OPTIMAL OPERATIONAL POLICIES IN BATCH DISTILLATION

by

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To my mother

"Father (PhD, 1971) and son (PhD, 1989) treaded the same path - only time and the Queen's Tower separated them"

- Author

ABSTRACT

Optimal operational policies are studied for batch distillation using a simulation model and optimal control techniques.

A realistic dynamic column model is used, which includes rigorous mass and energy balances, holdups, chemical reaction and general physical property models. The model consists of a mixed system of differential and algebraic equations (DAEs) which is solved without restructuring by a Gear type solver. The model is checked against several simulated and experimental data from the literature and found to be sufficiently accurate while requiring relatively modest computation times.

Optimal operational problems for batch distillation are formulated as optimal control problems using control variable parameterization and nonlinear programming optimisation which are solved by efficient SQP methods. Different kinds of objective functions and constraints are presented. Reflux ratio is chosen as the control variable which is discretized in different time intervals. Application of the method to realistic operational problems achieves large improvements in productivity compared to more conventional mode of operation.

The problem of choosing a recycle policy for an intermediate off-cut (whether to recycle, when and how much to recycle) for binary mixtures is formulated as a two level optimal control problem to minimize the overall distillation time. Also an efficient and robust one level optimal control problem is formulated for this purpose and the results are compared with those obtained by classical two level formulation. A measure, **q**, of "the degree of difficulty" of separation is used to decide whether or not an off-cut is needed.

A general optimal control formulation is considered for establishing optimal off-cut recycle policies in multicomponent batch distillation. Several difficulties in solving such problem are discussed. Some special cases are identified where the methods developed for binary mixtures can be applied to obtain recycle policies for multicomponent mixtures. The previously mentioned measure **q** of "the degree of difficulty" of separation is used to identify these special cases. A new approach for handling off-cut recycle in multicomponent distillation is proposed which is physically and thermodynamically sensible. In this approach each off-cut following a particular main cut from a given batch is recycled (if recycle is found to be worthwhile) by mixing it with the next batch just before the production of that particular main cut is initiated. The same procedure is followed for all the off-cuts in the process.

Application of the method to typical example problems demonstrates the idea and shows large improvement in column performance with such type of off-cut recycle.

The role of holdup has been the subject of controversy and conflicting unexplained effects on separation were reported in the past. Here a systematic study is performed of the effects of column holdup for binary mixtures. It is shown that increasing holdup in some cases improves column performance and that the effect is in other cases detrimental. Both effects are explained and correlated in terms of the previous measure **q** of "the degree of difficulty" of separation. This allows identifying an optimum holdup required to achieve the best performance of a column, for a given separation.

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Chapter 1

INTRODUCTION

1.1 Batch Distillation and its Operation

Batch distillation is, perhaps the oldest operation used for separation of liquid mixtures . Although for centuries batch distillation was used in a very wide field of applications from food, pharmaceutical to petroleum industries, continuous processes became the standard as it became necessary to produce large amount of products quickly and cheaply in the early forties. But because of renewed emphasis on the production of small amounts of products with high added value, batch processes are again becoming important nowadays. A recent survey by Parakrama (1985) shows that just in UK there are 99 batch processes in 74 companies. For a number of applications batch distillation offers advantages over continuous distillation. With only one batch distillation column it is possible to separate mixtures of n_c components that, with continuous distillation, require n_c-1 columns. Where it is necessary to treat economically small and variable quantities of materials with very different compositions and low throughputs, batch distillation becomes irreplaceable.

A batch distillation operation involves charging a still with the material to be separated and carrying out the fractionation until a desired amount has been distilled off. The overhead composition varies during the operation and usually a number of cuts are made. Some of the cuts are the desired products while others are intermediate fractions that can be recycled to subsequent batches to obtain further separation. A residual bottom fraction may or may not be recovered as a product.

1.1.1 General Operations:

The essential features of a conventional batch distillation column (Figure 1.1) are -

a) A reboiler which is charged with feed to be processed and which provides the heat transfer surface.

b) A rectifying column (either a tray or a packed column) superimposed on the reboiler, which is coupled with either a total condenser or a partial condenser system.

c) A series of product accumulator tanks connected to the product streams to collect the main and/or the intermediate fractions.

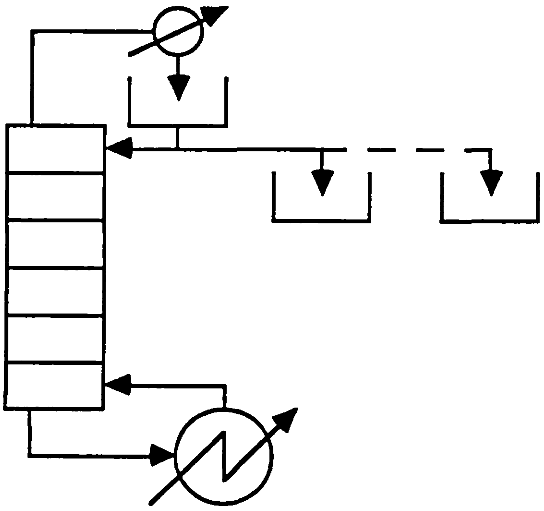


Figure 1.1 : A Conventional Batch Column

Operation of such a column can be conveniently described in two parts :

1. Start-Up period.
2. Product period.

1.1.1.1 Start-Up Period: In practice, an empty column is started up in the following sequence:

1) The reboiler is charged with the material to be processed and heat is applied to it to bring the material to its bubble point temperature.

2) Depending on the reboiler duty, a part of the material is vaporised and the vapor travels upward both through the plate holes and the downcomers and almost instantly reaches the condenser.

3) At this time, the coolant valve is opened and the condensed liquid is stored into a reflux drum. The reflux valve is opened when the liquid fills the condenser holdup tank. At this point some product may also be collected simultaneously.

4) The liquid begins to flow into the top plate and collects on the plate because of the retention made by the vapor flow. When the liquid level passes the weir height (thus filling the plate), the liquid begins to fall to the plate below and the same phenomenon is repeated until the reboiler is reached. In practice some liquid also trickles down from the plate holes when the flows are initially established.

5) If no product was withdrawn in step 3, the column is now run under total reflux operation until the unit is taken to a steady state or to a state when the distillate composition reaches the desired product purity.

The duration of the first step can usually be considered negligible compared to the overall batch distillation time, whereas the duration of steps 2-5 is important and in some cases it may take a long time to reach a steady state or the desired initial distillate composition (Holland and Liapis, 1983; Nad and Spiegel, 1987; Ruiz, 1988).

1.1.1.2 Product Period: Generally the product period starts when distillate removal from the process is begun. The operation in the product period and its duration depends on the requirements of the product. This period can be operated under the following conditions:

1. The start-up period is ended when the desired distillate purity is reached. Product take off is started and the product is collected at constant composition by varying the reflux ratio until a specified amount of distillate has been collected. This type of operation is known as "variable reflux operation" or "constant distillate composition operation". In this mode of

operation the reflux ratio is such that always produces on-specification material, stopping the fraction when the reflux ratio has climbed to some value considered to be "uneconomic".

2. The total reflux start-up period is ended when the unit reaches its steady state. Product is collected at some constant finite reflux ratio until the accumulated product composition reaches its desired purity. This type of operation is very common in practice and is known as "constant reflux operation". Under this operation mode the column is operated on a fixed reflux ratio for the whole fraction (cut), producing better than specification material at the beginning and distillate below specification at the end of the fraction.

The above two types of operations are referred to as "conventional" methods of operation in the literature.

3. A third type of operation is a trade off between the above two types of operation. Here an optimal reflux policy is chosen so that some objective function is satisfied (minimum time, maximum product, maximum profit etc.), subject to any constraints (product amount and purity) at the end of the process.

1.1.2 Specific Operations:

The column under consideration may be run during the start-up or product periods using various strategies :

1.1.2.1 Constant Vapor Boilup Rate : In this mode of operation the vapor rate out of the reboiler is held constant throughout the operation by continuously increasing the reboiler heat duty as the reboiler composition changes (Coward, 1967; Mayur et al., 1970; Mayur and Jackson, 1971; Kerkhoff and Vissers, 1978; Domenech and Enjalbert, 1981; Diwekar et al., 1987; etc.)

1.1.2.2 Constant Condenser Vapor Load: Vapor load to the condenser is kept constant throughout the operation in this mode. Nad and Spiegel (1987) maintained this constancy of vapor load in their experimental column by making an enthalpy balance around the condenser system and running the column at piecewise constant reflux ratio.

1.1.2.3 Constant Distillate Rate: This mode of operation demands constant rate of distillate throughout. This means that, for constant reflux ratio operation, the vapor load to the condenser is also constant. This type of operation was considered by Boston et al. (1980), Holland and Liapis (1983).

1.1.2.4 Constant Reboiler Duty: In this mode heat input to the reboiler is held constant throughout. In practice it is set to its maximum limit, the value of which depends on the heat exchange system to the reboiler. Domenech and Enjalbert (1974) used this mode of operation in their laboratory batch column.

Other types of column operation include:-

1.1.3 Cyclic Operations:

Cyclic operation is characterized by two modes of operation, called "transient total reflux" and "stripping". During the total reflux portion of the cycle, liquid reflux is returned to the column, but no product is withdrawn; and during the stripping portion of the cycle, the product is withdrawn; but no reflux is returned to the column. The models and calculation procedures are given by Barb (1967) and Barb and Holland (1967). The extreme difficulty of accurate measurement and control of small flow rates in laboratory columns strongly favor cyclic operation. The details of this operation were also presented by Holland and Liapis (1983).

1.1.4 Intermediate "cuts" and Recycle Operation:

Intermediate fractions (off-cuts) from a batch distillation operation which neither meet top nor bottom product composition requirements are often stored and recycled to subsequent batches for reprocessing. It was found in the past that the recycling of material improved the column performance to a great extent depending on the difficulty of separation of a given mixture in an existing column. This type of operation is particularly useful when using an existing column with perhaps a less than optimal number of stages. The main advantages with recycling of off-cuts are a potential reduction in distillation time and also recovery of valuable products. This type of operation was considered in the past by Liles (1966), Mayur et al. (1970), Christensen and Jorgensen (1987), Nad and Spiegel (1987) and Luyben (1988).

1.1.5 Reactive Distillation Operation

The use of batch distillation in which chemical reactions takes place is common practice in the chemical industries (Corrigan and Ferris, 1969; Egly et al., 1979; Cuille and Reklaitis, 1986; Wilson, 1987). Such an operation is particularly suitable when one of the reaction products has a lower boiling point than the others and the reactants. In fact, the higher volatility of this product results in a decrease in its concentration in the liquid phase, therefore increasing the reaction rate in case of irreversible reaction (due to the increased liquid temperature). In case of reversible reaction, this situation favors the forward reaction because the concentration of the product decreases due to its removal from the system by distillation. Therefore, in both cases higher conversion of the reactants is expected.

Apart from conventional batch distillation some developments were studied using some unconventional batch columns including the inverted batch column originally proposed by Robinson and Gilliland (1950). Various column configurations and operations were suggested to improve column performance and to retain temperature sensitive product qualities.

1.1.6 Inverted Batch Distillation

This type of batch distillation (Figure 1.2) combines the feed charge and the condenser reflux tank and operates in an all stripping mode with a minimum holdup reboiler.

This type of operation is supposed to eliminate the thermal decomposition problems of the products.

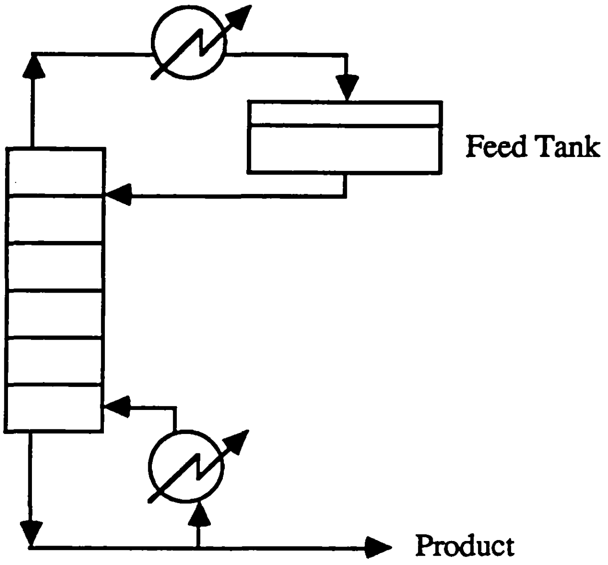


Figure 1.2 Inverted Batch Distillation (Feed Charged to Overhead Drum)

1.1.7 Sequential Steady State Operation

Abrams et al. (1987) considered a sequential steady state (S.S.S) operation (Figure 1.3) where each pass will remove one end product and leave the residual mixture held in a further storage, which becomes the feed tank for the next pass, except when separating the final binary mixture. The authors observed that when the number of theoretical plates was twice the minimum for steady state operation, there were substantial energy and productivity gains from "S.S.S" operation compared to conventional batch distillation.

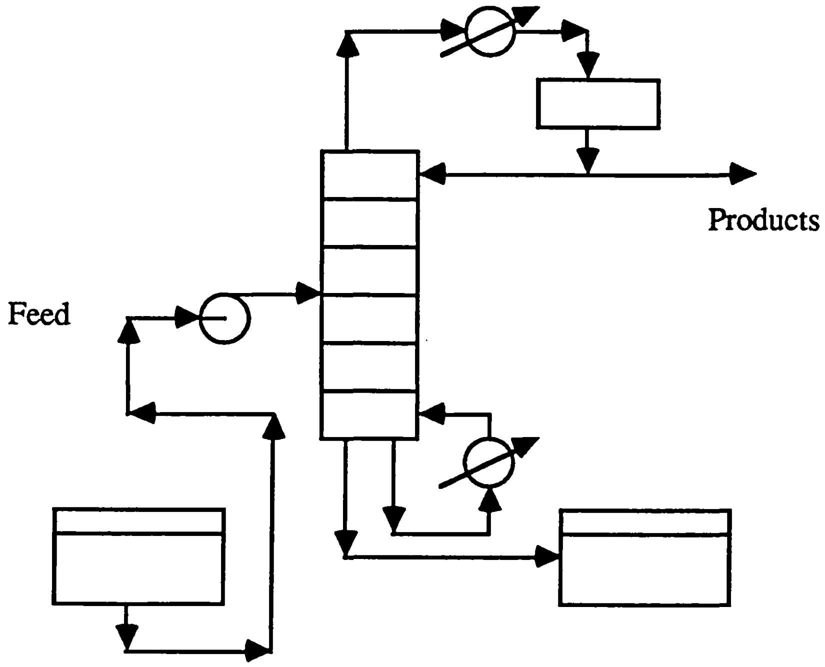


Figure 1.3 Sequential Steady State Operation (Tanks Alternate as Feed Tank and Intermediate Product Storage Tank, Except When a Final Product is Taken at Bottom)

1.2 Modelling and Simulation of Batch Distillation

Simulating the actual operation (both start-up and product period) of a batch distillation column has been the subject of much research for more than half a century. The main interest was usually to develop a model (consisting of mass and energy balances, hydraulic model, physical properties etc.) that could best predict the operation of the column.

Most of the early work in the field of batch distillation, beginning with the well known Rayleigh equation (Rayleigh, 1902) for the simplest case and continuing through the many graphical and empirical hand methods was reduced to academic interest with the advent of computers. With the development of high speed digital computers the main issues in modelling were whether and how to include energy balances, column holdup, plate hydraulics, accurate physical properties etc. to simulate the actual operation of batch columns. In many cases it was found that the models had to be simplified because of several reasons: size and complexity of the problem, capabilities of the computer, availability of suitable numerical methods to integrate the model, gain in accuracy in the prediction of real operation vs computation time etc.

1.2.1 Simulation of Start-up Period

Simulation of the actual start-up operation is very difficult unless there is a detailed rigorous model including plate hydraulics. However, simulation of the startup operation from step 5 (section 1.1.1.1) is fairly easy and can be done without considering a detailed hydraulic model and even with a very simple model. To do this the filling of holdups in step 3 and 4 (section 1.1.1.1) can be achieved in different ways: (a) directly with still pot liquid at the boiling point temperature (Converse and Huber, 1965; Mayur and Jackson, 1971; Holland and Liapis, 1983; Nad and Spiegel, 1987; Hitch and Rousseau, 1988) which consequently means plate and condenser compositions are initialized to the fresh feed composition for simulation; (b) operating the unit without reflux i.e., with only one rectification theoretical stage. Vapors from the reboiler are condensed and stored in the overhead equipment until liquid fills the condenser and the column holdups. This mode was suggested by Luyben (1971) and was used by Gonzalez-Velasco et al. (1987).

Step 5 (section 1.1.1.1) now simply requires to run the column at total reflux until the unit reaches steady state (Holland and Liapis, 1983; Nad and Spiegel, 1987) or until the instant distillate composition reaches the product composition so that the product can be collected at constant distillate composition (Coward, 1967; Kerkhoff and Vissers, 1978). A variation of this total reflux operation concept can also be found in the literature and in practice. In this variation only a part of the condensed liquid is returned to the column and the rest is taken out as product (product period starts from step 3 of section 1.1.1.1) (Converse and Huber, 1965; Mayur and Jackson, 1971).

Hitch and Rousseau (1988) simulated both the startup and product period for a packed column. He considered a detailed dynamic model which includes mass and energy balances, rigorous phase equilibria and mass tranfer relationships. Using a 3 component mixture they showed the effects of varying packing height, boilup rate, reflux ratio and condenser holdup on the column performance.

It is only recently that Ruiz (1988) considered the actual start-up procedure in his simulation by incorporating a very detailed model including plate hydraulics, weeping, channelling of vapor through downcomers, the actual liquid sealing of downcomers etc. He employed a generalised dynamic model (DYNAM), developed for continuous distillation by Gani et al. (1986), in batch distillation and considered the example of Holland and Liapis (1983) for this purpose. However, the results of Ruiz clearly showed that the initial period required to establish column holdup from an empty column (step 1 to 4 of the startup operation) was very small compared to the period required by step 5 of the startup operation. In fact, when the instant distillate composition profile obtained this way is compared to the composition profile obtained by Holland and Liapis (who initialized the column with fresh feed composition and simulated the startup operation straight from step 5) no notable difference is detected. Therefore inclusion of hydraulics in the model in such detail appears to be unnecessary and appears to only add complexity and much additional computation time. Ruiz and Holland and Liapis also simulated the product period. It must be observed that many other assumptions which are typically made, such as perfect mixing on a plate and equilibrium between liquid and vapor, are probably responsible for much more substantial deviation in the model response from actual behaviour.

1.2.2 Simulation of Product Period

In comparison, simulation of the product period is fairly easy and was considered by several authors in the past with different types of models (Huckaba and Danly,1960; Meadow, 1963; Domenech and Enjalbert, 1981; Ruiz, 1988).In this, research has typically paralleled that for continuous distillation. Some experimental simulations were also reported without modelling. Rose et al. (1950,1952) studied the effect of holdup for binary and ternary mixtures in a laboratory batch packed column and found increasing holdup was beneficial (product could be separated at higher purity) at low reflux ratio.

Huckaba and Danly (1960) modelled their batch distillation assuming constant weight holdup on trays and performing enthalpy balance they were able to simulate their experimental data. In addition the model was programmed to handle nonlinear equilibrium relationships and plate efficiency correlations. Although highly succesful from a simulation standpoint, the usefulness of the model was limited by the fact that it was applicable to binary systems only.

Meadow (1963) presented the first multicomponent batch distillation model in the published literature. The model was as rigorous as was then practical from computational standpoint. The main assumptions were perfect mixing on all trays, negligible vapor holdup, adiabatic operation, theoretical trays and constant volume holdup. Comparison of his results with constant molar holdup case with those with constant volume holdup case showed very little difference while the computation time was increased for the later case by an order of 17%. That was because of additional density equation required in the model in the constant volume holdup case.

Distefano (1968) used a similar model to Meadow and explored a number of non-stiff numerical techniques to integrate such models and discussed the limitations of such methods. Boston et al. (1980) also considered similar model to Meadow but used stiff methods to integrate the model and discussed the computational efficiency of such methods.

Corrigan and Ferris (1969) studied experimentally the effect of using batch distillation on the esterification reaction of methanol and acetic acid and showed that conversion could be increased by use of distillation.

Domenech and Enjalbert (1981) in their simulation of multicomponent mixtures used a simplified model using the assumptions of adiabatic column, negligible vapor holdup, constant or zero molar liquid holdup, theoretical trays, constant operating pressure, equimolal overflow and constant relative volatility. These simplifying assumptions reduced the dimension of the problem to a greater extent and eased the computational burden. The authors concluded that any loss in accuracy in the model by such simplifications could be balanced by a gain in the number of predictive calculations in shorter computation time. This type of model is therefore suitable for preliminary design studies.

Cuille and Reklaitis (1986) also considered a model similar to the one of Meadow but included chemical reactions on the plates, in the condenser and in the reboiler system. They simulated several example problems but were not able to demonstrate the need for reactive batch distillation with suitable example. Also there appear to be some problems with the solution method used, in particular with discontinuities when changing reflux ratio.

Diwekar and Madhavan (1986) considered a similar model of Domenech and Enjalbert (1981) but neglected the column holdup. They simulated their unsteady state model as a succession of a finite number of stationary states of short duration.

Nad and Spiegel (1987) and also Galindez and Fredenslund (1988) used the same assumptions as Domenech and Enjalbert but simulated the unsteady state behaviour using a similar approach to the one used by Diwekar and Madhavan (1986). Comparison of the simulated results of Galindez and Fredenslund with those by Boston et al. (1980), who used a more rigorous dynamic model and solution method, show significant variations. It was not possible to quantify the loss of accuracy in Nad and Spiegel's work because their experimental data were liable to error (within ± 10%) and to match the experimental data they had to adjust the reflux ratios in their simulation, the values of which were considerably different from those used in the experiment.

Luyben (1988) also considered a similar model to that used by Domenech and Enjalbert and simulated multicomponent batch distillation with recycle of intermediate fractions. The main purpose was to show the effect of different parameters (no of plates, relative volatilities) on the productivity.

As far as dynamic modelling of equilibrium processes are concerned, several models and their efficient methods of solution were developed in the past for continuous distillation (Gallun and Holland, 1979; Stewart et al., 1985; Gani et al., 1986, Pantelides et al., 1988) but only a few of the models were applied in batch distillation. Also, the effect of mechanical design on the control of dynamic continuous distillation were considered in the past (McGreavy and Tan, 1986) which is typically not yet considered in batch distillation.The computational time required for detailed simulation can be very substantial. Whether sufficient extra accuracy is obtained from the more complex models is an open question.

