the difference in operating time is reduced as N_{cyc} increases and the operating time with 20 cycles ($t_f = 10.77\ hr$) is only 2% longer than the operating time with 50 cycles ($t_f = 10.53\ hr$).

Some separations have 1 cycle as the optimum and others have an interior optimum in the interval $N_{cyc} \in [1, 50]$. The parameter ϵ usually increases with N_{cyc}. This is because as the number of cycles is increased, the need for maximum separation in each cycle is decreased and the total reflux period can be stopped earlier or at higher ϵ-values.

The optimal cycle time for the simplified policy. With the simplified cyclic policy the total reflux time, and thereby the cycle time, for each cycle is determined by the parameter ϵ. As the column is gradually depleted of the light component it will take increasingly longer to establish equilibrium in the column. This is illustrated in Figure 3.13 where the cycle time is given as a function of the cycle number and the corresponding amount of accumulated product as a function of operating time for Example 8 which has 11 cycles as optimum.

3.5.4 Summary

We have found that the cyclic policy can be recommended for difficult separations with small amounts of product recovered ($q > 0.7 - 0.8$ and $x_F < 0.6$). The optimal number of cycles is found to depend on the feed composition x_F. Using a large number of cycles (typically larger that 5-10) is found to be optimal for separations where $x_F > 0.5$ but

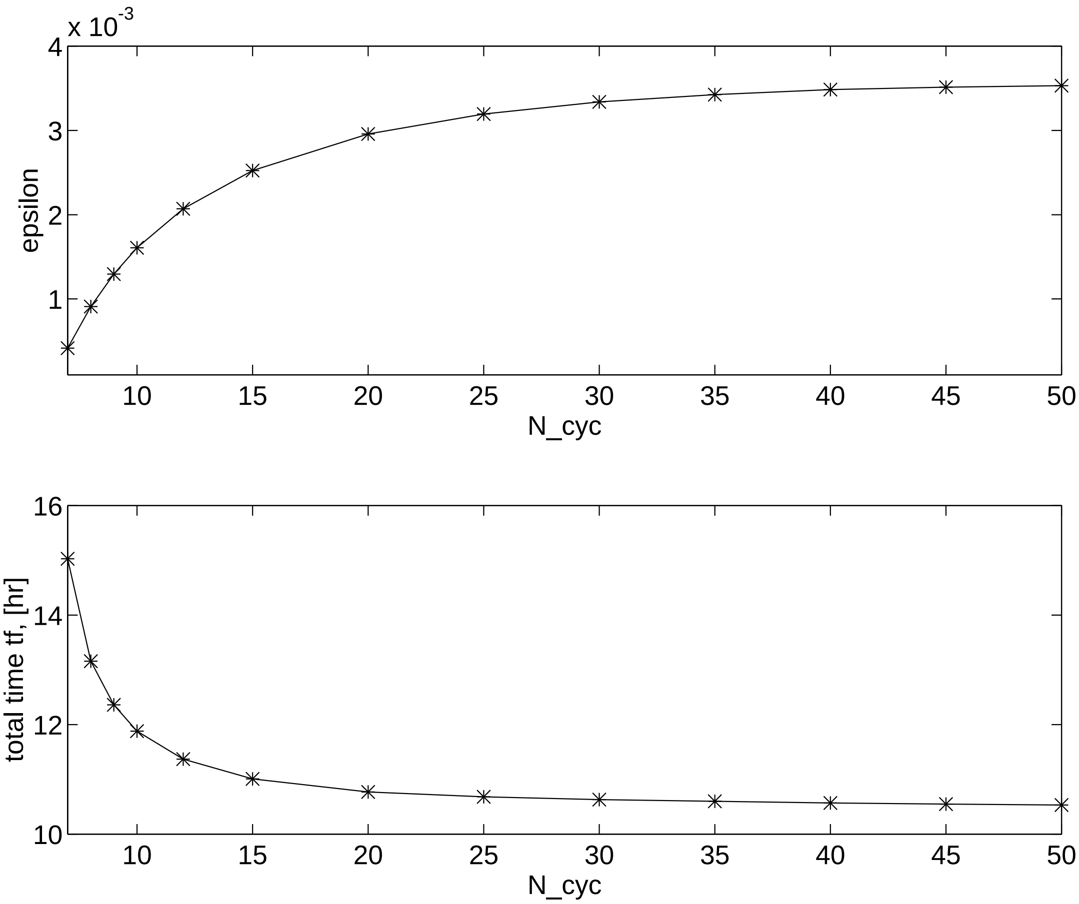

Figure 3.12: The parameter ϵ and the total operating time t_f as a function of number of cycles N_{cyc} for the simplified cyclic policy (Example 4: q=0.88).

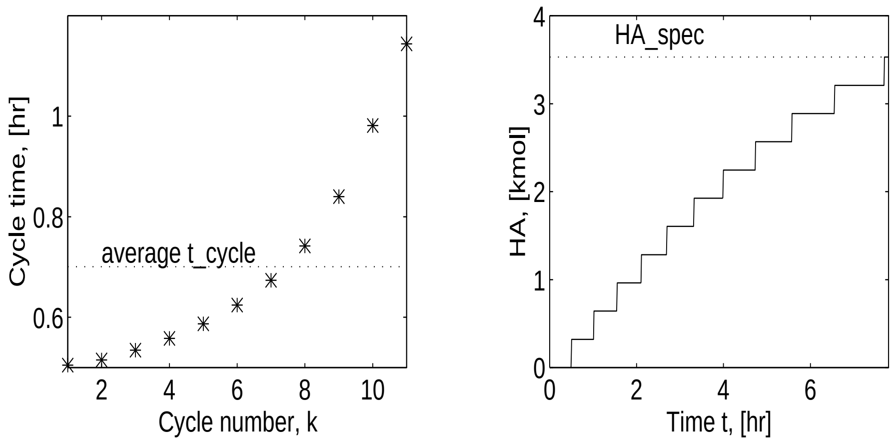

Figure 3.13: Cycle time t_{cyc} as a function of cycle number k and amount of accumulated product H_A as a function of operating time t. (Example 8: q=0.83).

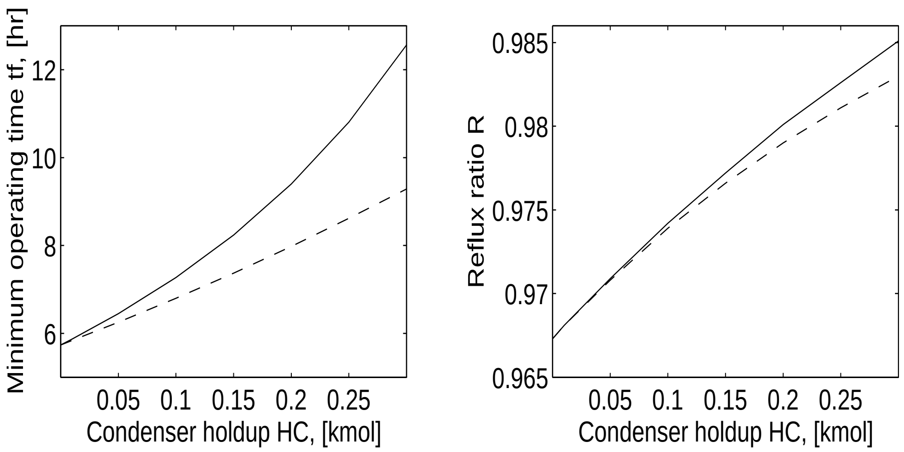

Figure 3.14: Optimal operating time t_f and corresponding constant reflux ratio R as a function of condenser holdup H_C. —-: the condenser holdup is drained to the reboiler and - - -: the condenser holdup is drained to the accumulator (Example 7: q=0.85).

will not be practical in real operation unless the operating time is long since it will be inconvenient to dump the condenser too often.

3.6 Effect of condenser holdup

In the results presented above, the condenser holdup was assumed negligible for the reflux policies. We find that a finite holdup will increase the operating time for these policies. The role of holdup on the performance of binary batch distillation was investigated by Mujtaba and Macchietto (1991) who mainly considered the effect of column tray holdup using constant and optimal reflux policies. Of more practical interest is the effect of condenser holdup. Mujtaba and Macchietto (1991) concluded that the condenser holdup should always be kept to a minimum. The reason was that for a total condenser, holdup only acts as an accumulator of material and not as a separation stage. According to Mujtaba and Macchietto, a larger condenser holdup means a longer operating time is always required to achieve a given separation using these policies.

However, in their study, Mujtaba and Macchietto (1991) assumed that the condenser drum holdup was drained back to the reboiler at the end of the separation. A second alternative, which seems more reasonable in practical cases, is to drain the condenser drum to the accumulator such that the condenser holdup becomes part of the accumulated product.

In Figure 3.14 we have plotted the minimum operating time and the reflux ratio as a function of condenser holdup for Example 7 using the constant reflux policy. Note that the internal column holdup is constant, $\sum^N H_j = 0.1\ kmol$. As expected from the results of Mujtaba and Macchietto (1991), the effect of condenser holdup is significant, and increasing H_C from 0 to 0.3 $kmol$ (from 0 to 3% of initial feed charge) results in an increase in the minimum operating time of 62 % when the condenser is drained to the accumulator and of 119 % when we follow Mujtaba and Macchietto and drain to the

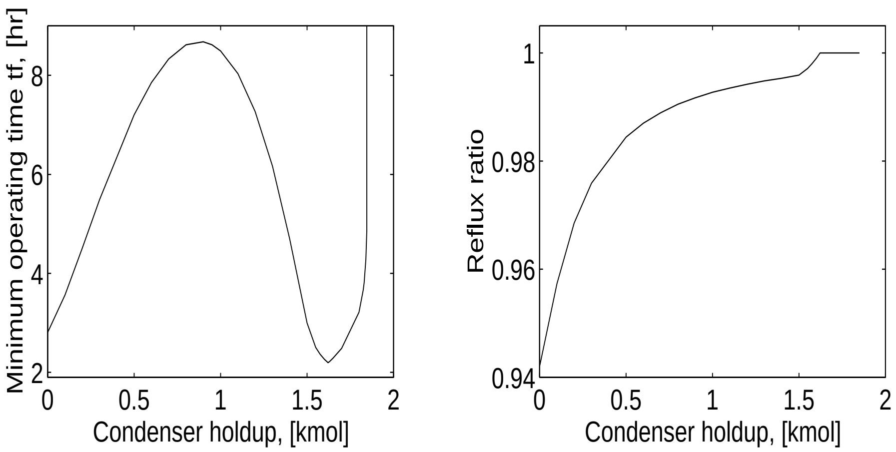

Figure 3.15: Optimal operating time t_f and corresponding constant reflux ratio R as a function of condenser holdup H_C. (Example 16: q=0.74).

reboiler. Thus, the increase is larger when the condenser holdup is drained to the reboiler. This is reasonable since the separation of the material in the condenser is "wasted" when the condenser is drain back to the reboiler at the end of the batch.

However, there are cases when a larger condenser holdup will in fact yield a shorter operating time. Consider Figure 3.15 where the operating time is given as a function of condenser holdup for Example 16 using a constant reflux ratio policy. (Recall from Figure 3.6 that for this example, the *cyclic policy* with one interval is the optimal operating policy.) We see from Figure 3.15 that for small holdups, the minimum operating time increases with increasing condenser holdup. However, for large holdups the operating time decreases and indeed reaches a minimum at a holdup equal to the holdup in the cyclic policy ($H_C = H_A^{spec}$). As expected, the operating time with this holdup is smaller than with a negligible condenser holdup. For even larger condenser holdups, the operating time increases again and approaches infinity as it becomes infeasible to meet the product specification.

The right plot in Figure 3.15 shows the corresponding optimal reflux ratio R. As expected, $R = 1$ (total reflux) when $H_C \geq H_A^{spec}$.

3.7 The cyclic policy in practice

No design changes are necessary for the column in order to run it according to the cyclic policy except that a larger condenser drum might be needed. One alternative is to use two condenser drums as mentioned in Section 3.3.2, such that the filling of the second is started at the same time as the dumping of the first one.

The main practical problem with the cyclic policy is that the feed composition must be known quite exactly since the amount of accumulate product H_A^{spec} is calculated based on this value. If H_A^{spec} is too low, less product is recovered. However, if H_A^{spec} is too large, it may not be possible to meet the specifications and the operating time goes to infinity as illustrated in Figure 3.15. A good estimate of the feed composition is therefore required. The tolerance for errors will depend on the purity specifications.

In practice, the best operating procedure would be to adjust the condenser holdup on-line based on e.g. measurements of distillate composition as a function of time. If the purity is lower than expected, the condenser holdup may be reduced and vice versa. In particular, the condenser holdup may be decreased towards the end to ensure that the specifications are met.

It is also possible to combine different operating policies. The column can be run with the cyclic policy for e.g. $N_{cyc} - 1$ cycles and then with a reflux policy after the last cycle until the specifications are met.

Once the condenser holdup has been fixed, several alternatives for the practical implementation of the cyclic policy may be suggested:

1. "Open-loop" operation where the times for each cycle $t_{cycle,k}$ (obtained from optimization) is specified. (Alternatively, an average cycle time $t_{cycle,average}$ can be used). This mode of operation is, however, very sensitive to changes and disturbances.

2. "Feedback" operation where the cycle is stopped when a given composition or temperature (obtained from optimization) is achieved.

3. One may consider a "feedback" operation where the cycle is stopped when a given approach to equilibrium is achieved. Since it may be difficult to measure $\epsilon = |x_1 - y_2|$ as we have defined it in this paper, it is probably better to stop when $dT_i/dt < \epsilon_T$. In words, when a temperature change in the column is less than a given bound. In this case, it is important to select a sensitive temperature (eg. a temperature in the middle of the column).

"Open-loop" operation is not recommended due to the high sensitivity to disturbances and changes in operating conditions. "Feedback" operation based on an optimal distillate composition profile is probably the best way of implementing the cyclic policy since this procedure ensures a high product purity, however, possibly at the expense of a longer operating time (this has been found to be the case in simulations).

3.8 Conclusions

In this paper we have presented the cyclic operating policy in detail and compared it with the conventional operating policies for a number of separations. The cyclic policy was found to be favorable for difficult separations where a small amount of product is to be recovered. Normally, batch columns are designed with $N \approx 2N_{min}$ ($q \approx 0.5$) and the constant reflux policy will be the most favorable. Batch columns are nevertheless often used for separations for which they are not designed ($q > 0.5$) and the cyclic policy may be favorable for these separations.

Notation

D	distillate flow	$kmol/hr$
H_A	liquid holdup in the accumulator	$kmol$
H_B	liquid holdup in the reboiler	$kmol$
H_C	liquid holdup in the condenser	$kmol$
H_F	amount of feed charge	$kmol$
H_j	liquid holdup on tray j	$kmol$
H_j^o	initial liquid holdup on tray j	$kmol$
H_R	liquid holdup in the residue in column and reboiler	$kmol$
K_P	gain for PI-controller	
L	reflux flow	$kmol/hr$
L_j	liquid flow from tray j	$kmol/hr$
L_j^o	initial liquid flow from tray j	$kmol/hr$
N	number of trays in the column section	
N_{cyc}	number of cycles in the cyclic policy	
N_{min}	minimum number of trays	
q	a measure for the degree of difficulty of separation $= N_{min}/N$	
R	internal reflux ratio, $= L/(L+D)$	
t	time	hr
t_0	initial time	hr
t_f	total operating time	hr
$t_{fill,k}$	filling period for cycle k	hr
$t_{totalreflux,k}$	total reflux period for cycle k	hr
$t_{dump,k}$	dumping period for cycle k	hr
$t_{cycle,k}$	cycle time $= t_{fill,k} + t_{totalreflux,k} + t_{dump,k}$	hr
V_j	vapor flow from tray j	$kmol/hr$
V_N	vapor flow from the reboiler	$kmol/hr$
x_A	mole fraction of light component in accumulator	
x_B	mole fraction of light component in reboiler	
x_D^s	set point for the mole fraction of light component in distillate flow	
x_F	mole fraction of light component in feed charge	
x_j	mole fraction of light component in liquid on tray j	
x_R	mole fraction of light component in residual in column/reboiler	
y_j	mole fraction of light component in vapor on tray j	
Greek letters:		
α	relative volatility	
ϵ	approach to equilibrium	
τ	hydraulic time constant	hr^{-1}
τ_I	integral time for PI-controller	hr
Scripts:		
k	cycle number	
^{spec}	specified value	
^{o}	initial value	

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Chapter 4

Comparison of Regular and Inverted Batch Distillation

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Abstract

Several new alternative strategies have been suggested to increase the productivity of batch distillation. One is the use of inverted columns where the feed is charged to the condenser drum instead of to the reboiler as for regular columns. In this paper we discuss the differences in dynamic behavior between a regular and an inverted batch column. Optimal results in terms of minimum operating time for separations in both columns are considered. Intuitively, one may think that an inverted column is best for separations with large amounts of light component in the feed. Surprisingly, we find the opposite: the inverted column is better in terms of less operating time when light component is present in only small amounts. The chapter concludes with a discussion of "inverted" separations; removal of light component in a regular column versus removal of heavy component in an inverted column.

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4.1 Introduction

Batch distillation is a very old unit operation in the chemical industry. During the last century, continuous distillation took over most of the processes where batch distillation was originally used. However, batch processes are becoming increasingly important again as a result of the expansion in the fine chemical and pharmaceutical industries. These industries are characterized by small amounts of products with high added value. Batch distillation has the advantage of being able to separate a number of products in a single column. A batch column can handle 1) multicomponent mixtures, 2) a wide range of feed compositions and 3) a varying degree of difficulty of separation (wide range of relative volatilities and product purities). The demand for efficiency and productivity is continuously increasing. However, the optimal operation is limited by the given operating strategy and the process equipment. Therefore there is a need for new or alternative strategies which can increase the productivity.

The use of inverted columns or "regular batch columns turned upside down" has been suggested earlier in the literature. In the inverted case, the feed is charged to the condenser drum and further continuously added to the top of the column. The liquid from the bottom of the column is vaporized in the reboiler and a fraction is removed as product. The products are taken out with the heaviest component first, then the second heaviest etc. The true inverse of a regular batch column requires both the feed charge stored in the condenser drum and the product taken out, to be in vapor phase which is very unlikely. It is therefore normally assumed that the feed and the product are in liquid phase. However, this configuration is not the true inverse of the regular column as we will show in this paper.

The use of an *inverted*, or stripping, column was originally proposed by Robinson and Gilliland (1950). They stated that the main advantage with this configuration was that the most volatile components would be collected in the condenser drum in high purity. They briefly discussed the possibility of running the column first in a normal batch fashion to remove the lighter components and then inverted to remove the heavier constituents. They also discussed a combined operation where the feed was added to the middle of the column and the light and heavy components were taken off simultaneously over the top and bottom respectively.

The combined operation was studied by Hasebe at al. (1992) who denoted this column configuration a *complex column*. They also discussed the difference between regular and inverted columns. They claimed that a regular column always has a better separation efficiency than an inverted column if the separation conditions are the same and the relative volatility is constant ("inverted" separations). The conclusion was based on a comparison between the equilibrium curves and operation lines for a regular and an inverted column with all other conditions equal.

Mujtaba and Macchietto (1992) discussed use of a complex column to improved the operation of reactive batch distillation. They found that this column configuration improved conversion and product yield significantly when the reaction products had two extreme boiling points (highest and lowest in the reaction mixture). The use of an inverted column for cases where the reaction product had a higher boiling point than the reactants was suggested but no examples where given. Mujtaba and Macchietto (1994) discussed use of inverted and complex columns for an example with reactive batch distillation. For this example the inverted column gave a lower conversion than the regular column. This was explained in terms of the difference in relative volatility between the heavy components compared to between the light ones.

Chiotti and Iribarren (1991) presented simplified models for binary batch distillation in both regular and inverted columns. They included an intermediate cut which was recycled to the next feed charge. Their models were based on the assumption of pseudo-steady-state in the column. Numerical integration was thereby avoided. They stated that the inverted column was more economical than the regular one for heavy products of high purity. They presented two numerical examples where they optimized the total annual cost for two separations in both an inverted and a regular column. Chiotti *et al.* (1993) extended these models to multicomponent mixtures. They also considered columns which could be used for both rectification (regular column) and stripping (inverted column) where the products were taken off both at the top and bottom of the column in given sequences. With their very simplified models they found that, for a ternary mixture, the inverted column was optimal when the purity specification for the heavy product was high. It should be noted that both in this paper and in the previous one, the optimal number of trays in the column was in most cases higher for the inverted column than for the regular one.

To the best of our knowledge, no comparison between regular and inverted columns in terms of optimal operation has been given in the literature so far.

In this paper, we will first briefly discuss the differences in dynamic behavior between a regular and an inverted batch column for separations with different feed compositions. Thereafter we consider the optimal operation of both columns in terms of minimum operating time for the same separations. Intuitively, one may think that an inverted column is best for separations with a large amount of light component in the feed, because one may somehow avoid to "boil off" this large amount. However, as we shall see the opposite conclusion is true: the inverted column is preferable to the regular column, in terms of shorter operating time, when the light component is present in small amounts in the feed mixture. At the end of the paper, we will discuss the operation of the two columns for "inverted" separations (removal of light component in a regular column versus removal of heavy component in an inverted column).

4.2 Comparison of dynamic behavior under total reflux

In this section we compare the behavior of an inverted and a regular column in terms of dynamic responses and rate of approach to equilibrium under total reflux operation (startup). The columns are shown in Figure 4.1. Note that the feed and the product are in liquid phase also in the inverted column. The ideal inverted column with feed and product in vapor phase is discussed at the end of this paper.

We will here compare the behavior when the same separation (in terms of feed composition x_F) is performed in both columns. "Inverted" separations ($x_{F,light}$ in the regular

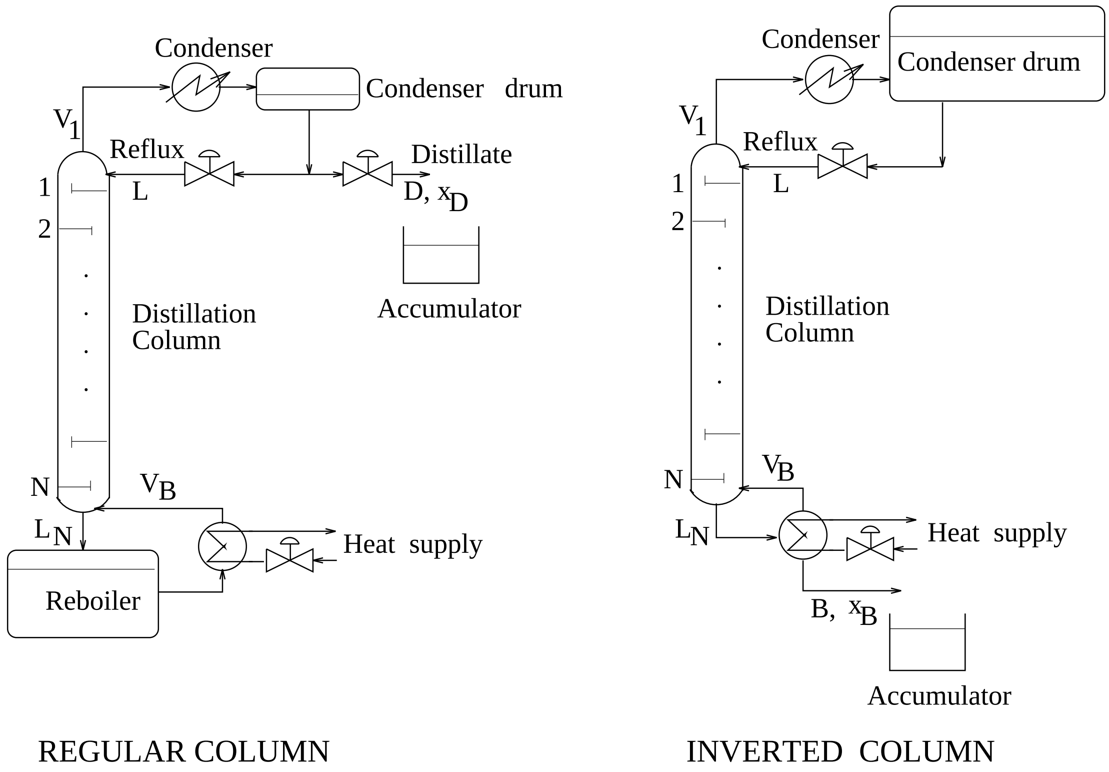

Figure 4.1: A regular and an inverted batch column.

Table 4.1: Operating parameters.

Column	N	α	V_B $kmol/hr$	H_F $kmol$	H_j $kmol$	H_B $kmol$	H_C $kmol$
Regular	10	2.0	10.0	10.0	0.001	$H_F - NH_j - H_C$	0.01
Inverted	10	2.0	10.0	10.0	0.001	0.01	$H_F - NH_j - H_B$

column equal to $x_{F,heavy}$ in the inverted column) will be discussed later. We will denote the first component taken off as the "withdrawn" component; i.e. light in the regular column and heavy in the inverted column. The "withdrawal" end of the column is the top for the regular column and the bottom for the inverted column. Mole fractions are given for the light component unless specifically stated otherwise.