Also none of the papers discussed so far raised the point of computation time spent in calculating thermophysical properties. Tyreus et al. (1976) studied the dynamics of a 40 tray binary tray distillation column (continous) and using an Euler integration scheme they required an order of 400000 iterative buble point calculations. This number may exceed several millions of property evaluations for multiple column configurations with nonideal mixtures. Even with more sophisticated integration method the portion of computation time spent in physical properties calculations can be high. Using a simple dynamic column model for continous distillation Chimowitz et al. (1985) showed that the use of local thermodynamic models (developed by Chimowitz et al. (1983)) speeded up the execution time, often by a factor of 5-10 when compared to algorithms that use rigorous thermodynamic evaluations. This approach was applied to batch distillation by Hillestad et al. (1988).

Table 1.1 summarizes the past works on the simulation of batch columns. In Table 1.1 rigorous and simple models are only differentiated by the energy balance. All the models include column holdup in some form (constant mass, molar or volume holdup). Details of the model types are presented in Tables 2.1 and 2.2 of chapter 2.

Table 1.1 Summary of the Past Work on Simulation

Reference	Model	Mixture	Phase Equilibria	Purpose of Simulation
Huckaba & Danly (1960)	Rigorous	Binary	Rigorous	Product Period
Meadow (1963)	"	Multicomponent	"	"
Distefano (1968)	"	"	"	"
Boston et al. (1980)	"	"	CRV	"
Domenech & Enjalbert (1981)	Simple	"	"	"
Holland & Liapis (1983)	Rigorous	"	Simple*	Start-up + Product Period
Cuille & Reklaitis (1986)	"	"	Rigorous	Product Period
Diwekar & Madhavan (1986)	Simple	"	CRV	"
Nad & Spiegel (1987)	"	"	Rigorous	Start-up + Product Period
Galindez & Fredenslund (1988)	"	"	"	Product Period
Luyben (1988)	"	"	CRV	"
Ruiz (1988)	Rigorous	"	"	Start-up + Product Period
Hitch and Rousseau (1988)	Rigorous	"	"	Start-up + Product Period

Key : CRV = constant relative volatility.

* using simple expression as a function of temperature only

1.3 Optimal Control of Batch Distillation

The optimal control of a process can be defined as a control sequence in time, which when applied to the process over a specified control interval, will cause it to operate in some optimal manner. The criterion for optimality is defined in terms of an objective function and constraints and the process is characterized by a dynamic model. The optimality criterion in batch distillation may have a number of forms, maximizing a profit function, maximizing the amount of product, minimizing the batch time etc. subject to any constraint on the system. The constraints which are most common in batch distillation are on the amount and on the purity of the product at the end of the process or at some intermediate point in time. The most common control variable of the process is the reflux ratio.

The earliest work on the optimal control of binary batch distillation was reported by Converse and Gross (1963) who maximised the amount of product produced in a fixed time subject to purity constraints (maximum distillate problem) using a simplified model. Converse and Huber (1965) studied the effect of column holdup in such an optimal control problem. Using a simple column model they concluded that holdup in all cases was detrimental and reduced the maximum amount of distillate compared to the no holdup cases, but they were not able to explain these results.

Diwekar et al. (1987) also solved the maximum distillate problem to obtain reflux policies for multicomponent mixtures. They employed an extended shortcut method of Diwekar and Madhavan (1986) to solve their model and used Pontryagin's maximum principle to obtain the optimal policy. Comparison of their binary results with those produced by Murty et al. (1980), who studied several optimization techniques for a simple batch column model show that the amounts of distillate obtained by shortcut and rigorous methods differ significantly (order of 22%) for columns with a small number of plates which are often used for this operation.

Coward (1967), Robinson (1969,1970) obtained optimal reflux policies for a problem in which the overall batch time to produce a given amount of distillate of specified purity was minimized (minimum time problem). Both authors used a simplified model. Robinson (1969) considered linearly increased vapor boilup rate in addition to optimal reflux ratio. Robinson (1970) noticed the effect of holdup in an industrial column and concluded that the holdup, however small, should not be neglected in the model.

Mayur and Jackson (1971) studied the effect of holdup in a three plate column for a binary mixture, having about 13% (of initial charge) plate holdup with no condenser holdup, although condenser holdup is more common in practice. They considered a time optimal control problem (minimum time problem) for a given separation, and found that for both constant reflux and optimal reflux operation, the batch time was about 15-20% higher for the holdup case compared to the no holdup case. Mayur and Jackson also considered a ternary mixture to obtain optimal reflux policy.

Egly et al. (1979) considered chemical reaction in their batch distillation model. The model was rigorous and included holdup, energy balance and rigorous physical properties. They also considered a time optimal control problem where the batch time was minimized but no details were given regarding input and kinetic data of the problem. Wilson (1987) studied the optimal design of batch distillation processes with a very simplified column model involving chemical reaction.

Hansen and Jorgensen (1986) considered the minimum time problem to obtain optimal reflux policy for both tray and packed column. However, since for packed column the mass transfer coefficient depends on the vapor and the liquid flowrates (Brown et al., 1950) the authors also considered vapor boilup rate as control variable in addition to reflux ratio. Using only reflux ratio as control variable the batch time saving over constant reflux policy was in the order of 3-4% for both tray and packed column. But the control of both reflux ratio and boilup rate yielded about 9-10% batch time saving for packed column compared to constant reflux operation.

Liles (1966) considered an optimal batch scheduling problem for binary and ternary mixtures with intermediate off-cut recycle and with a very simplified model. Only one off-cut was assumed for both binary and ternary cases which was collected and stored after the first product cut. This off-cut together with the final residue left in the reboiler was recharged to the next batch with the fresh feed. This process was continued until an optimal batch was found to purge the residue. The optimisation variable of the process was the amount and composition of the off-cut and the objective was to maximize a profit function.

Kerkhof and Vissers (1978) generated optimum reflux ratio policies by maximizing a profit function for binary mixtures using a simplified binary model. The maximization of the profit function was actually a combination of maximum distillate and minimum time problem as mentioned above and described in Chapter 3.

Mayur et al. (1970) considered recycle of off-specification materials (off-cut) in a binary batch columns. Using simplified model they were able to obtain optimal reflux policies by minimizing overall distillation time for a quasi-steady state operation mode. In a quasi-steady state operation mode, the off-cut recycled from the current batch is assumed to be identical (in amount and composition) to the off-cut from the previous batch. The overall distillation time includes the time to obtain the main product cut plus the time to obtain the off-cut which is going to be recycled to the next batch. With an example problem Mayur et al. were able to show that the recycling of the off-cut reduced the distillation time by 5% compared to the no recycle case.

Christensen and Jorgensen (1987) also considered recycle operation for binary mixtures and by defining a measure of "the degree of difficulty" of separation for tray columns they were able to show when and how much recycle was beneficial. They did not propose any such measure for packed column, but showed the benefit of recycling in several cases.

Table 1.2 summarizes the past work on the optimal control of batch distillation. In most of the cases the models used by the authors were simplified neglecting enthalpy balance and assuming constant relative volatilities. Column holdups were also neglected in the models except for those by Converse and Huber, Mayur and Jackson and Egly et al..

Table 1.2 Past Work on Optimal Control of Batch Distillation

Reference	Mixture	Optimisation Problem	No. of Controls	No.of Constraints
Converse and Gross (1963)	Binary	Maximum Product	1 (reflux)	1 (product purity)
Converse and Huber (1965)	"	"	"	"
Murty et al. (1980)	"	"	"	"
Diwekar et al. (1987)	Multicomponent	"	"	"
Coward (1967)	Binary	Minimum Time	"	2 (product amount and purity)
Robinson (1969)	"	"	1 (reflux)	"
Robinson (1970)	Multicomponent	"	1 (reflux)	"
Mayur and Jackson (1971) "		"	"	"
Egly et al (1979)	"	"	"	"
Hansen and Jorgensen (1986) "	Binary		"	2 (reflux + boilup)
Liles (1966)	Multicomponent	Maximum Profit	2 (off-cut amount and purity)	1 (product purity)
Kerkhof and Vissers (1978)	Binary	"	1 (reflux)	"
Mayur et al. (1970)	"	Minimum Overall Time With Recycle	"	2 (product amount and purity)
Christensen and Jorgensen (1987)	"	"	"	"

1.4 Conclusions

Investigation of different types of models used in the simulation and optimal control of batch distillation shows that there is still scope and need for improvement of the models used which should be detailed enough to include column holdups, energy balance, accurate physical property data etc. and at the same time should not yield an excessive computational burden. The investigation also shows that there is a large incentive to study improvements in column operation by using realistic dynamic models in optimal control studies.

Although the optimal off-cut recycle policies for binary mixtures have received some attention in the past, no attempt was made so far to obtain such policies for multicomponent mixtures. This clearly indicates a need for developing optimal off-cut recycle policies for multicomponent separation problems. Also there is a large scope for developing more robust and efficient formulations of the optimal off-cut recycle problem both in binary and multicomponent batch distillation.

Finally, column holdup presents a controversial issue. Few authors concluded that it was good for the operation while the rest held opposite views. Therefore, a systematic study of column holdup is required to explain these conflicting statements.

1.5 Present Work

In this work we restrict our attention to tray columns and to conventional batch distillation (Figure 1.1). However, the methods used in this work to obtain optimal operational policies can be easily extended to unconventional batch distillation. For packed columns the model equations are to be modified slightly and a suitable integrator is to be required.

The present work can be outlined as follows:

Chapter 2 - A classification of models available in the literature is made based upon the assumptions used to develop them. Different numerical techniques employed to solve the model are discussed with their merits and demerits. A realistic model is proposed and a robust and efficient solution method is selected for the proposed model. A number of simulations are carried out to check the performance of the model against available experimental data and simulated results from the literature.

Chapter 3 - This chapter considers the operation of a single batch distillation to obtain optimal reflux policies for such a column using a variety of objective functions, constraints and operations. An optimal control problem is formulated as a nonlinear programming problem and is solved by using an optimal control algorithm suitable for rigorous models.

Chapter 4 - In this chapter an one level optimal control formulation is proposed for an operation with intermediate off-cut recycle for binary mixtures, which is computationally much faster than the classical two level optimal control formulation. Results are obtained for both formulations using different mixtures and using variety of column specifications.

Chapter 5 - The separation of multicomponent mixtures in quasi-steady state intermediate recycle operation is considered. A general optimal control problem is formulated for such operations and possible difficulties in solving such problem are outlined. Areas are identified using the "degree of difficulty" measure of a given separation where the same formulation used for binary cases can be applied for multicomponent mixtures. A new operational strategy involving the recycle of off-cuts from a multicomponent batch distillation is proposed, which is physically and thermodynamically sensible. The implementation of the proposed operation is shown using typical 3 component mixtures.

Chapter 6 - This chapter examines the role of column holdup in batch distillation and explains the apparently contradictory behaviours observed in the past in terms of a "degree of difficulty" measure of a given separation. A number of binary mixtures are considered.

Chapter 7 - Finally this chapter concludes the work done in this thesis and outlines the future work.

Chapter 2

MODELLING AND SIMULATION

2.1 Introduction

This chapter briefly focusses on different types of batch distillation models and on the numerical solution techniques used for simulation. A realistic dynamic column model is chosen for use in simulation and optimal control studies. The model is expressed as a coupled system of differential and algebraic equations (DAEs) and an efficient algorithm for the solution of the model is selected which is based on Gear's work. The performance of the model is checked against several experimental data and previous simulation results reported in the literature.

2.2 Different Models Available in the Literature

Unlike continuous distillation, batch distillation is inherently an unsteady state process. Dynamics in continuous distillation are usually in the form of relatively small upsets from steady state operation, whereas in batch distillation individual species can completely disappear from the column, first from the reboiler and then from the entire column. Therefore the model describing a batch column is always dynamic in nature. A general model for batch distillation usually includes mass and energy balances, phase equilibria, plate hydraulics etc.

Depending on the choice of numerical techniques used for integration, the model equations can be reformulated and solved as a system of ordinary differential equations (ODEs) (Huckaba and Danly, 1960; Meadow, 1963; Distefano, 1968; Boston et al., 1980; Ruiz, 1988; etc.) or can be solved directly as a system of coupled nonlinear DAEs (Cuille and Reklaitis, 1986).

Table 2.1 summarizes the different types of model available in the literature (as mentioned in chapter 1) for batch distillation and Table 2.2 summarizes the literature references using those models.

Table 2.1 Types of Models Available in the Literature

	Types of Model						
	I	II	III	IV	V	VI	VII
Equimolal Overflow		√	√				
Equilibrium							
Const. Relative Vol.		√	√				
Rigorous	√	√		√	√	√	√
Liquid Holdup							
Constant Molar		√			√		
Const. Weight						√	
Const. Volume	√			√			√
Plate Hydraulics							√
Chemical Reaction				√	√		
Model Reformulated and Solved as							
ODEs	√	√	√		√	√	√
DAEs				√			
Steady-State		√					

Table 2.2 Summary of Different Models Used in the Literature

Type	Literature References
I	B5, D1, M6
II	C8, D2, D3, D5, G1, L3, M3, N1, R5
III	C5, C7, C10, H2, K1, L1, M4, R4
IV	C11
V	E1
VI	H7
VII	R10

2.3 Numerical Solution Methods and Their Limitations

The model equations were integrated by Huckaba and Danly (1960) using an explicit Euler method. Coward (1967) used an explicit fourth order Runge-Kutta method while Robinson (1969, 1970) used a 2nd order Runge-Kutta method for integrating the model equations in his optimal control study. Domenech and Enjalbert (1981) used the same method as Coward. Although such methods are very easy to use, they may become uneconomical because of the necessity to use small step sizes in order to maintain stability.

Meadow (1963) used what they called a "finite difference" method (essentially an explicit Euler integration method) to integrate the model equations which again suffers from the limitation of small step size to maintain stability and consequent large computation times. Although Meadow proposed an empirical relationship to calculate the step size to maintain stability, it does not predict accurately the maximum allowable size.

Distefano (1968) presented a most comprehensive study of the application of several predictor-corrector methods to batch distillation. It was concluded that of the methods tried, the Adams-Moulton-Shell predictor-corrector method possessed the most stability characteristics for numerical solution of transient distillation model. However, that even the most stable numerical integration techniques are not entirely satisfactory for the solution of transient distillation equations and other "stiff" differential equations. The forms of these differential equations are such that a small error in the calculated compositions will produce large errors in the derivatives, which tend to propagate as the calculations progress.

Quite often chemical engineering systems are encountered with widely different time constants, which give rise to both long-term and short-term effects. The corresponding ordinary differential equations have widely different eigenvalues. Differential equations of this type have come to be called *stiff systems*. In batch distillation, the system is frequently very stiff, owing either to wide ranges in relative volatilities or large differences in tray and reboiler holdups. Therefore, if methods for non-stiff problems are applied to stiff problems, a very small integration step must be used to ensure that the solution remains stable. This consequently means larger computation time.

Gear (1971a) defined "stiff-stability" and developed a method using backward differentiation formula (BDF) to solve stiff ODEs. This method allows larger step sizes which change to control integration error. Boston et al. (1980) employed Gear's method to integrate an ODEs model in conjunction with a suitable method for solving the system of algebraic equations that results when the integration formula is introduced. They used the "Inside-Out" algorithm of Boston (1978) to solve the resulting algebraic equations, which the author claimed to perform better than the Newton-Raphson method.

Holland and Liapis (1983) presented a batch distillation algorithms in which a modified trapezoidal integration formula was used in conjunction with the "theta" method to solve the resulting algebraic equations. This method also suffers from instability because of stiffness of the system.

Gear (1971b) implemented the idea of using stiff ODE methods (Gear, 1971a) for solving coupled nonlinear system of DAEs. He showed that the backward differentiation formula (BDF) methods could be as well used to solve stiff DAE systems. Gallun and Holland

(1982) used Gear's method to solve simultaneously the system of stiff DAEs resulting from unsteady state continuous distillation column. Recently Cuille and Reklaitis (1986) used the Gear type code LSODI (Hindmarsh, 1980) to solve the coupled system of DAEs resulting from their reactive batch distillation model.

Hansen and Jorgensen (1986) and Christensen and Jorgensen (1987) solved their model equations (for tray and packed column) in optimal control application with the use of orthogonal collocation techniques and claimed it to be fast compared to other integration methods, however, no mention was made about the stiffness of the problem and stability of the method.

Diwekar and Madhavan (1986) and Diwekar et al. (1987) assumed that the batch distillation column can be considered as continuous distillation column with changing feed. That is, for small intervals of time the batch column behaviour is analogous to a continuous column and they employed the widely used Fenske-Underwood-Gilliland short cut method to integrate the model. As the batch distillation processes are often very stiff, their method of solution was liable to significant errors (as discussed in chapter 1).

Galindez and Fredenslund (1988) also considered a similar approach to Diwekar and Madhavan but employed a modified Naphtali and Sandholm method for solving the quasi-steady-state approximation of dynamic model of short duration (for details see Naphtali and Sandholm, 1971; Christiansen et al., 1979). Their methods also suffers from the same limitations mentioned above and comparison of their results with Boston et al. (1980) shows significant differences. Although these methods have advantages over the rigorous methods in terms of computation time their use is therefore very much restricted to very preliminary studies. Nad and Spiegel (1987) also used similar models and using the same techniques tried to simulate their experimental data.

Hitch and Rousseau (1988) integrate their model for packed column by using a relaxation method and finite difference techniques to obtain the derivatives. This method also suffered from the limitation of small step size, for both time and space, to maintain stability.

The package, DYNAM, used by Ruiz (1988) includes both stiff and non-stiff methods of integrating ODEs. In this method of calculations the algebraic equations of the system are solved separately to determine the r.h.s of the ODEs needed by the numerical integrator.

It is clear from the above investigation that a stiff numerical integrator of either ODE type (as used by Boston et al or Ruiz) or DAE type (as used by Cuille and Reklaitis) is always desirable for inherently stiff batch distillation calculations. Since a DAE integrator does not need any rearrangement or reformulation of the model equations, it is preferred to a regular ODE integrator.

2.4 Model Used in this Work

Referring to Table 2.2 (Type III) it is observed that a large number of authors simplified their model by using assumptions of constant relative volatility, neglecting enthalpy balance and column holdups. These simplifying assumptions are sometimes very crude and it can be shown that the predicted dynamic behaviour of a system modelled this way is often quite different from the actual behaviour. In particular liquid holdup in the column has been found to have significant effects both in industrial and pilot plant columns (Rose and O'Brien, 1952; Robinson, 1970) and the detailed study presented later in chapter 6 suggests that holdup, however small, should not be neglected. Also some models require large computation time because of extra equations (density, plate hydraulics) which can be neglected without much affecting the predictions from the model to ease computational burden.

Considering all these factors we decided to use a moderately detailed dynamic model in this work, which includes the most important contributions like energy balances, column holdup and rigorous phase equilibria and which results nonetheless in reasonable computation time. The model assumes negligible vapor holdup, adiabatic plates,constant molar holdup on plates and in the condenser, perfect mixing on trays, fast energy dynamics, constant operating pressure and total condensation with no subcooling. The model also includes the option of chemical reaction on the plates, in the reboiler and in the condenser but not in the product accumulator. Refering to column configuration given in Figure 2.1a and to the tray model given in Figure 2.1b the model equations are presented in the following section.

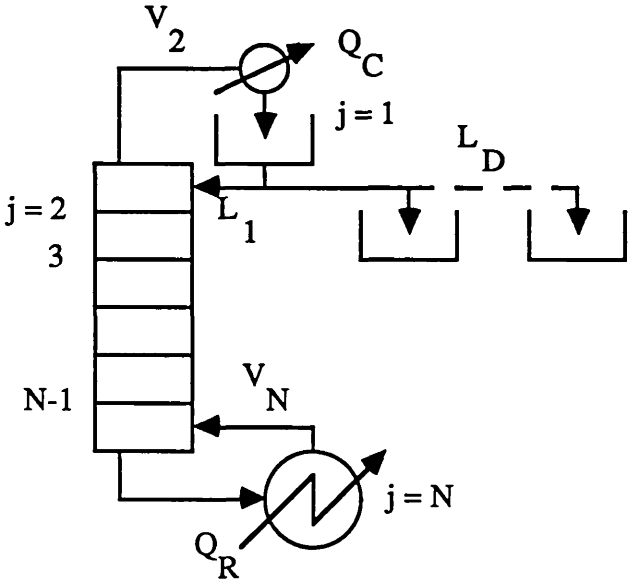


Figure 2.1a : A Typical Batch Column

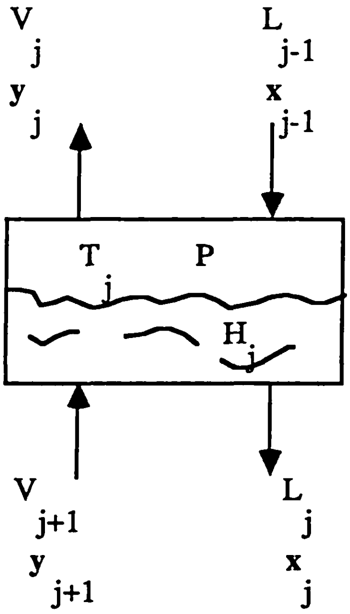


Figure 2:1b A Typical Equilibrium Stage

2.4.1 Model Equations

A. INTERNAL PLATES : $j = 2, N - 1$

Total Mass Balance :

$$0 = L_{j-1} + V_{j+1} - L_j - V_j + \Delta n_j H_j \tag{2.1}$$

Component Mass Balance :

$$H_j \frac{dx_{ji}}{dt} = L_{j-1} x_{j-1,i} + V_{j+1} y_{j+1,i} - L_j x_{ji} - V_j y_{ji} + \mathbf{r}_{ji} H_j \tag{2.2}$$

Energy Balance :

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

Equilibrium :

$$y_{ji} = k_{ji} x_{ji} \tag{2.4}$$

Restrictions :

$$\Sigma\, y_{ji} = 1 \tag{2.5}$$

Relations defining Physical Properties and reaction :

$$k_{ji} = k_{ji}\ (y_{ji}, x_{ji}, T_j, P) \tag{2.6}$$

$$h^L_j = h^L_j\ (x_{ji}, T_j, P) \tag{2.7}$$

$$h^V_j = h^V_j\ (y_{ji}, T_j, P) \tag{2.8}$$

$$\mathbf{r}_{ji} = \mathbf{r}_{ji}\ (\mathbf{k}^r, x_{ji}) \tag{2.9}$$

$$\Delta\, \mathbf{n}_j = \Sigma\, \mathbf{r}_{ji} \tag{2.10}$$

B. REBOILER : j = N

Total Mass Balance :

$$\frac{dH_N}{dt} = L_{N-1} - V_N + \Delta n_N H_N \tag{2.11}$$

Component Mass Balance :

$$H_N \frac{dx_{Ni}}{dt} = L_{N-1}(x_{N-1,i} - x_{Ni}) - V_N(y_N - x_{Ni}) + r_{Ni}H_N - \Delta n_N H_N x_N \tag{2.12}$$

Energy Balance :

$$0 = L_{N-1}(h^L_{N-1} - h^L_N) - V_N(h^V_N - h^L_N) + Q_R \tag{2.13}$$

The other equations are same as equations (2.4-2.10) where j is replaced by N.