4.2.1 Example studied

A binary mixture with constant relative volatility is assumed. The operating parameters are given in Table 4.1. The dynamic models are given in Appendix. SPEEDUP (1993) is used for the simulations. In the regular column, the feed is charged to the reboiler and in the inverted column to the condenser drum. In both cases the feed is assumed to be liquid at its boiling point. The time and energy input to heat the feed charge to its boiling point is neglected. In any case this time is the same for the two columns since the feed is in liquid phase in both cases, but for an ideal inverted column the feed charge has to be completely evaporated. Initially, the column is assumed filled with liquid at its boiling point with a composition equal to that of the feed. (This assumption is not important for the results, see Chapter 2 of this thesis.) It is assumed that the vapor flow V_B can be manipulated directly.

4.2.2 Results

We have considered three different cases for startup under total reflux:

1. The light component is present in only a small amount, $x_F = 0.1$
2. The feed is equimolar, $x_F = 0.5$
3. The light component is present in a large amount, $x_F = 0.9$

The total reflux responses for the light component are shown for the three cases in Figure 4.2. We see that in the regular column, the light component will be transported upwards and accumulates in the top whereas in the inverted column the heavy component is transported to the bottom. Note that the time to reach equilibrium is very short due to the low column and condenser holdups in this example.

In both cases, the dynamic responses are fastest in the withdrawal end of the column. That is, in the top of the regular column and in the bottom of the inverted column. Note that the initial responses in Figure 4.2 are equal but in opposite direction for the top and bottom of the columns ($dx_D/dt\ |_{t=0}$ in the inverted column equal to $dx_B/dt\ |_{t=0}$ in the regular column and vice versa).

For the case with equimolar feed, $x_F = 0.5$ (middle plots), the responses for the regular and the inverted column are very similar but not quite equal in terms of being mirror images of each other. This is because the inverted column studied here is not the true inverse of the regular column since the feed is in liquid and not in vapor phase. (This is discussed in Section 4.4.)

Rate of approach to equilibrium. Next consider the rate of approach to equilibrium. For this comparison the responses are best shown in terms of normalized variables:

$$x^{norm}(t) = \frac{x(t) - x^o}{x^{ss} - x^o} \tag{4.1}$$

From Figure 4.3 we see that both columns reach equilibrium slowest when the amount of "withdrawn" component is small. (For instance when $x_F = 0.1$ in the regular column and $x_F = 0.9$ in the inverted column). The reason for this is that it takes a long time for the small amount of the "withdrawn" component to moved from the feed end through the column, which is filled mostly with the other component, to the withdrawal end.

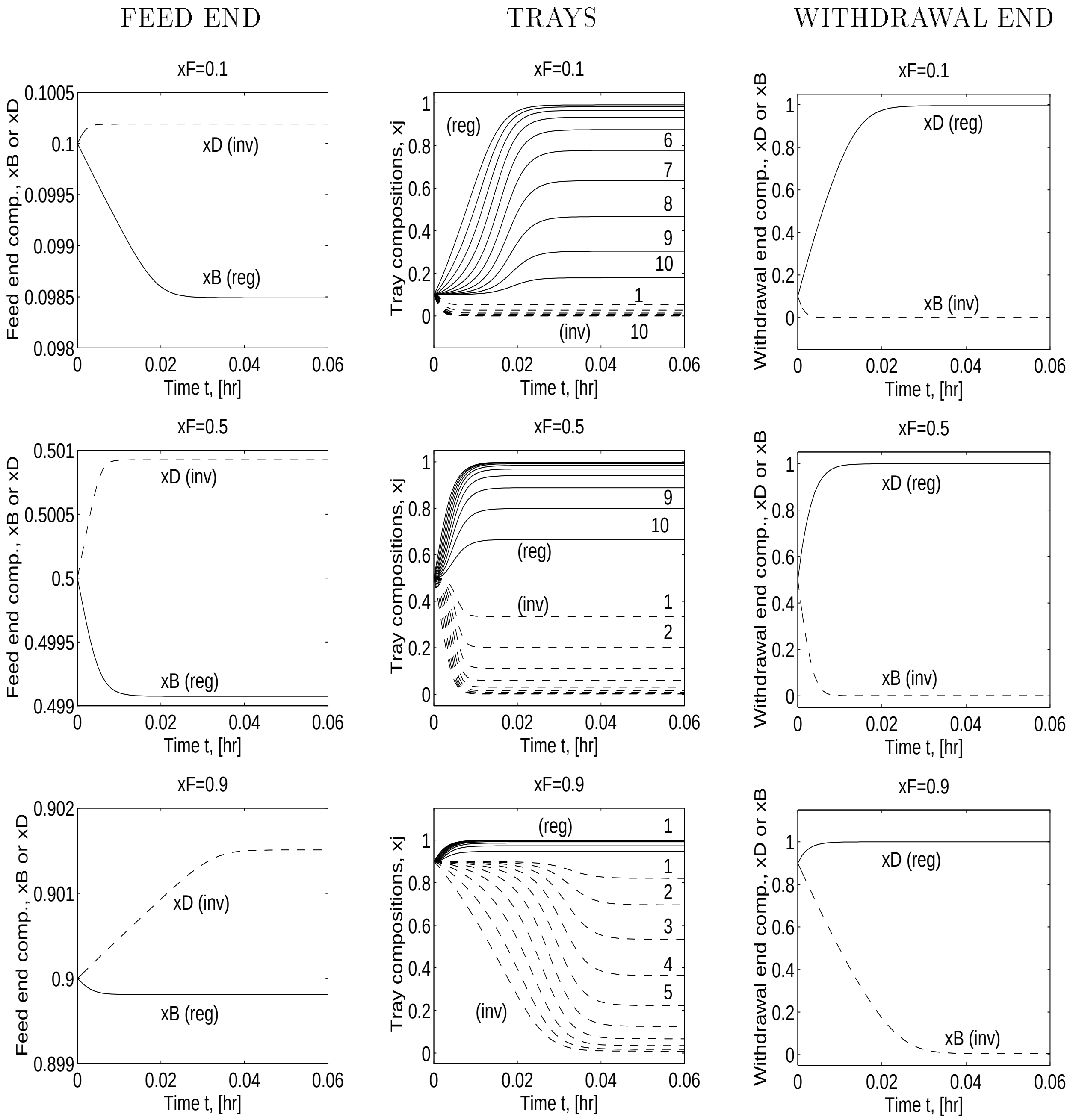

Figure 4.2: Time responses of compositions for regular and inverted column under total reflux.

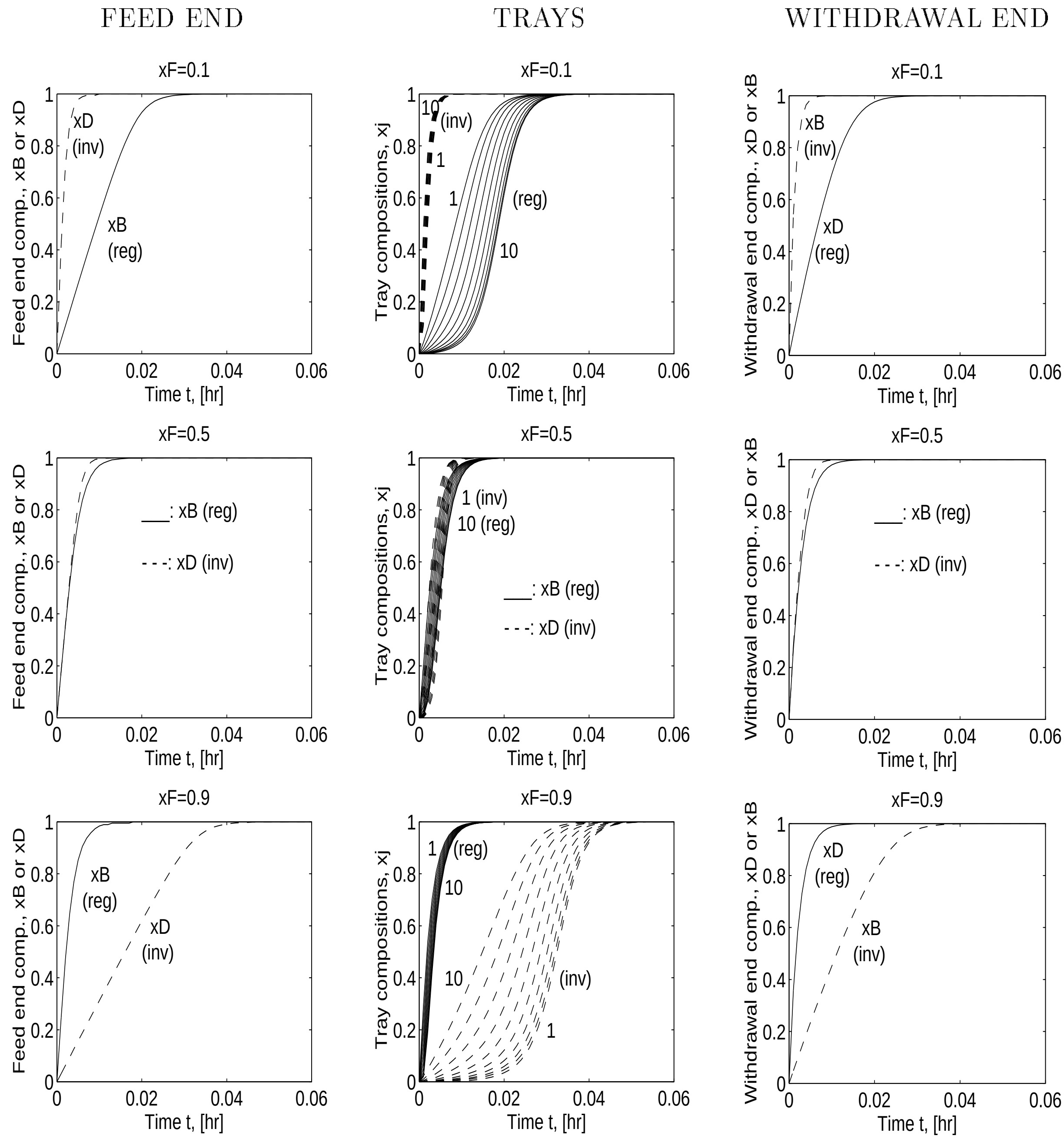

Figure 4.3: Time responses for normalized compositions for regular and inverted column under total reflux.

4.3 Optimal operation

In this section, we consider the optimal operation in both a regular and an inverted column in terms of minimum operating time for given separations. We will show that the inverted column is in fact better than the regular column for separations where the light component is present in a small amount in the feed mixture.

4.3.1 The minimum time problem

The optimal operation in terms of minimum operating time can be stated as:

$$\min_{R,R_B} t_f \tag{4.2}$$

subject to

$$x_A \geq x_A^{spec} \tag{4.3}$$
$$x_R \geq x_R^{spec} \tag{4.4}$$

In words, find the optimal reflux ratio R (for the regular column) or reboiler ratio R_B (for the inverted column) which minimizes the operating time t_f subject to constraints on the compositions of accumulated light and heavy product. In the following examples, we have selected to use the same purity specifications for the two products, that is

$$x_A^{spec} = 1 - x_R^{spec} = x^{spec} \tag{4.5}$$

The optimization parameters R and R_B are assumed piecewise continuously linear over two intervals giving a total of 5 optimization variables ($R(t_0)$, $R(t_1)$, $R(t_f)$, t_1 and t_f). The optimization program DAEOPT developed at Imperial College, London (Vassiliadis, 1993) is used. It should be noted that since more than one optimization variable is involved, several local minima may exist.

4.3.2 Results

We consider 13 cases with different values for the feed composition and product specifications. Other operating parameters are as given before (Table 4.1). The minimum operating time is presented for the 13 cases in Table 4.2 and shown graphically in Figure 4.4. The corresponding optimal reflux and reboiler ratios are shown for three of the cases in Figure 4.5 and details on how the compositions change with time are shown in Figure 4.6 for one case.

From Table 4.2 and Figure 4.4 we see that the inverted column has indeed a shorter operating time than the regular column for some separations. More surprisingly, the inverted column is better for cases where the products are to be recovered at high purity from a feed *low in light component* whereas the regular column if found to be best for cases where the products are to be recovered at high purity from a feed *rich in light component*. The regular column is found to be better than the inverted column for all symmetrical separations, $x_F = 0.5$.

Table 4.2: Minimum operating time t_f for varying purity specifications x^{spec} for regular and inverted column (Same purity specification x^{spec} for both products).

Purity specification x^{spec}	Minimum operating time inverted column t_f, hr	regular column t_f, hr
Feed composition $x_F = 0.1$		
0.925	1.043	**0.409**
0.950	1.369	**1.024**
0.975	**2.264**	2.940
0.980	**2.532**	4.448
0.990	**4.041**	>100
Feed composition $x_F = 0.5$		
0.925	3.022	**2.200**
0.950	3.807	**2.610**
0.990	10.010	**5.905**
Feed composition $x_F = 0.9$		
0.925	**0.709**	1.248
0.950	1.791	**1.630**
0.975	4.691	**2.410**
0.980	7.856	**2.462**
0.990	>100	**3.192**

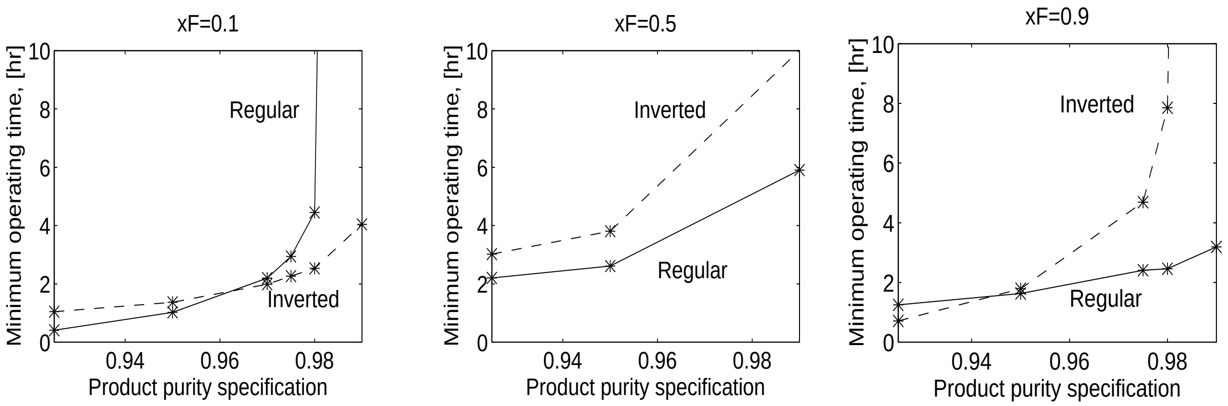

Figure 4.4: Minimum operating time t_f as a function of purity specifications x^{spec} for a regular and an inverted column.

However, more importantly, for some specifications the inverted column gives a moderate operating time when the separation is not even possible in the regular column ($x_F = 0.1$ and $x^{spec} = 0.999$).

The reason why the inverted column is best when x_F is small is, as shown previously in Figure 4.3, that it takes a long time to "move" a small amount of light component from

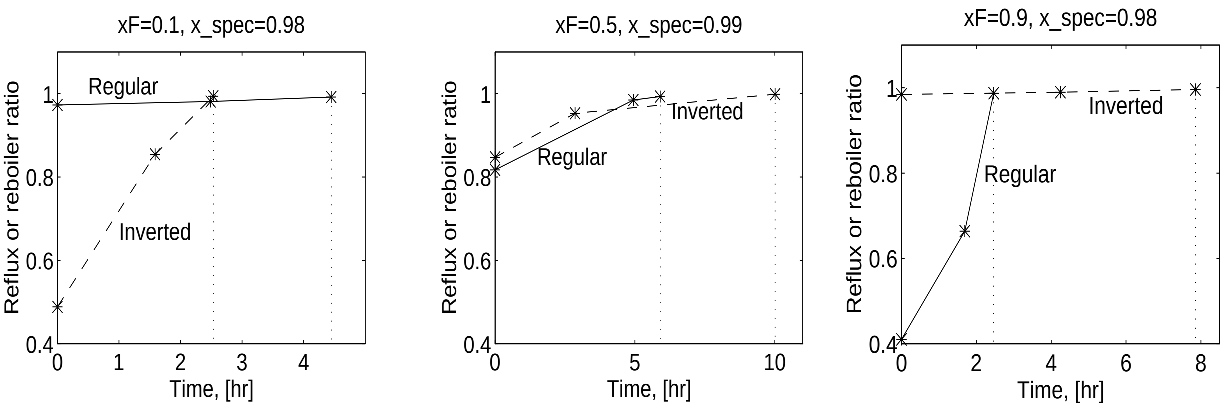

Figure 4.5: Optimal reflux ratio R and reboiler ratio R_B.

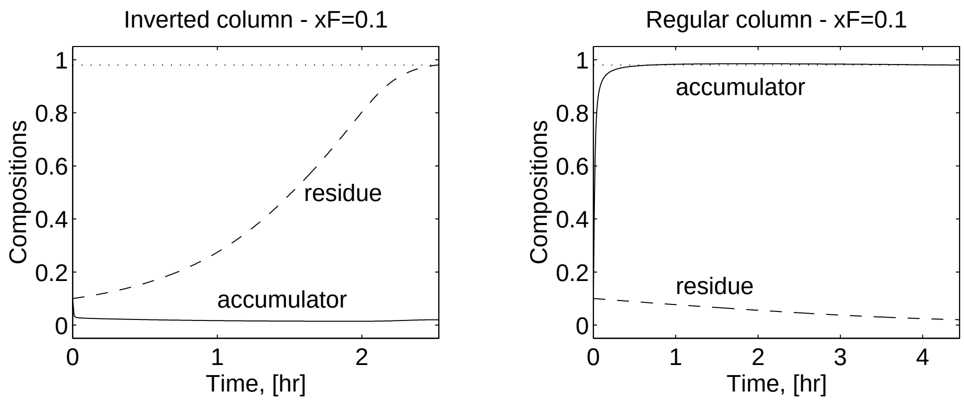

Figure 4.6: Composition of product in accumulator or residue ($x_F = 0.1$ and $x^{spec} = 0.98$.)

the reboiler to the top of the column when a regular column is used. In some cases the separation is not even possible in a regular column.

The reflux and reboiler ratios in Figure 4.5 are very close to 1 for separations where a small amount of product is to be withdrawn from the column leaving a large amount of the other product as residue (e.g. $x_F = 0.1$ in the regular column and $x_F = 0.9$ in the inverted column). In the opposite case, where a large amount is withdrawn, the ratios are low initially but increase with time as the column is gradually depleted of the "withdrawn" product (e.g. $x_F = 0.9$ in the regular column and $x_F = 0.1$ in the inverted column).

The compositions of the products in the residual and the accumulator are given in Figure 4.6 for a case when the inverted column is best ($x_F = 0.1$ and $x^{spec} = 0.98$). For the regular column, the accumulator composition is slightly higher than the specification during most of the run and reaches the specified value at the final time. In the inverted column, the composition in the residue is slowly increasing as the heavy component is removed in the bottom.

4.3.3 Practical considerations

For cases with a small amount of light component, when an inverted column generally is optimal, the inverted column may also be an easier solution in practice because the end of the batch can be easily detected from temperature measurements. The temperature in the residue will decrease steadily during the batch (see Figure 4.6). For the same separation in the regular column, the end of the batch is difficult to detect based on temperature measurements at the ends of the column alone.

However, there are two main disadvantages with the inverted column. The first is that the reboiler holdup must be very low since the reboiler holdup in an inverted column has the same effect on operating time as the condenser holdup in a regular column, i.e. the operating time will normally increase with reboiler holdup (see Chapter 3). A sufficiently low holdup may be difficult to achieve in practice. Secondly, the holdup in the reboiler must be controlled using the reflux flow from the condenser drum as manipulated variable since the liquid flow from the column section to the reboiler can not be controlled directly. (In our simulations the liquid flow dynamics were neglected and the level control was assumed perfect.)

4.4 The performance of "inverted" separations

So far we have considered the separation of the same feed mixture in a regular and an inverted column. The problem one is faced with in practice it the decision as to which column configuration is the best for a given feed mixture. Nevertheless, in this section we will compare what we denote *"inverted" separations*: the removal of light component from a regular column compared to the removal of heavy component from an inverted column or vice versa. The feed compositions are mirror images of each other x_F(regular column)$=1-x_F$(inverted column). This is done in an attempt to more fully understand the behavior of inverted columns.

Let us first go back to the optimal results in the previous section. By comparing the shape of the plots in Figure 4.4 it can be seen that the separation in the inverted column (dotted line in left plot) has a similar operating time profile as the separation in the regular column (solid line in right plot). The same comparison can be made for the reverse case. Also, the optimal reflux and reboiler ratios have a similar shape and value for "inverted" separations (dotted line in left plot similar to dotted line in right plot in Figure 4.5). Nevertheless, the operating time for "inverted" separations have different values, for example:

$$\begin{array}{lll} \text{Inverted column:} & t_f = 2.264\ hr & \text{for } x_F = 0.1 \text{ and } x^{spec} = 0.975 \\ \text{Regular column:} & t_f = 2.410\ hr & \text{for } x_F = 0.9 \text{ and } x^{spec} = 0.975 \end{array}$$

In the following we will compare the regular and the inverted column for "inverted" separations in terms of dynamic behavior under total reflux and during production. We will argue that the difference is due to the fact that both the feed and the product are in liquid and not in vapor phase in the inverted column. The inverted column studied so far is therefore not the true inverse of the regular column.

4.4.1 Comparison of dynamic behavior under total reflux

In Figure 4.7 we have plotted the dynamic responses in composition of the "withdrawn" component for "inverted" separations corresponding to the same examples as in Section 4.2 (light component in the regular column and heavy component in the inverted column). The tray responses (middle plots) are therefore from the bottom and up for increasing light composition for the regular column but from the top and down for increasing heavy composition for the inverted column.

We see that the steady state profiles are equal but the approach to equilibrium is different. The case with a small amount of light component in the regular column is found to approach equilibrium faster than the "inverted" case with a small amount of heavy component in the inverted column ($x_{F,withdrawn} = 0.1$, upper plots). However, when the amount of "withdrawn" component is increased, the rate of approach to equilibrium becomes fastest for the inverted column (bottom plots).

4.4.2 Operation of "inverted" separations

Above we found that the steady state profiles under total reflux are the same for a regular and an inverted column for a given separation. However, the approach to equilibrium was different. How will this affect the product removal in the two columns? To answer this, consider the two following two cases:

1. A small amount of "withdrawn" component is removed from the feed charge ($x_F = 0.1$ for the regular column and $x_F = 0.9$ for the inverted column).

2. A large amount of "withdrawn" component is removed from the feed charge ($x_F = 0.9$ for the regular column and $x_F = 0.1$ for the inverted column).

The withdrawal and feed end compositions profiles are given in Figure 4.8 and 4.9 for the two cases. In both cases an initial total reflux period of 0.05 hr is used to reach steady state before the production period. Constant product flows are assumed. The specification for the withdrawal end composition is for the "withdrawn" component in both cases, $x_{D,light}^{spec} = x_{B,heavy}^{spec}$.

From Figure 4.8 it can be seen that, for the case with a small amount of "withdrawn component", the product composition for the inverted column drops below the specification $x_{B,heavy}^{spec} = 0.9$ very quickly. For the regular column however, the separation is satisfactory. For this case, the regular column has the fastest responses under total reflux as shown in Figure 4.7.

From Figure 4.9 we see that for the case with a large amount of "withdrawn" product, the specification for the product composition is held longest for the inverted column. For this case, the inverted column has the fastest responses under total reflux as shown in Figure 4.7. However, note that if the product specification had been lower, for example 0.85, the regular column would have been best.

An explanation for the difference in dynamic behavior between the two columns can be found by considering the equilibrium and operating lines as suggested by Hasebe *et al.* (1992). Consider Figure 4.10 where the equilibrium and operating lines are plotted

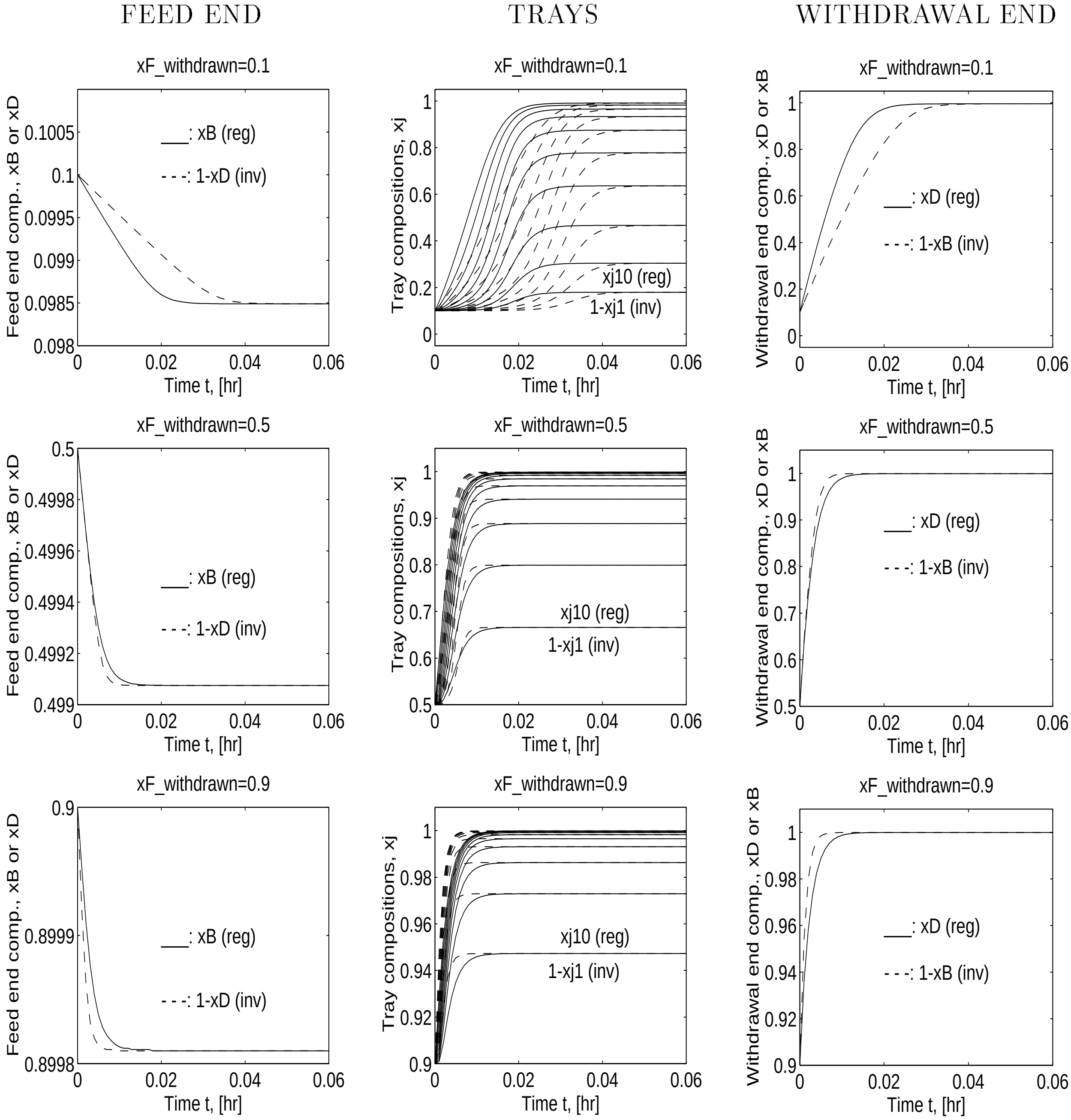

Figure 4.7: Time responses of compositions of withdrawn component for a regular and an inverted column under total reflux (inverted separations).

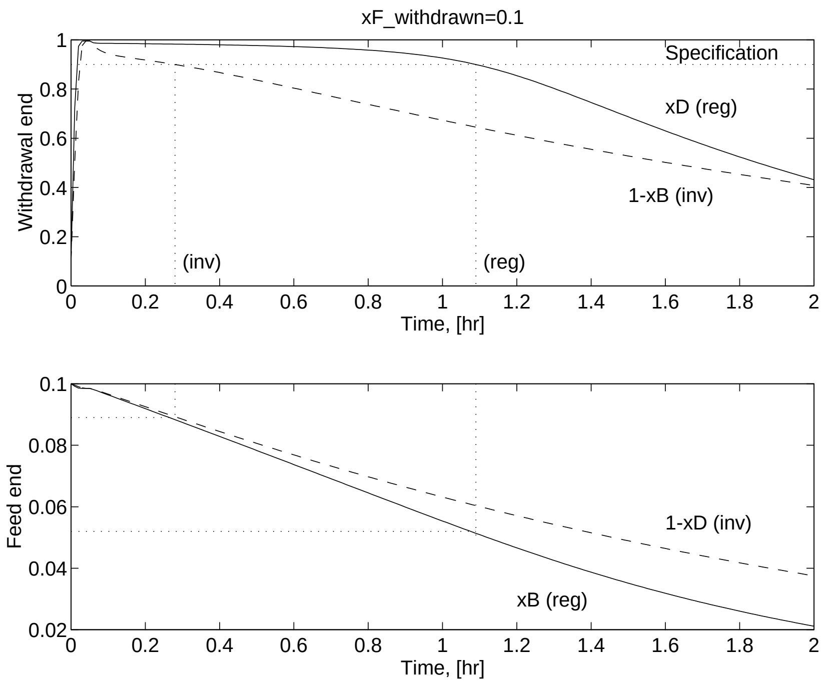

Figure 4.8: Withdrawal and feed end compositions profiles for the withdrawn component. ($x_F = 0.1$ for the regular column and $x_F = 0.9$ for the inverted column, $x_{D,light}^{spec} = x_{B,heavy}^{spec} = 0.9$ and $D = B = 0.5\ kmol/hr$.)

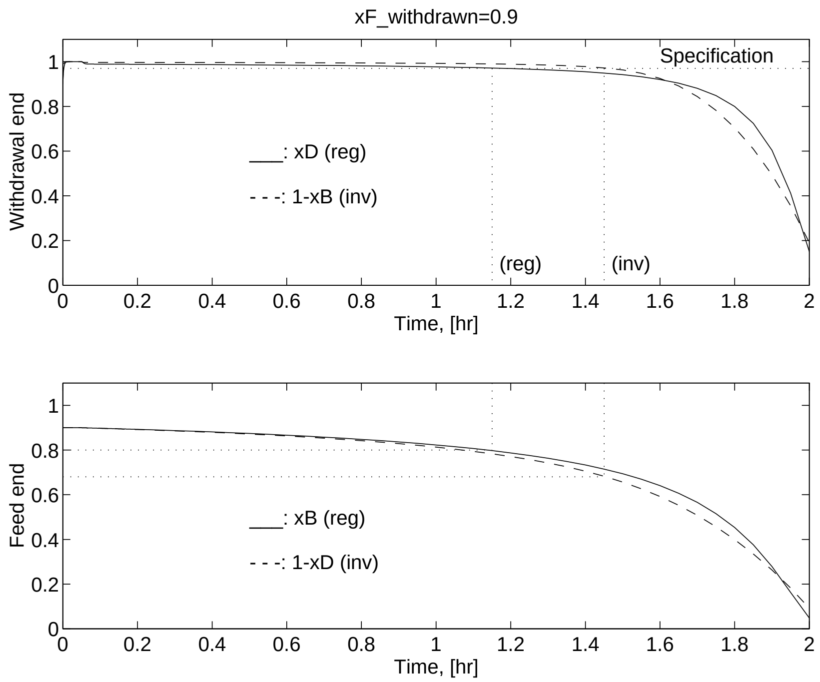

Figure 4.9: Withdrawal and feed end compositions for the withdrawn component. ($x_F = 0.9$ for the regular column and $x_F = 0.1$ for the inverted column, $x_{D,light}^{spec} = x_{B,heavy}^{spec} = 0.97$ and $D = B = 5\ kmol/hr$.)

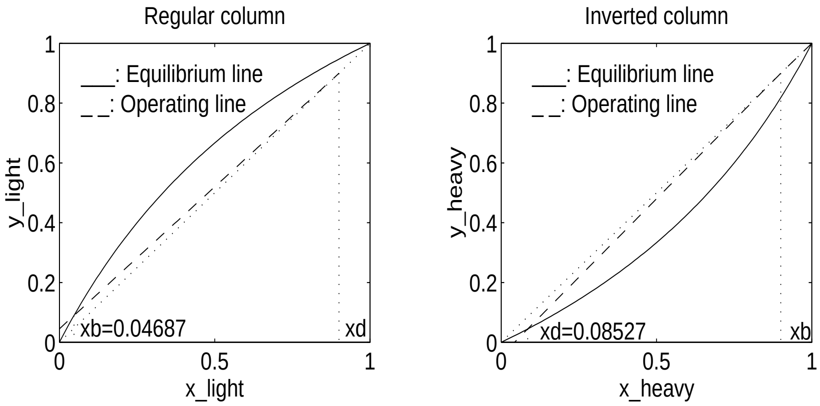

Figure 4.10: Equilibrium and operating lines for a regular and an inverted column ($x^{spec}_{D,light} = 0.9$ for the regular column and $x^{spec}_{B,heavy} = 0.9$ for the inverted column).

for the case with a small amount of "withdrawn" component (Figure 4.8). In the plots, it is assumed that the columns are run with a constant product composition (constant distillate operating policy). Since the compositions are varying (see Figure 4.8), a new plot should be made for each instantaneous value of x_D or x_B. However, Figure 4.10 still gives a good representation of the separation taking place in Figure 4.8.

For the regular column, the product specification ($x^{spec}_{D,light} \geq 0.9$) will be satisfied as long as the feed end composition of the light component is higher than $x_{B,light} = 0.047$. For the inverted column however, the feed end composition of the heavy component must be higher than $x^{spec}_{D,heavy} = 0.085$ for the product specification ($x^{spec}_{B,heavy} \geq 0.9$) to be satisfied.

The same discussion on equilibrium and operating lines can be made for the case with a large amount of "withdrawn" component (not shown).

Summary

In this section we have found that:

- The regular column has a faster approach to equilibrium than the inverted column for "inverted" separations with a *small* amount of "withdrawn" component in the feed (light in the regular column and heavy in the inverted column). Also, the regular column is able to maintain a high product composition for a longer period which will again result in either a larger amount of on-spec product or a shorter operating time for this column.

- For "inverted" separations with a *large* amount of "withdrawn" component in the feed (light in the regular column and heavy in the inverted column), the situation is the opposite. A faster approach to equilibrium and a longer on-spec period is found for the inverted column. The conclusion by Hasebe *et al.* (1992) that the regular column always has a better separation efficiency than the inverted column is thereby found to be wrong.

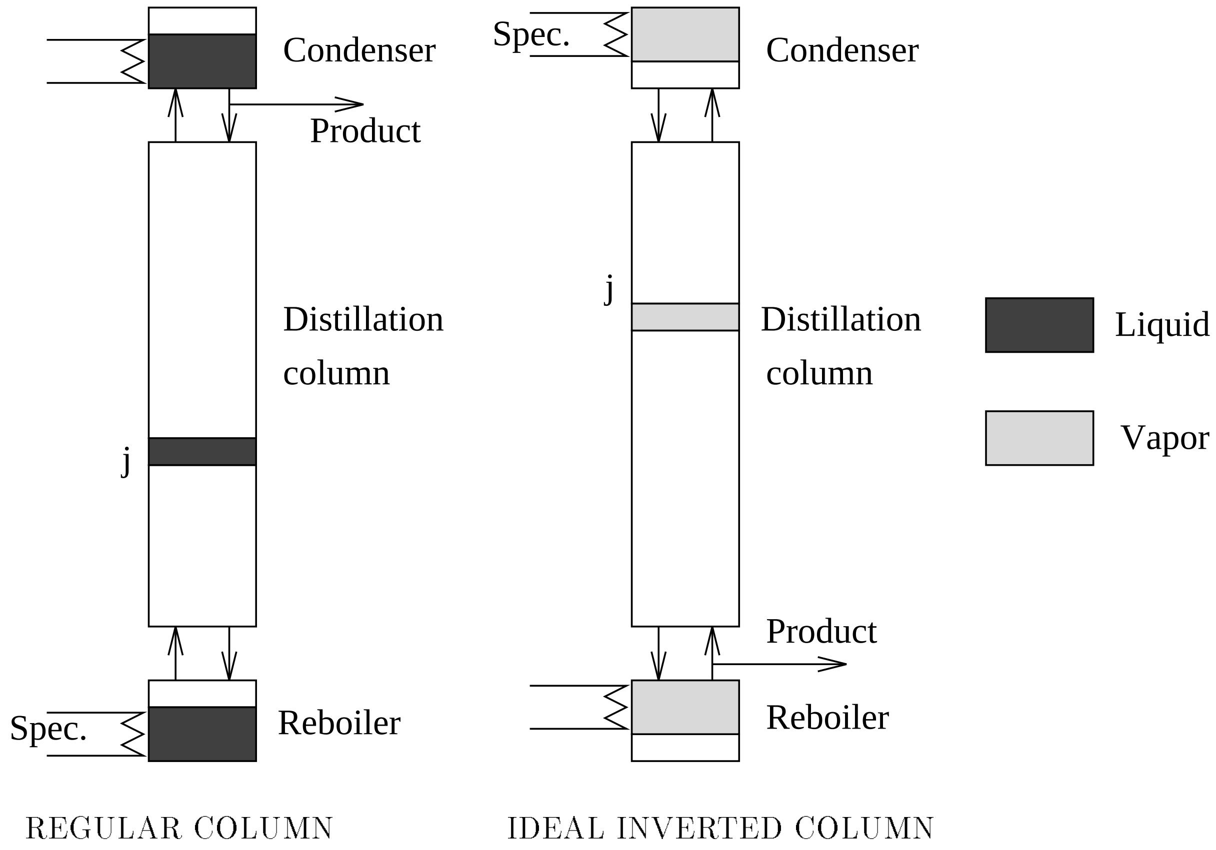

Figure 4.11: A regular and an ideal inverted batch column.

4.4.3 Ideal inverted column

Hasebe *et al.* (1992) explained the difference between regular and inverted columns to be due to the fact that the function $f(x) = y(\alpha, x) - x$ where $y(\alpha, x)$ is the vapor-liquid equilibrium equation, is not symmetrical around the vertical line $x = 0.5$. They stated that the regular column always has a better separation efficiency than the inverted column when the relative volatility is constant and the separation conditions are the same (here meaning "inverted"). However, this is not the case as shown in the previous section. The main difference between the two columns is due to the fact that the inverted column is *not the true inverse of the regular column* since the feed and product is in liquid and not in vapor phase.

The true inverse of a regular batch column is denoted an ideal inverted column in the following. It is a column where the feed is charged as *vapor* to the condenser drum. This vapor is continuously condensed in a total condenser and fed to the top of the column as liquid. The liquid from the bottom of the column is completely evaporated in the reboiler. A fraction of this vapor is returned to the column and some is taken off as product.

In Figure 4.11, a regular column is compared to an ideal inverted column. The column configurations are clearly equivalent provided:

- The liquid feed in the reboiler of the regular column is vapor feed in the condenser of the ideal inverted column

- The liquid holdup in the condenser of the regular column is vapor holdup in the reboiler of the ideal inverted column
- The product is taken out as liquid in the top of the regular column but as vapor from the bottom of the ideal inverted column
- The trays are filled with liquid in the regular column but with vapor in the ideal inverted column
- The vapor holdup is neglected in the regular column but the liquid holdup is neglected in the ideal inverted column (with our model assumptions)
- The vapor flow is specified in the regular column but the liquid flow is specified in the ideal inverted column

Finally, for constant relative volatility the separation is equivalent provided the liquid mole fraction of light component in the regular column is the mole fraction of heavy component in the inverted column. This follows since

$$\frac{y_{light}/x_{light}}{y_{heavy}/x_{heavy}} = \alpha = \frac{x_{heavy}/y_{heavy}}{x_{light}/y_{light}} \tag{4.6}$$

However, having both the feed charge and the product as vapor will not be very convenient. A more practical solution is therefore to have the feed as liquid in the condenser drum and to remove the product from the reboiler as liquid. This is the column studied so far in this paper and which we denote a *practical inverted column*. Note that the number of theoretical stages is equal for the regular and the two inverted columns:

Column:	Theoretical stages:	
Regular	N trays + reboiler	$= N+1$
Ideal inverted	N trays + condenser	$= N+1$
Practical inverted	N trays + reboiler	$= N+1$

Note that one may directly obtain operational data for the ideal inverted column from data for a regular batch column. One then needs to reverse the data as follows:

$$L^{inv} = V^{reg}, \quad y_F^{inv} = 1 - x_F^{reg}, \quad y_A^{inv} = 1 - x_A^{reg}, \quad y_R^{inv} = 1 - y_R^{reg} \tag{4.7}$$

(We use y_F, y_A and y_R to denote that these are vapor phase compositions.)

The data in the right column of Table 4.2 which are for a regular column with vapor flow $V_B = 10$ $kmol/hr$, will therefore correspond to data for an ideal inverted column with reflux flow $L = 10$ $kmol/hr$. For example, for the case with $x_F = 0.9$ and $x_A^{spec} = 1 - x_R^{spec} = 0.99$ the minimum operating time is 3.19 hr when vapor flow is fixed at $V_B = 10$ $kmol/hr$. Thus, with $y_F = 0.1$ (feed is vapor), $y_A^{spec} = 1 - y_R^{spec} = 0.01$ and *reflux flow* L fixed at 10 $kmol/hr$, the minimum operating time in an ideal inverted column is 3.19 hr. (If we instead fixed the vapor flow at 10 $kmol/hr$ then the operating time would be even shorter.) It is interesting to note from Table 4.2 that in a practical inverted column this separation needs 4.04 hr which is significantly higher.

4.5 Conclusions

In this paper we have compared a regular and an inverted column in terms of dynamic behavior and optimal operation. The inverted column is found to yield the shortest operating time for separations where the light component is present in a small amount in the feed. This is because in a regular column, the relatively small amount of light component must be moved through the column from the feed end to the withdrawal end which is more time consuming since a very high reflux ratio must be used.

Note that Robinson and Gilliland (1950), who originally proposed the use of inverted columns, stated that the main advantage was that the light component would be collected in the condenser drum in high purity. This is confirmed in this paper.

We have found that the steady state profiles are equal, but the approach to equilibrium is different, for "inverted" binary separations (removal of light component in a regular column and of heavy component in an inverted column). This is because the inverted column is not the true inverse of the regular column since the feed and product are in liquid and not in vapor phase.

The use of an ideal inverted column is, however, more of academical interest. In practice, one is faced with the decision as to whether to use a regular or a practical inverted column configuration for a given feed mixture. We believe that this paper has at least indicated the answer to this question.

Notation

B	bottom flow for inverted column	$kmol/hr$
D	distillate flow for regular column	$kmol/hr$
H_A	liquid holdup in accumulator	$kmol$
H_B	liquid holdup in reboiler	$kmol$
H_C	liquid holdup in condenser	$kmol$
H_F	amount of initial liquid feed	$kmol$
H_j	liquid holdup on tray j	$kmol$
H_R	amount in residual	$kmol$
L	reflux flow	$kmol/hr$
L_j	liquid flow from tray j	$kmol/hr$
N	number of trays in the column section (excl. reboiler and condenser)	
R	internal reflux ratio $=L/V_1$ for regular column	
R_B	reboiler reflux ratio $=V_B/L_N$ for inverted column	
t	time	hr
t_0	initial time	hr
t_1	switching time for operating variables	hr
t_f	final operating time	hr
V_j	vapor flow from tray j	$kmol/hr$
V_B	vapor flow from the reboiler to the column section	$kmol/hr$
x_A	mole fraction of light component in accumulator	
x_B	mole fraction of light component in reboiler	
x_D	mole fraction of light component in distillate	
x_F	mole fraction of light component in feed	
x_j	mole fraction of light component in liquid leaving tray j	
x_R	mole fraction of light component in residual	
y_B	mole fraction of light component in vapor leaving reboiler	
y_D	mole fraction of light component in vapor distillate	
y_j	mole fraction of light component in vapor leaving tray j	
Greek letters		
α	relative volatility	
Scripts		
inv	inverted column	
j	tray number	
liq	liquid	
norm	normalized	
o	initial conditions	
reg	regular column	
spec	specified value	
ss	steady state	
vap	vapor	
withdrawn	withdrawn component (light in regular and heavy in inverted)	

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Appendix. Dynamic models

The dynamic models used in this study are valid under the following assumptions:

1) Staged batch distillation column (trays numbered from the top and down)
2) Perfect mixing on all trays
3) Negligible vapor holdup
4) Constant stage pressures and tray efficiencies
5) Equal vapor flows in the column
6) Total condensation with no subcooling in the condenser
7) Constant relative volatility
8) Constant molar liquid holdup on all trays (liquid dynamics are neglected)
9) Constant molar condenser holdup in the regular column (perfect control)
9) Constant molar reboiler holdup in the inverted column (perfect control)

The following differential and algebraic equations result (see Figure 4.1):

	Regular column	Inverted column
reboiler	$\mathbf{dH_B/dt} = L_N - V_B$ $\mathbf{d(H_Bx_B)/dt} = L_N x_N - V_B y_B$	$\mathbf{0} = L_N - V_B - \mathbf{B}$ $\mathbf{H_B dx_B/dt} = L_N x_N - V_B y_B - \mathbf{Bx_B}$ $\mathbf{R_B = V_B/L_N}$
column tray	$0 = L_{j-1} + V_{j+1} - L_j - V_j$ $H_j dx_j/dt = L_{j-1}x_{j-1}$ $+V_{j+1}y_{j+1} - L_j x_j - V_j y_j$ $V_j = V_{j+1}$	$0 = L_{j-1} + V_{j+1} - L_j - V_j$ $H_j dx_j/dt = L_{j-1}x_{j-1}$ $+V_{j+1}y_{j+1} - L_j x_j - V_j y_j$ $V_j = V_{j+1}$
cond-enser	$\mathbf{0} = V_1 - L - \mathbf{D}$ $\mathbf{H_C dx_D/dt} = V_1 y_1 - L x_D - \mathbf{Dx_D}$ $\mathbf{R = L/V_1}$	$\mathbf{dH_C/dt} = V_1 - L$ $\mathbf{d(H_Cx_D)/dt} = V_1 y_1 - L x_D$
accum-ulator	$dH_A/dt = \mathbf{D}$ $d(H_A x_A)/dt = \mathbf{Dx_D}$	$dH_A/dt = \mathbf{B}$ $d(H_A x_A)/dt = \mathbf{Bx_B}$
equil-ibrium	$y_j = \frac{\alpha x_j}{1+(\alpha-1)x_j}$	$y_j = \frac{\alpha x_j}{1+(\alpha-1)x_j}$

The reflux ratio R for the regular column is the internal reflux ratio L/V_1, not the usual external ratio L/D. (This is better solved numerically since $R_{internal} \in [0,1]$ whereas $R_{external} \in [0, \infty >$.) The reboiler ratio in the inverted column, R_B, is also defined as an internal ratio V_B/L_N. The external ratio is in this case given as $R_{B,external} = V_B/B$.

It is assumed that the vapor flow V_B can be manipulated directly. The heavy product flow B in the inverted column is assumed to be taken out from the reboiler as liquid with a composition equal to x_B.

Chapter 5

Control Strategies for Reactive Batch Distillation

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Abstract

A batch reactor may be combined directly with a distillation column by distilling off the light component product. This is done in order to increase the reactor temperature or to improve the product yield of an equilibrium reaction. The same amount of the light product should be removed as the amount being formed by the reaction at any time. A linearized model has been developed which describes the process behavior satisfactory for control analysis purposes. Controllability of a combined batch reactor/batch distillation column is found to depend strongly on operating conditions and on the time during the run. In general, controlling only the reactor temperature (one point bottom control), is difficult since the set point

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must be specified below a maximum value in order to avoid break-through of intermediate component in the distillate. This maximum value is usually not known a priori. For the example considered in this study, control of both reactor temperature and distillate composition (two-point control) is also found to be difficult due to large interactions in the column. As with one point bottom control, the reactor temperature must be specified below a maximum value. However, energy can be saved since the heat duty can be decreased with time. Controlling the temperature on a tray in the column (one point column control) is found to give good performance for the given process with no loss of reactant and a high reactor temperature although no direct control of the reactor temperature is obtained.

5.1 Introduction

Batch distillation is used in the chemical industry for the production of small amounts of products with high added value and for processes where flexibility is needed, for example, when there are large variations in the feed composition or when production demand is varying. A batch reactor can be combined with a distillation column as shown in Figure 5.1. The objective is to increase the reaction temperature and to improve the product yield of equilibrium reactions in the reactor by distilling off one or more of the products, thereby driving the equilibrium towards the products.

The control objective when considering batch processes is usually either i) to minimize the operating time or ii) to maximize the product yield or quality. Most of the papers

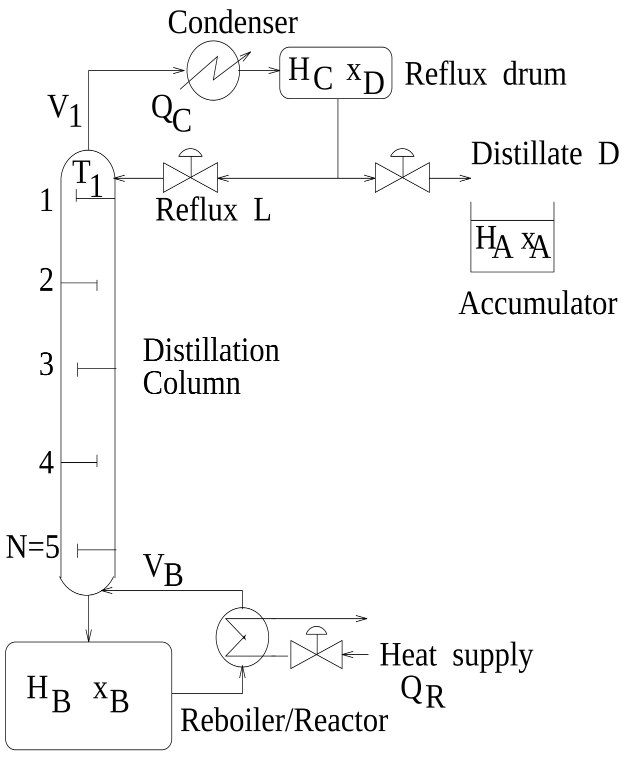

Figure 5.1: Batch distillation column/reactor.

published on batch distillation focus on finding optimal reflux ratio policies. However, sometimes the control objective is simply to obtain the same conditions in each batch. This was the case for the specific industrial application which was the starting point for our interest in this problem and which is to be presented later.