C. CONDENSER AND ACCUMULATOR SYSTEM : j = 1

Accumulator Total Mass Balance :

$$\frac{dH_a}{dt} = L_D \tag{2.14}$$

Component Mass Balance :

a) Accumulator

$$H_a \frac{dx_{ai}}{dt} = L_D(x_{Di} - x_{ai}) \tag{2.15}$$

b) Condenser Holdup Tank

$$H_c \frac{dx_{Di}}{dt} = V_2 y_2 + r_{1i}H_c - (V_2 + \Delta n_1 H_c)x_{Di} \tag{2.16}$$

Energy Balance :

$$0 = V_2 h_2^V - (V_2 + \Delta n_1 H_c)\, h_1^L - Q_C \tag{2.17}$$

Other Equations :

$$L_1 = r\,(V_2 + \Delta n_1 H_c) \tag{2.18}$$

$$L_D = (V_2 + \Delta n_1 H_c)\,(1 - r) \tag{2.19}$$

$$T_1 = T_1(x_{Di}, P) \tag{2.20}$$

$$h_1^L = h_1^L\,(x_{Di}, T_1, P) \tag{2.21}$$

The other equations are the same as equation (2.9-10) with j is replaced by 1.

For a column with N equilibrium stages (including reboiler and condenser) and a mixture of n_c components the model results in a set of $[6N+(4N-1)\ n_c+1]$ differential and algebraic equations. It is to be noted that since a total enthalpy balance is considered heat of reactions are not used explicitly in the energy balance equations.

The model presented above is general in the sense that it includes energy balances, column holdups, rigorous phase equilibria and chemical reaction. The only variations from other models of comparable detail (Boston et al., Cuille and Reklaitis, Ruiz etc.) are (1) the assumption of constant molar instead of constant volume holdup (2) the exclusion of plate hydraulics (3) the assumption of fast energy dynamics. However, these assumptions only reduce the number of equations to some extent (which also reduces the computational time) while keeping realisticity. The inclusion of these additional equations could be done fairly easily if needed.

2.5 Integration of the Model Equations

The model equations presented in the previous section constitute a coupled system of DAEs of index one. Index is simply defined by the maximum number of differentiations required to reduce a DAE system to an ODE system. DAE system of index exceeding unity occur in many areas of chemical engineering modelling. For example if we remove the assumption of total condensation in the condenser and write material and energy balance for a fixed condenser volume, the resulting DAEs will be of index two. This was shown in detail by Pantelides et al. (1988). Solution of such DAE systems is sometimes difficult and requires a consistent initialization of the variables in the system. Recently Pantelides (1988) developed a procedure for consistent initialization of such DAE systems. Classification of DAE system according to their index was also given in some detail in Morison (1984).

Systems of index zero or one are ODE systems or simple DAE systems, and should cause no problem when integrated by existing methods (for details see Morison, 1984). In this work the proposed model is solved by the computer code DAEINT developed by Morison for index one DAE systems which was based on Gear's BDF method (Gear, 1971b). Implementation of the method will be given here in brief (for details see Morison).

BDF methods are predictor-corrector methods. At each step a prediction is made of the state at the next point in time. A correction procedure corrects the prediction. If the difference between the predicted and corrected states is less than the required local error, the step is accepted. Otherwise the step length is reduced and another attempt is made. The step length may also be increased if possible and the order of prediction is changed when this seems useful. The following sections briefly point out some important features in the implementation of BDF methods.

2.5.1 Estimation and Testing of the Local Error

In BDF methods at each step a k^{th} order prediction of the state is made using information obtained on the previous steps. Corrections are made to the prediction until the corrections have become sufficiently small. The corrected steps are then compared with the predicted values, an estimate of the local error is made and this is tested. It is assumed that, because of the stability of the methods, the global error is related to the local error. But in solving DAE systems it is useful to examine the propagation of global error and base the local error test on it.

2.5.2 Convergence Criteria

The convergence test must be related to the local error test. The level of convergence must be such that the local error estimate is not significantly affected. Hindmarsh (1980) converged the variables to within 10% of the local error. Morison (1984) used the same scheme for convergence.

2.5.3 Discontinuities

There are two types of discontinuities. One is time dependent while the other is state dependent. Time dependent discontinuities occur when there is a discontinuity in an input. For example, in batch distillation, piecewise constant reflux ratio in different time intervals causes discontinuities. These should always be specified by the user or automatically detected by optimal control algorithm so that the integration proceeds upto the discontinuity and is restarted at this point. Otherwise the integration routine will perform a crude root finding problem at great expense. State dependent discontinuities are usually defined by a state variable, or a function of the state variables, reaching a critical level. For example, when a liquid level reaches a critical height a pump starts up.

2.5.4 Initialization of the DAE system

The initialization of the system is important. While, in systems of ODEs all of the state variables must be initialized, in DAE systems only some of the variables need to be initialized, which is the same as the number of differential variables for index one system. The other variables can be determined using the algebraic equations. It is inconvenient for the user to be required to initialize all of the variables as this might require the solution of a set of nonlinear algebraic equations. The code DAEINT incorporates this initialization procedure so that it requires the minimum amount of information from the user.

2.6 Model Validation

A couple of tests are carried out using published experimental data and simulated results from literature to validate the proposed model.

2.6.1 Example Problem 1

2.6.1.1 Problem Description: Domenech and Enjalbert (1974) carried out a series of experimental tests in a laboratory batch distillation column. A binary mixture of Cyclohexane and Toluene was considered for the purpose. The experimental equipment used was a perforated plate column, with 4 trays and a 60 litre reboiler heated with a heat transfer coefficient of 3 kw. The experimental results obtained by Domenech and Enjalbert together with column input data are presented in Table 2.3a, where x_{N0} refers to initial composition of cyclohexane and x_D refers to instant distillate composition of the same component. Since the overall stage efficiency is 75%, the number of theoretical plates in our simulation was 3. The column was initialized at its total reflux steady state values.

In this problem we simulated the experimental operation using the Soave-Redlich-Kwong (SRK) model for the VLE property calculations. For this purpose we used the implementation of Maduabueke (1987) and Macchietto et al. (1988) which also provides analytical derivatives of k values with respect to all its arguments. Vapor pressures are calculated using Antoine's vapor pressure equation. Vapor phase enthalpies are calculated using ideal gas heat capacity values and the liquid phase enthalpies are calculated by subtracting heat of vaporization from the vapor enthalpies. The input data required to evaluate these thermodynamic properties are taken from Reid et al. (1977).

2.6.1.2 Results and Discussion : The results for four different operating conditions are presented in Table 2.3b and are also plotted in Figure 2.2 together with the experimental results. It is quite clear from Table 2.3b and Figure 2.2 that the results obtained by our model are in very good agreement with those of original reference. The simulation time for each case study was about 1 minute (CPU) on an IBM 4341 system.

Table 2.3a Input and Experimental Data from Domenech and Enjalbert (Example 1)

Case 1		Case 2		Case 3		Case 4	
r = 3 x_{N0} = 0.62		r = 4 x_{N0} = 0.55		r = 6 x_{N0} = 0.42		r = 9 x_{N0} = 0.30	
H_a (lbmol)	x_D	H_a	x_D	H_a	x_D	H_a	x_D
0.0	0.960	0.0	0.945	0.0	0.920	0.0	0.900
21.0	0.940	10.0	0.930	10.0	0.900	5.0	0.860
29.0	0.930	20.0	0.910	25.0	0.875	12.5	0.850
50.0	0.920	29.0	0.905	39.0	0.865	23.0	0.840
60.0	0.910	40.0	0.900	60.0	0.830	36.0	0.800
75.0	0.895	50.0	0.895	80.0	0.730	43.0	0.770
90.0	0.875	60.0	0.885	89.0	0.630	51.0	0.690
110.0	0.825	81.0	0.850	100.0	0.475	63.0	0.560
129.0	0.700	93.0	0.830			75.0	0.400
140.0	0.550	104.0	0.770			83.0	0.300
		118.0	0.650				
		123.0	0.530				
		131.0	0.370				

Components : Cyclohexane and Toluene

No of Plates : 4

Total Condenser

Initial Charge : 200 lbmol

Holdup
Internal Plates = 2.5 lbmol Condenser = 2.5 lbmol

Stage Efficiency : 75% Pressure = 1 atm.

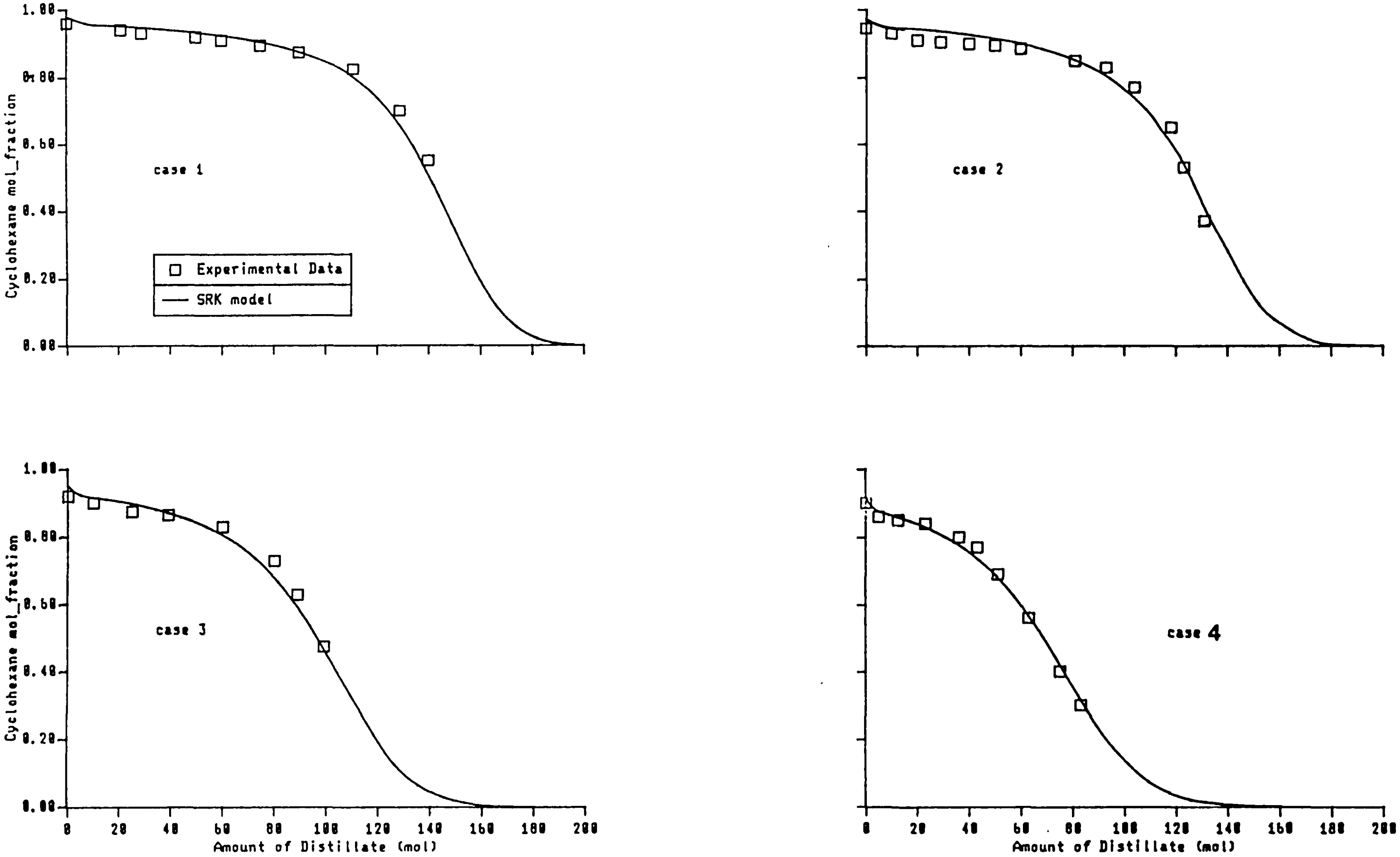


Figure 2.2 Simulation and Experimental Results of Domenech and Enjalbert (1974)

Table 2.3b Simulation Results by Our Model (Example 1)

Case 1		Case 2		Case 3		Case 4	
H_a (lbmol)	x_D	H_a	x_D	H_a	x_D	H_a	x_D
0.0	0.979	0.0	0.971	0.0	0.951	0.0	0.915
21.0	0.950	10.0	0.940	10.0	0.915	5.0	0.875
29.0	0.943	20.0	0.930	25.0	0.895	12.5	0.860
50.0	0.932	29.0	0.926	39.0	0.868	23.0	0.827
60.0	0.920	40.0	0.923	60.0	0.805	36.0	0.772
75.0	0.900	50.0	0.910	80.0	0.675	43.0	0.734
90.0	0.876	60.0	0.900	89.0	0.590	51.0	0.688
110.0	0.805	81.0	0.850	100.0	0.470	63.0	0.560
129.0	0.680	93.0	0.810			75.0	0.410
140.0	0.500	104.0	0.790			83.0	0.288
		118.0	0.640				
		123.0	0.530				
		131.0	0.380				

2.6.2 Example Problem 2

2.6.2.1 Problem Description: In this problem we consider the complex separation [taken from Boston et al., 1980], involving a quarternary mixture, 5 operational steps (3 main cuts), with the addition of a secondary charge after CUT 1. The first two operational steps form CUT 1, where the most volatile component, propane, is removed from the system. Then the main accumulator is dumped and a secondary charge containing 40 percent butane and 60 percent hexane is added to the reboiler instantaneously, before beginning of the third step. The third step (CUT 2) is a production step which produces butane. The accumulator is dumped again before the beginning of the next two steps (CUT 3) when pentane is removed from the system, leaving hexane as the bottom product. The problem is defined in detail in Table 2.4 and the results obtained by Boston et al are presented in Table 2.5.

We initialized the column at the total reflux steady state and simulated this problem for a time period of 30.24 hr as was done by Boston et al. The duration of individual operation steps was also set equal to those considered by the original authors (see Table 2.5).

Equilibrium k values were calculated using Antoine's vapor pressure equation and enthalpies by the same procedure mentioned in the previous example.

Table 2.4 Input Data for Example 2

No. of Internal Stages	8
Total Condenser	1
Partial Reboiler	1
Total	N = 10
Internal Stage Holdup	= 4.93 E-3 lbmol
Condenser Holdup	= 4.93 E-2 lbmol
Operating Pressure	= 1.013 bar
Reboiler Feed	= 100 lbmol
Components :	Propane , n-Butane , n-Pentane , n-Hexane
Composition :	< 0.1 , 0.3 , 0.1 , 0.5 > mole fraction
Intermediate Charge (to the reboiler)	= 20 lbmol
Composition :	< 0.0 , 0.4 , 0.0 , 0.6 >
Time of Charge :	After Propane Removal Step (after step 2)

<u>2.6.2.2 Results and Discussion :</u> The simulation results by our model are presented in Table 2.6 and the corresponding accumulated and distillate composition curves are presented in Figures 2.3 and 2.4. Comparison of the results by our model with those in the original reference shows a very good agreement again. The small deviations of our results from Boston et al. in the instant distillate composition might be due to the assumptions of constant relative volatility by the authors. It was found that the relative volatility (with respect to pentane) of propane ranges from 8.892 to 17.832, that of butane from 2.830 to 4.045 and that of hexane from 0.258 to 0.369 over the whole operation period. Therefore, the use of average values of relative volatility may lead to different instant distillate composition profile although the effect is evened out in the accumulated distillate composition. The simulation time for this example

Table 2.5 Simulation Results by Boston et al.(1980)

OPERATION STEP	1	2	3	4	5
PURPOSE	C_3 REMOVAL		C_4 PROD.	C_5 REMOVAL	
Reflux Ratio	5	20	25	15	25
Vapor Flowrate	12	42	52	32	52
Distillate Rate	2	2	2	2	2
Time (hr)	4.07	1.81	18.27	4.31	1.78
PRESENT DIST.					
Propane	0.800	0.015	---	---	---
Butane	0.200	0.985	0.164	---	---
Pentane	---	---	0.836	0.800	0.016
Hexane	---	---	---	0.200	0.984
ACCUM. DIST.					
Propane	0.988	0.849	------	---	---
Butane	0.012	0.151	0.990	0.006	0.004
Pentane	---	---	0.010	0.987	0.791
Hexane	---	---	---	0.007	0.204
[lb mol]	8.132	11.760	36.543	8.613	12.167
STILL POT					
Propane	0.021	---	---	---	---
Butane	0.325	0.319	0.001	---	---
Pentane	0.109	0.113	0.133	0.023	0.002
Hexane	0.545	0.567	0.866	0.977	0.998
[lb mol]	91.737	88.124	71.600	62.998	59.398

Table 2.6 Simulation Results by Our Model (Example 2)

OPERATION STEP	1	2	3	4	5
PURPOSE	C_3 REMOVAL		C_4 PROD.	C_5 REMOVAL	
Reflux Ratio	5	20	25	15	25
Vapor Flowrate	12	42	52	32	52
Distillate Rate	2	2	2	2	2
Time (hr)	4.07	1.81	18.27	4.31	1.78
PRESENT DIST.					
Propane	0.754	0.031	---	---	---
Butane	0.246	0.969	0.254	---	---
Pentane	---	---	0.745	0.613	0.091
Hexane	---	---	---	0.387	0.909
ACCUM. DIST.					
Propane	0.981	0.850	----	----	----
Butane	0.019	0.150	0.988	0.017	0.012
Pentane	---	---	0.012	0.940	0.778
Hexane	---	---	---	0.043	0.210
[lb mol]	8.139	11.760	36.548	8.619	12.180
STILL POT					
Propane	0.021	---	---	---	---
Butane	0.325	0.319	0.001	---	---
Pentane	0.109	0.113	0.133	0.023	0.002
Hexane	0.545	0.567	0.866	0.977	0.998
[lb mol]	91.86	88.240	71.680	63.061	59.380

Figure 2.3 Accumulated Distillate Composition (Example 2)

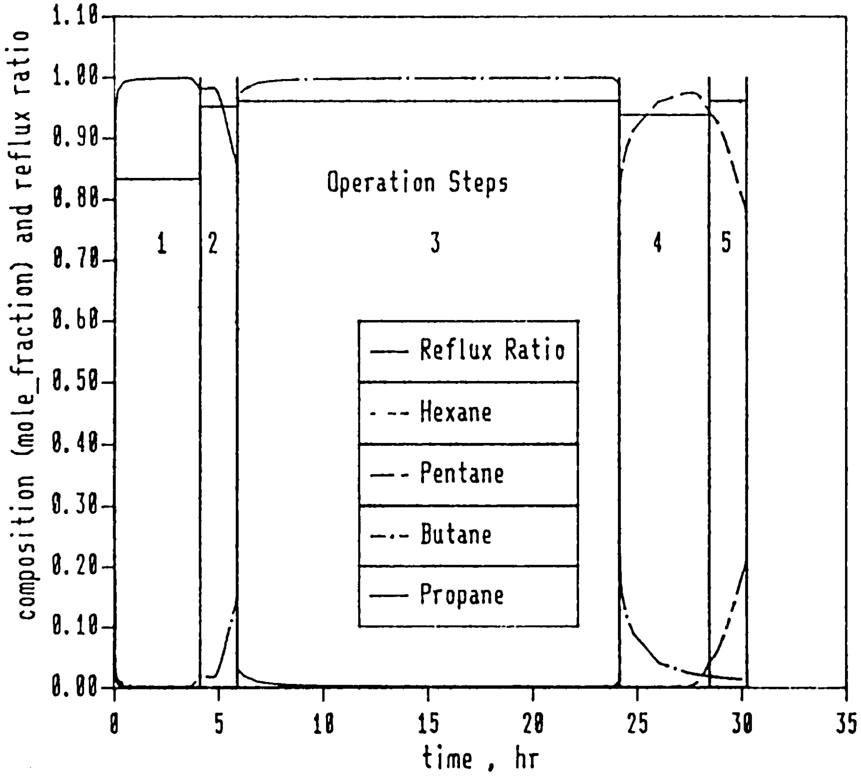
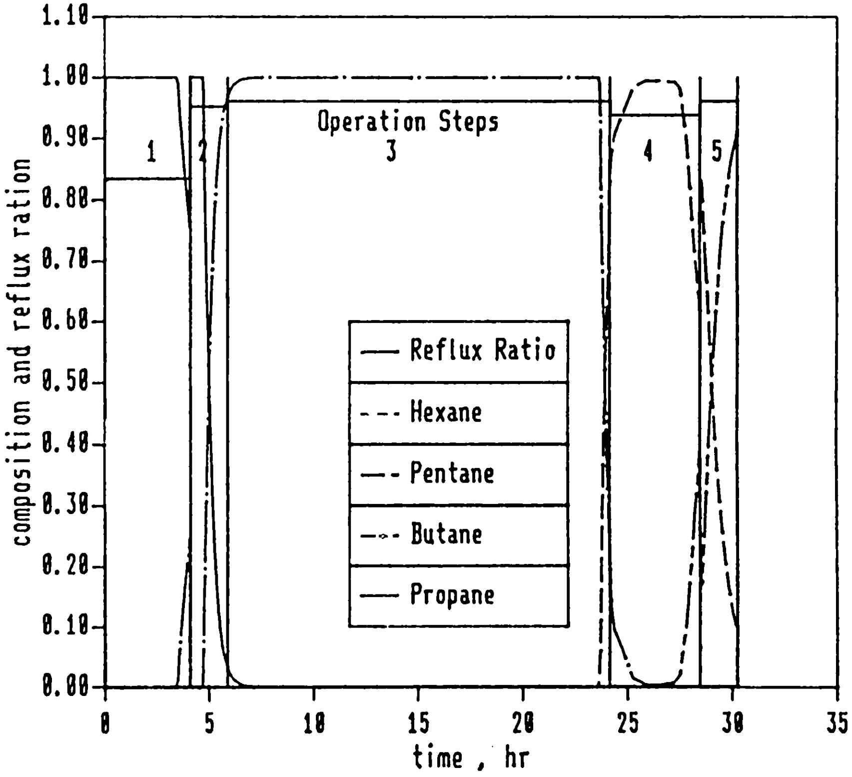


Figure 2.4 Instant Distillate Composition (Example 2)



problem was about 7 CPU minutes on an IBM 4341 system.