Few authors have considered the operation of batch distillation with chemical reaction although these processes are inherently difficult to operate and control. The analysis of such systems in terms of controllability has so far only been considered by Sørensen and Skogestad (1991, 1992). Roat *et al.* (1986) have developed a methodology for designing control schemes for continuous reactive distillation columns based on interaction measures together with rigorous dynamic simulation. However, no details about their model were given.

Modeling and simulation of reactive batch distillation has been investigated by Cuille and Reklaitis (1986), Reuter *et al.* (1988) and Albet *et al.* (1991). Cuille and Reklaitis (1986) developed a model and solution strategies for the simulation of a staged batch distillation column with chemical reaction in the liquid phase. Reuter *et al.* (1988) incorporated the simulation of a PI-controller in their model of a batch column with reaction only in the reboiler. The controller was used to control the top tray temperature using the distillate flow as manipulated variable. They stated that their model could be used for the investigation of control structure with the aid of Relative Gain Array analysis (RGA) but no details were given. Albet *et al.* (1991) presented a method for the development of operational policies based on simulation strategies for multicomponent batch distillation applied to reactive and non-reactive systems.

Egly *et al.* (1979, 1983) considered *optimization and operation* of a batch distillation column accompanied by chemical reaction in the reboiler. Egly *et al.* (1979) presented a method for optimization of batch distillation based on models which included the non-ideal behavior of multicomponent mixtures and the kinetics of chemical reactions. The column operation was optimized by using the reflux ratio as control variable. Feeding one of the reactants during the reaction was also considered. In a later paper (1983), they also considered control of the column based on temperature measurements from different parts of the column. The optimal reflux ratio policy was achieved by adjusting the distillate flow using a non-linear control system. However, no details where given about neither the column/reactor nor the control system.

The purpose of this paper is to give an understanding of the dynamic behavior of reactive batch distillation and to show how this process differs from batch distillation without reaction. Also we want to investigate the possible difficulties in controlling a reactive batch distillation process and give some alternative control strategies based on an industrial example.

First, an industrial process, consisting of a batch reactor with a rectifying column on top, is presented. In this process the reaction is limited to the reactor/reboiler. Based on a staged equilibrium model, we present special features of reactive batch distillation. We show how a linearized model of the process can be used to describe the behavior in different operating points. Using this linear model we present a simple controllability analysis based on step responses. Several control strategies are presented. We consider *two-point control*, where both the top and the bottom part are controlled, as well as *one point control*, where only one part of the column/reactor is controlled. A Relative Gain Array (RGA) analysis is used in the investigation of control structures in two point control.

5.2 Process example

In this study we have developed a dynamic model for a combined batch reactor/batch distillation process consisting of a total mass balance, the mass balances for each component, the energy balance, linear tray hydraulics and Raoult's law for the vapor-liquid equilibrium. A staged equilibrium model is used for the distillation column and the condenser holdup is assumed to be perfectly controlled. The dynamic model is given in Appendix A. It was solved and linearized using SPEEDUP (1992).

The basis for this study is an industrial equilibrium esterification reaction of the type

$$\xi_1 R_1(aq) + \xi_2 R_2(l) + \xi_3 R_3(l) \rightleftharpoons P(s) + W(l)$$

where R_1 is a dibasic aromatic acid (Therephthalic acid with $T_b = 767^0C$), R_2 and R_3 are glycols (1,2-Propanediol with $T_b = 188^0C$ and 1,6-Hexanediol with $T_b = 243^0C$), P is the solid polymer product and W is the side-product (Water with $T_b = 100^0C$). The overall reaction rate is described by the following rate equation:

$$r_B = r_{R_1} = k_1 c_{R_1}(c_{R_2} + c_{R_3}) - k_2 c_W \tag{5.1}$$

where

$$k_r = k_{0,r} \exp\left(-\frac{E_r}{R_g}\left(\frac{1}{T_B} - \frac{1}{T_0}\right)\right) \tag{5.2}$$

The determination of the kinetic parameters ($k_{0,r}$, E_r and T_0) based on the industrial case study is given in Appendix B. The activation energy for the reverse reaction is the same as for the forward reaction, i.e. $E_1 = E_2$. The frequency factor for the reverse reaction $k_{0,2}$ is one fourth that of the forward reaction $k_{0,1}$. However, the reverse reaction is usually negligible as the concentration of W in the reactor, c_W, will in most cases be very small (typically about 0.005 kmol/m^3 compared to about 4.7 kmol/m^3 for R_1) since the water is constantly removed by distillation. The reverse reaction will only have some influence at the very end of the batch when most of the reactants have been used up and also c_{R_1}, c_{R_2} and c_{R_3} are small. A summary of the process data is given in Table 5.1. Note that some water is added initially to dissolve the heavier components. The physical properties data for the components (P_{ji}^{vap}, h_{ji}^L, h_{ji}^V and ρ_i) were taken from Daubert and Danner (1985).

The amount of reactant R_2 in the feed in the industrial unit was 20 % higher than necessary to yield complete conversion of the reaction. This was done to account for the possible loss of reactant in the distillate with the existing operating procedure. In this study no excess of R_2 was used.

This process has been studied earlier by Sørensen and Skogestad (1991, 1992, 1994). Compared to the process model used by Sørensen and Skogestad (1991, 1992), in this study the number of stages in the column is reduced from 6 to 5 and the excess of

Table 5.1: Process data for the industrial case study.

Column:	reboiler + 5 trays + total condenser + accumulator
Reaction:	$1.0\ R_1 + 0.7\ R_2 + 0.3\ R_3 \rightarrow P(s) + 2.0\ W$
Activation energies:	$E_1 = E_2 = 18$ kcal/mol $= 7.5 \cdot 10^4$ kJ/kmol
Frequency factors:	$k_{0,1} = 0.06$ and $k_{0,2} = 0.015$
Reference temperature for k_r:	$T_0 = 500.0$ K
Pressure in column/reactor:	P=1 atm
Maximum heat input:	$Q_{R,max} = 0.85 \cdot 10^6$ kJ/hr = 236.1 kW
Hydraulic time constant:	τ=0.0018 hr =6.5 s
Condenser holdup:	H_C= 1.7 kmol
Initial holdup on the trays:	H_j^0= 0.1 kmol
Initial feed:	R_1: 10.40 kmol (Acid)
	R_2: 7.28 kmol (Alcohol)
	R_3: 3.12 kmol (Alcohol)
	P: 0.00 kmol (Polyester)
	W: 2.20 kmol (Water)

R_2 is no longer included. Furthermore, the model is extended to also include energy balances and a temperature- and concentration dependent reaction rate (see Appendix A). Optimization and on-line operation of the process using one-point column control was studied by Sørensen and Skogestad (1994). (An extended version of this paper is presented in Chapter 6 of this thesis).

The existing operating procedure was to use *one-point top control*; the temperature at the top of the column T_1 was kept constant at about 103^oC using the reflux flow L as manipulated variable. Simulations with the dynamic model in Appendix A gives an average distillate composition of 0.004 (about 2 weight%) of the reactant R_2 and thereby a loss of this component (about 1 % in total). Due to disturbances, such as varying heat input or pressure, this loss was often higher in practice. The vapor flow was maximized by using maximum constant heating of the reactor $Q_{R,max}$ and the condenser level was controlled by the distillate flow D. The simulated temperature profile with this control strategy for the ideal case with no disturbances is given in Figure 5.2. The reactor temperature T_B is almost constant at the beginning but increases slightly as reaction proceeded. There is a sharp increase in temperature at the end of the reaction when the light reactant R_2 is almost used up. The conditions on tray 4 and 5 are practically equal since the column has more stages than needed for the desired separation. The simulated conversion at the end point (t=10.5 hr) is 0.957 due to the loss of R_2.

With the existing control scheme there is no direct control of the reactor temperature T_B and more severely, there is a varying loss of the reactant R_2. This leads to a varying quality of the product $P(s)$ between batches.

For an equilibrium reaction, combining the reactor with the distillation column has two effects:

1. Increase the reactor temperature by removing the light component W as distillate.

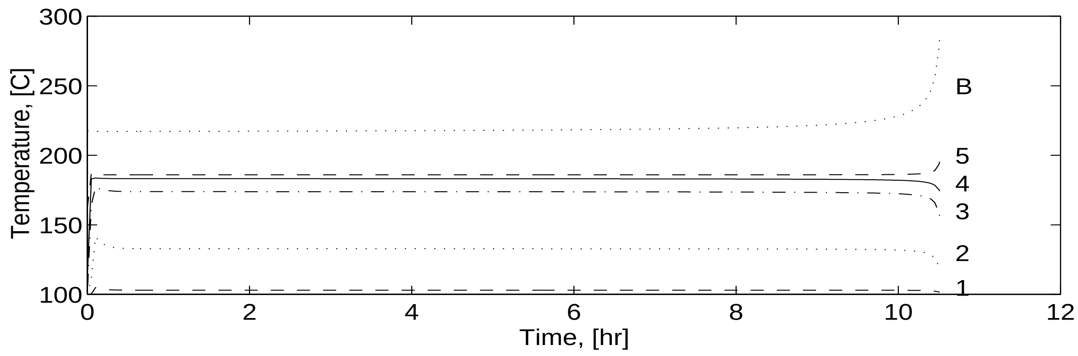

Figure 5.2: Temperature profile with existing control strategy (one-point top control) without disturbances.

This will also increase the rate constants and thereby the reaction rate.

2. Drive the equilibrium reaction towards the products P and W by removing the side-product W as distillate.

For the example in this study, the rate of the reverse reaction is in most cases very low due to the low concentration of W in the reactor. The main effect will therefore be the increase in reactor temperature.

In operating the process, one of the main objectives is to keep the reactor temperature as high as possible in order to reduce the reaction time. One way of achieving this is to run the process under pressure but this was not possible for the industrial process studied in this example. However, the reactor temperature must not exceed 240^0C since this may lead to decomposition of the polymer product P.

5.3 Understanding reactive batch distillation

The conventional operating policies for normal batch distillation columns are:

1. Constant distillate composition x_D (or temperature T_1)

2. Constant reflux ratio R

3. Time varying or optimal reflux ratio $R(t)$

The constant distillate composition policy is usually implemented using feedback control with either reflux flow or distillate flow as manipulated variable. The reflux ratio policies are open loop, that is, predefined values are used without feedback from the process.

As long as the set point for the controlled variable x_D or the set variable R is high enough compared to the heat input Q_R, the separation in a regular batch column will be satisfactory. Overspecifying the set point, e.g setting the reflux ratio to a higher value than necessary, will merely result in a slightly longer operating time but the products being distilled will be purer than specified.

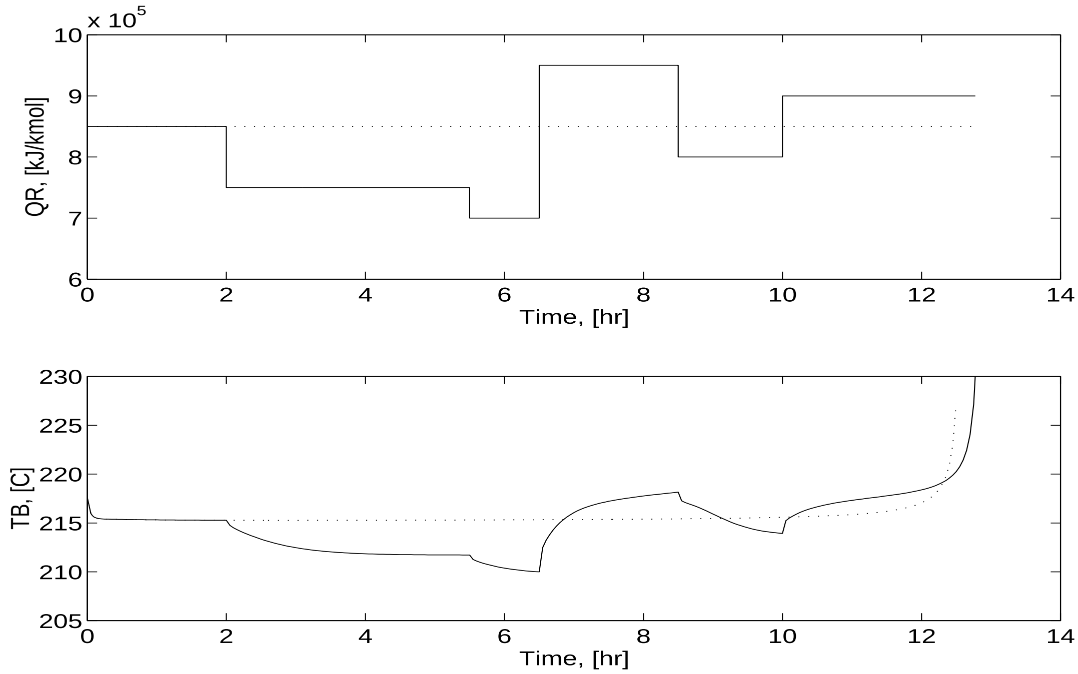

Figure 5.3: Constant reflux ratio policy ($R = 0.92$). Top: Disturbances in heat input Q_R. Bottom: Reactor temperature profile T_B with (-) and without (··) disturbance in heat input Q_R.

In the case of reactive batch distillation, where a reaction is taking place either only in the reboiler/reactor or in the whole column, some of the components will be removed and some will be formed in addition to what is removed or fed as part of the separation. The obvious question is then: Can the same operating policies still be used as for regular batch distillation? The answer is yes for the constant distillate composition policy but no for the open-loop reflux policies and we will in this section explain why.

Constant reflux policy

The temperature profile for the reactor when the process is operated according to the constant reflux ratio policy is given in Figure 5.3. Simulations with and without disturbances are given. The disturbance is assumed to be in the reboiler heat duty Q_R (max. ± 18 %). The temperature profile for the case with disturbances (solid line) deviates significantly from the ideal one without disturbances (dotted line). It should be noted that the loss of reactant R_2 is only $3.7 \cdot 10^{-5}$ and $9.1 \cdot 10^{-5}$ kmol for the undisturbed and disturbed case respectively. However, the reboiler temperature deviates up to $6^o C$ from the undisturbed one which will result in a varying quality of the polymer product P between batches.

In the case of *reactive* batch distillation, an open-loop operating policy (like reflux ratio policies) is therefore not good enough. Even though the separation in the column section may be satisfactory, the product quality might still vary (little reactant R_2 was lost in either case in Figure 5.3). Because of the reaction in the reactor, we *need some sort of feedback control* with the reactor/column conditions in order to reduce the effect

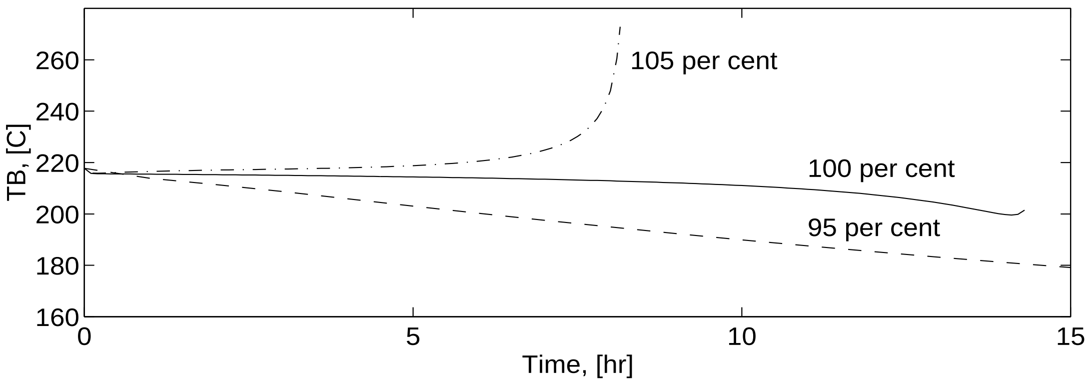

Figure 5.4: Reactor temperature profile when the removal of light side-product W in the distillate is 95 %, 100 % and 105 % of the amount formed at any time (simulations).

Table 5.2: Results for varying removal of light side-product W in the industrial case study.

removal as % of formation	loss of reactant R_2, kmol	conversion of R_1	final batch time, hr	product P, kmol (monomer block)	energy consumption, kJ
105.00	1.091	0.764	8.16	7.946	$6.9 \cdot 10^6$
100.00	$2.2 \cdot 10^{-5}$	0.990	14.30	10.296	$12.2 \cdot 10^6$
95.00	$3.8 \cdot 10^{-6}$	0.990	328.04	10.296	$278.8 \cdot 10^6$

of possible disturbances on the reaction.

Effect of product flow

To understand the above results we consider the effect of the distillate (product) flow. Ideally, the same amount of light product should be removed in the separation as the amount being formed by the reaction at any time. If less is removed in the distillate, too much light product will be left in the column and the overall reaction rate will decrease due to the reduced temperature. If the distillate flow is too high, all the light product formed will be removed but in addition also some of the light reactants. Some of the reactants are thereby lost and the conversion will be reduced. Also in some cases these reactants must be removed from the distillate later.

This is shown in Figure 5.4 for the reversible reaction considered in this study. The reaction is run either until 99 % conversion or until less than $1 \cdot 10^{-4}$ kmol R_2 is left in the reactor (note that these are simulated results). The loss of R_2, the conversion, the final operating time, the amount of product and the total energy consumption for each of the cases shown in Figure 5.4 are given in Table 5.2.

From Figure 5.4 it can be seen that when the distillate flow is too high (105 % removal), not only the light product W will be removed but also some of the reactant R_2. The reactor temperature increases as the reactant is removed and the reaction stops when all the light reactant is either reacted or removed. This loss of one of the reactants will not only result in a reduced amount of polymer product P, but also in a polymer which will have a different molecular structure than the specified product.

When the distillate flow is too low (95 % removal) not all of the light product W formed in the reactor is removed. The reactor temperature therefore decreases and the reaction becomes very slow. After 15 hr the conversion is still only 63 %.

For the case when the exact amount is removed (100 % removal), the temperature is decreasing during the batch. This is due to the amount of side-product W in the reboiler initially which is added to dissolve the heavier components. Since only the amount formed by the reaction is removed, the mole fraction of water in the reboiler will increase as the reaction proceeds and the reactants are used up. The temperature will therefore decrease. In fact, removing some of the initial water together with what is formed will speed up the reaction by increasing the temperature. However, care must be taken so that no reactant R_2 is lost (not shown).

For the reaction considered here the temperature dependency of the reaction rate is the dominant effect. If the reaction is considered to be irreversible ($r_B = k_1 c_{R_1}(c_{R_2} + c_{R_3})$) the same results as presented in Figure 5.4 are obtained (not shown).

5.4 Controllability analysis

In this section we will describe some important features considering the dynamic behavior of reactive batch distillation using the process example. A linear model is developed for this purpose. Measurements, manipulated variables and disturbances are defined and the effect of operating point and of disturbances on the measurements are discussed.

5.4.1 Linear model

In order to investigate the controllability of a process using available tools, a linear model is needed. Based on the non-linear model described in Appendix A, a linear model can be developed by linearizing the equation system at a given operating point. For continuous processes normally only one operating point is considered; that of the steady state conditions. The linear model is then found by linearizing around this operating point and will be valid for small deviations from steady state. When considering batch processes there is no such steady state; the conditions in the reactor or column are changing with time and the model is linearized along a trajectory. A linearized model of the process can be described by the following equations:

$$d\mathbf{x}/dt = A\mathbf{x} + B\mathbf{u} + E\mathbf{d} \tag{5.3}$$

$$\mathbf{y} = C\mathbf{x} + D\mathbf{u} + F\mathbf{d} \tag{5.4}$$

Where

$$\begin{aligned}
\mathbf{x} &= [\text{all states}]^T \\
\mathbf{y} &= [\Delta T_B, \Delta x_{D,2}, \Delta T_c, \Delta T_1, \Delta T_2, \Delta T_3, \Delta T_4, \Delta T_5]^T \\
\mathbf{u} &= [\Delta D, \Delta L, (\Delta Q_R)]^T \\
\mathbf{d} &= [\Delta P_B, (\Delta Q_R), \Delta \xi_2, \Delta E_1, \Delta k_{0,1}, \Delta k_{0,2}]
\end{aligned}$$

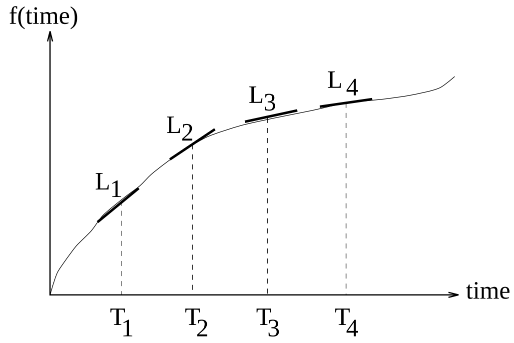

Figure 5.5: Linearization of a function $f(time)$ at $t = T_1$, T_2, T_3 and T_4.

Note that the reboiler heat duty Q_R is assumed to be either a control variable or a disturbance. Laplace transformation yields:

$$\mathbf{y}(s) = G(s)\mathbf{u}(s) + G_d(s)\mathbf{d}(s) \tag{5.5}$$

All variables represent deviations from the nominal trajectory or the "natural drift" along a trajectory with the flows and disturbances constant. It is the *deviation* from the natural drift which is important for control since this is the only thing the control system can affect. However, since this is a batch process which is never at a steady-state, the *way* we have reached the linearization point is also of importance since this will affect the states in the model. (Even when, at a given time, the values of the disturbances and outputs are identical). In this section we have derived the linear model from the trajectories given by the idealized case of 100 % removal of side-product W as presented earlier. The linear model is valid for small deviations from the nominal trajectory at the linearization time. This is illustrated in Figure 5.5 where a function $f(time)$ is linearized at $t = T_1$, T_2, T_3 and T_4. The resulting linear models are denoted L_1, L_2, L_3 and L_4.

In defining the linear model we have assumed that the temperature in the reboiler T_B can be measured in addition to the amount of reactant R_2 in the distillate flow $x_{D,2}$. These are the two variables we are mostly interested in controlling. In addition we also assume that the temperature in the condenser and on all the trays can be measured. The manipulated variables are the distillate flow D, the reflux flow L and in some cases the heat input to the reboiler Q_R. The heat input is normally at its maximum value to reduce the operating time. In the study of one-point control it is assumed that Q_R is a possible disturbance. Other main disturbances are the pressure in the reactor P_B and the reaction parameters (stoichiometric coefficient, activation energy and frequency factors).

The linear and non-linear models are compared for a 10 % step in reflux flow L at time $t = 6$ hr in Figure 5.6. The linear model is seen to describe the responses well up til $t \approx 0.1$ hr or 6 min. The linear model can therefore be used for control analysis purposes since in that case only the initial responses are of interest.

5.4.2 Effect of operating point

In Section 5.3 it was shown how the system changes with different product flows. It is rather obvious that the conditions in the column/reactor change when the operating

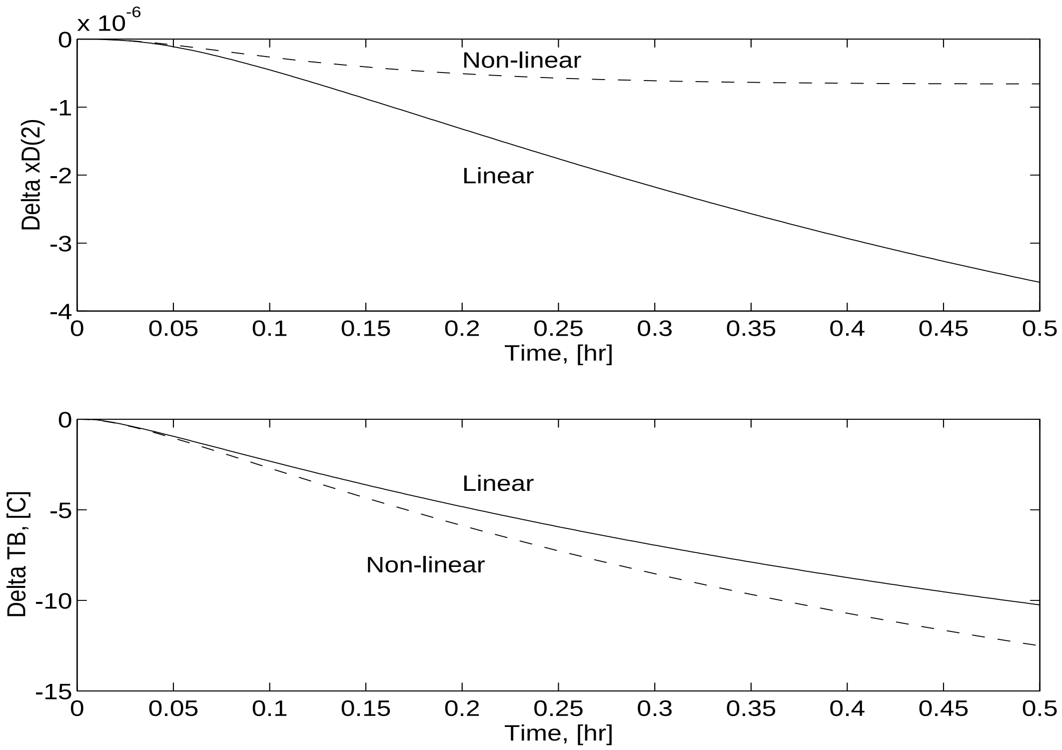

Figure 5.6: Effect of 10 % step in reflux flow L for linear and non-linear model on composition of R_2 in the distillate $x_{D,2}$ and reactor temperature T_B (at time $t = 6$ hr with 100 % removal of side-product W).

parameters, such as product flow, change. However, the conditions also change *during* the run.