2.6.3 Example Problem 3

2.6.3.1 Problem Description: In this problem we consider the ternary separation [taken from Nad and Spiegel, 1987] of a cyclohexane-heptane-toluene mixture. The experimental column of Nad and Spiegel consists of 20 theoretical stages including condenser and reboiler. The feed to the column was 2.93 kmol of which 1.9% was total column holdup and 1.2% was condenser holdup. The column underwent an initial total reflux operation for about 2.54 hr before any product was collected. After then the mixture was separated into 3 main product cuts with 2 intermediate cuts in between, leaving a final product in the reboiler.

In the work of Nad and Spiegel experimental data was included. Such data was reproduced in that work using the computer programme DISBATCH of Galindez and Fredenslund (1988). The authors obtained reasonably good agreement in their calculation. However, to do so, they had to use a reflux profile considerably different from that reported as the experimentally measured one. They investigated the influence of a number of input variables on the simulated behaviour of the column and tried with different reflux ratio profiles until the best agreement with experiment was obtained.

In this work we simulate their example for the first product cut using a reflux ratio profile very close to that used by Nad and Spiegel in their own simulation and a nonideal phase equilibrium model (SRK). The purpose of this is to show that a better model and better integration method achieves an even a better fit to their experimental data. Also the problem is simulated using ideal phase equilibrium model (Antoine's equation) and computational details are presented. The input data to the problem are given in Table 2.7. Enthalpies were calculated using the method mentioned in the previous examples. Initialization of the plate and condenser compositions (differential variables) was done using the fresh feed composition.

Table 2.7 Problem Definition (example 3)

No. of Internal Stages	= 18
Total Condenser	= 1
Partial Reboiler	= 1
Total	N = 20
Internal Stage Holdup	= 3.093 E-3 kmol
Condenser Holdup	= 35.160 E-3 kmol
Vapor Load to the condenser	= 2.75 kmol/hr
System Pressure	= 1.013 bar
Reboiler Feed	= 2.93 kmol
Components :	Cyclohexane , n-Heptane , Toluene
Composition :	< 0.407 , 0.394 , 0.199 >

2.6.3.2 Results and Discussions : Figure 2.5 shows the simulated instant distillate composition profiles by our model and by that of Nad and Spiegel. The figure also includes experimentally obtained instant distillate composition data and the adjusted reflux ratio profiles used by Nad and Spiegel and us.

We may observe from figure 2.5 that the reflux ratio profile considered results in a a better fit to the experimental data than the simulated profiles of Nad and Spiegel.

Figure 2.6 shows the accumulated distillate compositions using nonideal and ideal phase equilibrium models. Figure 2.7 shows a similar comparison for instant distillate composition profiles. The results clearly show that the extent of variation on the product composition depends on when the cut is made. However, in this example, at the end of the cut, the product composition does not widely differ in the two cases. The results, together with the solution statistics are given in Table 2.8.

Figure 2.5 Simulation Results by Our Model and Comparison with Experiment and Simulation of Nad and Spiegel (Example 3)

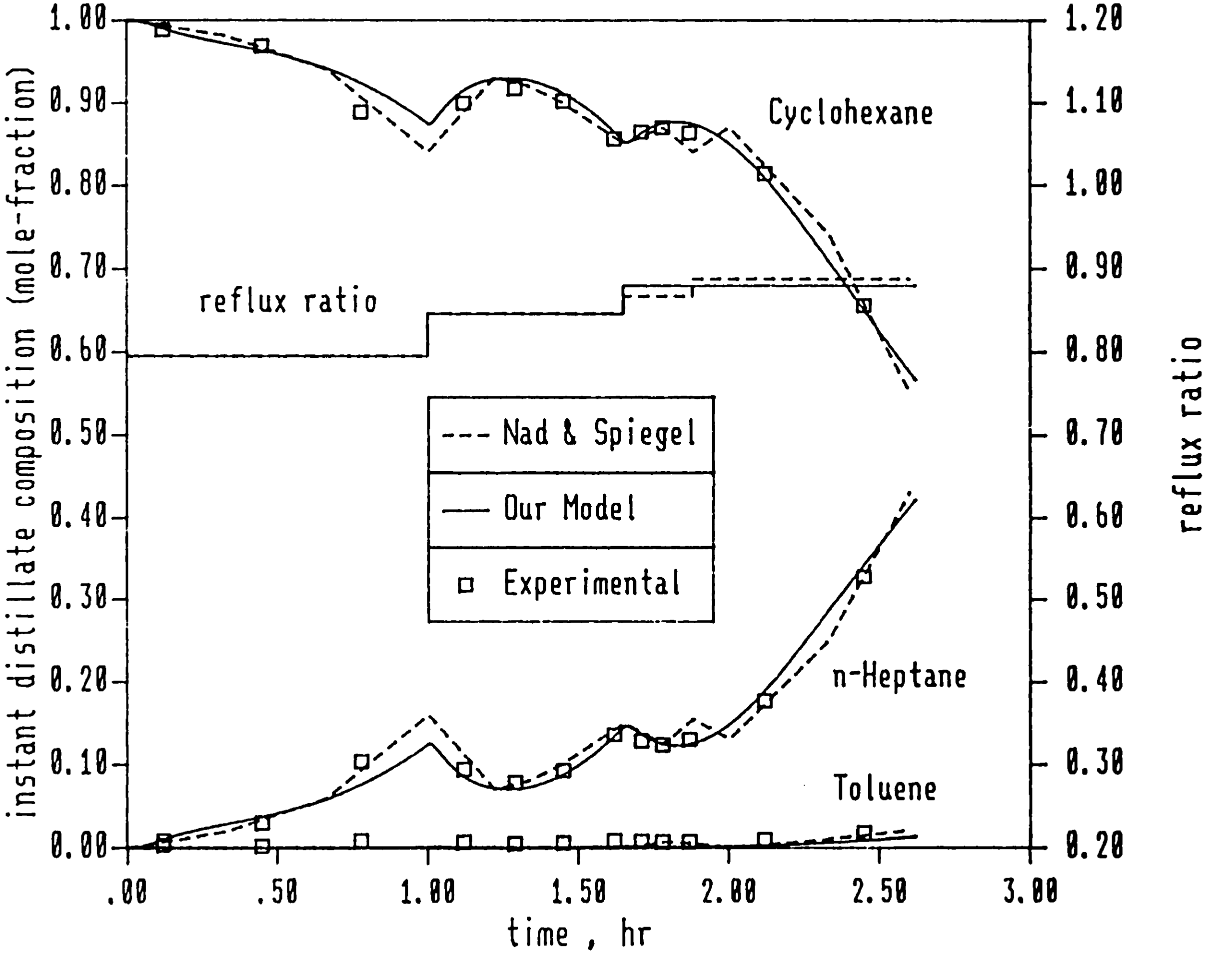


Figure 2.6 Accumulated Distillate Composition Using Rigorous and Simple Phase Equilibrium Models (Example 3)

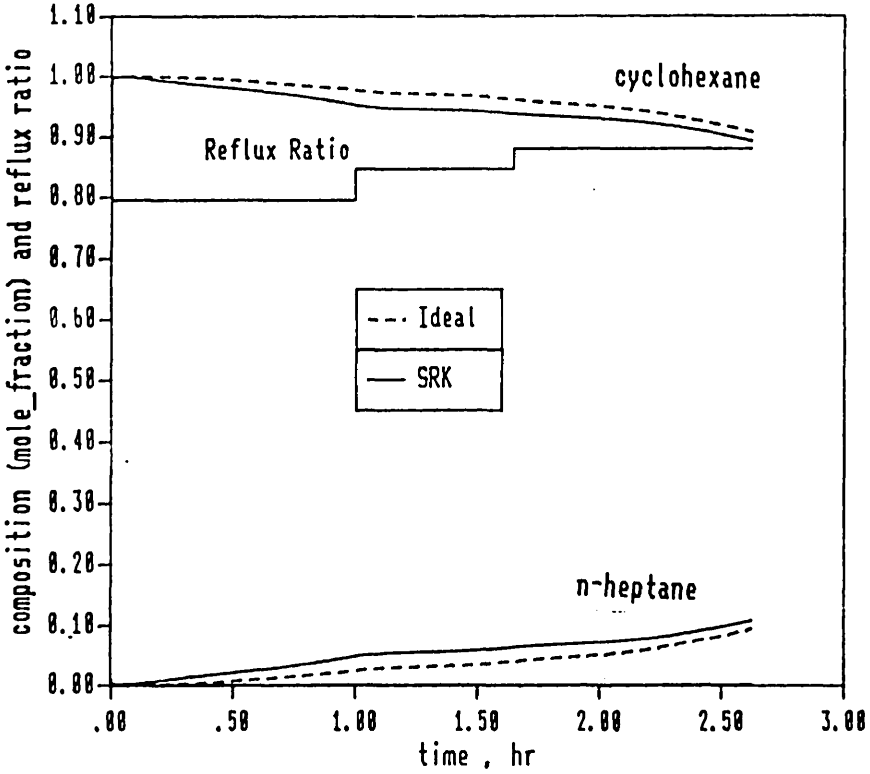


Figure 2.7 Instant Distillate Composition (Example 3)

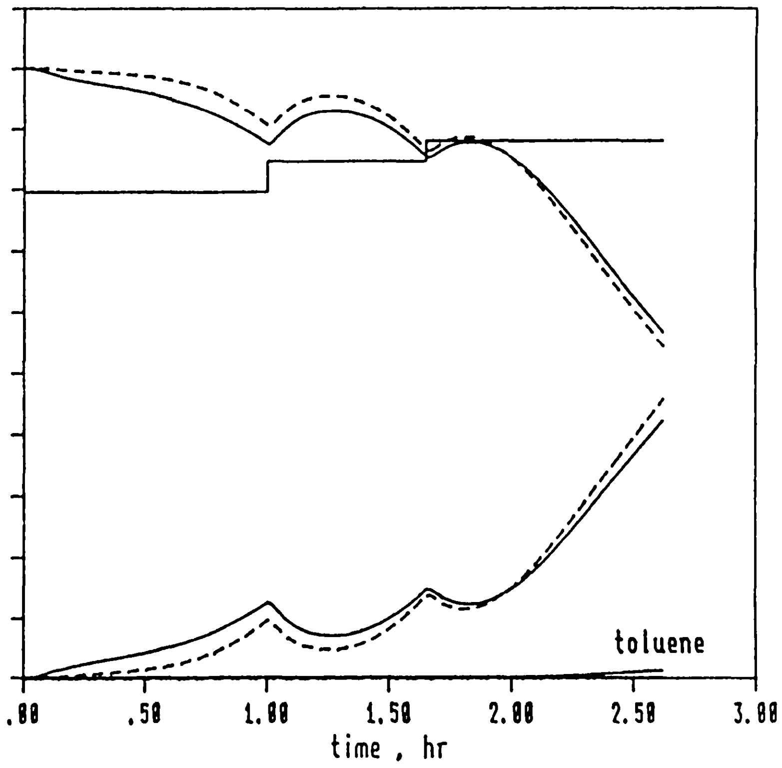


Table 2.8 shows that the product composition obtained by both ideal and nonideal phase equilibrium model are very close to the experimentally obtained value. However, the computation times for the two cases are considerably different. As can be seen from the table about 67% time saving (compared to nonideal case) is possible when ideal equilibrium is used.

Table 2.8 Experimental and Simulated Results of Example 3

Experimental Results of Nad and Spiegel (1987)

Accumulated Distillate (cyclohexane) = 1.16 kmol
Cyclohexane composition = 89.5 %
Operation Time (including total reflux period) = 5.16 hr

Simulation Results by our Model

1. Nonideal case

Accumulated Distillate (cyclohexane) = 1.16 kmol
Cyclohexane composition = 89.1 %
Operation Time (including total reflux period) = 5.16 hr

Total simulation time (approx) = 4.9 mins
Total time in physical properties calc. = 3.3 mins
% of total time spent in phys. prop. = 67.0 %

1. Ideal case

Accumulated Distillate (cyclohexane) = 1.16 kmol
Cyclohexane composition = 90.6 %
Operation Time (including total reflux period) = 5.16 hr

Total simulation time (approx) = 1.6 mins
Total time in physical properties calc. = 0.4 mins
% of total time spent in phys. prop. = 24.0 %

Time saved by using ideal phys. prop. = 67.0 %

Key: Time saved is based on the time required by non-ideal case.

Nad and Spiegel justified using a pseudo-steady state solution procedure for solving their model equations on the basis of saving computation time, but the results presented in Table 2.8 show that computation time even for such a big problem using rigorous integration and physical properties is not too high to be uneconomical.

2.6.4 Example Problem 4

2.6.4.1 Problem Description: We consider a simulation with chemical reaction in this example. No suitable example was found in the literature for reactive batch distillation to consider and compare in this work. Egly et al. studied the reversible reaction scheme $A + B \rightleftharpoons C + D$ and employed optimal control techniques to obtain an optimal operation but no details were given regarding input and kinetic data. Cuille and Reklaitis (1986) considered the esterification of acetic acid with 1-propanol in a reactive batch distillation but their example was not suitable for the purpose. Since 1-propanol (one of the reactants) is more volatile component in the system, the removal of species by distillation would mean removal of the reactant from the column thus producing low conversion. We simulate their example using some of the data given by those authors to test the above fact. It was found that the conversion of acetic acid decreased significantly compared to the operation without any distillation. Therefore, for this type of reaction system distillation means additional cost.

The example we illustrate is the esterification reaction of acetic acid and methanol. This type of reaction system was considered by Corrigan and Ferris (1969) in a batch distillation column (no direct comparison with their results is possible here because of insufficient data) and Sawistowski and Pilavakis (1988) in a continuous distillation column. The reaction products are methyl acetate and water. Since the reaction scheme is reversible (Barbosa and Doherty, 1988) and methyl acetate is highly volatile in the system, the use of distillation should certainly improve the conversion of the reactants. We assume there is only one liquid phase for this reaction system (Sawistowski and Pilavakis also did not experience multiple liquid phase problems for this kind of reaction system). The esterification reaction together with rate equations and rate constants are given in the following :

$$\underset{(A)}{CH_3\text{-}COOH} + \underset{(B)}{CH_3OH} \rightleftharpoons \underset{(C)}{CH_3COOCH_3} + \underset{(D)}{H_2O}$$

The rate equation in terms of formation of component A can be given by -

$$r_A = -k^r_1\, x_A x_B + k^r_2\, x_C x_D$$

where, k^r_1 and k^r_2 are the forward and backward rate constants. We used temperature independent $k^r_1 = 2$ and $k^r_2 = 1$ for convenience. Also we used Antoine's vapor pressure equation to calculate vapor liquid equilibria for convenience. Enthalpies are calculated by the same procedure mentioned in the previous examples. These simplifications, however, will not hamper the conclusions drawn from this example. Table 2.9 presents the input data for the problem.

Table 2.9 Input Data for Example 4

No. of Internal Stages	8
Total Condenser	1
Partial Reboiler	1
Total	N = 10
Internal Stage Holdup	= 1.0 E-3 kmol
Condenser Holdup	= 1.0 E-2 kmol
Operating Pressure	= 1.013 bar
Reboiler Feed :	
Initial Feed	= 2.5 kmol
Components :	Acetic acid , Water
Composition :	< 0.90 , 0.10 > mole fraction
Intermediate Feed (after 1 hr operation at total reflux)	= 2.5 kmol
Components :	Methanol , water
Composition :	< 0.90 , 0.10 > mole fraction
Condenser Vapor Load	= 2.5 kmol/hr

Liquid compositions on the trays and condenser are initialized to that of fresh feed which is a mixture of 90% acetic acid and 10% water (mol fraction). The column is run with this feed for about an hour at total reflux condition before methanol is charged to the column. After 1 hour the intermediate charge containing methanol (0.9 mol fraction) and water (0.1 molfraction) is charged to the column and reaction thus starts. The column is then run at total reflux condition for sufficiently long time (about 4 hrs) to reach steady state (case 1). Column condition at this point of time will give an indication about the conversion of the reactant without any removal of product from the system. Two cases are studied at different constant reflux ratio values after this total reflux operation. In both cases it was decided to produce 2.25 kmols of product. Case 2 operates at reflux ratio 0.9 for about 9 hrs and case 3 operates at reflux ratio 0.95 for about 18 hours. The purpose of these two case studies are to show the effect of reflux ratio on the conversion of the reactant and the purity of the product.

2.6.4.2 Results and Discussions: The results obtained are summarized in Table 2.10. The effect of distillation followed by chemical reaction is very clear from the results of Table 2.10. By removing one of the reaction products by distillation it was possible to increase the overall conversion from about 53% to 73%. The simulation results also show the effect of reflux ratio on the conversion (Figure 2.8) and purity of the distillate obtained by the reactive distillation process. It was possible to increase conversion by operating the column at higher reflux ratio. This was because at high reflux ratio, a very little of the light reactants (methanol) was removed from the system. But since at lower reflux a substantial amount of the reactant was removed from the system it was likely to have low conversion. Purity of the distillate product was also improved at higher reflux ratio. Figure 2.8 clearly shows that after 5 hours of operation the reaction reaches its equilibrium meaning no further conversion of the reactants. As the reaction product (methyl acetate) is removed from the system at finite reflux ratio (case 2 and 3) forward reaction now becomes favorable and further conversion of the reactants is possible. When there are no methanol and ester in the system the reaction stops meaning no further conversion as can be seen in Figure 2.8 at the end of the process. The simulation time for case 2 was about 4 minutes and for case 3 it was 6 minutes using IBM 4341 system.

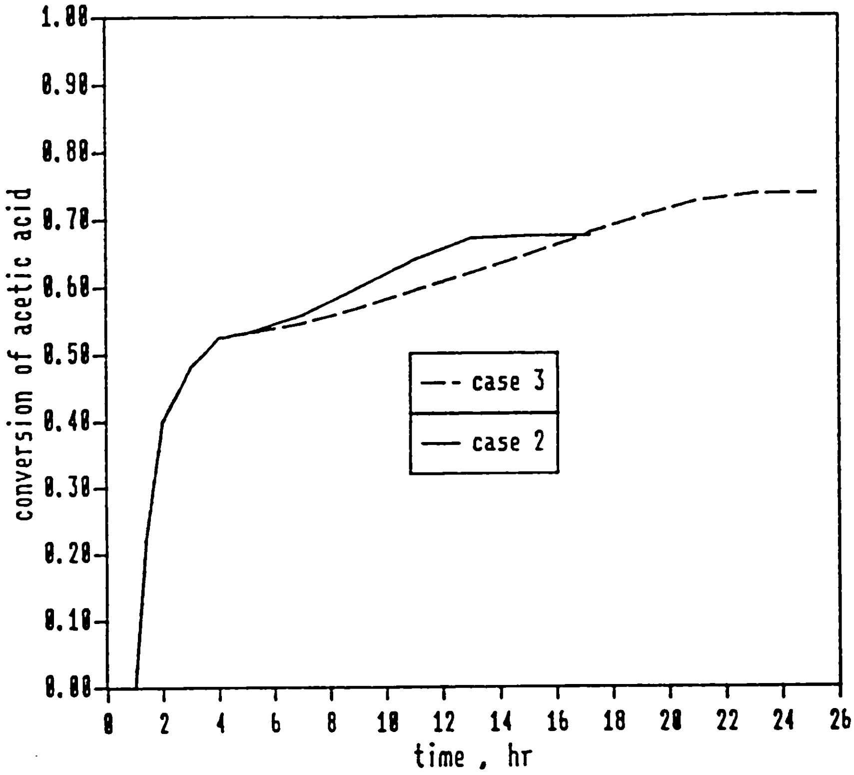
Corrigan and Ferris (1969) and Sawistowski and Pilavakis (1988) experienced an azeotropic mixture of alcohol and ester (35%-65%) in the product when the column was operated at low initial concentration of acetic acid. However, at higher initial acetic acid concentration they were able to avoid this situation and were able to produce ester at higher concentration. Although no value for this initial concentration of acetic acid was reported, using equal concentration of acetic acid and methanol we were able to produce ester at higher composition compared to the azeotropic composition reported by Corrigan and Ferris (1969).

Table 2.10 Results of Example 4

	Case 1 (total reflux)	Case 2	Case 3
Reflux Ratio	1.0	0.90	0.95
Operation Time , hr	5.0	9.0	18.0
Present Distillate			
Acetic Acid	-------	0.004	0.002
Methyl Alcohol	0.066	0.182	0.140
Methyl Acetate	0.934	0.106	0.059
Water	-------	0.708	0.799
Accum. Dist.			
Acetic acid	------	-------	-------
Methyl Alcohol	------	0.291	0.238
Methyl Acetate	------	0.681	0.742
Water	------	0.028	0.020
[kmol]	0.0	2.25	2.25
Still Pot			
Acetic Acid	0.211	0.267	0.219
Methyl Alcohol	0.201	0.006	0.002
Methyl Acetate	0.242	0.003	------
Water	0.346	0.724	0.779
[kmol]	5.00	2.75	2.75
Acetic Acid Charged		2.25 kmol	
Acetic Acid Reacted , kmol	1.195	1.516	1.648
% Conversion	53.11	67.38	73.23

The results obtained clearly indicate that by application of a proper reflux ratio control policy operation of such reactive distillation column can be substantially improved.

Figure 2.8 Conversion Profile of Acetic Acid (Example 4)



2.7 Conclusions

The performance of the batch distillation model presented in this chapter on a number of test cases from the literature clearly shows its capability for use in simulation and control studies. The proposed model is detailed enough to consider major issues like column holdup, general and rigorous physical property models, energy balance etc. and simple enough to reduce computation time by using constant molar holdup assumption and by neglecting plate hydraulics. Simulation times are reported for individual case studies, however, this measure will be directly dependent on the number of plates, number of components and physical property model used and on the length of the simulated operation. A general purpose batch distillation code has been written based on this model with several configurational and operational options and also with the provision of generating analytical derivatives in sparse matrix form required by the integration routine. This code is used in the next few chapters for finding optimal operational policies in batch distillation.