Linear responses for the distillate composition and the reboiler temperature after a step in reflux flow are given at different operating points or selected times in Figure 5.7 (t = 0.5, 2, 4, 6, 8, 10, 12 and 14 hr). Note that the responses are the *deviation* from the nominal trajectory. The nominal case with 100 % removal of light side-product W is assumed. The effect on temperature increases with time and especially towards the end of the batch ($t = 14$ hr). The responses in distillate composition vary considerably with the largest response at the beginning of the batch ($t = 0.5$ hr). However, the responses in distillate composition are similar but differ in magnitude if plotted on log-scale (not shown). The difference in responses is due to the changing conditions in the column/reactor. There is a light/heavy front in the column section which is lowered with time thus giving the reduced responses in distillate composition and the increased responses in reactor temperature.

The responses for all tray temperatures after the same step in reflux flow are given in Figure 5.8. The effect by the step is largest for tray 4 and 5 and almost negligible for the top of the column. Remember that the existing operating procedure was to control the temperature on tray 1 using the reflux flow as manipulated variable which is seen to be difficult due to the very small responses. The responses on tray 5 in different operating points are more or less equal except at the beginning and end of the batch (Figure 5.8,

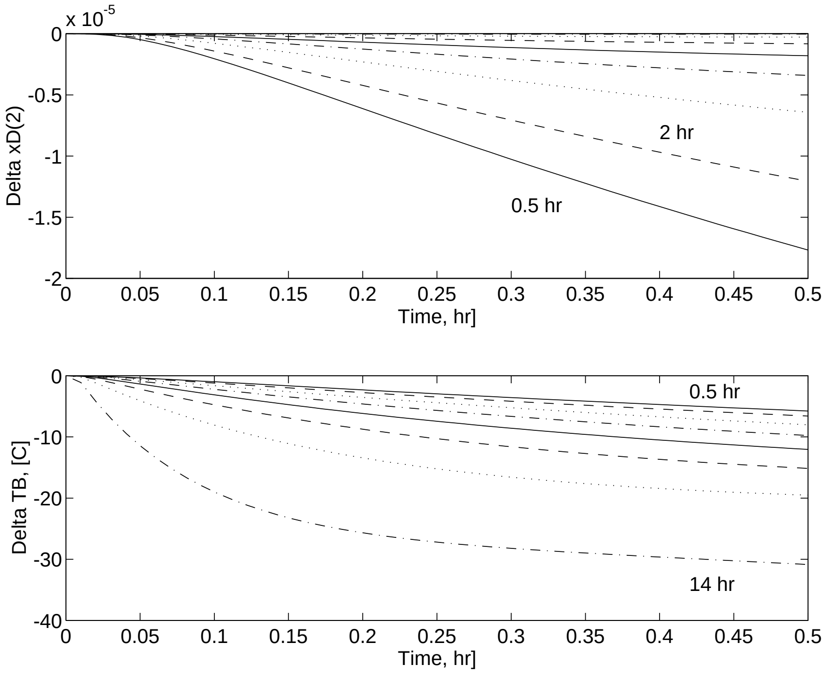

Figure 5.7: Effect of 5 % step in reflux flow L for linear model on distillate composition $x_{D,2}$ and reactor temperature T_B at different operating points (t = 0.5, 2, 4, 6, 8, 10, 12 and 14 hr for the case with 100 % removal of side-product W).

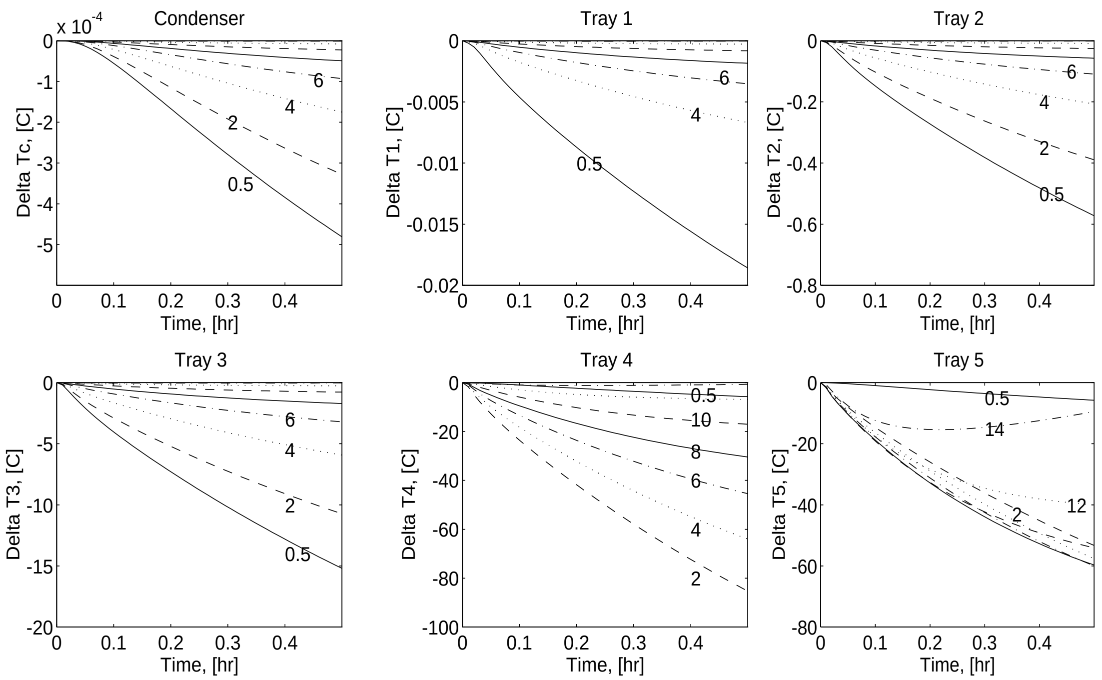

Figure 5.8: Effect of 5 % step in reflux flow L on condenser and tray temperatures for linear model at different operating points (t = 0.5, 2, 4, 6, 8, 10, 12 and 14 hr for the case with 100 % removal of side-product W). (Note that the scaling differs).

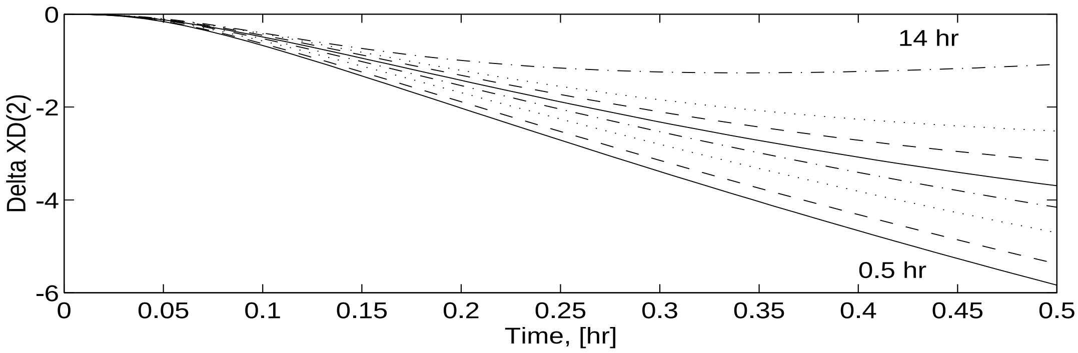

Figure 5.9: Effect of 5 % step in reflux flow L on the *logarithmic* distillate composition $X_{D,2}$ for linear model at different operating points (t = 0.5, 2, 4, 6, 8, 10, 12 and 14 hr for the case with 100 % removal of side-product W).

bottom right plot). For all the other trays the responses decrease constantly with time.

It must be stressed that the responses in Figure 5.8 depend strongly on the nominal conditions at the operating point. Here we found the linearized models based on the case with 100% removal of side-product W. Different nominal conditions may give other responses than the ones in Figure 5.8. (See Chapter 6 where tray 3 is found to have the largest responses for optimal nominal conditions.) However, the top tray is always found to have a very limited response to steps in the manipulated variables.

Reducing the non-linearity in distillate composition

An interesting feature in Figure 5.7 is that the responses in distillate composition $x_{D,2}$ to step changes have a similar initial shape if plotted on a log-scale. This is actually a general property for binary distillation (Skogestad and Morari, 1988). The inherent nonlinearities in this variable can therefore be reduced by using a log transformation on the distillate composition x_D:

$$X_{D,light} = -\ln\left(1 - x_{D,light}\right) \tag{5.6}$$

which in deviation variables becomes

$$\Delta X_{D,light} = \frac{\Delta x_{D,light}}{1 - x^*_{D,light}} \quad \text{and} \quad \Delta X_{D,heavy} = \frac{\Delta x_{D,heavy}}{x^*_{D,heavy}} \tag{5.7}$$

where $x^*_{D,light}$ and $x^*_{D,heavy}$ are nominal values. The responses for the distillate composition $X_{D,heavy} = X_{D,2}$ after the transformation is given in Figure 5.9. These initial responses, which are of primary importance for control, are very similar for the various operating points and the non-linearity is thereby reduced compared to Figure 5.7. From Figure 5.7 there is no obvious transformation that can be suggested to deal with the non-linear effect for the reactor temperature.

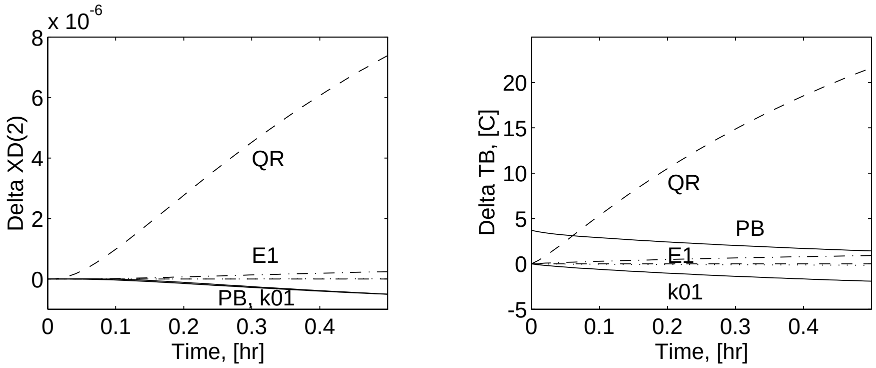

Figure 5.10: Effect on distillate composition $X_{D,2}$ and reactor temperature T_B of 10 % steps in various disturbances (at t=6 hr with 100 % removal of side-product W).

5.4.3 Effect of disturbances

We have identified one of the main disturbances to be the reboiler heat duty Q_R in those cases where it is not used as a manipulated variable (one-point top or column control). Also the pressure in the reactor P_B and the reaction parameters; the stoichiometric coefficients for the alcohols (ξ_2 and $\xi_3 = -1 + \xi_2$), the activation energy for the reaction ($E_1 = E_2$) and the frequency factors for the forward and reverse reaction ($k_{0,1}$ and $k_{0,2}$) are assumed to be disturbances. The selected reaction parameters are used to represent uncertainty in the reaction mechanism.

Linear step responses in reactor temperature and distillate composition by 10 % steps in the disturbances are given in Figure 5.10. For this operating point, the reboiler heat duty Q_R is the largest disturbance with the largest expected effect. Also the pressure P_B and the frequency factor for the forward reaction $k_{0,1}$ have some effect. Similar responses are also found for the other operating points (not shown).

5.4.4 Important considerations for control

In the previous sections we have shown the importance of feedback control for reactive batch distillation and explained why conventional batch distillation operating policies can not be used in the reactive case. Also we have identified the following problems in operating a reactive batch distillation process:

1. The same amount of side-product being formed should be removed at any time.

2. The system's response to changes vary with time as the conditions in the column/reactor change.

3. The trays in the column have different and varying sensitivity to changes.

4. The responses are generally non-linear.

These factors must be taken into account when deciding the control strategy.

5.5 Control strategies

The varying loss of reactant R_2 in the distillate and the lack of direct control of the reactor temperature were the major problems with the existing operating practice using one-point column control. In the control part of this study the following alternative control strategies are considered:

- One-point bottom control (controlling the reactor temperature directly)
- Two-point control (controlling both the distillate composition and the reactor temperature)
- One-point column control (controlling the temperature on a tray in the column)

The control parameters for the PI-controllers used in the simulations are given in Table 5.3. All controllers were tuned (using the linearized model at t=6 hr with 100 % removal of side-product W) such that the closed-loop gain was one at frequency 60 rad/hr, giving a response time of 1 min. However, for two-point control the $X_{D,2} - L$ loop had to be de-tuned to a response time of 3 min to reduce the interactions in the column. Note that the logarithmic composition $X_{D,2}$ was used instead of $x_{D,2}$ for two-point control.

Table 5.3: Control parameters used in simulations for the study of control strategies.

bottom control:	$K_p = 3.0$ and $\tau_I = 0.1$ hr	$(T_B \rightarrow L)$
two-point control:	$K_p = -2.15$ and $\tau_I = 0.1$ hr	$(X_{D,2} \rightarrow L)$
	$K_p = -0.12$ and $\tau_I = 0.1$ hr	$(T_B \rightarrow Q_R)$
column control:	$K_p = 0.25$ and $\tau_I = 0.1$ hr	$(T_5 \rightarrow L)$

5.5.1 One-point bottom control

The objective with one-point bottom control is to keep the reactor temperature constant at the highest possible temperature as this will maximize the rate of reaction for a temperature dependent reaction. The reflux flow L is used as manipulated variable and the heat input to the reactor is kept at its maximum value ($Q_R = Q_{R,max} = 0.85 \cdot 10^6\ kJ/kmol$). Consider the following cases:

I $T_B^{set} = 210^oC$
II $T_B^{set} = 215^oC$
III $T_B^{set} = 220^oC$

The temperature profiles for each case are given in Figure 5.11. The loss of R_2, the conversion, the final operating time, the amount of product and the total energy consumption for each of the cases shown in Figure 5.11 are given in Table 5.4.

For cases I and II the conditions are more or less constant with time, whereas case III has a changing temperature profile with large variations at the beginning of the run but more or less stabilizing midway through. For case I the front between light and heavy

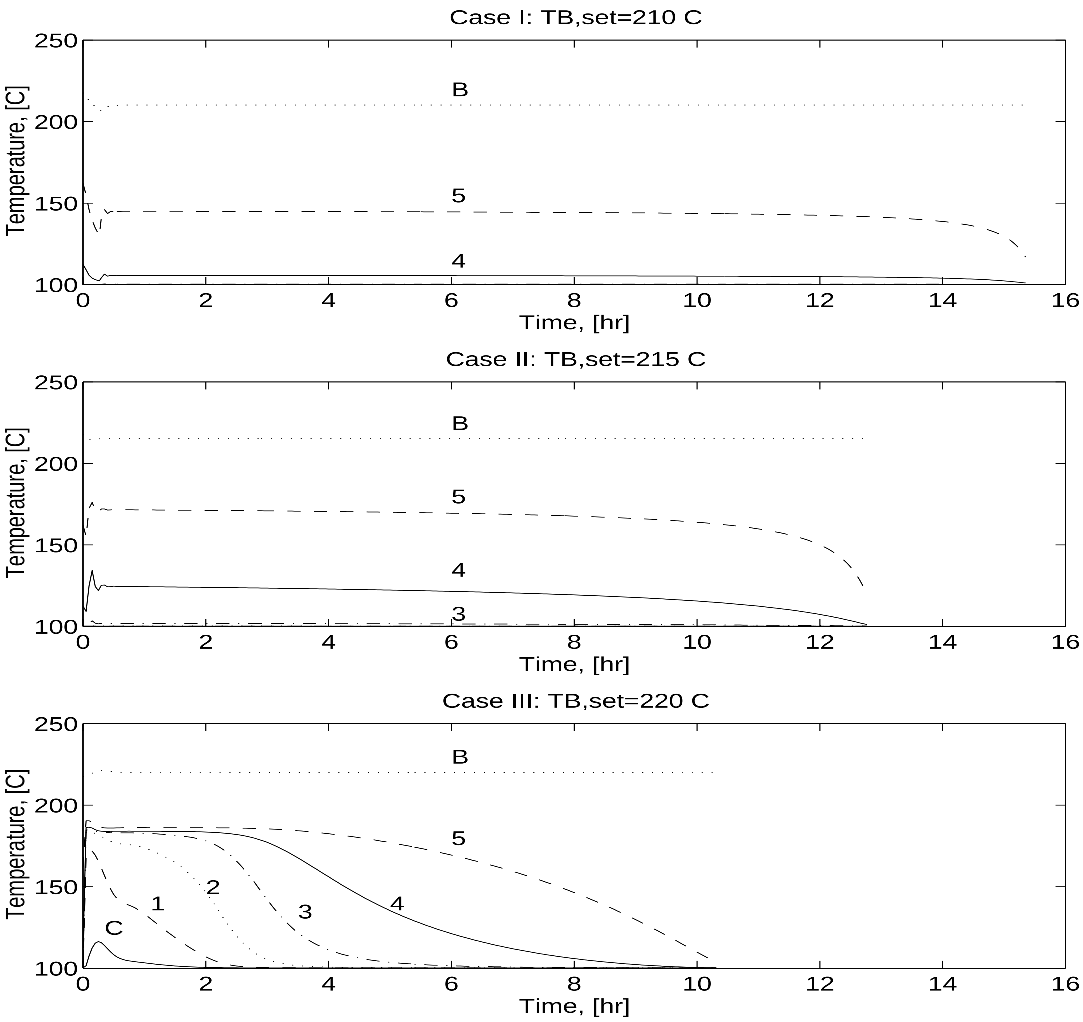

Figure 5.11: One-point bottom control. Temperature profiles for different set point.

Table 5.4: Results for one-point bottom control for cases with different set points for the reactor temperature T_B^{set}.

case	set point reactor temperature, ^0C	loss of reactant R_2, kmol	conversion of R_1	final batch time, hr	product P kmol (mon. block)	energy consumption, kJ
I	210.0	$4.3 \cdot 10^{-6}$	0.990	15.35	10.296	$13.05 \cdot 10^6$
II	215.0	$3.0 \cdot 10^{-5}$	0.990	12.77	10.296	$10.85 \cdot 10^6$
III	220.0	0.749	0.897	10.31	9.329	$8.77 \cdot 10^6$

component is low in the column, more light component than necessary is held back and the reaction rate decreases. For case II the front between light and heavy component is higher in the column and the reaction rate therefore increased. For case III the set point for the reactor temperature is too high and there is a loss of reactant R_2 during the first couple of hours of the run. This is the same effect as described in Section 5.3.

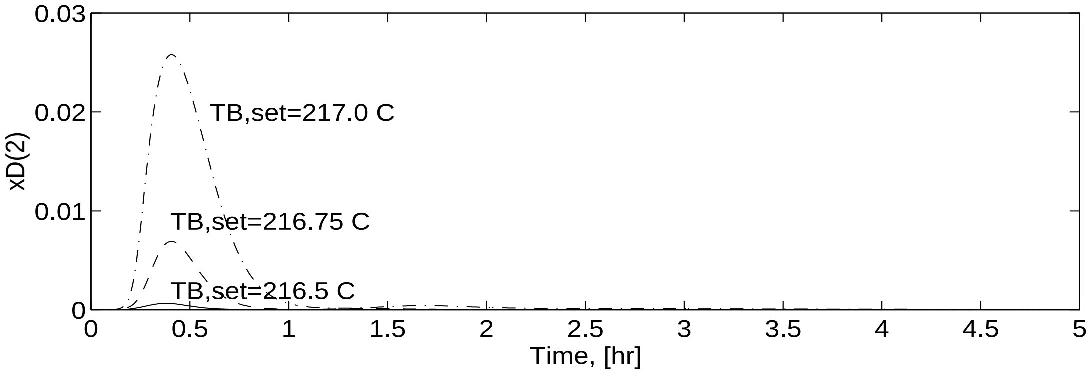

Figure 5.12: One-point bottom control. Composition of R_2 in distillate $x_{D,2}$ at different T_B^{set} for the first five hours of the batch.

If the reactor temperature is too low and too little side product is removed, the reaction becomes slow as shown in Table 5.4 (case I). If it is too high and too much is removed, the conversion is reduced due to the loss of reactant (case III).

In other words, there is a maximum set point for the reactor temperature in order to maximize the reaction rate but at the same time avoid loss of the reactant R_2. This maximum value will depend on the relative amounts of the reactants in the feed, on the reaction mechanism and on the disturbances in the system. This is illustrated in Figure 5.12 which shows how the mole fraction of R_2 in the distillate $x_{D,2}$ changes when the set point for the temperature controller in the reactor is increased from $T_B^{set} = 216.5^oC$ to $T_B^{set} = 217.0^oC$. An increase of 0.5^oC causes the mole fraction of reactant R_2 to increase by a factor of 25. However, the loss of reactant is only temporary and $x_{D,2}$ is reduced to an acceptable level after about 1 hr. The break-through is caused by the fact that when the specified temperature is above a certain maximum value where most of the light component W is removed, a further increase in temperature is only possible by removing the light reactant R_2. If the set point temperature is specified below the maximum value, in this case $T_B^{set} \approx 216.0^oC$, good control of the system is achieved ($T_B \approx T_{N,set}$ and $x_{D,2} < 1 \cdot 10^{-5}$). The system can, however, become unstable at the end of the batch depending on the choice of control parameters in the PI-controller. This is because the controllers are tuned using the linear model at $t = 6$ hr. Since the conditions are changing with time, the system may become unstable when the conditions are very different from this operating point.

Another alternative for raising the reaction temperature, and thereby the reaction rate for a temperature dependent reaction, is to let the set point follow a given trajectory, e.g. a linear increase with time. Again, the maximum reactor temperature to avoid break-through will limit the possible increase and break-through is inevitable if it is specified too high (not shown).

5.5.2 Two-point control

By using *two-point control* it may be possible to control both the top and the bottom part of the column by implementing two single control loops in the system. In this way energy consumption can be reduced since it will no longer be necessary to keep the reboiler heat duty at its maximum value. In the case of the esterification process, it is desirable to control not only the reactor temperature T_B but also the composition of the distillate $x_{D,2}$, i.e. the loss of reactant R_2.

Two different control configurations are considered for the batch column:

LV**-configuration** Controlling the condenser level H_C using the distillate flow D leaving the reflux flow L and the reboiler heat duty Q_R to control the distillate composition $X_{D,2}$ and the reactor temperature T_B:

manipulated variable:	controlled variable:
D	H_C
L	$X_{D,2}$ or T_B
Q_R	$X_{D,2}$ or T_B

DV**-configuration** Controlling the condenser level H_C using the reflux flow L leaving the distillate flow D and the reboiler heat duty Q_R to control the distillate composition $X_{D,2}$ and the reactor temperature T_B:

manipulated variable:	controlled variable:
L	H_C
D	$X_{D,2}$ or T_B
Q_R	$X_{D,2}$ or T_B

Note that the logarithmic distillate composition $X_{D,2}$ is used in both cases.

Analysis of two point model

Open-loop step responses for both configurations are given in Figure 5.13 and 5.14. The term "open-loop" should here be put in quotes because we are not talking about an uncontrolled column, but assume that the condenser level is perfectly controlled ($H_C \leftrightarrow D$ or $H_C \leftrightarrow L$). We consider the effect of the remaining independent variables on the distillate composition and reactor temperature. From Figure 5.13 it can be seen that, for the LV-configuration, the responses in $X_{D,2}$ and T_B to the chosen steps in L (+1 $kmol/hr$) and Q_R (+0.05 kJ/hr) are similar but in opposite direction. For the DV-configuration in Figure 5.14, the responses to a step in D are similar to the step in L for the LV-configuration but in the opposite direction. However, the responses to a step in Q_R are very small and also there is an inverse response in $X_{D,2}$. This is a general property for distillation (Skogestad, 1992).