Chapter 3

OPTIMAL OPERATION OF A BATCH DISTILLATION COLUMN (SINGLE BATCH)

3.1 Introduction

In this chapter optimal operational policies for distillation of a single batch are developed using a variety of objective functions (minimum time, maximum product, maximum profit etc.) and of constraints (amount of product, purity of the product etc.). An optimal control problem is formulated, which uses the rigorous and realistic column model described in chapter 2. A general purpose batch distillation front end translator is developed which offers several configurational and operational options and permits setting up a problem with greater ease. Optimal profiles of key control variable are obtained using a general purpose optimal control and optimisation code. Application of the optimal control policies to several operational problems show that large improvements can be obtained over a more conventional mode of column operation (constant reflux ratio operation).

Depending on the number of components in a mixture to be processed, usually a number of cuts are made in a single batch operation. Some of the cuts are the desired products while others are the intermediate fractions that can either be thrown away or can be recycled to subsequent batches to obtain further separation. The amount and composition of each cut, and the total time required to process each batch (hence productivity) depend on the column operation, and in particular on the policy adopted for the reflux ratio. Since for many separations even a modest change in batch time has a significant economic impact, there is a very large incentive for improving the operation of such columns.

An optimal reflux rate policy, which is essentially a trade-off between constant distillate composition and constant reflux methods, is based on the ability to yield the most profitable operation. Early studies showed that although the policy of changing the reflux ratio to maintain product purity specification (constant distillate composition operation) was within few percent of the optimal policy, the constant reflux ratio policy was far from the optimal (Coward, 1967; Robinson [1969,1970]; Kerkhof and Vissers, 1979). In most cases reflux ratio was used as control variable of the process and in some cases (packed columns) the vapor boilup rate was used together with the reflux ratio (Hansen and Jorgensen, 1986; Cristensen and Jorgensen,1987).

Most of the work to date on optimal control of batch columns (chapter 1) used Pontryagin's maximum principle applied to very simplified column models expressed as an ODE system except for Converse and Huber (1963) and Liles (1966) who used the dynamic programming method. Since it was decided in this work to use the model presented in chapter 2, which is an index 1 system of DAEs, therefore, it was necessary to use an optimal control algorithm that can handle DAEs system.

Recently Morison (1984) and Renfro et al. (1987) developed optimal control algorithms and applied them to processes described by a system of DAEs.

Morison (1984) used a sequential model solution and optimisation strategy which is commonly known as Feasible Path Approach. In this approach, the process variables are partitioned into dependent variables and independent variables (decision variables), and for each choice of the decision variables, the simulator is used to converge the process equations (DAEs). Therefore, the method includes two levels, the first level performs the simulation to converge all the equality constraints and to satisfy the inequality constraints and the second level performs the optimisation. The resulting optimisation problem is thus an unconstrained nonlinear optimisation problem or a constrained optimisation problem with simple bounds for the associated decision variables plus any interior or terminal point constraints (e.g. amount and purity of the product at the end of the cut).

The advantage of this approach is that the resulting optimisation problems are in general small in terms of the number of variables. Since each search point is feasible, if the process is terminated at the optimisational level before reaching a solution, the terminating point is still feasible and may be acceptable as a practical, although sub-optimal, solution of the problem.

However, since the feasible path approach requires a complete solution of DAEs for each trial value of the decision variables this may be expensive. The simulation level may fail since a feasible region may not even exist for certain value of the decision variables.

Renfro et al. (1987) proposed a method, using piecewise constant functions for the independent variables, that combined quasi-Newton optimisation algorithms and global spline collocation to simultaneously optimise and integrate systems described by DAEs. Application of the collocation techniques to DAEs results in a large system of algebraic equations which appear as constraints in the optimisation problem. This approach is generally known as the Infeasible Path Approach which results in a large sparse optimisation problem.

The main advantage of this approach is that it avoids repeated simulations for each optimisation iteration and hence eliminates the possibility of premature termination due to a convergence failure of the simulation. Also, this approach is faster in terms of computation time compared to feasible path approach but at the expense of increasing the size of the optimisation problems in terms of variables.

However, in practice, a chemical process may not be physically well defined over a wide range of constraints. Therefore, some precautions must be taken to identify and to satisfy such constraints (e.g. non-negative flows) when an infeasible path method is used. Also, this approach is less familiar to design engineers since each point may not be feasible and acceptable, and a large number of variables may require to be initialized. In addition, it is difficult to identify the effect of the decision variables on the objective function in the course of optimisation and thus providing diagnostic information becomes a more serious issue for the infeasible path approach (see Chen, 1988 for more details on Feasible and Infeasible Path approach in optimisation).

Although the method proposed by Renfro et al. appears to have much potential, the choice was made to use the algorithm developed and implemented by Morison. This is because of several reasons : the algorithm is general, can optimise design variables, initial conditions of the states in addition to control and control switching times. The optimisation of control switching times together with final time is most desirable to obtain time optimal operation in batch distillation. The optimisation of the initial conditions is also desirable in developing off-cut recycle policies (presented in chapter 4 and 5). Also the implementation of Morison's algorithm includes other options: fixed switching times and final time, variable switching times but fixed final time (which is desirable to obtain maximum distillate operational policy) etc.

The algorithm of Morison was based on the work of Joffe and Sargent (1972) and Sargent and Sullivan (1979). The optimal control problem for DAE system is formulated as a nonlinear programming problem and controls are parameterized giving a set of parameters including control parameters (controls levels and switching times), design parameters, initial conditions and final time. These were optimised to give a minimum of the objective function subject to any constraints present. A sequential quadratic programming (SQP) method is used for optimisation of parameters. Optimal control formulation and optimisation technique are discussed in brief in the next section and are presented in more detail in Morison (1984).

3.2 Formulation of the Optimal Control Problem as a Nonlinear Programme

The batch distillation process described by the system of DAEs in chapter 2 is written as:

$$f(t\,,\dot{x}_1(t)\,,x(t)\,,u(t)\,,v)=0\,,\qquad [t_0,t_F] \qquad (3.1)$$

where t is the independent variable (time), $x(t) \varepsilon R^n$ is the set of all the variables (not to be confused with the liquid compositions), $\dot{x}_1(t)$ denotes the derivative of $x_1(t)$ with respect to time, $x(t) = [x_1(t), x_2(t)]$; $x_2(t)$ being algebraic variables, $u(t) \varepsilon R^m$ is a vector of control variables, such as reflux ratio, and $v \varepsilon R^p$ is a vector of time invariant parameters (design variables), such as molar holdups on each plate, efficiency etc. The time interval of interest is $[t_0,t_F]$ and the function $f:RxR^nxR^nxR^mxR^p \text{-->} R^n$ to all its arguments and is subject to restriction $[f_{\dot{x}_1} : f_{x_2}]$ is nonsingular. $f_{\dot{x}_1}$ and f_{x_2} are the derivatives of the function f with respect to $\dot{x}_1$, x_2.

Since the DAE system represented by equation (3.1) is an index 1 system, consistent initialization only requires $x_1(t)$ to be initialized. If these initial conditions $x_1(t_0) = x_{10}$,are to be optimised together with control and design variables then they must satisfy the bounds

$$\begin{gathered} a^0 \leq x_{10} \leq b^0 \\ a^u(t) \leq u(t) \leq b^u(t), \quad t \varepsilon [t_0, t_F] \\ a^v \leq v \leq b^v \end{gathered} \tag{3.2}$$

where $a^u(t)$ and $b^u(t)$ are given continuous functions of time on $[t_0,t_F]$. Also, in general, we have interior or terminal constraints at time t_p of the form :

$$a^F \leq F(t_p, \dot{x}_1(t_p), x(t_p), u(t_p), v, x_{10}) \leq b^F, \tag{3.3}$$

where $F(t,\dot{x}_1,x,u,v,x_{10}) \varepsilon R^q$ and is continuously differentiable with respect to its arguments and $t_0 \leq t_p \leq t_F$. At the terminal point $t_p = t_F$. The system performance is measured in terms of a scalar objective function to be minimized or maximized

$$J = F_0(t_F, \dot{x}_1(t_F), x(t_F), u(t_F), v, x_{10}) \tag{3.4}$$

The general optimal control problem is to choose an admissible set of controls u(t), initial conditions x_{10}, design variables v and final time, t_F ,to minimize (or maximize) the objective function, J , subject to the bounds on the control, initial condition, design variables and to all other constraints.

3.2.1 Parameterization of the Controls: To pose the optimal control problem as a nonlinear programming problem the controls u(t) are approximated by a finite dimensional representation. The time interval $[t_0,t_F]$ is divided into a finite number of subintervals, each with a set of basis functions involving a finite number of parameters

$$u(t) = \phi^j(t , z_j) \ , \ t \, \varepsilon \, [(t_{j-1} , t_j) , j = 1,2, ... M \ ] \qquad (3.5)$$

where $t_M = t_F$. The functions of $\phi^j (t , z_j)$ are assumed to be continuously differentiable with respect to t and z_j, and the derivatives are uniformly bounded. The control is thus defined by the parameters z_j and the switching times t_j , j=1,2,....,M. The control constraints now become

$$a^u(t) \leq \phi^j (t, z_j) \leq b^u(t) \ , \ t \, \varepsilon \, [(t_{j-1}, t_j), \ j = 1,2,...,M] \qquad (3.6)$$

The set of decision variables for the nonlinear programme can now be written as

$$y = \{ x_{10,} z_1, z_2,. . . . ..., z_M, t_1, t_2, ..., t_F, v \} \qquad (3.7)$$

(again this y is not to be confused with vapor compositions)

3.2 2 Optimisation: The optimisation problem presented in the previous section can be written more formally as :

Find the set of decision variables, y (given by eqn 3.7) which

minimize J (y) (3.8)

subject to Equality constraints (eq. 3.1)

Inequality constraints (eq. 3.2, 3.3 and 3.6)

This constrained nonlinear optimization problem is solved using the successive quadratic programming (SQP) algorithm of Powell (1978) who based it on the work of Han (1976). The efficiency and robustness of the algorithm was discussed by Morison. The difficulties arising from the use of an other optimisation code (MINOS/AUGMENTED) for the optimal control of DAE system were discussed by Morison and are not reported here.

Very recently Chen (1988) has developed and implemented an improved SQP algorithm which was found to be more robust and efficient compared to that modified and implemented by Morison. The algorithm of Chen requires in general smaller number of expensive function evaluation and is useful for large scale optimisation. Since his work was not available until most of the work in this thesis was done, therefore, his code will only be used in the later part of this thesis(chapter 4 and 5)

Each "function evaluation" of the optimiser (eqn 3.8) requires a full integration of the DAE system (eqn 3.1) and this is achieved using a Gear's type integrator as mentioned in chapter 2. The solution of the optimisation problem requires the derivatives of the objective function and constraints with respect to all the arguments of y (eqn 3.7). These are evaluated in an efficient way using adjoint variables, based on a variational analysis. Full detail of these procedure are described in Morison (1984).

3.3 Optimal Control Formulation for Single Batch Distillation (single cut)

The optimal operation of a batch column depends on the objective to achieve at the end of the process. Depending on the objective function and any associated constraints, the optimal control problem can be of several types. A brief formulation of each type is presented in this section. Situations in which each formulation may be applied are also discussed. All the formulations use the nomenclature defined in Figure 3.1 and in all cases the initial state (x_{10}) and design variables (v) are assumed to be fixed in advance and are therefore not optimised.

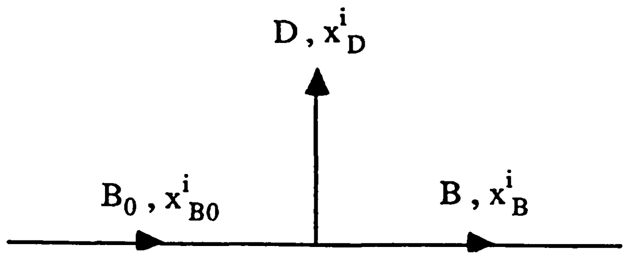


Figure 3.1 A Typical Batch Operation (single cut)

Figure 3.1 shows a typical batch operation for a particular "cut" i, where the component i is being produced as overhead. B_0, x^i_{B0} are the amount of reboiler charge and its composition with reference to component i; D, x^i_D are the amount and purity of overhead distillate; and B, x^i_B are the amount and composition of the residue left after the operation. The batch time for the operation is given by t^i. The batch time, here, simply means the time to produce a cut. It excludes the charging and cleaning time before and at the end of the batch. For simplicity we avoid the use of superscript (i) in the following sections.

3.3.1 Minimum Time Problem: Coward (1967), Robinson (1969,1970), Mayur and Jackson (1971), Hansen and Jorgensen (1986) minimized the batch time to yield a given amount and composition of distillate. This type of problem is also defined as the time optimal control problem. A time optimal operation is very often desirable when the amount of product and its purity are somehow fixed but a reduction in batch time means a saving of operating costs (steam, operator) and an increase in the number of batches which can be processed. Mathematically the optimal control problem can be written as:

$$\begin{aligned} &\underset{r(t)}{\text{Min}} && J = t_F \\ &\text{subject to} && (D - D^*)^2 \leq \varepsilon_1 \\ &&& x^*_D - \varepsilon_2 < x_D < x^*_D + \varepsilon_2 \end{aligned} \qquad (3.9)$$

where D , x_D are the amount of distillate and its composition at the final time t_F ; D^*, x^*_D are the specified amount of distillate product and its purity and ε_1 and ε_2 are very small positive numbers. $r(t)$ denotes all the optimisation variables [controls (reflux ratio) and their switching times].

Using simplified column models (ODEs) a time saving of more than 10% compared to constant reflux ratio operation was reported by the earlier workers.

3.3.2 Maximum Distillate Problem : Converse and Gross (1963), Diwekar et al. (1987) considered an optimisation problem which maximizes the amount of distillate product of specified purity for a given time of operation t^*_F. This type of operation is often useful when a fixed period is assigned to a particular unit in the process flowsheet for a particular job. The optimisation problem can be written as:

$$\begin{array}{ll} \underset{r(t)}{\text{Max}} & J = D \\ & \\ \text{Subject to} & x^*_D - \varepsilon_2 < x_D < x^*_D + \varepsilon_2 \\ \text{and} & t_F = t^*_F \end{array} \qquad (3.10)$$

where D, x_D, x^*_D , $r(t)$ and ε_2 are as described in section 3.3.1. In previous work using simplified column models an optimal policy resulted in product yields of up to 3-7% over those obtained with conventional policies.

3.3.3 Feasible Trajectory Problem: During the early stage of this work we proposed a different kind of optimal control formulation for batch distillation which neither minimizes the batch time nor maximizes the amount of distillate. The objective was set as to minimize a quadratic function which closely satisfies the amount of product. The purity of the product was set as an inequality constraint to be satisfied at the end of the process. In that formulation both reflux ratio and time were varied. The problem can be written as :

$$\underset{r(t)}{\text{Min}} \qquad J = (D - D^*)^2 \qquad (3.11)$$

Subject to $x^*_D - \varepsilon_2 < x_D < x^*_D + \varepsilon_2$

where D, x_D, x^*_D, r(t) and ε_2 are as described in section 3.3.1. In fact, what is required is just to find a feasible combination of reflux ratio profile and processing time. This kind of formulation does not necessarily minimize the batch time, but obtains a feasible sub-optimal column operation. In spite of not explicitly requiring an "optimal" solution from the point of view of operation time, we found that usually the solution of this problem saves plant operation time to a great extent compared to constant reflux operation. Results of the application of the method are presented later in this chapter.

3.3.4 Maximum Profit Problem : Kerkhof and Vissers (1978) combined the minimum time and the maximum distillate problems to maximize a profit function, using simple binary model and maximum principle solution. Both the amount of distillate and the time of operation were free and the only constraint arose from the purity of the product. Reflux ratio was used as the control variable of the process. The problem can be written as:

$$\max_{r(t)} \quad P = \frac{\text{added value to product}}{\text{time}} - \text{operating cost}$$

$$= \frac{C_1 D(t_F) - C_2 B_0}{t_F + T_s} - C_3 \qquad (3.12)$$

Subject to $x^*_D - \varepsilon_2 < x_D < x^*_D + \varepsilon_2$

where P is the profit (£/hr), C_1 is the product value (£/lbmole or kmol), C_2 is the raw material cost (£/lbmole or kmol), C_3 is the operating cost (energy, wages, deprcciation etc.) (£/hr), T_s is the charging and cleaning time between batches. Kerkhof and Vissers considered C_3 as fixed operating cost, but in practice, these may also vary with different boilup rate. Therefore, more general expression could be used to evaluate the operating cost per hour.

At the optimum profit t_F corresponds to the minimum time for D (t_F) and D (t_F) is the maximum distillate in time t_F. x_D, x^*_D, r(t) and ε_2 are same as before. The authors showed that for difficult separations optimal reflux control policy yields upto 5% more distillate, corresponding to 20-40% higher profit, than either constant overhead or constant reflux control policies. When $C_1 = 1.0$, $C_2=C_3=0.0$, the maximum profit problem reduces to a <u>maximum productivity</u> problem.

In all the above four types of optimal control formulations, the model equations with appropriate bounds on control variables (as mentioned in section 3.2) act as constraints to the problems.

3.4 Optimal Control Formulation for Single Batch Distillation (multicut)

Having more than one cut in batch distillation is very common when processing a multicomponent mixture. A general optimal control problem formulation for such operation will exactly be the same as that presented in section 3.2. The cut specifications in this case will appear as interior constraints of the nonlinear optimisation problem. The end of each cut, however, may well be defined by a finite number of control intervals. In this section we will discuss about the type of interior point constraints that should be considered in such general formulation.

Before discussing the type of interior point constraint it is worth at this point to define the concept of "cut component" which is frequently used in this work. It is to be noted at this point that the components are numbered in such a way that component 1 is the lightest component and component n_c is the heaviest is in the mixture. In a particular cut (1,2,3,.......) the "cut component" simply refers to the component numbered by the same number as used to identify that particular cut. For example, in cut 1 (main-cut or off-cut) the "cut component" is component 1, in cut 2 the "cut component" is the component 2 and likewise. It is sometimes useful to refer components onward of the "cut component" with respect to the "cut component". For example, in cut 2, the 3rd component in the mixture can be referred to as 2nd component with respect to the "cut component". The usefullness of the above definition can be realised in the following and also in chapter 5 where optimal recycle policies for multicomponent batch distillation are discussed.

The optimisation problems mentioned in section 3.3 can be extended to multicut operation and the interior point constraints will be typically the purity specification of the "cut component" at the end of each cut. This holds true even for minimum time problem. If the amount of product is specified at the end of each cut together with a purity specification, the general formulation is automatically decomposed into a series of independent time optimal control problems and the summation of the minimum times over all the cuts will be the global minimum time for the whole problem.

However, as the number of components increase, a general multicut optimal control problem will result in a large number of optimisation variables. This is because of increasing number of control intervals. The number of variables will be doubled when the switching times are also to be optimised. Furthermore, inclusion of the interior point constraints will add greater complexity to the problem.

However, there are cases, when both the amount and purity of the product at the end of each cut are known or required to be satisfied and the objective is to improve the operation of such columns. In this work we restrict ourselves to such cases and consider the minimum time and feasible trajectory problems for such a case (see example 3 in section 3.6). Because of the above specifications at the end of each, the problem is decomposed into a series of independent time optimal control problems. Initialization of each of the problems is done by using the solution of the previous problem.

The minimum time problem for a multicut operation can be defined as follows: given the amount and composition of fresh feed (B_0, x^i_{B0}), determine the optimal reflux ratio policy to obtain the given amount and composition of the main product cuts (D^*_C, x^*_D) and the given amount and composition of the final bottom product (B^*_C, x^*_B) [where C identifies a particular cut] in a minimum time. Mathematically the problem can be written as:

$$\underset{r(t)}{\text{Min}} \qquad J = t^1 + t^2 + \; = t_F \tag{3.13}$$

where t^1, t^2 ... etc. are the minimum times for individual cuts and t_F is the global minimum for the multicut operation. t^1, t^2 can be obtained using the minimum time formulation given in section 3.3.1. Similar formulations can be presented for other types of problems.

3.5 Implementation of Optimal Control in Batch Distillation

The use of the optimal control package for DAE systems developed by Morison (1984) requires the user to supply :

1. All the model equations in residual form.
2. Derivatives of the equations with respect to differential, algebraic, design and control variables in sparse matrix form.
3. Physical property models.
4. Objective function and constraints.
5. A suitable DAE integrator.
6. A suitable optimisation routine.

As part of the present work a general batch distillation code was written with several configurational, operational and a general physical property model options. The user can select

1. Any number of plates and components.
2. Reboiler and condenser system, but no intermediate plates.
3. Total condenser with or without holdup tank.
4. Any physical property model.
5. Vapor load to the condenser or reboiler duty as design variable.
6. Reflux ratio as control variable.
7. Any type of objective function and constraints (in particular those mentioned in section 3.3) for single and multicut operation.

The physical property code (Maduabueke, 1987; Macchietto et al., 1988) used throughout this work has the option of providing analytical derivatives of k-values and enthalpies with respect to composition, temperature and pressure which are interfaced to the batch distillation code using common blocks. Thermodynamic input data are also interfaced with the physical property model through common blocks. However, in cases, where analytical derivatives are not provided these can be generated by using finite difference techniques.

Other options such as use of a partial condenser, of vapor boilup rate as a design or control variable and of chemical reaction terms in the model equations (for optimal control study) are not presently available but could be very easily implemented in the code.

Given any suitable combination of the options given above the code automatically generates:

1. All the model equations.
2. All the derivatives (analytical) in sparse matrix form.

3.6 Example Problems

Three example problems are chosen in this work to demonstrate the application of different types of optimal control problems in batch distilllation. The first two examples will consider single cut operations while the third one will consider a multicut operation.

3.6.1 Example Problem 1 (single cut):

3.6.1.1 Problem Description: We choose the example problem by Boston et al. (1980) presented in chapter 2. Input data regarding configuration and operation of the column for this problem were presented in detail in Table 2.4 of chapter 2. The problem involves a complex separation of quarternary mixture in 5 operational steps. The simulation results for the problem were given in Table 2.6 of the previous chapter.

All the four types of optimal control problems described in section 3.3 are considered here to obtain optimal control profiles (reflux ratio) for the first operation step of the problem as shown in Table 2.6.