In a distillation column there are large interactions between the top and the bottom part of the column. A change in conditions in one end will lead to a change in the other end as well. Because of these interactions, a distillation column can be difficult or almost impossible to control. The interactions in a system can be analyzed by various tools (see e.g. Wolff *et al.*, 1992), amongst them the RGA, or Relative Gain Array. Systems with no interactions will have a RGA-value of 1. The larger the deviation is from 1, the larger

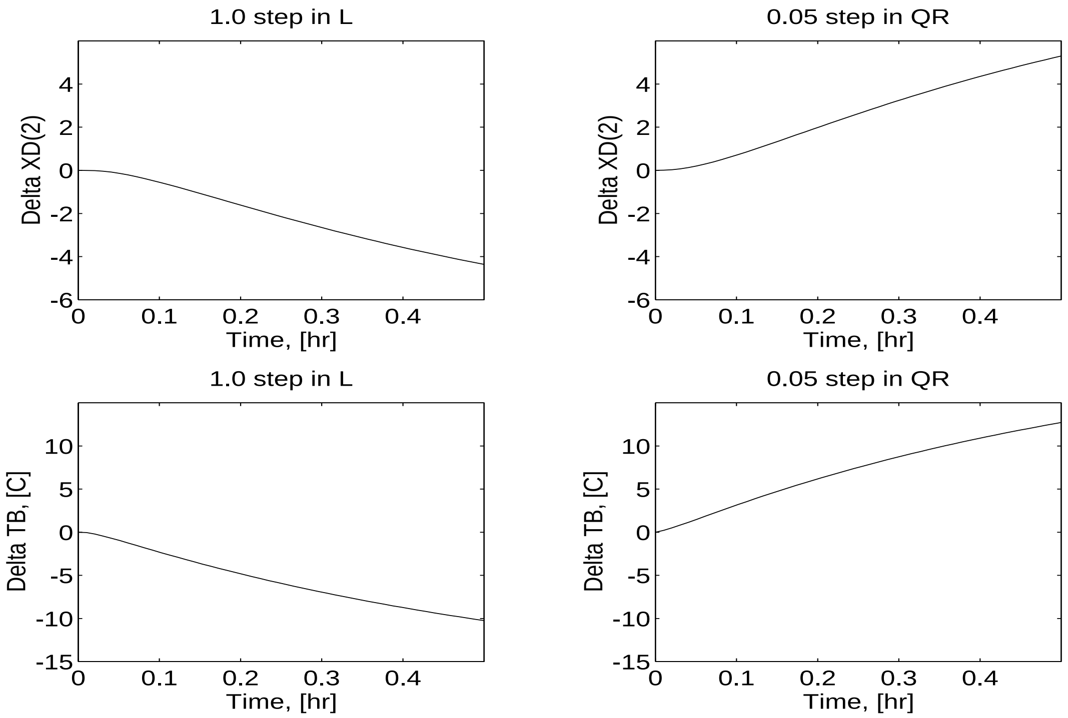

Figure 5.13: Linear open-loop step responses for LV-configuration (at t=6 hr for 100 % removal of side-product W).

are the interactions and the more difficult is the process to control. Pairing control loops on steady-state RGA-values less than 0 should be avoided.

The magnitude of the 1,1-element of the RGA for both the LV- and DV-configuration is given as a function of frequency in Figure 5.15. For the LV-configuration the RGA is very high at low frequencies (when the linear system is approaching a steady state). This shows that the interaction reduces the effect of the control input (L, Q_R) and makes control more difficult. RGA for the DV-configuration is generally lower at all frequencies. This difference between configurations is the same as one would observe in a continuous distillation column.

Nevertheless, the control characteristics from the RGA-plot for the LV-configuration are not quite as bad as they may seem. For control, the steady-state values are generally of little interest (particularly in a batch process since the process will never reach such a state). The region of interest is around the system's closed-loop bandwidth (response to changes), which is in the frequency range around 60 rad/hr corresponding to a response time of about 1 min. We note that the RGA is closer to 1 at these frequencies and also the difference between the two configurations is much less.

From the high-frequency RGA, which is close to 1, we find that for decentralized control, the loop pairing should always be to use the reboiler heat duty to control the reactor temperature. Either the reflux flow or the distillate flow should be used to control the distillate composition or the loss of reactant R_2. This is in agreement with physical intuition. The following possible pairing is suggested:

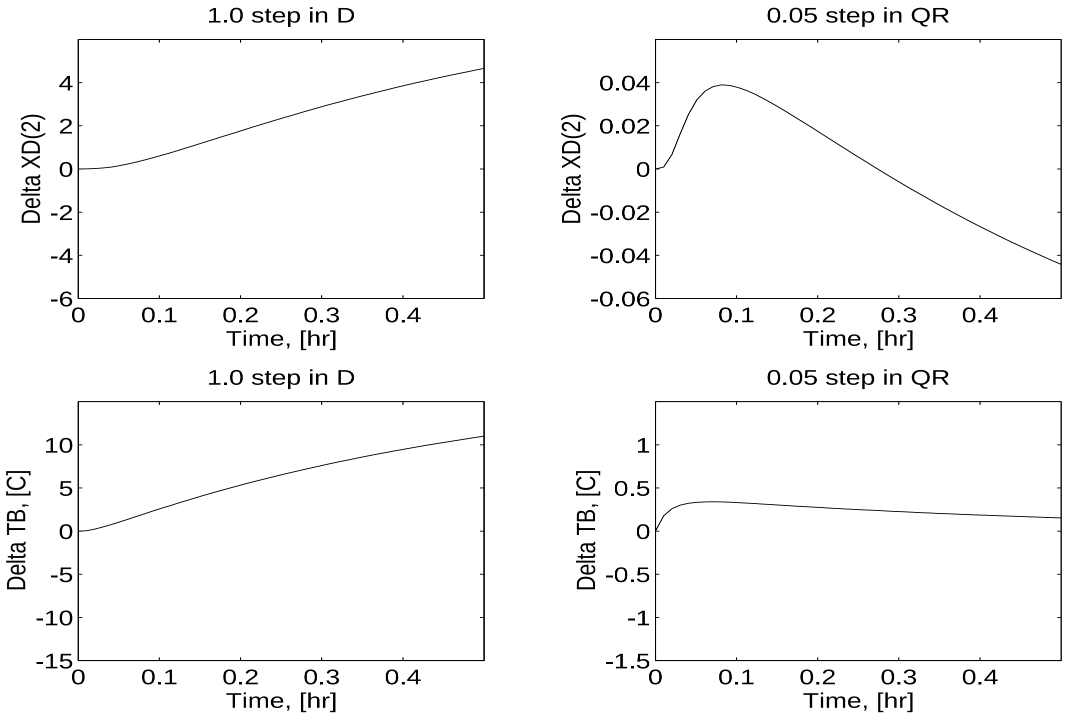

Figure 5.14: Linear open-loop step responses for DV-configuration (at t=6 hr for 100 % removal of side-product W). Note that the y-axis scaling is 100 and 10 times smaller for changes in Q_R compared to the LV-configuration.

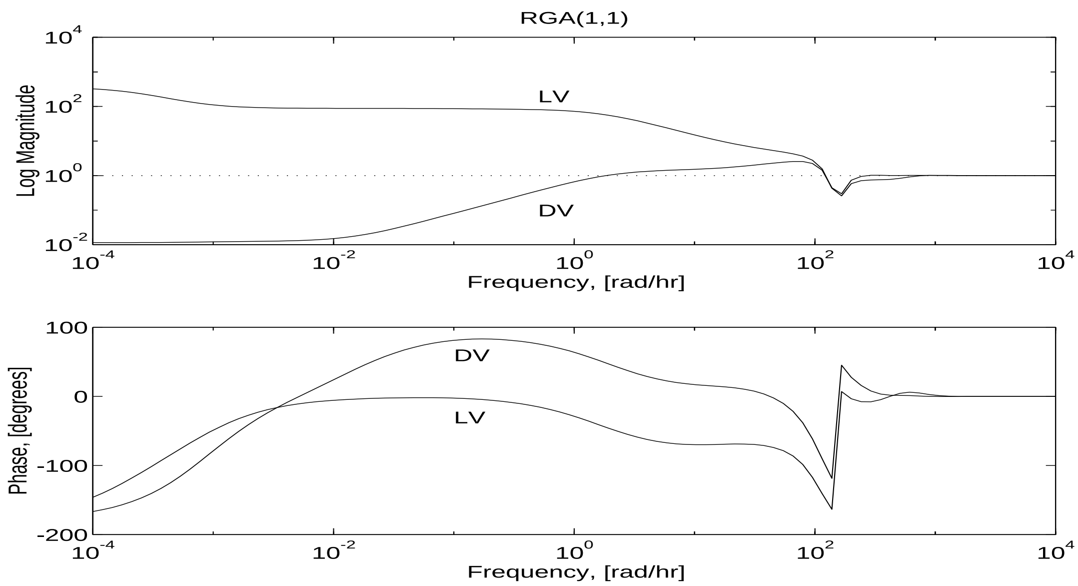

Figure 5.15: RGA(1,1) for LV- and DV-configuration for linear model (at t=6 hr for 100 % removal of side-product W).

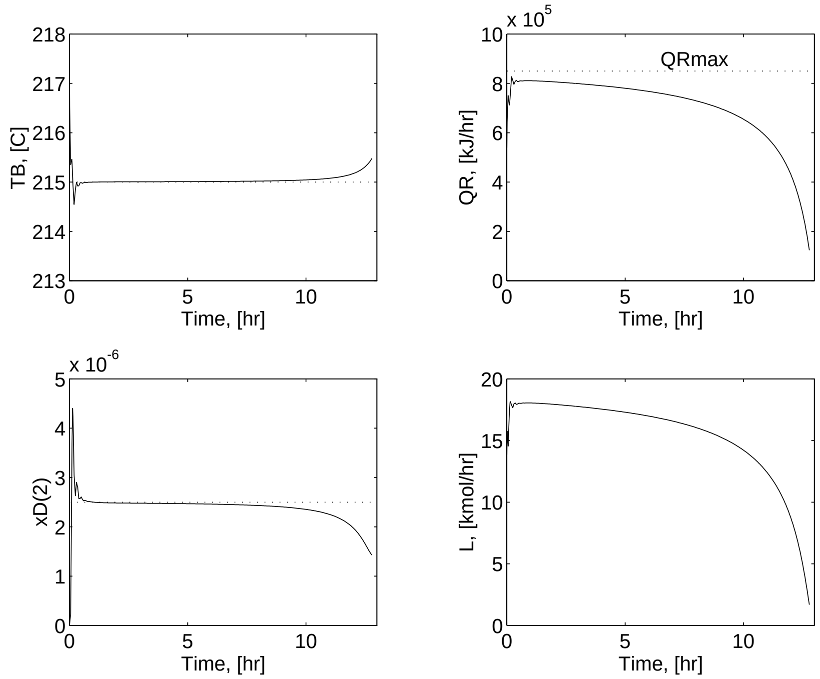

Figure 5.16: Two-point control. Reactor temperature T_B, distillate composition $X_{D,2}$, heat input Q_R and reflux flow L for LV-configuration (set points $T_B^{set} = 215^0C$ and $x_{D,2}^{set} = 2.5 \cdot 10^{-6}$).

manipulated variable:	controlled variable:
L or D	H_C
L or D	$X_{D,2}$
Q_R	T_B

Non-linear simulation of two-point model

Closed-loop simulations confirm that two-point control may be used if fast feedback control is possible. However, as in the case for one-point bottom control, we still have the problem of specifying a reasonable set-point for the bottom temperature to avoid breakthrough of reactant R_2 in the distillate. An example of two-point control of the process using the LV-configuration is given in Figure 5.16 and Table 5.5 with the following set point for the controllers: $T_B^{set} = 215^0C$ and $x_{D,2}^{set} = 2.5 \cdot 10^{-6}$. (In order to reduce the non-linearity in the model we control the transformed distillate composition $X_{D,2}$ instead of $x_{D,2}$.)

Table 5.5: Results for two-point control (LV-configuration).

set point reactor temperature, ^0C	set point distillate composition	loss of reactant R_2, kmol	conversion of R_1	final batch time, hr	product P kmol (mon. block)	energy consumption, kJ
215.0	$2.5 \cdot 10^{-6}$	$4.9 \cdot 10^{-5}$	0.990	12.79	10.296	$9.04 \cdot 10^6$

It can be seen that only a minor break-through of reactant occurs during the run. Both the distillate composition $x_{D,2}$ and the reactor temperature T_B are kept approximately at their set points through most of the batch but deviate towards the end. It should be noted how the heat duty decreases with time which shows that energy can be saved using two-point control. (Compare the results in Table 5.5 with case II in Table 5.4 where $T_B^{set} = 215^oC$ and Q_R is fixed at $0.85 \cdot 10^6$ kJ/kmol and the total energy consumption is $10.9 \cdot 10^6$ kJ with approximately the same operating time as in Table 5.5).

It is difficult to achieve tight control of both ends of the column at the same time not only due to the interactions in the column (RGA$\approx$ 5 at frequency 60 rad/hr). Another problem is that the set points for the two loops can not be specified independently. If the reactor temperature is specified too high, the control system will not be able to maintain the specified purity in the top and the column will become unstable (not shown). "Decoupling" the column sections by introducing more trays in the column will reduce the interaction problem.

5.5.3 One-point column control

With the existing operating practice presented in Section 5.2 the temperature at the top of the column was controlled. The set point was $T_1^{set} = 103\ ^0C$ which gave a composition of 0.4 % of reactant R_2 in the distillate. By lowering the set point to e.g 100.1 ^0C the distillate would be purer, but the column would become very sensitive to measurement noise, and this system would not work in practice.

One alternative is to measure the composition $x_{D,2}$ and use this for feedback as in two-point control in the previous section. However, implementing an analyzer (or possibly an estimator based on the temperature profile) is costly and often unreliable. A simpler alternative is to place the temperature measurement further down in the column since this measurement is less sensitive to disturbances. Also, the response to changes in the manipulated variable is larger (see Figure 5.8). In this investigation the temperature on tray 5, the tray above the reactor, is chosen as the new measurement to be used instead of the one on the top tray. This is because the effect of changes in the manipulated variable L is large and most constant with time for this tray (see Figure 5.8).

The reboiler heat duty is kept fixed at its maximum value ($Q_{R,max} = 0.85 \cdot 10^6$ kJ/hr). With this control configuration ($T_5 \leftrightarrow L$) there is no direct control of the reactor temperature. However, with an appropriate choice of set point T_5^{set}, loss of reactant R_2 can easily be avoided and one of the main causes of the operability problems is thereby eliminated.

The temperature profile for one-point column control with set point $T_{5,set} = 170^0C$ is shown in Figure 5.17 and the results are given in Table 5.6.

Table 5.6: Results for one-point column control.

set point temperature, ^0C	loss of reactant R_2, kmol	conversion of R_1	final batch time, hr	product P, kmol (monomer block)	energy consumption, kJ
170.0	$3.6 \cdot 10^{-5}$	0.978	12.22	10.171	$10.39 \cdot 10^6$

The conditions are "stable", i.e. no break-through of reactant R_2, throughout the

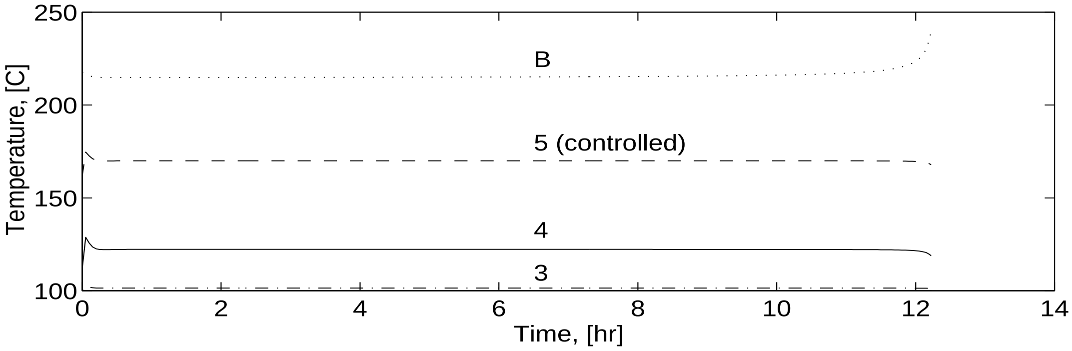

Figure 5.17: One-point column control. Temperature profile with $T_5^{set} = 170^oC$.

batch. The reactor temperature increases towards the end but reaches the maximum reactor temperature before complete conversion. This can be avoided by adding a small excess of reactant R_2 initially to account for the fact that some of this reactant will be located on the trays in the column and will therefore not be accessible for the reaction. Note that this control procedure, with Q_R fixed at its maximum, will yield the highest possible reactor temperature. This may be important in some cases when the reaction is slow.

5.5.4 Comparison of control strategies

In this section three alternative control strategies have been presented:

1. One-point bottom control (controlling the reactor temperature directly)

2. Two-point control (controlling both the distillate composition and the reactor temperature)

3. One-point column control (controlling the temperature on a tray in the column)

It was found that with the *one-point bottom* strategy, good control of the reactor temperature was achieved if the set point temperature was specified below a maximum value. This is illustrated in Figure 5.18 for the case with disturbances in the reboiler heat duty. The disturbances are the same as in Figure 5.3. The maximum reactor temperature is however, difficult to estimate a priori. In practice the set point must therefore be specified low enough to ensure an acceptable performance. This will of course result in a slower reaction and a longer operating time.

Two-point control allows both the reactor temperature and the distillate composition to be controlled. Energy will be saved compared with one-point control as the heat duty can be reduced. Two point control of the process including the disturbances in reboiler heat duty is given in Figure 5.19. Also in this case good control is achieved. However, both the reactor temperature and the distillate composition deviate from their set points towards the end of the batch.

The existing operating practice, controlling the temperature at the *top of the column*, is poor, sensitive to noise and leads to a varying loss of reactant R_2 and thereby varying

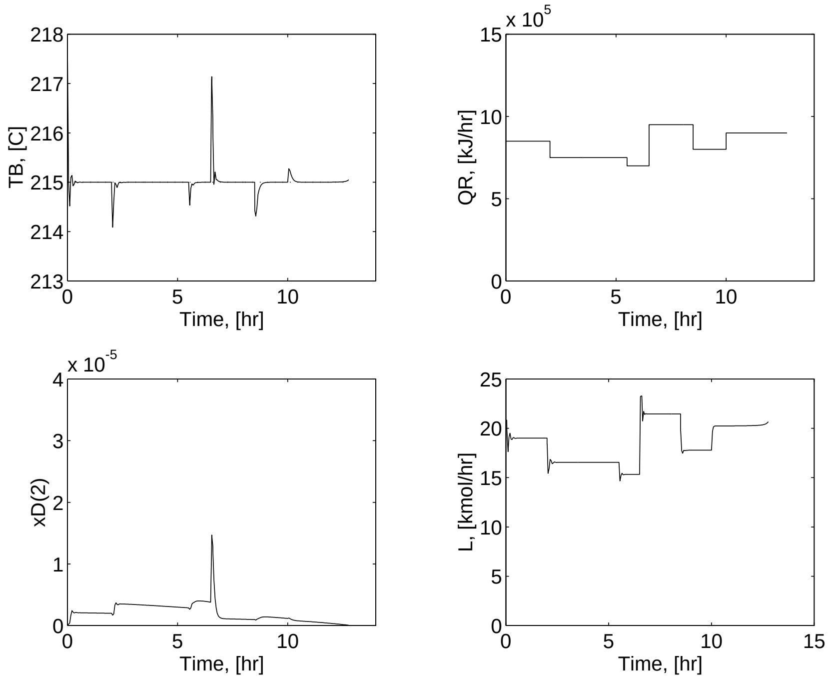

Figure 5.18: One point bottom control with disturbances in Q_R. Reactor temperature T_B, distillate composition $x_{D,2}$, heat input Q_R and reflux flow L with set point $T_B^{set} = 215^0C$. (Dotted line: Set point.)

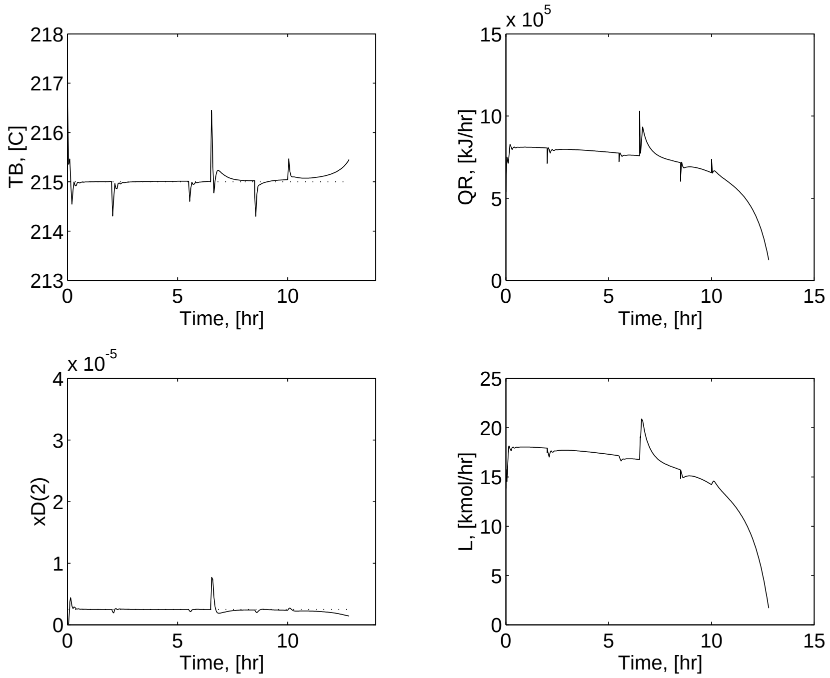

Figure 5.19: Two point control (LV-configuration) with disturbances in Q_R. Reactor temperature T_B, distillate composition $x_{D,2}$, heat input Q_R and reflux flow L with set point $T_B^{set} = 215^0C$ and $x_{D,2}^{set} = 2.5 \cdot 10^{-6}$. (Dotted lines: Set points.)

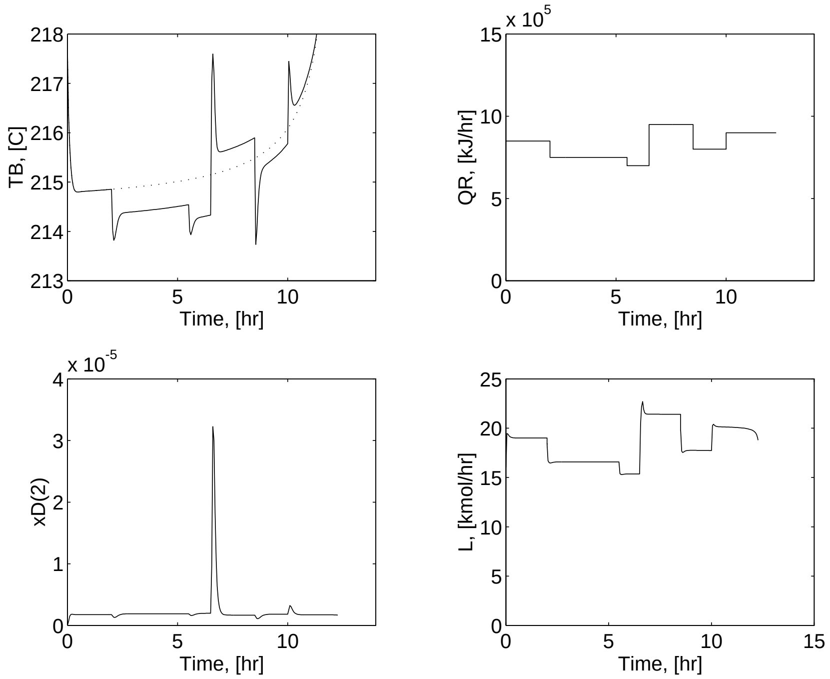

Figure 5.20: One point column control with disturbances in Q_R. Reactor temperature T_B, distillate composition $x_{D,2}$, heat input Q_R and reflux flow L with set point $T_5^{set} = 170^0C$. (Dotted line: Undisturbed case).

product quality. The measuring point should therefore be moved from the top tray and further down in the column. The proposed new strategy of *one-point column control*, where the temperature on tray 5 is controlled, has several advantages:

- No loss of reactant R_2 (compared to controlling the top temperature)
- Need not worry about maximum attainable reactor temperature (compared to controlling the reactor temperature directly by one-point bottom control)
- No interactions with other control loops (compared to two point control)

The temperature profile for one-point column control with disturbances in the system is given in Figure 5.20. It can be seen that also with this control strategy, the deviations in the reactor temperature are small compared with the open-loop strategy in Figure 5.3. The deviations are larger than for one point bottom or two point control. Nevertheless, if the reactor temperature can be allowed to deviate about $\pm 0.5^0C$, one point column control is the simplest and most efficient control strategy.

5.6 Conclusions

A combined batch reactor/distillation process has been presented in this paper. The need for feedback control for reactive batch distillation has been discussed. It has been shown that the same amount of light product should be removed in the separation as the amount being formed by the reaction at any time.

A linearized model has been found to describe the process behavior satisfactory for control analysis purposes. The effect of operating point and disturbances has been discussed based on the linearized model. It has been found that the system's response to changes vary with time as the conditions in the column/reactor change. Also the responses are generally non-linear.

Different control strategies have been presented and were found to give good control of the process also in the presence of disturbances. Controlling the temperature on a tray in the column (one point column control) is found to give good performance for the given process with no loss of reactant and a high reactor temperature although no direct control of the reactor temperature is obtained.

Notation

A	system matrix	
B	system matrix	
C	system matrix	
c_i	concentration of component i in the reactor	$kmol/m^3$
D	system matrix	
D	distillate flow	$kmol/hr$
$\mathbf{d}$	disturbance vector	
E_r	activity coefficient for reaction r	$kJ/kmol$
E	system matrix	
F	system matrix	
$G(s)$	transfer function	
$G_d(s)$	transfer function for disturbances	
h_j^L	liquid enthalpy on tray j	$kJ/kmol$
h_{ji}^L	liquid enthalpy of component i on tray j	$kJ/kmol$
h_j^V	vapor enthalpy on tray j	$kJ/kmol$
h_{ji}^V	vapor enthalpy of component i on tray j	$kJ/kmol$
H_A	liquid holdup in the accumulator	$kmol$
H_j	liquid holdup on tray j	$kmol$
H_j^0	initial liquid holdup on the trays =0.1)	$kmol$
H_N	liquid holdup in the reactor/reboiler	$kmol$
H_C	liquid holdup in the condenser	$kmol$
k_r	rate constant for reaction r	
$k_{0,r}$	frequency factor for reaction r	
K_p	controller gain	
L	reflux flow	$kmol/hr$
L_j	internal liquid flow	$kmol/hr$
L_j^0	initial liquid flow (=15)	$kmol/hr$
N	number of theoretical stages	
n_c	number of components	
n_W	amount of water in reactor	$kmol$
P	pressure	atm
P_B	reactor/reboiler pressure	atm
P_{ji}^{vap}	vapor pressure of component i on tray j	atm
Q_C	condenser heat duty	kJ/hr
Q_R	reboiler heat duty	kJ/hr
r_B	reaction rate	$kmol/hr$
R_g	ideal gas constant = 8.314	$kJ/kmol, K$
R	internal reflux ratio = L/V_N	
t	time	hr
t_f	final operating time	hr
T_b	boiling point temperature	K
T_B	reactor/reboiler temperature	K
T_j	temperature on tray j	K
T_0	reference temperature	K

$\mathbf{u}$	control vector	
V_j	vapor flow from tray j	$kmol/hr$
$\overline{V}$	average reactor volume	m^3
$\mathbf{x}$	state vector	
$x_{j,i}$	mole fraction of component i in liquid on tray j	
$x_{D,i}$	mole fraction of component i in distillate	
$X_{D,2}$	logarithmic mole fraction of comp. R_2 in distillate $= -\ln(1 - x_{D,2})$	
$y_{j,i}$	mole fraction of component i in vapor from tray j	
$\mathbf{y}$	measurement vector	

Greek letters

Δ	deviation from nominal operating point	
τ	hydraulic time constant	h^{-1}
τ_I	integral time for controller	hr
ξ_i	stoichiometric coefficient of component i	
ρ_i	liquid density of component i	$kmol/m^3$

Scripts

B	reactor/reboiler
C	condenser
i	component number
j	tray number
r	reaction number
set	set point
*	nominal value

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Appendix A. Dynamic model

The equations used in this study to describe the dynamic behavior of the combined batch reactor/batch distillation column consist of the total mass balance, the mass balance for each component, the energy balance, tray hydraulics and phase equilibrium and are valid under the following assumptions:

1. Staged batch distillation column with trays numbered from the top

2. Perfect mixing and equilibrium on all stages

3. Constant stage pressures and tray efficiencies
4. Negligible vapor holdup
5. Negligible changes in liquid enthalpies ($dh^L/dt \approx 0$) (algebraic energy balance)
6. Total condensation with no subcooling in the condenser
7. Chemical reaction limited to the reboiler
8. Raoult's law for the vapor-liquid equilibrium
9. Immediate heat input
10. Constant molar holdup in the condenser (perfectly controlled)
11. Linear tray hydraulics

Below, j denotes the stage number and i the component number. The following differential and algebraic equations result:

reboiler, $i = 1, n_c$ and $j = B$:

$$dH_B/dt = L_{N-1} - V_B + \sum_{i=1}^{n_c} \xi_i r_B \tag{5.8}$$

$$d(H_B x_{Bi})/dt = L_{N-1} x_{N-1,i} - V_B y_{Bi} + \xi_i r_B \tag{5.9}$$

$$0 = L_{N-1}(h_{N-1}^L - h_B^L) - V_B(h_B^V - h_B^L) + Q_R \tag{5.10}$$

$$r_B = r_B(c_i, T_B) \tag{5.11}$$

$$c_i = \frac{x_{Bi} H_B}{\sum_{i=1}^{n_c} \frac{x_{Bi} H_B}{\rho_i}} \tag{5.12}$$

column tray, $i = 1, n_c$, $j = 1, N$:

$$dH_j/dt = L_{j-1} + V_{j+1} - L_j - V_j \tag{5.13}$$

$$d(H_j x_{ji})/dt = L_{j-1} x_{j-1,i} + V_{j+1} y_{j+1,i} - L_j x_{ji} - V_j y_{ji} \tag{5.14}$$

$$0 = L_{j-1}(h_{j-1}^L - h_j^L) + V_{j+1}(h_{j+1}^V - h_j^L) - V_j(h_j^V - h_j^L) \tag{5.15}$$

condenser, $i = 1, n_c$ and $j = C$:

$$0 = V_1 - L - D \tag{5.16}$$

$$H_C \cdot d(x_{D,i})/dt = V_1 y_{1,i} - L x_{D,i} - D x_{D,i} \tag{5.17}$$

$$0 = V_1 h_1^V - L h_C^L - D h_C^L - Q_C \tag{5.18}$$

$$L = R \cdot V_1 \tag{5.19}$$

accumulator, $i = 1, n_c$ and $j = A$:

$$dH_A/dt = D \tag{5.20}$$

$$d(H_A x_{Ai})/dt = D x_{Di} \tag{5.21}$$

linearized tray hydraulics, $j = 1, N$ and B:

$$L_j = L_j^0 + \frac{H_j - H_j^0}{\tau} \tag{5.22}$$

equilibrium, $i = 1, n_c$, $j = 1, N$ and B:

$$y_{ji} = k_{ji} x_{ji} \tag{5.23}$$

restriction, $i = 1, n_c$, $j = 1, N$ and B:

$$\sum_{i=1}^{n_c} y_{ji} = 1 \tag{5.24}$$

physical properties, $i = 1, n_c$, $j = 1, N$ and B:

$$k_{ji} = \frac{P_{ji}^{vap}(T_j)}{P} \tag{5.25}$$

$$h_j^L = \sum_{i=1}^{nc} x_{ji} \cdot h_{ji}^L(x_{ji}, T_j) \tag{5.26}$$

$$h_j^V = \sum_{i=1}^{nc} y_{ji} \cdot h_{ji}^V(y_{ji}, T_j) \tag{5.27}$$

$$\rho_i = \rho_i(T) \tag{5.28}$$

The solid polymer product forms a separate phase and is therefore not included in the model. The initial conditions are total reflux and no reaction. Note that the reflux ratio is given as the *internal* reflux ratio $R = R_{internal} = L/V_1$ since this is better solved numerically ($R_{internal} \in [0, 1]$ while $R_{external} \in [0, \infty >$).

Appendix B. Kinetic parameters

Very little data about the reaction were given by the chemical company who provided the case study. Several hundred reactions were assumed to take place in the reactor during the batch. Although, in this study only the overall reaction is considered. The rate of the overall reaction was assumed to be approximately equal to the total amount distilled off divided by the total operating time. The kinetic parameters used in this study were selected based on literature data for reactions similar to the one studied here.

Activation energies for esterification between aromatic carboxylic acid and alcohol are reported in the range 13.8-27.5 kcal/mole for mono acids and alcohols and excess alcohol during reaction (Table of Chemical Kinetics, 1951). For the reactions most similar to the reaction investigated in this study, the activation energy is $E_1 \approx 18$ kcal/kmol. Therefore

$$E = 18 \cdot 4.184 \cdot 10^3 = \underline{7.5 \cdot 10^4 \ \ kJ/kmol}$$

The *frequency factor* for the overall reaction can be found from the rate equation (Eq. 5.1) assuming only forward reaction since $k_{1,0} = k_1$ when $T = T_0$. Only the forward reaction is considered since most of the water is removed by distillation and the reverse reaction will therefore be very slow. The reference temperature used is:

$$T_0 = \underline{500.0 \ \ K}$$

The rate of reaction was found from experiments (chemical company) to be:

$$\begin{aligned}
\frac{dn_W}{dt} &= \xi_W r_B V \\
\frac{\Delta n_W}{\Delta t_f} &\approx \xi_W r_B \overline{V} \\
r_B &= \frac{\Delta n_W}{\Delta t_f} \frac{1}{\xi_W \overline{V}} \\
r_B &= 1.25 \cdot \frac{1}{2 \cdot 1} = 0.625 \ \ kmol/hr
\end{aligned}$$

Here $\overline{V}$ is the average volume in the reactor, ξ_W the stoichiometric coefficient for water and $\Delta n_W / \Delta t_f$ the total amount of water distilled off divided by the total operating time.

The initial concentrations have been calculated from the reactor feed charge and average densities and are given in Table 5.7.

Table 5.7: Initial reactor concentrations.

Comp.	M_W $kg/kmol$	T_m K	T_b K	density $kmol/m^3$	amount $kmol$	volume m^3	concentration $kmol/m^3$
R_1	166.133	700.15	1040.00	9.1	10.4	1.099	4.481
R_2	76.095	213.15	460.75	11.0	7.28	0.664	3.137
R_3	118.176	315.15	516.15	6.6	3.12	0.470	1.344
W	18.015	273.15	373.15	51.2	2.2	0.004	0.948
Total					23.0	2.321	

The frequency factor at initial conditions is

$$\begin{aligned}
k_{0,1} &= \frac{r_B}{c_{R_1}\left(c_{R_2} + c_{R_3}\right)} \\
&= \frac{0.625}{4.48\left(3.14 + 1.34\right)} \\
k_{0,1} &= 0.03
\end{aligned}$$

However, it is assumed that the concentrations in the reactor will decrease with time. The following frequency factor is therefore used in this study:

$$k_{0,1} = \underline{0.06} \tag{5.29}$$

(Note that in Chapter 6, $k_{0,1} = 0.03$).

The given data indicate that a temperature increase of $\Delta T = 19^0$ C is necessary in order to double the reaction rate for the forward reaction.

No expression or data for the reverse reaction has been given in the literature. Since the amount of water in the reboiler will be very low due to the distillation, the rate is usually assumed to be negligible. In this study however, the reverse reaction is to be included. The following assumptions are made:

1. The rate of the overall reverse reaction can be expressed as

$$r_{reverse} = k'_{0,2} c_W c_P \approx k_{0,2} c_W$$

   where the approximation holds since water will be the limiting reactant.

2. The activation energy for the reverse reaction is the same as for the forward reaction

$$E_2 = E_1 = 7.5 \cdot 10^4 \ kJ/kmol$$

3. The frequency factor for the reverse reaction is one fourth that for the forward reaction

$$k_{0,2} = 0.25 k_{0,1} = 0.015$$

The last two assumptions are the same as those made for the esterification reaction between acetic acid and ethanol (Holland, 1981).

The heat of polymerization for poly(butylene terephthalate) is ΔH (kJ/kmol) ≈ 0 (Encyc. of Pol. Sci. and Eng., 1990). It is therefore assumed that the heat of polymerization for poly(propylene/hexylene terephthalate), which is assumed to be the polymer product P, is approximately equal to zero.

Chapter 6

Optimal Control and On-line Operation of Reactive Batch Distillation

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Abstract

While both the dynamic modeling, and to a lesser extent, the optimization of reactive batch distillation have been studied in the past, issues related to control and on-line operation of such a process have not been properly addressed. In this study, a-priori optimal profiles of the operating variables are established for an industrial reactive distillation process. The optimal profiles are found either by maximizing the profitability or minimizing the operating time subject to constraints on the reboiler temperature and the accumulator composition. The control properties at optimal and non-optimal conditions are assessed. An one-point column control scheme with a PI-controller is designed to implement the optimal profiles. The controller performance is tested using a simulation model including disturbances in the reaction model and reboiler heat duty. The fully automatic operation of the process according to the optimal policies is demonstrated in conjunction with both a continuous controller and a real time control system.

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6.1 Introduction

Batch distillation is used in the chemical industry for the production of small amounts of products with high added value and for processes where flexibility is needed. A batch reactor can be combined with a distillation column as shown in Figure 5.1. This combined unit operation, called *reactive batch distillation*, especially suits those chemical reactions where reaction equilibrium limits the conversion in a normal reactor to a low-to-moderate level. By continuously separating low-boiling products from the reactants and other products while the reaction is in progress, the reaction can proceed to a much higher level of conversion. Also, removal of a light product results in a decrease in its concentration in the liquid phase which will increase the temperature and thereby the reaction rate.

While both the dynamic modeling of reactive batch distillation, and to a lesser extent, its optimization has been studied in the past (Egly *et al.* (1979), Cuille and Reklaitis (1986), Albet *et al.* (1991), Mujtaba and Macchietto (1992)), issues related to controllability and control of such a process have not been properly addressed. Some attention to the implementation of optimal control profiles for batch distillation has been given by Quintero-Marmol and Luyben (1992) and Bosley and Edgar (1992). So far, only Sørensen and Skogestad (1992) have considered controllability of reactive batch distillation (see also Chapter 5 of this thesis).

This study addresses several aspects of optimal control of a reactive batch distillation process with reference to a specific industrial application. The control objective is either

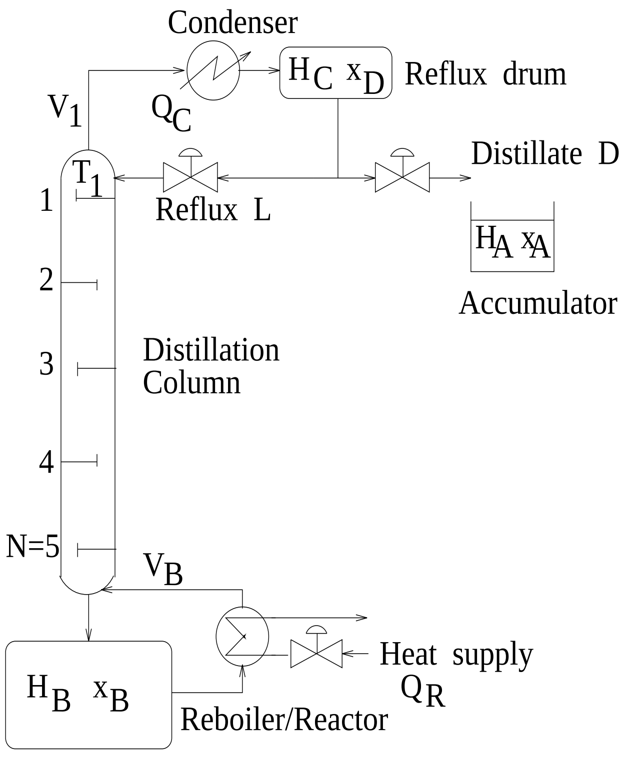

Figure 6.1: Batch distillation column/reactor.

to maximize profitability or to minimize operating time. There are constraints on the maximum allowable reboiler temperature to avoid thermal decomposition of the product and on the minimum loss of volatile reactant in the distillate. Manipulated variables for control are the heat input to the reactor and the internal reflux ratio, Q_R and R respectively. It is assumed that the condenser duty, Q_C, is used for pressure control and the distillate flow, D, for condenser level control. Of particular interest are:

1. Establishing a-priori optimal profiles of the operating variables,
2. Assessing the controllability properties at the optimal conditions,
3. Designing controllers to implement the optimal profiles and
4. Verifying stability and control performance of the controller.

6.2 Reactive batch distillation model

The dynamic model used in this study is valid under the following assumptions:

1. Staged batch distillation column with trays numbered from the top down (accumulator= A, condenser = C, reboiler B)
2. Perfect mixing and equilibrium on all trays
3. Constant stage pressures and tray efficiencies
4. Negligible vapor holdup
5. Negligible changes in liquid enthalpies ($dh^L/dt \approx 0$) (algebraic energy balance)
6. Total condensation with no subcooling in the condenser
7. Chemical reaction limited to the reboiler
8. Raoult's law for the vapor-liquid equilibrium
9. Immediate heat input
10. Constant molar holdup in the condenser (perfectly controlled)
11. Constant molar holdup on the trays (optimizations)
12. Linear tray hydraulics (simulations)

The molar holdup on the trays was assumed constant for the open-loop optimization while a more complex model including linear tray hydraulics was used for simulations and control studies. The linear hydraulics model has the form $L_j = L_j^o + (H_j - H_j^o)/\tau$, where L_j^o and L_j are the initial and current liquid flows and H_j^o and H_j the initial and current molar tray holdup respectively. The parameter τ is the hydraulic time constant.

Table 6.1: Process data for the industrial case study.

Column:	reboiler + 5 trays + total condenser + accumulator
Reaction:	$1.0\ R_1 + 0.7\ R_2 + 0.3\ R_3 \leftrightarrow P(s) + 2.0W$
Reaction rate:	$r_N = r_{R_1} = k_1 c_{R_1}(c_{R_2} + c_{R_3}) - k_2 c_W$
Rate constants:	$k_r = k_{r,0} \exp\left(-\frac{E_r}{R_g} * \left(\frac{1}{T_B} - \frac{1}{T_0}\right)\right)$
Activation energies:	$E_1 = E_2 = 18\ kcal/mol = 7.5 \cdot 10^4\ kJ/kmol$
Frequency factors:	$k_{0,1} = 0.03$ and $k_{0,2} = 0.25 \cdot k_{0,1} = 0.0075$
Reference temperature:	$T_0 = 500.0\ K$
Pressure in column and reactor:	$P = 1\ atm$
Hydraulic time constant:	$\tau = 0.0018\ h = 6.5\ s$
Initial holdups (reboiler, trays, and condenser):	20.6, 0.1, 1.7 $kmol$
Initial reboiler composition:	$x^o(W, R_2, R_3, R_1) = [0.0097, 0.3444, 0.1505, 0.4854]$
Initial tray compositions, $j = 1, N$:	$x^o(W, R_2, R_3, R_1) = [0.999999, 0.000001, 0.0, 0.0]$
Value of product P:	$C_{prod} = 50\ \$/kmol$
Cost of steam:	$C_{heat} = 3\ \$/10^6\ BTU = 2.844 \cdot 10^{-6}\ \$/kJ$
Total cost of raw material:	$C_{raw} = 200\ \$$
Charging and cleaning time:	$t_s = 0.5\ hr$
Total reaction time:	specified or optimized, $t_f\ hr$
Reboiler heat duty:	specified or optimized, $Q_R \in [0, 0.9] \cdot 10^6\ kJ/hr$
Reflux ratio:	specified or optimized, $R \in [0, 1.0]$

The reaction considered is a condensation polymerization reaction between a dibasic aromatic acid (R_1: Terephthalic acid) and two glycols (R_2: 1,2-Propanediol and R_3: 1,6-Hexanediol) which form a polymer (P: Poly(propylene/hexylene)-terephthalate) and Water (W). The polymer forms a separate, solid phase and is therefore not included in the distillation model. High conversion is achieved by distilling off the water. The physical properties data for the liquid components were taken from Coulson and Richardson (1983) for Water and 1,2-Propanediol and from Daubert and Danner (1985) for 1,6-Hexanediol and Terephthalic acid. A summary of the process data are given in Table 6.1.

The same process was also studied in Chapter 5 of this thesis. It should be noted that the parameters used in this chapter are slightly different from the ones in Chapter 5. The rate constants are only half the values used in Chapter 5; $k_{0,1} = 0.03$ here instead of 0.06. Also the initial amounts are slightly different.

6.3 Optimal a-priori operation

The optimal reflux ratio and reboiler heat duty can be found based on some optimization criterion. In this study the optimization problem has been solved according to a maximum profit or minimum time control objective. In general, an optimal control problem can be stated as (Vassiliadis, 1992):

$$\min_{\dot{x}(\cdot),x(\cdot),y(\cdot),u(\cdot),v,x(t_0),t_f} \Phi(\dot{x}(t_f), x(t_f), y(t_f), u(t_f), v, t_f) \tag{6.1}$$

subject to

$$\begin{aligned} g(\dot{x}(t), x(t), y(t), u(t), v, t_f, t) &= 0 & (6.2)\\ h_1(\dot{x}(t_f), x(t_f), y(t_f), u(t_f), v, t_f) &= 0 & (6.3)\\ h_2(\dot{x}(t_f), x(t_f), y(t_f), u(t_f), v, t_f) &\geq 0 & (6.4) \end{aligned}$$

where g are the dynamic model equations and h_1 and h_2 are the equality and inequality constraints respectively. In addition there are upper and lower bounds on the state (x, y), design (v) and control (u) variables and also on the final time (t_f). The optimal control problem is solved by parameterization of the control vector u into a finite number of variables which are optimized using a standard nonlinear optimization technique (SQP). The method includes two levels. The first level performs an integration of the differential and algebraic equations for fixed values of the decision variables. The second level optimizes these decision variables and satisfies end point constraints. A complete solution of the dynamic model for each trial value of the decision variables is required. Note that the term *optimal control* does not involve control in the sense we discuss control later in this paper. The solution to the optimal control problem is a set of *optimal operating parameters* which may subsequently be used as set points for the actual controllers in on-line operation.

In the following we will consider two formulations of the maximum profit problem where zero cost of raw material is assumed in the first but not in the latter. Also, a minimum time control objective is considered.

6.3.1 Maximum profit problem I - zero cost of raw material

The optimal operating policy in terms of maximum profit can be determined by optimizing the overall hourly profit [$/hr]:

$$\max_{R,Q_R,t_f} P^{\$} = \frac{\left(C_{prod}A_{prod} - C_{heat}\int_0^{t_f} Q_R(t)dt\right)}{t_f} - C_{fixed} \tag{6.5}$$

where C_{prod} is the value of product P ($\$/kmol\ monomer\ block$), A_{prod} is the amount of product P at t_f ($kmol$), C_{heat} is the cost of steam to the reboiler ($\$/kJ$), Q_R is the reboiler heat duty (kJ/hr), t_f is the final time (hr), C_{fixed} are the fixed costs ($\$/hr$) and $P^{\$}$ is the profitability ($\$/hr$). Zero cost of raw material is assumed (see Section 6.3.2 where this cost is included). Note that since the initial charge is fixed, and we assume all unreacted feed is recycled to the next batch at no cost, this will not affect the location of the optimum. We use $C_{fixed} = 0$ since the optimum will also be independent of the fixed costs. A single discretization interval is used with constant parameters resulting in three variables (R, Q_R and t_f) for the optimization. The constraints are:

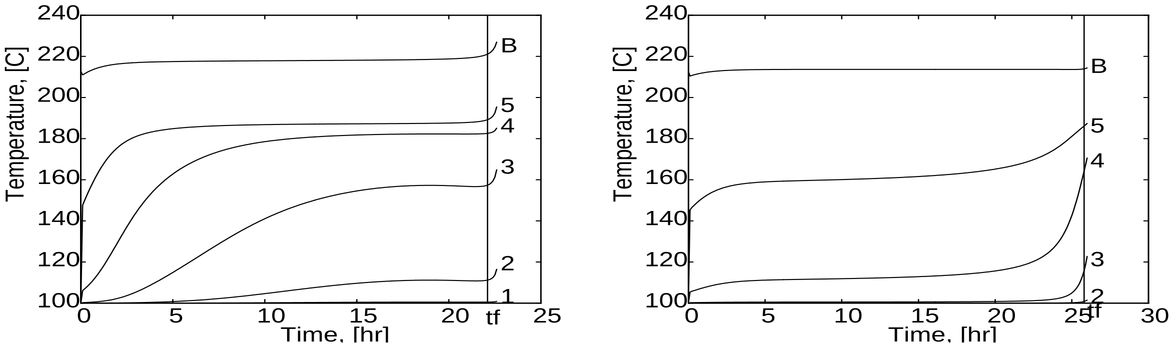

Figure 6.2: Temperature profiles for a) optimal and b) sub-optimal conditions (Maximum profit problem I - zero cost of raw material).

1. Negligible loss of reactant R_2 in the accumulated distillate

$$x_{A,W}(t_f) \geq 0.999 \tag{6.6}$$

2. Reboiler temperature below $240^oC = 513.15\ K$ to avoid thermal decomposition of the end-product

$$T_B(t_f) \leq 513.15 \quad K \tag{6.7}$$

Simulations have shown that the reboiler temperature is highest at the end of the reaction, so keeping the endpoint temperature constraint is sufficient. If this was not the case, the constraint on the reboiler temperature could be added as a path constraint instead of as an end-point constraint.

With the process data given in Table 6.1 the optimal values are:

Optimal parameters			Optimal results			
R	Q_R kJ/hr	t_f hr	$P(t_f)$ $\$/hr$	$x_{A,W}(t_f)$	$T_B(t_f)$ K	$\chi_{R_1}(t_f)$
0.950	$0.747 \cdot 10^6$	22.1	19.9	0.9997	494.4	0.97

The parameter χ_{R_1} is the conversion of reactant R_1. Solutions with different values of the monomer product, e.g. $C_{prod} = 100\ \$/kmol$ or $C_{prod} = 25\ \$/kmol$, give almost the same optimal results.

The temperature profiles for the process, given the optimal operating parameters, are presented in Figure 6.2.a. The reboiler temperature is more or less constant during the whole batch but the temperature in the top section of the column increases slowly. There is a sudden increase in temperature on all trays immediately after the optimal final time. The end of the reaction in the real plant can therefore be easily detected using temperature measurements on any one of the trays or in the reboiler.

When using sub-optimal values for R and Q_R (e.g. $R = 0.954$ and $Q_R = 0.691 \cdot 10^6$ kJ/hr), a different response is obtained (Figure 6.2.b). The constraints are still satisfied but the previous optimal conversion is only reached at t=25.8 hr. The profitability is

15% lower, at 16.9 $\$/hr$. The temperatures for this sub-optimal operation are lower than under optimal conditions showing that we are over-refluxing the column. Also, detection of the termination conditions based on temperature alone is much more difficult.

Note that the optimum with respect to time is very flat. Indeed, ending the batch after 16 hr (25% time saving) still yields 99% of the optimal profitability with a conversion of R_1 of 0.71. The reason for this insensitivity to the value of t_f, is that we have assumed that there is no cost involved in recycling the unreacted feed since the cost of raw material is neglected.