For the minimum time problem, the amount of distillate product and its composition at the end of first operation step ($D^* = 8.139$ kmol, $x^*_D = 0.981$, see Table 2.6) are set as constraints and the batch time is minimized.

For the maximum distillate problem, the batch time is fixed ($t^*_F = 4.07$ hr, see Table 2.6) and the product composition ($x^*_D=0.981$) at the end of this time is set as a constraint and the amount of product is maximized.

For the feasible trajectory problem, the amount of product ($D^* = 8.139$ kmol) is set as the objective to attain at the end of the process. The product composition ($x^*_D=0.981$) is set as constraint to be satisfied at the end of the process.

For the maximum productivity problem, the product composition ($x^*_D=0.981$) is set as constraint and the optimal control problem finds the optimal amount of product and batch time to maximize the productivity.

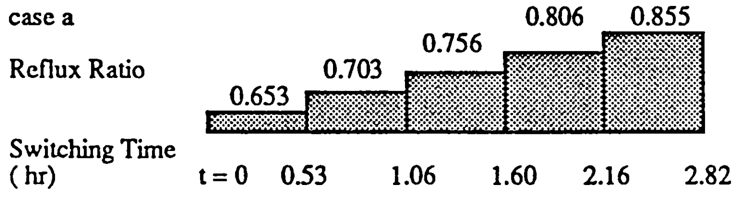
We use 5 control intervals in each of the above problems. In each case the results are also compared with those obtained using single constant reflux ratio. For all types of problems we use $\varepsilon_1 = 0.0001$ and $\varepsilon_2 = 0.001$. The bounds on the control are $0.3 < r < 1.0$.

3.6.1.2 Results and discussions: The results for each type of problems are presented in Table 3.1-3.4. Comparison of the results for all cases using both optimal variable reflux ratio and optimal single constant reflux ratio are presented in Table 3.5. The results clearly show that the constraints on purity in all formulations are satisfied within tolerance (ε_1 and ε_2). Also the optimal reflux ratio profiles are all increasing with time except the feasible

Table 3.1 Results of Minimum Time Problem (Example 1)

	Optimal R.R Policy (5 intervals) [case a]	Constant R.R Policy [case b]
Batch time (hr)	2.82	4.01
Propane Production. (lbmol)	8.117	8.117
Product Conc. (%)	98.1	98.1

case a



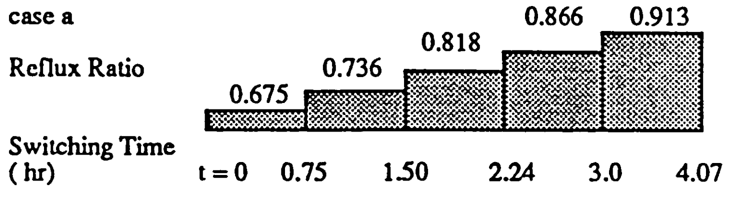
case b

Optimal constant Reflux Ratio = 0.831

Table 3.2 Results of Maximum Distillate Problem (example 1)

	Optimal R.R Policy (5 intervals) [case a]	Constant R.R Policy [case b]
Batch time (hr) (fixed)	4.07	4.07
Propane Production. (lbmol)	9.26	8.15
Product Conc. (%)	98.1	98.1

case a



case b

Optimal constant Reflux Ratio = 0.833

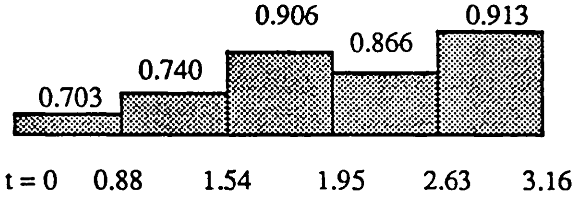
Table 3.3 Results of Feasible Trajectory Problem (Example 1)

	Variable R.R Policy (5 intervals) [case a]	Constant R.R Policy [case b]
Batch time (hr)	3.16	4.05
Propane Production. (lbmol)	8.141	8.139
Product Conc. (%)	98.1	98.1

case a

Reflux Ratio

Switching Time (hr)



case b

Optimal constant Reflux Ratio = 0.832

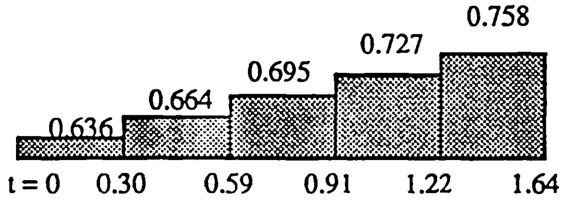
Table 3.4 Results of Maximum Productivity Problem (Example 1)

	Optimal R.R Policy (5 intervals) [case a]	Constant R.R Policy [case b]
Batch time (hr)	1.64	1.75
Propane Production. (lbmole)	5.88	5.67
Product Conc. (%)	98.1	98.1

case a

Reflux Ratio

Switching Time (hr)



case b

Optimal constant Reflux Ratio = 0.730

Table 3.5 Comparison of the Productivity for STEP 1 Operation of Example 1 by Different Optimal Control Formulations.

Problem	Productivity by optimal variable reflux ratio	Productivity by optimal constant reflux ratio
Minimum Time	2.881	2.025
Maximum Distillate	2.275	2.002
Feasible Trajectory	2.574	2.012
Maximum Profit	3.587	3.240

Productivity obtained in the simulation (Table 2.6) = 1.999

trajectory problem. The reason is probably due to the nature of the objective function of this problem. In all cases attempted it was found that an optimal reflux policy is advantageous over a conventional constant reflux ratio operation mode.

3.6.2 Example Problem 2 (single cut)

3.6.2.1 Problem description: In this problem we consider the ternary separation example of Nad and Spiegel (1987). The problem was described in Table 2.7 and the simulation results were presented in Table 2.8 of chapter 2.

Here we consider the minimum time problem to obtain the same amount of product at the specified composition (D^*= 1.16 kmol, x^*_D= 0.906) as were obtained in the simulation, using an ideal phase equilibrium model (see Table 2.8). The simulation used 4 reflux ratio levels including initial total reflux operation. In this problem we also use 4 control intervals (reflux ratio levels) to compare the simulation results. The bounds on the control and the tolerances on the constraints are same as mentioned in the previous example.

The simulation results presented in the previous chapter used 2.54 hr of initial total reflux operation (also used in the experimental column by Nad and Spiegel, 1987) before any product was withdrawn from the column. In this problem it is also the aim to find out whether an initial total reflux period is at all required or not for the given separation and if it is, for how long the column is to be operated under total reflux condition.

In this work we also consider the maximum profit problem with some arbitrary cost functions. The aim is to produce cyclohexane at the specified composition ($x^*_D = 0.906$) most profitably. The cost functions used are $C_1 =$ £100.0 /kmol, $C_2 =$ £20.0 /kmol, $C_3 = 10.0$/hr and $T_s = 0.5$ hr.

3.6.2.2 Results and Discussions : Results are summarized in Table 3.6 for the minimum time problem, together with detailed solution statistics. Figure 3.2 and 3.3 show the corresponding accumulated and instant distillate composition profiles together with the optimal reflux ratio profiles. The reflux ratio profile is increasing with time as expected. The results clearly show the benefit of optimal control of the reflux ratio. As can be seen from Table 3.6 the operation time is reduced by at least 50% compared to that in the simulation and experimental runs. The results also show that for the given separation an initial total reflux operation is not at all required and the product can be collected from the very beginning of the process.

Table 3.6 Results of Minimum Time Problem and Detailed Solution Statistics (Example 2)

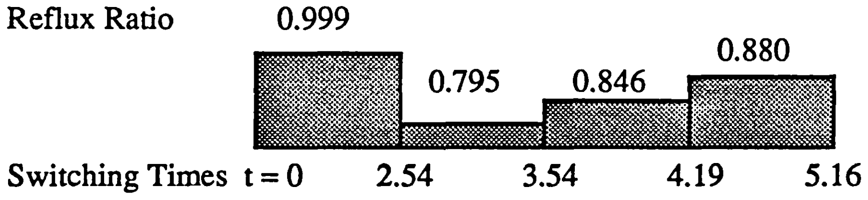
	Objectives Set D , x	Objectives attained
Amount of Cyclohexane	1.16 kmol	1.159
Cyclohexane Conc.	90.60 %	90.61
Batch time	(5.16 hr) @	2.61

% Operation time saved (compared to simulation) = 50.58

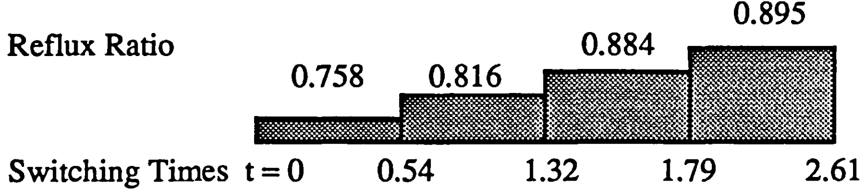
Solution Statistics :

No. of function evaluations = 17
No. of gradient evaluations = 13
Time for each function evaluation (approx.) = 1.3 mins. (CPU)
Time for each gradient evaluation = 2.1 mins.
Total time used in physical properties = 11.8 mins
Total time (including translator and optimiser) = 49.9 mins.

Initial Reflux Ratio Profile:



Optimal Reflux Ratio Profile:



@ batch time as used in simulation and also in experiment (see chapter 2)

Figure 3.2 Accumulated Distillate Composition (Minimum Time Problem, Example 2)

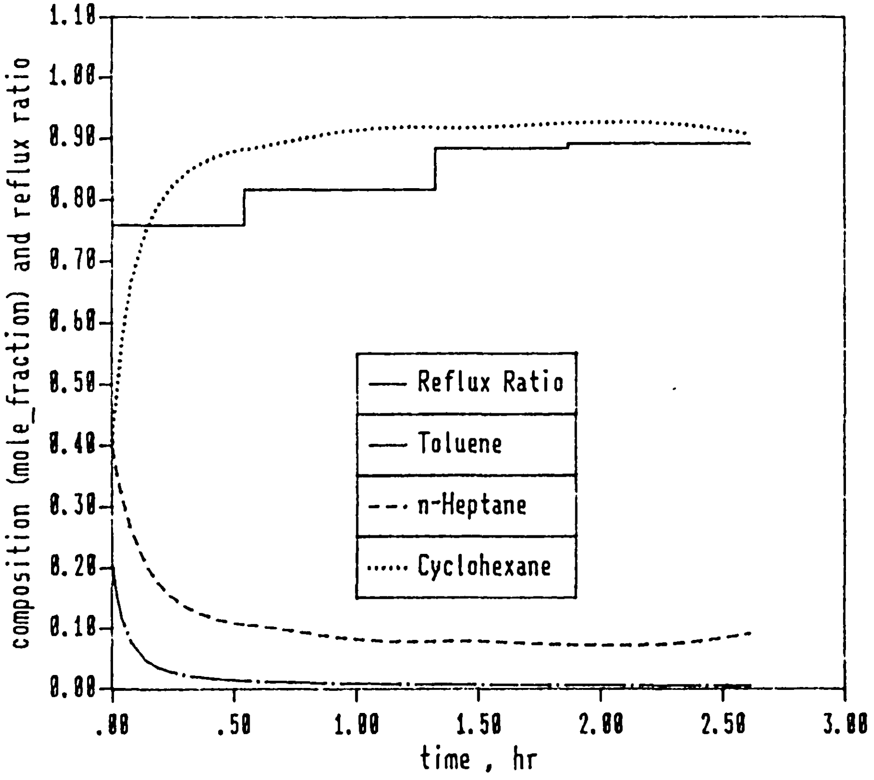
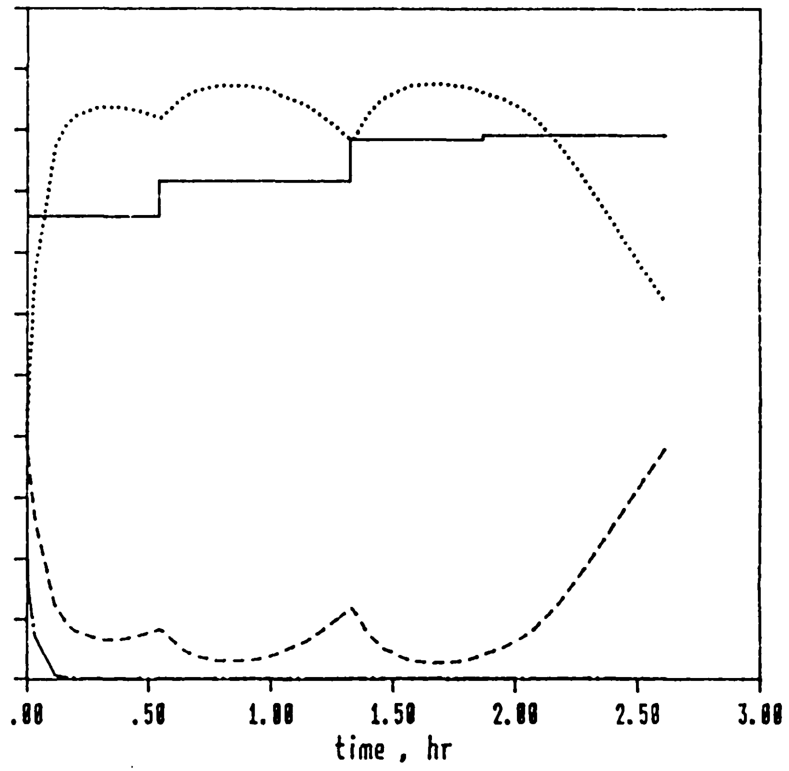


Figure 3.3 Instant Distillate Composition (Minimum Time Problem, Example 2)

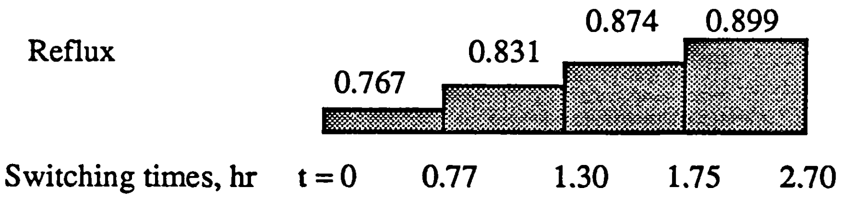


For the maximum profit problem the results are summarized in Table 3.7. It shows that for maximum profit the optimal amount of cyclohexane to be obtained is 1.18 kmol at the desired purity of 90.6%. Using the cost functions considered in this case, the amount of cyclohexane (1.16 kmol) set for the minimum time problem results in a profit of £8.46 / hr which is slightly below the maximum profit. The optimal reflux profile is increasing all along as can be seen from Table 3.7.

Table 3.7 Results of Maximum Profit Problem (Example 2)

Optimal amount of cyclohexane = 1.18 kmol
Composition of cyclohexane (%) = 90.6
Batch time required = 2.70 hr
Maximum profit = £8.56 / hr

Optimal reflux ratio profile :



3.6.3 Example Problem 3 (multicut)

3.6.3.1 Problem description: We again choose the example problem by Boston et al. (1980) presented in chapter 2 and also in section 3.6.1 of this chapter. As mentioned before, the problem involves 5 operational steps producing 3 main product cuts, with the addition of a secondary charge after the first cut. Operation Step 1 and 2 in Table 2.6 form CUT 1, where propane is removed from the system. Further to the addition of a second charge (see Table 2.4 for input details of the problem) a butane production step follows (CUT 2). The remaining two steps (CUT 3) remove pentane from the system, leaving hexane as the bottom product. The total time for the operation was 30.24 hr (simulation, see Table 2.6) using single constant reflux ratio in each operation step.

Here we consider the minimum time problem for this multicut operation. We specify both the amount and composition of the product (set to the values presented in Table 2.6) at the end of each operation step. For the first 4 steps these refer to the accumulated distillate, for step 5 these refer to the amount left in the still pot. Optimal control profiles (reflux ratio) in each operation step are then obtained independently of other step with the final state of each step being the initial state of the next step. We use five control intervals in each step and the bounds on the control and tolerances on the constraints are kept as before. Optimum results are also obtained for a single control interval in each step, yielding the optimal constant reflux policy.

At the early stage of this work we applied the feasible trajectory problem in this multicut operation. The results are also presented here for comparison with those obtained using the minimum time formulation. Some of these results were also presented in Mujtaba and Macchietto (1988).

3.6.3.2 Results and discussions: Product amounts and compositions attained at the end of each operation step for the minimum time and feasible trajectory problem using the optimal reflux ratio profiles and constant reflux ratio are given in Table 3.8. Comparison of the results are given in Table 3.9. The actual control profiles for the minimum time fomulation are shown in Figures 3.4-3.5 for optimal variable reflux operation and in Figures 3.6-3.7 for optimal constant reflux operation. These also show the accumulated distillate and instant distillate composition curves for each case. Similar profiles are presented for feasible trajectory problem in Figures 3.8-11.

It is clear from Table 3.8 that the results obtained are in very good agreement with the objectives set for each individual problem. Table 3.9 also cleary shows the advantages of optimal control reflux policies by different types of optimal control problem over the conventional constant reflux operation. Table 3.9 shows that the time optimal control policy

Table 3.8 Objectives Attained by Variable and Constant Reflux Policies (Minimum Time and Feasible Trajectory Problems, Example 3)

		Objectives set	Objectives attained *			
		D^*, x^*_D	Minimum Time Problem		Feasible Trajec. Problem	
			case a	case b	case a	case b
Operation						
STEP 1	Propane accum	8.139 lbmol	8.117	8.117	8.141	8.139
	Propane conc.	98.1 %	98.1	98.1	98.1	98.1
	Batch time	(4.07 hr) @	2.82	4.01	3.16	4.05
STEP 2 and CUT 1	Propane prod.	11.760	11.737	11.737	11.737	11.731
	Conc.	85.0	85.0	85.0	85.0	85.0
	Batch time	(1.81)	1.37	1.56	1.40	1.47
STEP 3 and CUT 2	Butane Prod.	36.548	36.567	36.566	36.550	36.601
	Conc.	98.8	98.8	98.8	98.81	98.82
	Batch time	(18.27)	2.57	9.20	12.40	17.49
STEP 4	Pentane accum	8.619	8.620	8.620	8.620	8.630
	Conc.	94.0	94.1	94.1	94.1	94.1
	Batch Time	(4.31)	2.83	3.49	3.59	3.52
STEP 5 and CUT 3	Hexane Prod.	59.38 (B^*)	59.44	59.44	59.40	59.20
	Conc.	99.8 (x^*_B)	99.72	99.72	99.72	99.68
	Batch time	(1.78)	1.54	1.55	1.50	1.55

* case a represents optimal variable reflux policy
case b represents optimal constant reflux policy

@ Batch time within brackets are those used in the simulation (Table 2.6, chapter 2)

Table 3.9 Comparison of the Results of Minimum Time and Feasible Trajectory Problems (Example 3)

Basis : 100 lbmole of product

	Batch Time [hr]	Productivity [lbmole/hr]
Non optimal operation (simulation and Boston et al.)	30.20	3.31
Minimum time formulation		
Optimal constant reflux ratio	19.81	5.05
Optimal variable reflux ratio	11.13	8.99
Feasible Trajectory formulation		
Constant reflux ratio	28.04	3.55
Variable reflux ratio	22.05	4.53

(variable reflux) saves about 63% of the operation time compared to that of simulation results and about 50% of that obtained by the feasible trajectory formulation. Even the time optimal constant reflux policy saves about 33% of the operation time compared to the original simulation and about 28% compared to the feasible trajectory formulation. Even using the feasible trajectory formulation the operation time saved by the variable reflux ratio policy is about 27% compared to the simulation and about 22% compared to the constant reflux operation. Table 3.9 also compares the results in terms of productivity. Since in this problem we separate all the components in different fractions the productivity calculations are based on the total charge to the column. The Table clearly shows that the time optimal control policy (variable reflux) achieves a productivity twice that obtained by feasible trajectory formulation and almost three times over the original simulation case.

The reflux ratio profiles in Figure 3.4 are all along increasing with time as expected, except for a low reflux value at the end of operation step 2 and step 5. The reason could be as

Figure 3.4 Accumulated Distillate Composition (Minimum Time Problem, Case a, Example 3)

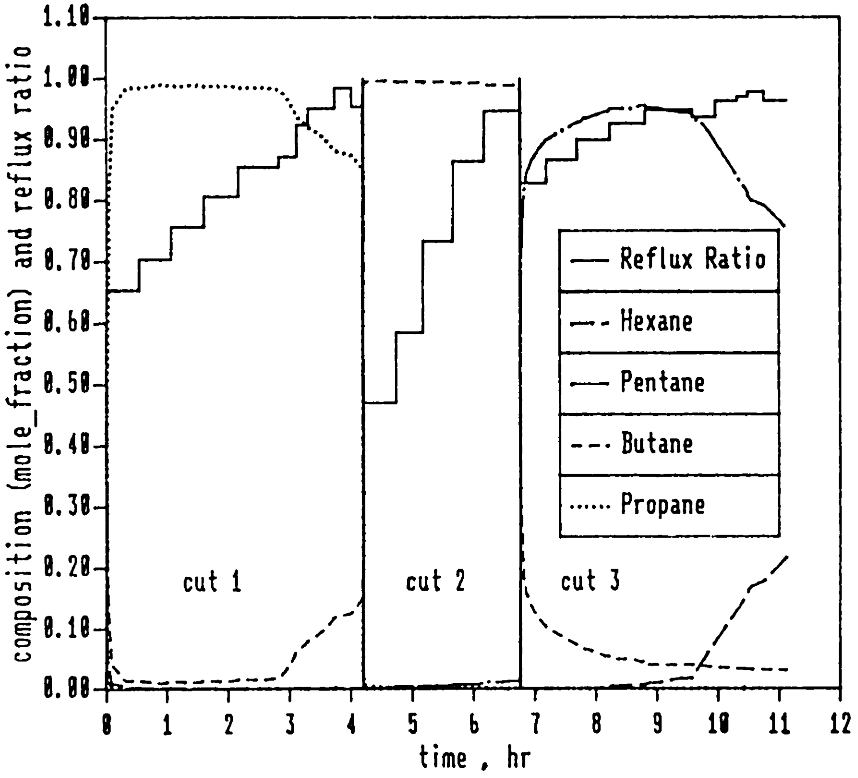


Figure 3.5 Instant Distillate Composition (Minimum Time Problem, Case a, Example 3)

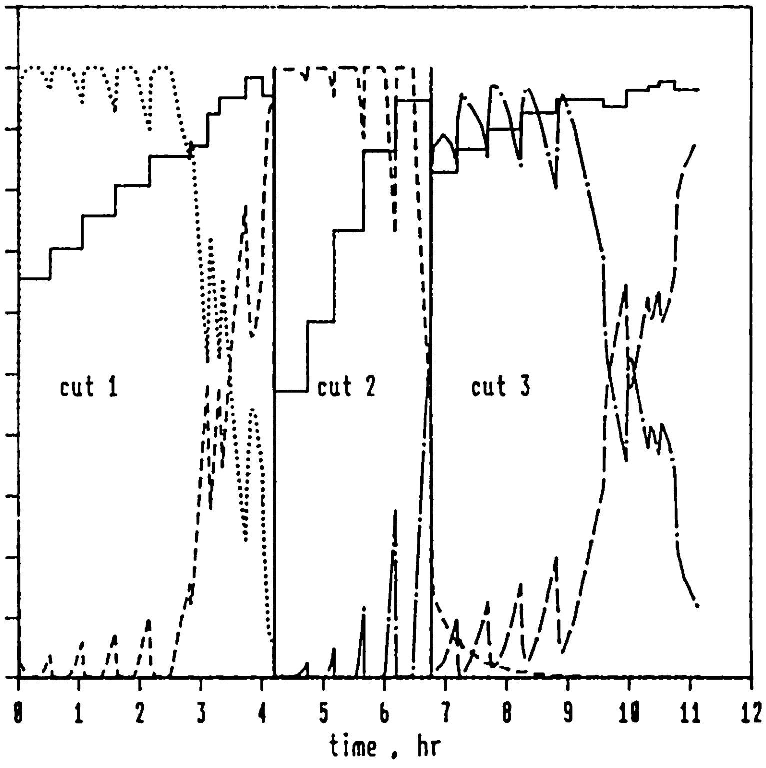


Figure 3.6 Accumulated Distillate Composition (Minimum Time Problem, Case b , Example 3)

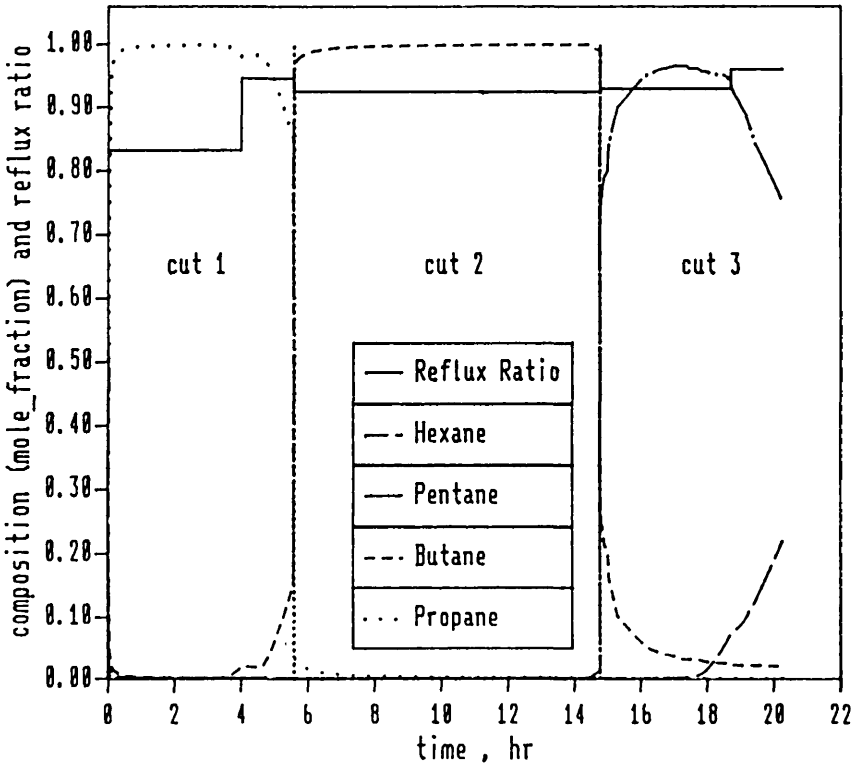


Figure 3.7 Instant Distillate Composition (Minimum Time Problem, Case b, Example 3)

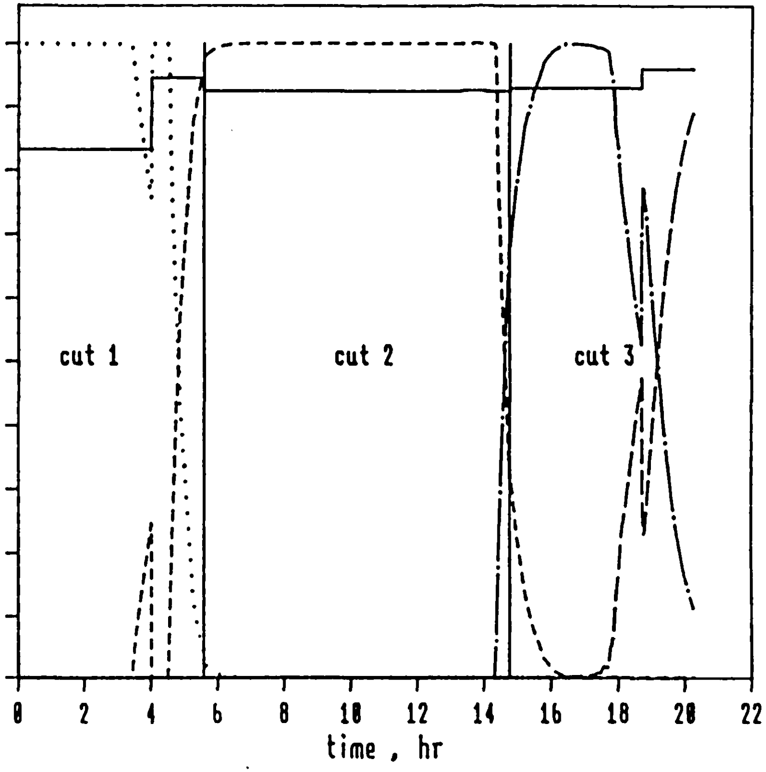


Figure 3.8 Accumulated Distillate Composition (Feasible Trajectory Problem, Case a, Example 3)

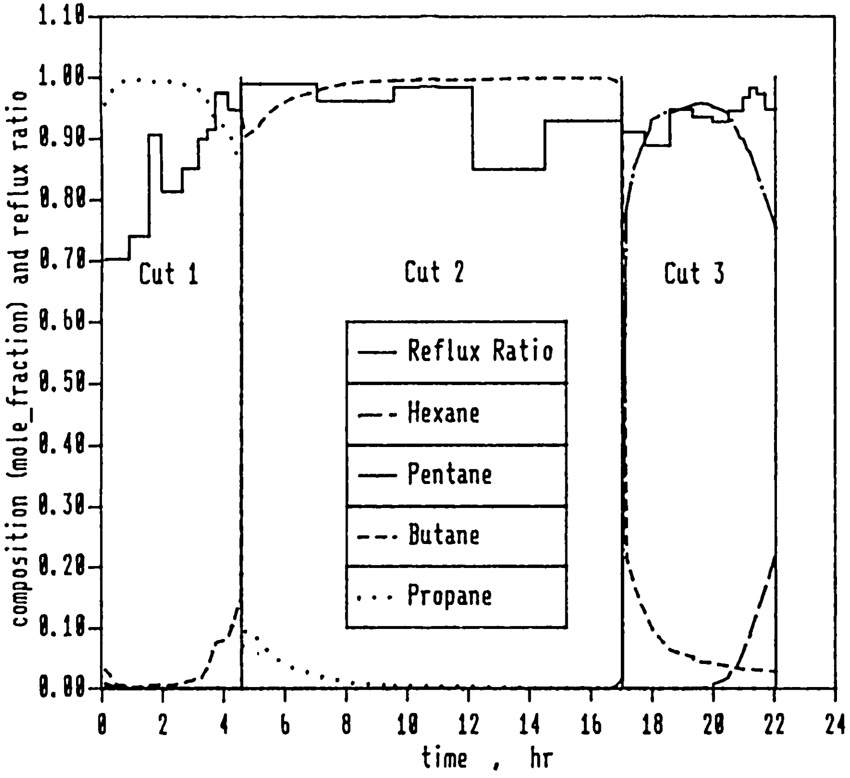


Figure 3.9 Instant Distillate Composition (Feasible Trajectory Problem, Case a, Example 3)

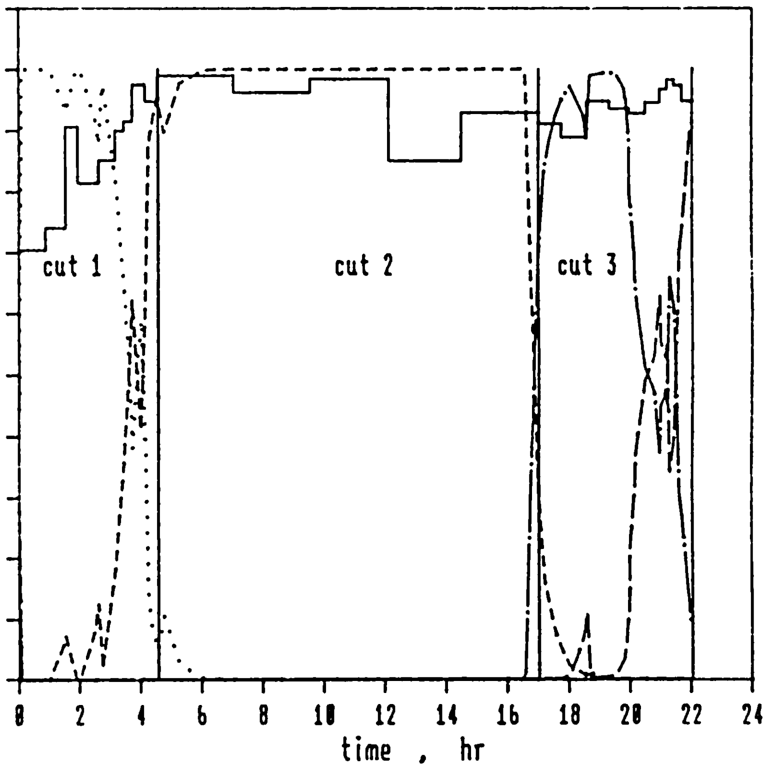


Figure 3.10 Accumulated Distillate Composition (Feasible Trajectory Problem, Case b, Example 3)

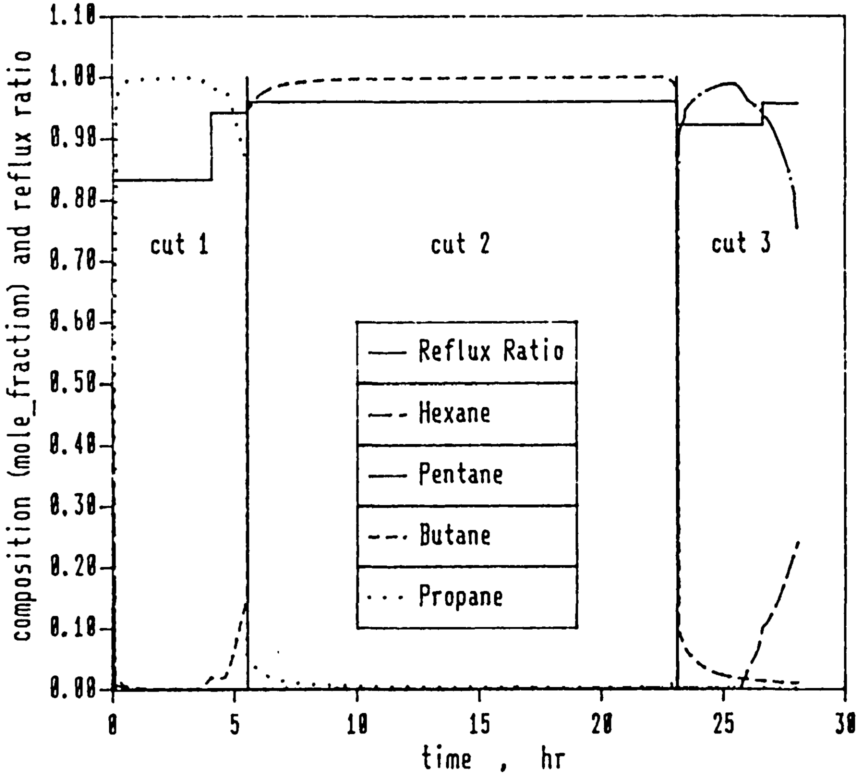
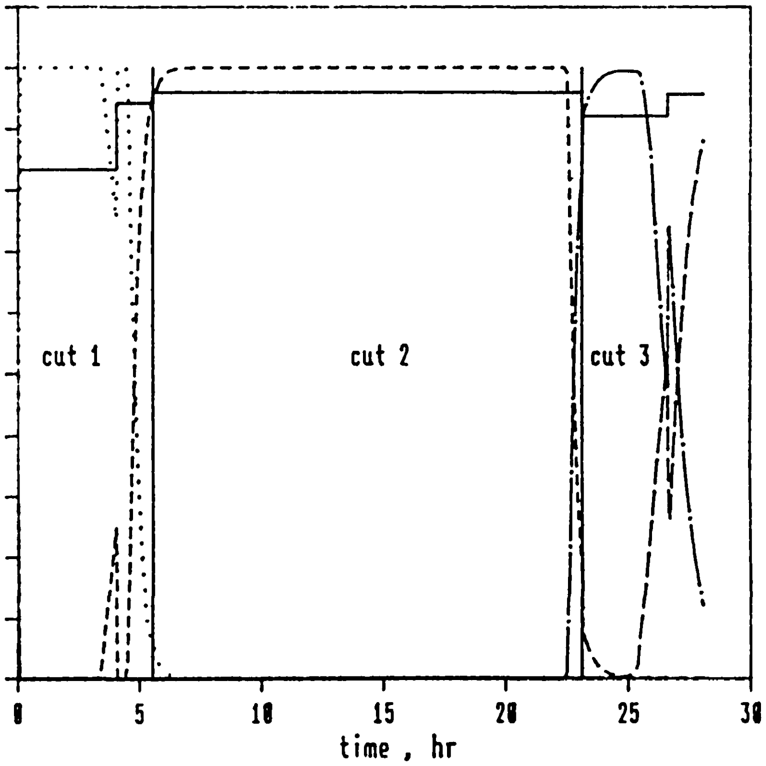


Figure 3.11 Instant Distillate Composition (Feasible Trajectory Problem, Case b, Example 3)



follows: Since propane is removed almost completely from the system after the second operation step, initial increasing reflux ratio for that operation helped to get the lighter component (propane) on the top of the column. During the last control interval of that operation step reflux takes a lower value to quickly collect the lighter component from the top to meet the desired specification and at the same time bringing the next component (butane) quickly to the top of the column which is to be collected in the next operation step. The same is true in operation step 5. However in this case the fall in reflux ratio in the last interval collects quickly the lighter component (pentane) from the column, thus leaving the final bottom product at its specified purity. Similar nature of the reflux profile was not observed in other operation steps, because there was enough material left of the component to be removed in a particular operation step in the column after the operation step is completed.

The instant distillate composition curves in Figure 3.5 clearly shows that a fall in concentration of the component which is being separated is followed by an automatic increase in reflux ratio to ensure product quality at the end of the process. The reflux ratio profiles obtained by the feasible trajectory formulation (Figure 3.8) do not follow any regular pattern mentioned above. The reason is probably due to the nature of this objective function as mentioned in section 3.6.1.2. Because the objective was set to minimize a quadratic function that closely obtain the required amount of product, the control policy merely chooses arbitrary combinations of reflux ratios and completion time which only satisfy the purity constraint at the end of the operation step.

3.7 Conclusions

Optimal variable reflux ratio policies were obtained for a single batch distillation for a variety of optimal control problems using various configurational, operational and physical property model options. These optimal policies were obtained for both single cut and multicut operation using a realistic column model. Application of the method to various operational problems clearly show the robustness and flexibility of the codes used.

Comparison of the results using optimal variable reflux ratio with conventional constant reflux ratio operation shows large improvements of column performance can be achieved. The results obtained by different types of optimal control formulation show the relative advantages of one over another. In the present work we only used 4 or 5 control intervals within each operation. Even further improvements may be expected using a larger number of control intervals.

Detailed solution statistics regarding the number of function and gradient evaluations, total time for physical property calculations and the total CPU time were presented in Table 3.6 for example 2. Example 2 is the biggest problem solved in this chapter and resulted in a 278 differential and algebraic equations. The initial reflux ratio profiles for the problem were those used in the simulation (shown in Table 3.6). These are widely different from the optimal ones. The optimal control problem took 17 function evaluations and 13 gradient evaluations and the total CPU time was about 50 minutes using an IBM 4341 computer. The translator time and the time spent in the optimisation routine for each call of the optimser are very small (only few seconds of CPU). However, both function and gradient evaluation numbers and the CPU time could be greatly reduced by more than 50% using good initial reflux ratio profiles. Although detailed solution statistics are not presented for each of the problems a typical value for the number of function evaluations varies from 10-25 and that of gradient evaluations from 7-20.

Chapter 4

OFF-CUT RECYCLE IN BINARY BATCH DISTILLATION

4.1 Introduction

In this chapter we consider the operation of a distillation column with production of main product cuts and intermediate cuts (off-cuts). A method is presented for calculating optimal off-cut recycle policies for binary batch distillation using the realistic column model described in chapter 2. Constraints and objective functions are formulated for a classical two level optimal control problem in which the optimal amount and composition of the recycle cut and the reflux policies are established. Application of the method to several examples shows the advantages of recycling. The method also evaluates when and how long an initial total reflux operation is needed. A measure of "the degree of difficulty" of separation is found to give a priori a qualitatively correct indication of whether recycling an off-cut is worthwhile or not. Later in this chapter a new method is proposed where an one level optimal control formulation is presented for calculating the optimal off-cut recycle policies. This approach has been found to be more robust and at least 5 times faster than the two level one.

In a mixture of n_c components if each component is to be recovered in a separate fraction, there are in principle n_c-1 main distillate cuts, one bottom residue fraction and n_c-1 intermediate cuts (Figure 4.1 and 4.2). Thus for binary mixtures there is usually one main-cut and one off-cut. This intermediate off-cut in binary separation problem can either be thrown away or can be recycled to the next batch and thus a periodic operation (see Figure 4.4) can be established (Liles, 1966; Mayur et al., 1970; Nad and Spiegel, 1987; Christensen and Jorgensen, 1987 and Luyben, 1988).

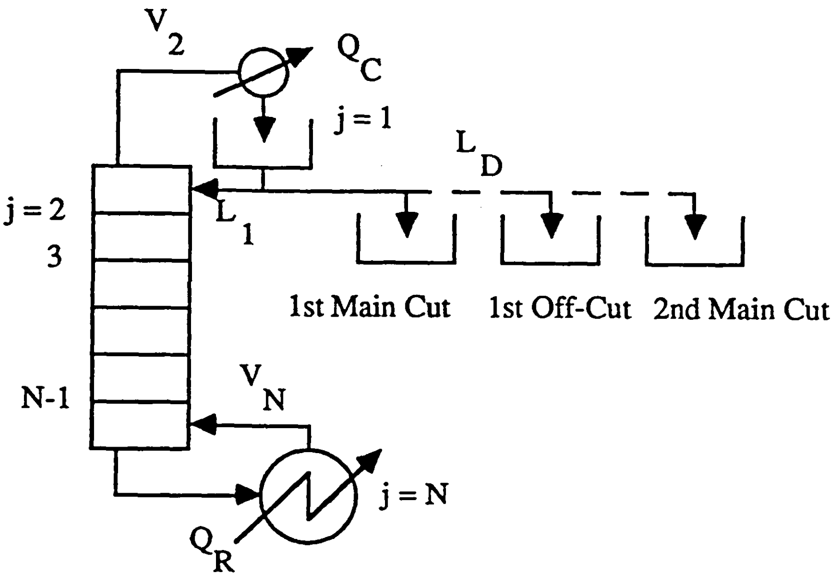


Figure 4.1 : A Typical Batch Column with Intermediate Cuts

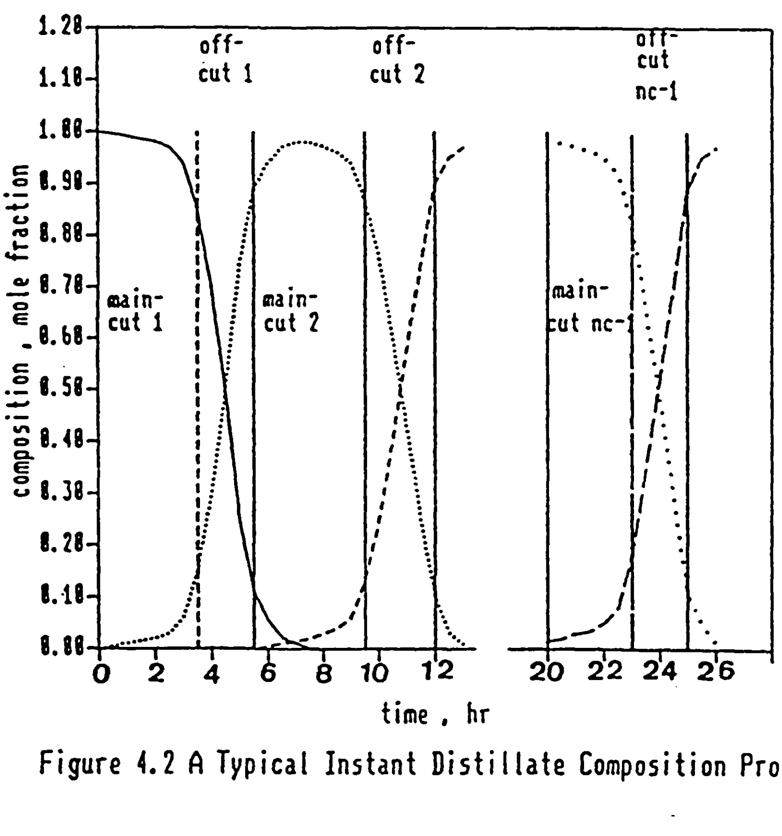


Figure 4.2 A Typical Instant Distillate Composition Profile

An optimal control of this type of periodic operation was first attempted and reported in the literature by Mayur et al. (1970), who considered the initial charge to the reboiler as a fresh feed stock mixed with the recycled off-specification material from the previous distillation. Each batch cycle is then operated in two distillation steps. During the first distillation a quantity of overhead distillate meeting the light product specification is collected. The residue is further distilled in the second distillation step until it meets the bottom product specification. The overhead during this second distillation step meets neither specifications and is recycled as part of the charge for the next batch. As the batch cycle is repeated a quasi-steady state mode of operation is attained which is characterised by the identical amount and composition of the recycle (from the previous batch) and the off-cut (from the current batch). Luyben (1988) indicates that the quasi-steady state mode is achieved after three or four such cycles.