6.3.2 Maximum profit problem II - fixed cost of raw material

Next consider a slightly different objective function where the cost of raw material is taken into consideration. Also a charging and cleaning time of $t_s = 0.5\ hr$ is included:

$$\max_{R,Q_R} P^{\$} = \frac{\left(-C_{raw} + C_{prod}A_{prod} - C_{heat}\int_0^{t_f} Q_R(t)dt\right)}{t_f + t_s} - C_{fixed} \tag{6.8}$$

where C_{raw} is the total cost of raw material ($) and t_s is the charging and cleaning time (hr). With this objective function and the process data given in Table 6.1 the optimal values are given below for the case with one discretization interval and constant or linear operating parameters where $t_o = 0$ is the start of the batch and t_f the final time:

	Optimal parameters			Optimal results			
	R	Q_R kJ/hr	t_f hr	$P(t_f)$ $\$/hr$	$x_{A,W}(t_f)$	$T_B(t_f)$ K	$\chi_{R_1}(t_f)$
constant	0.933	$0.545 \cdot 10^6$	23.0	10.99	0.9990	508.5	1.00
linear	$0.9317(t_o) \rightarrow 0.9325(t_f)$	$0.533(t_o) \rightarrow 0.548(t_f) \cdot 10^6$	23.0	10.993	0.9990	508.0	0.998

The temperature profiles for both cases are given in Figure 6.3. With the cost of raw material included in the objective function, the optimum is no longer flat. When compared to the optimal results from the previous problem (Eq. 6.5), it can be seen that both the reflux ratio and the reboiler heat duty are reduced. However, the total operating time is increase from 22.1 hr to 23 hr. The reboiler heat duty is reduced by more than 25%. This is because the relative influence of the energy consumption is larger when the cost of raw material is included with the same costs as in the previous problem. Note that the reaction is now run almost to complete conversion. As a result, the final reactor temperature has increased.

The optimal solutions for constant or linear operating parameters are very similar. However, the energy consumption is slightly reduced for linear parameters. Optimal solutions with up to 4 intervals have also been found with optimal profitabilities slightly higher than those presented here (not shown).

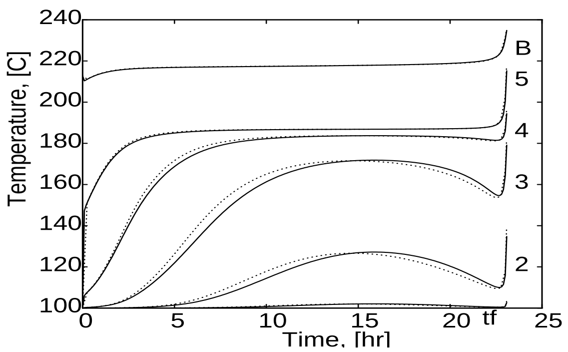

Figure 6.3: Temperature profiles for optimal conditions with constant (dotted lines) and linear operating parameters (solid lines) (R and Q_R) (Maximum profit problem II- fixed cost of raw material).

6.3.3 Minimum time problem

Above the optimal performance of the process was defined in terms of maximum profit. Alternatively, a minimum operating time can be used as optimization criterion:

$$\min_{R,Q_R} t_f \tag{6.9}$$

where t_f is the minimum operating time, subject to the same constraints on the maximum loss of reactant R_2 and maximum reactor temperature (Eq. 6.6-6.7). In addition there is a constraint on the minimum conversion

$$\chi_{R_1} \geq 0.99 \tag{6.10}$$

With the process data given in Table 6.1, the optimal solution to the minimum time problem is given below for the case with one discretization interval and constant operating parameters:

Optimal parameters			Optimal results			
R	Q_R kJ/hr	t_f hr	$P(t_f)$ $\$/hr$	$x_{A,W}(t_f)$	$T_B(t_f)$ K	$\chi_{R_1}(t_f)$
0.957	$0.9 \cdot 10^6$	22.1	10.576	0.9990	508.5	0.99

In Figure 6.4, the temperature profile for the minimum time solution is compared to the optimal profile from the maximum profit problem with fixed cost of raw material (Eq. 6.8). The reboiler heat duty is now at its maximum value which is as expected since the objective is to run the batch in minimum time regardless of costs. As a result, the reflux ratio is also increased to maintain the specified product purity. However, the profitability is slightly decreased which is mainly due to the higher energy consumption but also to the lower conversion ($\chi_{R_1}(t_f) = 0.990$ versus $\chi_{R_1}(t_f) = 0.998$ for the maximum profit problem). The temperatures profiles in the column are slightly increased compared to the maximum profit solution.

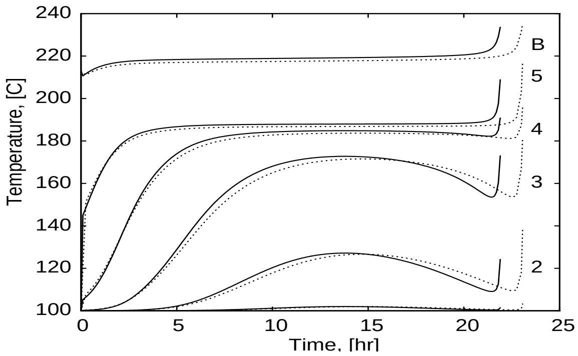

Figure 6.4: Temperature profiles for optimal conditions with constant linear operating parameters R and Q_R. ($-$: Minimum time problem, $\cdot\cdot$: Maximum profit problem II - fixed cost of raw material).

6.4 Implementation of the optimal policies

Since the formation of product resembles a continuous feed to the reboiler (Sørensen and Skogestad, 1992), the issues involved in control of a reactive batch column are more similar to conventional continuous distillation control than to batch distillation control. In a conventional batch column one may usually implement the optimal values of R and Q_R in an "uncontrolled" open-loop fashion. However, for reactive distillation the operation is usually much more sensitive to the exact value of the reflux ratio R. A slightly too low R may result in a distillate flow which is larger than the amount of light component (W) formed in the reaction, and the difference must be provided by loss of the intermediate boiling reactant (R_2). Therefore there is a need to implement the optimal policy in some feedback fashion which maintains control of the distillate composition.

We propose to keep Q_R constant at its optimal value and adjust R to control the temperature on some tray inside the column. The temperature measurement should be placed on a tray with a large response to changes in the controller input. By controlling the temperature on one of the trays one will also to some extent indirectly control the reactor temperature since there is a strong coupling between the compositions in the column (Sørensen and Skogestad, 1992). (See Chapter 5 of this thesis for a discussion of controllability and control strategies for reactive batch distillation.)

6.5 One-point column control

We now consider implementing the optimal solution to the maximum profit problem I (where cost of raw material was neglected). Various disturbances are introduced in the simulation model to test the sensitivity to process changes (Figure 6.5.a). The disturbances are assumed to be in:

1. Reboiler heat duty Q_R

2. Frequency factors for the forward and reverse reaction, $k_{0,1}$ and $k_{0,2}$

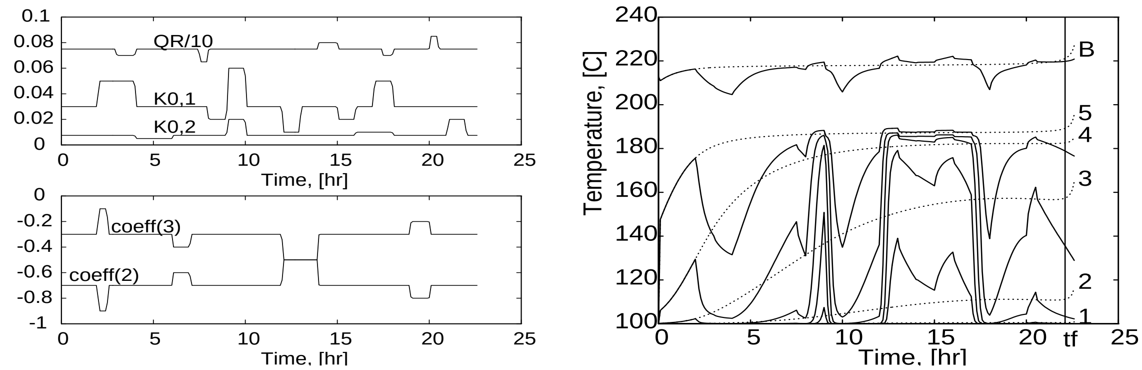

Figure 6.5: a) Disturbances in the process and b) temperature profiles for the uncontrolled process (dotted lines: undisturbed profiles).

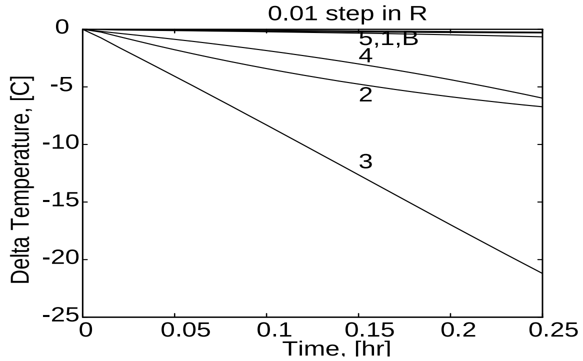

Figure 6.6: Linear temperature responses for a 1% step in reflux ratio R (dynamic model linearized at $t = 15\ hr$ with the optimal temperature profile in Figure 6.2.a.)

3. Stoichiometric coefficients of the alcohols (reactant 2 and 3) in the rate equation

Disturbances 2 and 3 are used to represent some uncertainty in the reaction mechanism.

First consider the uncontrolled case where R is kept constant. The temperature profiles (Figure 6.5.b solid lines) deviates significantly from the optimal profiles (Figure 6.5.b dotted lines). The disturbances yield breakthrough of the intermediate component resulting in some loss of reactant R_2 ($x_{A,W} = 0.989$ for the accumulated distillate) and the batch therefore does not satisfy the specifications. More severely, the reboiler temperature deviates up to 12^o C from the optimal one which may result in a varying quality of the product P between batches.

Next consider implementing the optimal policy in a feedback fashion by varying the reflux ratio R to keep a selected tray temperature constant. The reflux ratio R is the manipulated variable and Q_R is kept at its optimal value. The temperature measurement is placed on tray 3. The selection of measurement point is based on a comparison of the responses in tray temperatures to changes in the controller input R. Figure 6.6 illustrates typical responses at a given operating point. (See Chapter 5 of this thesis for a further discussion of controllability of reactive batch distillation.) Note that the tray responses found here are different from the ones presented in Chapter 5 where it was recommended

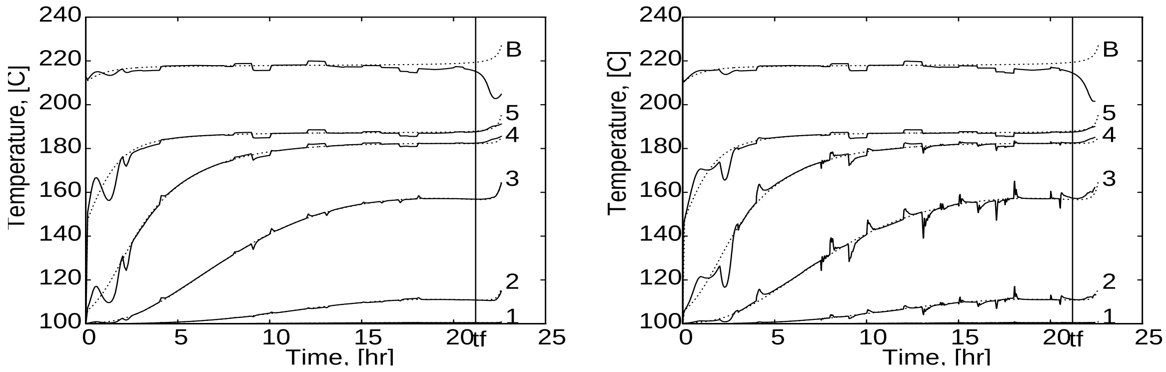

Figure 6.7: Temperature profiles using a) continuous controller and b) real time control system.

to place the temperature measurement on tray 5. This is because the nominal temperature profiles in Figure 6.6 are different from the ones studied in Chapter 5.

The set point for the temperature controller is then the optimal temperature profile for tray 3 as a function of time (fitted to a polynomial). A time delay of 15 seconds was added to the temperature measurement and the controller settings used are $K_p = -0.01$ and $\tau_i = 0.1$. The controller is tuned using a linearized model at given operating points (see Chapter 5).

As can be seen from the results in Figure 6.7.a. (solid lines), good control is achieved. The deviation in the reboiler temperature from the optimal profile (dotted lines) is small during most of the batch, but increases up to 20^o C towards the end. The optimal conversion ($\chi_{R_1} = 0.97$) is reached at $t = 21.2$ hr. After this time the reboiler temperature starts drifting away from the optimal value. For the controlled process (solid lines), the reboiler temperature is decreasing instead of increasing as for the optimal process (dotted lines). This is because the controller maintains the temperature on tray 3 at its set point as given by the optimal profile. The uncontrolled temperature on tray 3 increases with time as the reaction proceeds. But due to the disturbances, the reaction rate is quicker than under optimal conditions. Keeping the temperature at the optimal set point will therefore hold light component back in the lower part of the column causing the reboiler temperature to decrease.

Detecting the end of the reaction based on the criteria from the optimal temperature profile, that the reaction is ended when the temperatures start rising rapidly, can not be applied in this case. However, the fact that the reboiler temperature is drifting away from the optimal in either direction is an indication that the reaction is ended. The end point can therefore be estimated to when the reboiler temperature deviates from the optimal value on either side by an amount greater than, for example 5^oC.

The simulated process was also interfaced to an industrial real-time control system Paragon (1991). All controllers were implemented using Paragon's own facilities. Instead of using a polynomial to describe the simulation temperature set point profile, only 16 measurement points were used. The sampling time was 1 minute. As shown in Figure 6.7.b, good control is achieved and the responses are very similar to those obtained with the simulated continuous controller.

6.6 Conclusions

The optimal operating values for reflux ratio and reboiler heat duty have been determined according to the maximum profit and minimum time control objective for a batch distillation column with reaction in the reboiler. The optimal policy must be implemented using feedback control to avoid loss of reactant ("break-through") in the distillate waste product. We select to control an intermediate column temperature which location is selected based on a sensitivity analysis of the optimal temperature profiles. The column has been controlled using both a simple continuous PI-controller and a real time control system, with disturbances in the reaction parameters and in the reboiler heat duty. Good performance has been achieved in both cases. For this industrial application it has been found that the end of the reaction can be detected based on temperature measurements in the reboiler. An alternative to feedback control is an open-loop policy with over-refluxing but this may lead to significant cost penalties.

Notation

A_{prod}	amount of product P	$kmol$
c_i	concentration of component i in the reactor	$kmol/m^3$
C_{fixed}	fixed costs	$\$/hr$
C_{heat}	cost of steam	$\$/kJ$
C_{prod}	value of product P	$\$/kmol$
C_{raw}	total cost of raw material	$\$$
E_r	activity coefficient for reaction r	$kJ/kmol$
g	dynamic model equations in the general optimization problem	
h_1	equality constraints in the general optimization problem	
h_2	inequality constraints in the general optimization problem	
H_j	liquid holdup on tray j	$kmol$
H_j^o	initial liquid holdup on the trays =0.1	$kmol$
k_r	rate constant for reaction r	
k_{0r}	frequency factor for reaction r	
K_p	PI-controller gain	
L	reflux flow	$kmol/hr$
L_j	internal liquid flow	$kmol/hr$
L_j^o	initial liquid flow =15	$kmol/hr$
L_D	distillate flow	$kmol/hr$
N	number of theoretical stages	
n_c	number of components	
P	pressure	atm
$P^{\$}$	profitability	$\$/hr$
Q_R	reboiler heat duty	kJ/hr
r_N	reaction rate	$kmol/hr$
R_g	ideal gas constant = 8.314	$kJ/kmol, K$
R	internal reflux ratio = L/V	
t	time	hr
t_o	start time	hr
t_f	final operating time	hr
t_s	charging and cleaning time	hr
T_B	temperature in reactor	K
T_0	reference temperature	K
u	control variables in the general optimization problem	
v	design variables in the general optimization problem	
V_j	vapor flow from tray j	$kmol/hr$
x^o	initial composition	
$x_{A,W}$	mole fraction of component W in the accumulator	
x	state variables in the general optimization problem	
y	algebraic variables in the general optimization problem	

Greek letters

Δ	deviation from nominal operating point	
τ	hydraulic time constant	h^{-1}
τ_I	integral time for PI-controller	hr
Φ	objective function in the general optimization problem	
ξ_i	stoichiometric coefficient of component i	
χ_{R_1}	conversion of reactant R_1	

Scripts

i	component number
j	tray number
r	reaction number

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Chapter 7

Discussion, Conclusions and Directions for Future Work

7.1 Discussion

This thesis addresses different aspects of operation and, to a lesser extent, control of batch distillation columns. The research has been motivated by the need for more economical operating policies for batch columns. With an increased demand for flexibility and productivity in the fine chemical and pharmaceutical industries, optimal operation of batch distillation has become increasingly important. However, the operating policy and the process equipment limit the attainable performance. This thesis has therefore aimed at finding other ways of operating conventional batch columns. Alternatives to the traditional column configuration have also been considered.

Increased product quality has been an incentive to gain a better understanding of the behavior of reactive batch distillation, which subsequently may lead to improved control of this integrated process. A batch reactor is combined with a distillation column in order to increase productivity. However, combined processes are often difficult to operate due to interactions between the units. In this thesis, the dynamic behavior and controllability of an industrial case study has been studied. It suggests alternative control strategies to the conventional top-temperature control strategy which is found to be less suitable for batch columns. The thesis concludes with a discussion of optimal operation of the same case study. On-line operation according to an optimal policy is demonstrated in conjunction with both a continuous and a real time control system.

In this thesis, relatively simple dynamic models have been considered. This is mainly due to limitations in the optimization software. No commercial optimization programs have been available, and the program used is complicated and not very user friendly. However, modeling issues have not been a priority as long as the models have sufficed to illustrate important principles.

Controllability and control strategies of reactive batch distillation have only been considered in relation to a specific industrial process. It can be argued that at least controllability should be considered for other applications as well. However, there was not enough time to treat this in full.

7.2 Conclusions

The main contributions of this thesis are summarized below:

Chapter 2 discusses optimal startup of batch distillation columns. The actual duration of the startup period is found to be of limited significance unless the startup time is very different from the optimal value. A short cut formula is presented which can provide an estimate for the required startup time based on operating parameters which are normally known a priori. Alternative methods of reducing the startup time, by changing the operating procedure or the process equipment, have been considered. Neglecting the initial filling of the column section and the condenser is a normal assumption in the optimization literature. It is established that this is acceptable in most cases if the objective is to describe the duration and not the actual trajectories of the startup period.

Chapter 3 is the first major work to investigate the cyclic operating policy. This policy is compared to conventional operating policies and is found to significantly reduce the operating time for difficult separations. Both an optimal and a simplified cyclic policy is presented. The simplified policy is easy to calculate and implement and is found to yield solutions very close to those from the very time consuming rigorous dynamic optimization. The effect of condenser holdup on operating time is discussed.

Chapter 4 presents a comparison between regular and inverted batch distillation. In an inverted column, the feed is added to the condenser drum and the product is withdrawn from the reboiler. This column configuration is found to yield a shorter operating time than the conventional regular configuration for separations where the light component is present in a small amount in the feed mixture. For some mixtures where the separation is not possible in a regular column, good performance can be achieved in an inverted column. The operation is viable, but more work is needed.

Chapter 5 is the first work to treat controllability and control strategies for reactive batch distillation. An industrial esterification process is used as an example. When a reactor is combined with a distillation column, the reaction will act as a continuous feed during the batch. The same amount of light component should be removed as the amount being formed by the reaction at any time. The controllability of the process is studied, based on linearized models at different operating points. It is found to be very non-linear and also to depend strongly on the operating conditions and on the time during the batch. The importance of feedback control is demonstrated. Several control strategies are presented as alternatives to the conventional top-temperature control strategy which is found to be less suitable for batch columns.

Chapter 6 treats optimization according to the maximum profit and minimum time control objective of the same reactive batch distillation example as in Chapter 5. The optimal policy is implemented using feedback control with disturbances included in the dynamic model to study the performance of the controller. Both a continuous PI-controller and a real time control system have been used and good performance has been achieved in both cases. For this industrial application, the end of the reaction can be detected based on temperature measurements in the reboiler.

An overall conclusion for Chapters 2, 3 and 4 is that conventional batch distillation is not well suited for difficult separations or for separations where the amount of light component in the feed charge is small. For such separations, inverted columns or cyclic operation may be preferred.

7.3 Directions for future work

7.3.1 Continuation of the work in this thesis

Optimal operation and control of batch distillation columns have been considered in this thesis. However, several aspects have not been treated in full. The following topics should be further investigated:

1) The short cut formula for estimation of startup time needs to be evaluated for industrial separations. Improvements in the derivation of the formula are most probably needed. (Chapter 2.)

2) Filling the condenser drum with light product initially to reduce the startup time was considered for one case problem using a constant reflux ratio policy. The possible benefits from this procedure need to be elaborated further (Chapter 2.)

3) In the discussion of the cyclic operating policy, the influence of condenser holdup was briefly considered but can be further investigated. (Chapter 3.)

4) A further discussion is needed on the practical implementation of the cyclic policy. (Chapter 3.)

5) The use of inverted columns was discussed for one set of operating parameters but must be further investigated before definite statements can be made about its merit. (Chapter 4.)

6) Only simple control strategies were considered for the control of a reactive batch distillation example. However, it would be worthwhile to also consider other strategies, for example cascade control or gain scheduling. (Chapters 5 and 6.)

7.3.2 Optimization of batch distillation

This thesis has discussed possible operating modifications for improving the optimal operation of a single batch column. Both the boilup rate and the condenser holdup have been used as degrees of freedom. However, there are still many aspects of optimal operation that have not been considered.

1) Many applications, and especially azeotropic separations, are run under variable pressure. This parameter clearly affects the optimal operation of the process and should be considered for optimization.

2) Off-cuts are often produced in batch distillation. These are either recycled to the feed of the next batch or re-processed separately. Even for simple mixtures there are

several alternative operations possible. With complex trade-offs as a result of the many degrees of freedom available, there is ample scope for optimization.

3) Addition of feed during the batch is often done in practice. Optimization of the amount of additional filling and the charging times should be considered. This also applies to the addition of reactant during the course of reactive batch distillation.

4) Batch columns are often used for mixtures for which they are not designed. The product must in some cases be concentrated in several steps when the separation abilities of the given column is not sufficient. Again, there are several trade-offs and a proper procedure for optimal operation may increase the productivity of this type of separations.

5) The column characteristics have been pre-specified in all the work in this thesis. The process equipment clearly limits the optimal performance and there is therefore ample scope for optimal design of batch columns.

7.3.3 Alternative operating policies and configurations

The optimal operation of a given column is limited by the operating policy and the process equipment. Alternative operating policies and configurations should therefore be considered.

1) In this thesis a new operating cyclic policy has been presented. However, this policy is sensitive to variations in the feed mixture. A combination of operating policies may be a possible way to exploit the benefits from the cyclic policy and at the same time ensure a flexible operation.

2) Other column configurations have been presented in the literature. In *complex columns*, the feed is added to the middle of the column and the products are withdrawn from the top and the bottom either simultaneously or alternately. This configuration should be compared to the conventional one in terms of flexibility and productivity. Also, control aspects must be considered. Placing several columns in series, where the top flow of one column acts as a bottom feed to the next, may be beneficial for multicomponent mixtures. However, this configuration is very sensitive to disturbances. Good operating and control strategies are therefore needed.

7.3.4 Controllability and control of batch distillation

This thesis has discussed controllability and control strategies for reactive batch distillation. An industrial process was used as an example. However, there is a need for further research in this area.

1) Dynamic behavior and controllability of reactive batch distillation where the reaction takes place in the whole column, should be considered and in particular, the influence of the reaction parameters.

2) Controllability of regular batch distillation has been studied briefly by the author, although no results have been presented in this thesis. The controllability is found to depend on the operating conditions and the time during the batch as for the reactive example studied in this thesis. However, a systematic analysis is needed in order that general conclusions can be made.

3) The choice of type and placement of measurements is clearly of importance but is seldom given much consideration. Placement of temperature measurements, for example, should be based on a controllability analysis of the process.

4) Most industrial batch columns have rather simple control systems. More advanced systems, such as multivariable feedback controllers, Model Predictive Control etc., might increase the operability of the columns.

7.3.5 Application of methods to industrial problems

Research on specific processes is of limited use if the methods developed can not be applied in practice. Although experiments on operating columns often yield case-specific results, it is important to confirm results found from simulations by performing experiments on a real column. In particular, the results on the cyclic operating policy and the inverted column configuration presented in this thesis need to be verified by experiments. In addition, one may in some cases discover new and interesting effects through experiments which may have been overlooked in a simulation model. Nevertheless, one should also be aware of the possibility for alternative separations methods which may be more beneficial than batch distillation for certain separations.

7.3.6 Azeotropic batch distillation

This thesis has been limited to a discussion of ideal mixtures. However, batch distillation is often used in industry for separation of azeotropic mixtures which are inherently difficult to separate. The order of product fractions may also vary with feed composition. There is obviously a need for research in this area. Of particular importance is the selection of entrainers which may be added to break the azeotropes and the decision where and when these should be added.