Mayur et al. measured the benefits of recycling in terms of a potential reduction in batch time. However the benefits may be measured as follows:

1. Reduction in batch time: For a given fresh feed and a given separation, the column performance is measured in terms of minimum batch time required to achieve a desired separation (specified top and bottom product purity for binary mixture). Then an optimal amount and composition of recycle, subject to physical bounds (maximum reboiler capacity, maximum allowable purity of the off-cut) are obtained in an overall minimum time to produce the same separation (identical top and bottom products as in the case without recycle). The difference in the minimum times obtained with and without recycle shows the benefit of recycling.

2. Increase in productivity : For a fixed reboiler charge it is wished to obtain the optimal amount of fresh feed and the composition of the recycle (note: optimal amount of feed will automatically determine the optimal amount of recycle) to produce a given separation (specified top and bottom product purity for binary mixture) in a minimum batch time. This will give the productivity in terms of fresh feed processed per unit time. For the same given separation and no recycle, the productivity is obtained by processing a charge of fresh feed equal to the reboiler capacity. The difference in these productivities with and without recycle on a fixed

reboiler charge basis will show the benefit of recycling. However, the second measure has not yet been applied.

Mayur et al. (1970) formulated a two level optimal control problem to obtain optimal amount and composition of the off-cut recycle for the quasi-steady state operation which would minimize the overall distillation time for the whole cycle. For a particular choice of the amount of off-cut R_1 and its composition x_{R1} (Figure 4.4) they obtained a solution for the two distillation steps which minimizes the distillation time of the individual steps by selecting an optimal reflux policy. The optimum reflux policy is described by a function $r_1(t)$ during the first stage of distillation when a mixed charge (B_C, x_{BC}) is separated into a distillate (D_1,x^*_{D1}) and a residue (B_1,x_{B1}), followed by a function $r_2(t)$ during the second stage of distillation, when the residue is separated into an off-cut (R_1,x_{R1}) and a bottom product (B_2,x^*_{B2}). Both $r_1(t)$ and $r_2(t)$ are chosen to minimize the time for the respective stage of operation. However, these conditions are not sufficient to completely define the operation, because R_1 and x_{R1} can take many feasible values. Therefore the authors used a sequential simplex method to obtain the optimal values of R_1 and x_{R1} which minimize the overall distillation time. The authors showed for one example that the inclusion of a recycled off-cut reduced the batch time by 5% compared to the minimum time for a distillation without recycled off-cut.

The second published reference on the optimal treatment of off-cut recycles in batch distillation appeared only recently, when Christensen and Jorgensen (1987) investigated the possible economic impact of off-cut recycling on some difficult binary separations. For tray column they defined a measure of "the degree of difficulty" (q) of a given separation in a given column. With several examples for binary mixtures they showed that this measure could qualitatively predict the profitability of using an off-cut recycle. They also measured the benefits of recycling in terms of reduction in batch time. They found that the greater the measure of the difficulty of separation, the larger the benefits of using off-cut recycle.

It is quite clear from the previous works that recycling of off-specification material is particularly interesting if a given separation is to be performed in an existing column. The number of trays in the column may not be quite appropriate for the distillation task at hand, and running the column in a conventional manner (without recycling) may need a very long time. In such a case recycling offers a possibility of reducing the distillation time. In addition, recycling an off-cut may be used advantageously to reduce capital investment for a given batch separation by allowing a smaller column to be used than would otherwise be necessary with ordinary operation.

The problem of choosing whether and when to recycle each off-cut and the size of the cut is a difficult one. It has been tackled in the past either by repeated simulation (Luyben, 1988) or by a dynamic programming approach (Liles, 1966) or as an optimal control problem (Mayur et al., 1970; Christensen and Jorgensen, 1987). Most of the work to date on optimal control of batch distillation (including Mayur et al. and Christensen and Jorgensen) used Pontryagin's maximum principle applied to very simplified column models expressed as an ODE system as mentioned in chapter 1.

In this work therefore, we use a more realistic dynamic column model as presented in chapter 2. The measure of "the degree of difficulty" of the separation (Christensen and Jorgensen, 1987) is used to decide whether or not an off-cut is needed, and the optimal control algorithm of Morison (1984) is used to develop operational policies for reflux ratio profiles and amount and timing of off-cuts which minimize the distillation time. Several examples illustrate the method and the advantage of the optimal recycle policies for binary mixtures. Christensen and Jorgensen suggest that recycling the off-cut is beneficial for a value of q (degree of difficulty) ≥ 0.60. This criterion will also be assessed in this work.

It is quite obvious that a two level optimal control formulation is very expensive in terms of computation time. This is because for any particular choice of R_1 and x_{R1} a complete solution (sub-optimal) of the two distillation steps are required. The same is true for each gradient evaluation with respect to the decision variables (R_1 and x_{R1}). In the present work we proposed a faster one level optimal control formulation for the recycle problem which eliminates the requirement to calculate any sub-optimal or intermediate solution. In this formulation we minimize directly the total distillation time satisfying the separation requirements for the first distillation step as interior point constraints and for the second distillation step as final time constraints. It has been found that the proposed formulation is much more robust and at least 5 times faster than the classical two level formulation.

In all the case studies presented in this chapter it is assumed that the amount of fresh feed to be processed in the long production campaign is fixed for every batch cycle, but the reboiler is oversized to some extent. The optimal amount of recycle is obtained within this bound so that maximum benefit can be achieved out of a given column.

4.2 Two Level Optimal Control Formulation

Before going to a detailed formulation of the problem the following discussion is worthwhile:

Consider a problem with no recycle as shown in Figure 4.3 (this figure is similar to that presented in Figure 3.1 but is reproduced here with different nomenclature for convenience of discussion), where an initial charge to the column is B_0 with composition x_{B0}. The charge is separated into two fractions, the overhead as (D_1, x_{D1}) and the bottom as (B_2, x_{B2}).

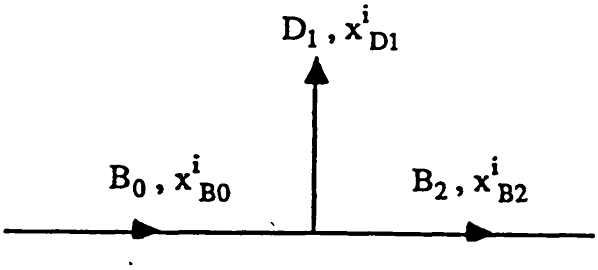


Figure 4.3 A Typical Single Batch Operation

The overall mass balance is therefore :

$$B_0 = D_1 + B_2 \tag{4.1}$$

$$B_0 x^1_{B0} = D_1 x^1_{D1} + B_2 x^1_{B2} \tag{4.2}$$

$$\sum x^i_{D1} = 1 \tag{4.3}$$

$$\Sigma x^i_{B2} = 1 \tag{4.4}$$

where i = 1,2 indicates the particular component.

For a given initital charge (B_0, x_{B0}) the unknown variables in the above system of equations are : D_1, x^1_{D1}, x^2_{D1}, B_2, x^1_{B2}, x^2_{B2}.

Therefore we are left with a degree of freedom (DF) = 2. One of the choices of design variables could therefore be (x^1_{D1} and x^1_{B2}). Since we deal with only binaries in this chapter we drop out the superscripts to indicate the component number. From now on (x^1_{D1} and x^1_{B2}) will be expressed as (x^*_{D1} and x^*_{B2}) meaning these variables are specified. With these specification we can now easily formulate a time optimal control problem for the no recycle case mentioned above. The problem can be stated as :

Starting with a given fresh feed of (B_0, x_{B0}) select a reflux ratio profile r(t) to achieve an overhead product (D_1, x^*_{D1}) in a minimum time (t_F) leaving the final bottom product (B_2,x^*_{B2}). This can be formulated as:

$$\min_{r(t)} \quad J = t_F \tag{4.5}$$

s.t. constraints

$$x_{D1}(t_F) = x^*_{D1}$$

$$x_{B2}(t_F) = x^*_{B2}$$

and all distillation model equations.

Now we can remove the 2nd constraint from the above formulation and substitute it by the relation :

$$D_1(t_F) = \underline{D}_1,$$

where $\underline{D}_1$ is the solution of equations (4.1-4.4) for a given charge B_0, x_{B0} and specifications x^*_{D1} and x^*_{B2}.

Since the two specifications above are equivalent we end up with the same optimal solution.

The necessity of the above discussion will now be realised. Figure 4.4 shows a quasi steady state mode of operation, with off cut recycle. A fresh charge B_0, of composition x_{B0} is mixed with the off cut R_1, x_{R1} from the previous batch to produce a mixed charge to the reboiler (B_C, x_{BC}). The main cut $\underline{D}_1$, x^*_{D1} is produced over the time t_1 (first period), leaving a residue B_1, x_{B1}. At this time the distillate is simply diverted to a second receiver, and further distillation over the second period for time t_2 produces the off cut and the final bottom product $\underline{B}_2$, x^*_{B2} ($\underline{B}_2$ is the solution of equations (4.1-4.4) as mentioned before)

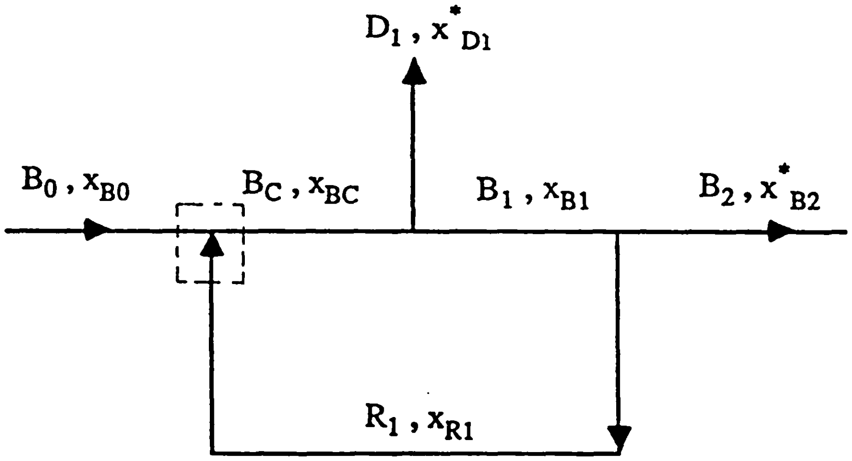


Figure 4.4 Quasi Steady-State Recycle Operation For Binary Batch Distillation

The following optimisation problem is considered. Given a batch charge B_0, x_{B0}, a desired amount of distillate $\underline{D}_1$ of specified purity x^*_{D1} and a final desired bottom product $\underline{B}_2$ of specified purity x^*_{B2} determine the amount and composition of the off-cut (R^*_1, x^*_{R1}) and the reflux rate policy r(t) which minimize the overall distillation time. Writing a mass balance equation around the mixer of Figure 4.4 we can easily show that there are again two degrees of freedom left. So one of the possible choices could be to specify R_1 and x_{R1} or to optimize these variables as mentioned above. A two level optimal control problem is therefore formulated as :

$$\begin{aligned} \text{P0} \qquad & \underset{R^*_1 ,\, x^*_{R1}}{\text{Min}} \quad J = t_1 + t_2 \\ \text{subject to} \qquad & a_R < R^*_1 < b_R \\ & a_{x_R} < x^*_{R1} < b_{x_R} \end{aligned} \tag{4.6}$$

where t_1 is obtained from

$$\begin{aligned} \text{P1} \qquad & \underset{r(t)}{\text{Min}} \quad t_1 \\ \text{subject to} \qquad & (D_1 - \underline{D}_1)^2 < \varepsilon_1 \\ \text{and} \qquad & x^*_{D1} - \varepsilon_2 < x_{D1} < x^*_{D1} + \varepsilon_2 \end{aligned} \tag{4.7}$$

and t_2 is obtained from

$$\begin{aligned} \text{P2} \qquad & \underset{r(t)}{\text{Min}} \quad t_2 \\ \text{subject to} \qquad & (R_1 - R^*_1)^2 < \varepsilon_1 \\ \text{and} \qquad & x^*_{R1} - \varepsilon_2 < x_{R1} < x^*_{R1} + \varepsilon_2 \end{aligned} \tag{4.8}$$

where ε_1 and ε_2 are small positive numbers and a_R, b_R, a_{x_R} and b_{x_R} are bounds on the amount of recycle cut and on its composition. In addition, all differential and algebraic model equations must be added as equality constraints to problem P1 and P2, with suitable boundary conditions. These are mentioned in detail in chapter 3.

Thus the multiperiod optimisation problem is formulated as a sequence of two independent optimal control problems (P1 and P2), with the total time minimized by a proper choice of the off cut variables in an outer problem (P0) and the quasi steady state conditions appearing as a constraint in P2. For each iteration of P0 a complete solution of P1 and P2 is required. Thus, even for an intermediate sub-optimal off cut recycle, a feasible quasi-steady state solution is calculated. The gradients of the objective function with respect to each decision variable (R_1 or x_{R1}) in problem P0 are evaluated by a finite difference scheme which again requires a complete solution of problem P1 and P2 for each gradient evaluation. This actually makes the classical formulation very unattractive.

Reflux ratio r(t) is the control variable which governs the operation during each period. This is discretised into a small number of intervals (typically 3 to 10) with a constant value during each interval. The optimal values in each interval and the switching times for each of the inner problems are again obtained using the optimal control algorithm of Morison (1984) with optimisation based on a successive quadratic programming (SQP) as described in chapter 3. The outer problem P0 is also solved using an SQP algorithm.

4.3 "The degree of difficulty", **q**,of a Given Separation

Christensen and Jorgensen (1987) proposed a measure **q** of "the degree of difficulty"of separation for batch distillation of binary mixtures, defined as:

$$q = \int_{x_{B2}}^{x_{B0}} \frac{(N_{min} + 1)\, dx_B}{(x_{B0} - x_{B2})(N_T + 1)} \qquad (4.9)$$

where x_{B0} and x_{B2} are defined in Figure 4.3 and x_B is an intermediate value of the reboiler composition between x_{B0} and x_{B2}.

This measure was chosen based upon the ratio of the necessary minimum number of trays N_{min} (averaged over the reboiler composition) in the column at total reflux and constant product composition (x_{D1}) to the actual number of trays in the given column N_T. Christensen and Jorgensen assumed a constant relative volatility α and evaluated N_{min} using the Fenske equation:

$$N_{min} + 1 = \frac{\ln\left(\frac{x_{D1}}{1-x_{D1}} . \frac{1-x_B}{x_B}\right)}{\ln \alpha} \qquad (4.10)$$

In this work eqn.(4.9-4.10) for **q** are maintained but α values are calculated at the top and bottom of the column from rigorous equilibrium data and a different geometric average α is used in eqn. (4.10) as x_B changes. The value of **q** increases with decreasing relative volatilities, increasing high purity demands and decreasing number of stages in excess of the minimum. It ranges from 0 (infinite number of stages) to 1 (minimum number of stages).

4.4 Example Problems Using the Two Level Formulation

4.4.1 Problem Description

Two typical binary mixtures and a variety of separation specifications are used to demonstrate how optimal recycle policies can be obtained and to assess the validity of "the degree of difficulty" measure. The mixtures are 1) Benzene-Toluene, 2) Butane-Pentane. For simplicity ideal equilibrium and Antoine's vapor pressure equations are used here. The light component is always the first one.

Column configuration, initial charge to the column and separation requirements for several cases are presented in Table 4.1. The minimum time required for a given separation (D_1, x^*_{D1}) is reported in Table 4.1 as t_r for the case with off cut recycle (solution of P0) and as t_{nr} for the case with no recycle (solution of P1 with R_f=0). In both cases the reflux ratio was discretised into 3 control intervals during the main cut period and a single interval during off cut production.The bounds on the reflux ratio $0.2 \leq r \leq 1.0$. and the tolerances $\varepsilon_1 = 0.0001$ and $\varepsilon_2 = 0.001$ are used for all test problems. In all the cases 4% of the fresh feed was used as total column holdup, half of it in the condenser, the rest equally distributed on the plates.

Liquid compositions of trays, condenser holdup tank and accumulator (differential variables) at time t=0 are set equal to the fresh charge composition (x_{B0}) to the reboiler. It is also possible to set these values to mixed charge composition (x_{BC}). Reboiler holdup and compositions are initialized to the mixed charge (B_C,x_{BC}) at each iteration of P0. The differential and algebraic equation system is solved at time t=0 to initialize all other variables. The first product (D_1,x_{D1}) (see Figure 4.4) is drawn off starting from t = 0. For the second distillation no reinitialization is required. The distillate is simply diverted to a different product accumulator and integration is continued.

4.4.2 Results and Discussion

Results are summarised in Table 4.1 and Figures 4.5-4.9 which show the optimal reflux ratio profiles and the corresponding accumulated and instant distillate composition curves. The optimal reflux ratio profiles presented in Figures 4.5-4.9 are different from case to case.

With difficult separations (Figure 4.6 and 4.7 , no recycle cases) a high value of the reflux ratio is obtained initially to obtain a high distillate concentration of the desired product. The reflux is then lowered, and increased again at the end. A period of total reflux operation is automatically determined, if found to be optimal, which is the case for difficult separations (Figure 4.7, no recycle case). With an easy separation (Figure 4.5, 4.6, 4.8, recycle case) the reflux ratio profile is usually increasing all along as expected. This is because the effect of recycling the off-cut is to make the separation for the main cut easier, for a given column configuration. But for more difficult separations (Figure 4.7 and 4.9) even the recycle cases take an initial high reflux although this is comparatively lower than for cases without recycle (Figure 4.7 and 4.9). The optimal reflux ratio during off cut production is fairly low in all cases and in agreement with practical experience (Rose, 1985).

The benefits of recycling are also quite clear. In some cases the recycle of off-specification materials results in operation time savings of over 70% of the optimal time required without recycle. Also the size of the optimal off-cut increases (and so does its composition) as the separation becomes more difficult (higher q). Table 4.1 shows that recycling is beneficial for **q** values greater than approximately 0.6. However, it is noticed that the extent of the benefit for similar values of **q** varies from case to case. Comparison of case 4 and case 10 which have the same and quite high **q** shows that the time savings are extremely different. Although high **q** values suggest that recycling should be used the extent of time savings depends on various other factors (type of mixture, amount and purity required and vapor boilup rate etc.)

Table 4.1 Input Data and Results (ideal cases)

Case	Binary	N_T	B_0	x_{B0}	$\underline{D}_1$	x^*_{D1}	V_c	q	t_{nr}	t_r	% Time	Optimal	
	Mixture		kmol		kmol		kmol/hr		hr	hr	Saved	R_1(kmol)	x_{R1}
1	1	8	5.0	0.60	3.0	0.960	3.0	0.483	3.90	----	----	----	----
2	1	4	5.0	0.60	3.0	0.900	3.0	0.597	3.10	2.94	5.03	0.50	0.607
3	1	3	5.0	0.60	3.0	0.900	3.0	0.746	4.27	3.46	18.97	1.07	0.654
4	1	3	5.0	0.60	3.0	0.912	3.0	0.799	5.57	3.91	29.90	1.22	0.676
5	1	3	5.0	0.60	3.0	0.920	3.0	0.836	7.22	4.25	41.10	1.50	0.679
6	2	7	5.0	0.60	3.0	0.970	3.0	0.400	2.74	----	----	----	---
7	2	3	5.0	0.60	3.0	0.900	3.0	0.500	1.87	----	----	----	---
8	2	3	5.0	0.60	3.0	0.940	3.0	0.637	2.80	2.51	10.29	0.50	0.571
9	2	3	5.0	0.60	3.0	0.955	3.0	0.706	3.88	2.95	23.88	0.99	0.649
10	2	3	5.0	0.60	3.0	0.970	3.0	0.799	13.06	3.62	72.26	1.34	0.665

Key: V_C is the condenser vapor load.

% Time saved is based on t_{nr}.

Figure 4.5a Accumulated Distillate Composition

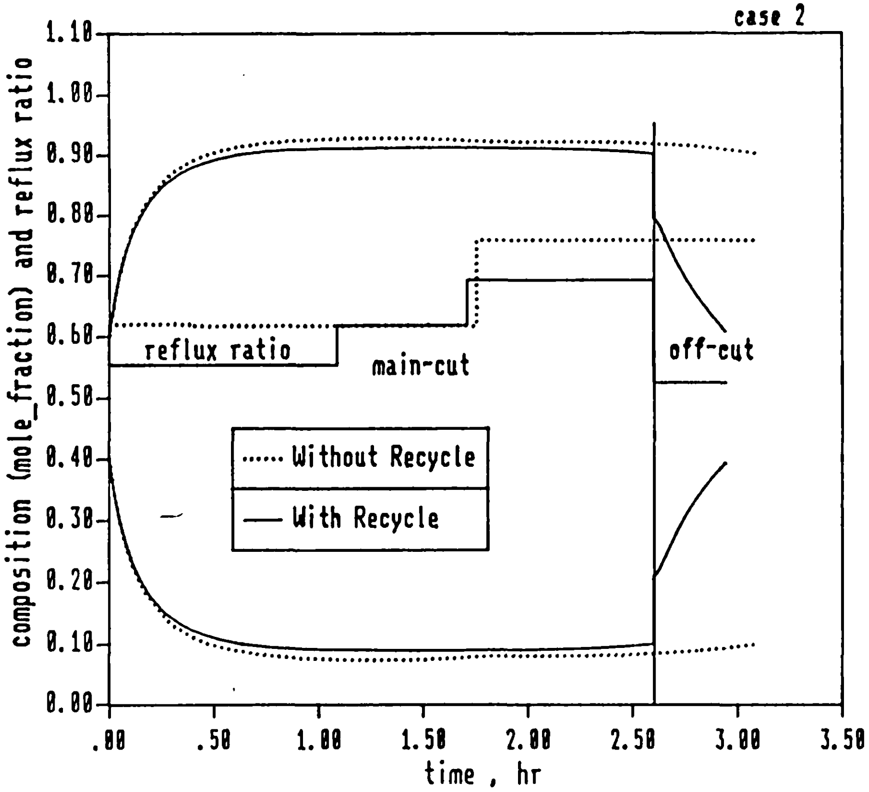


Figure 4.5b Instant Distillate Composition

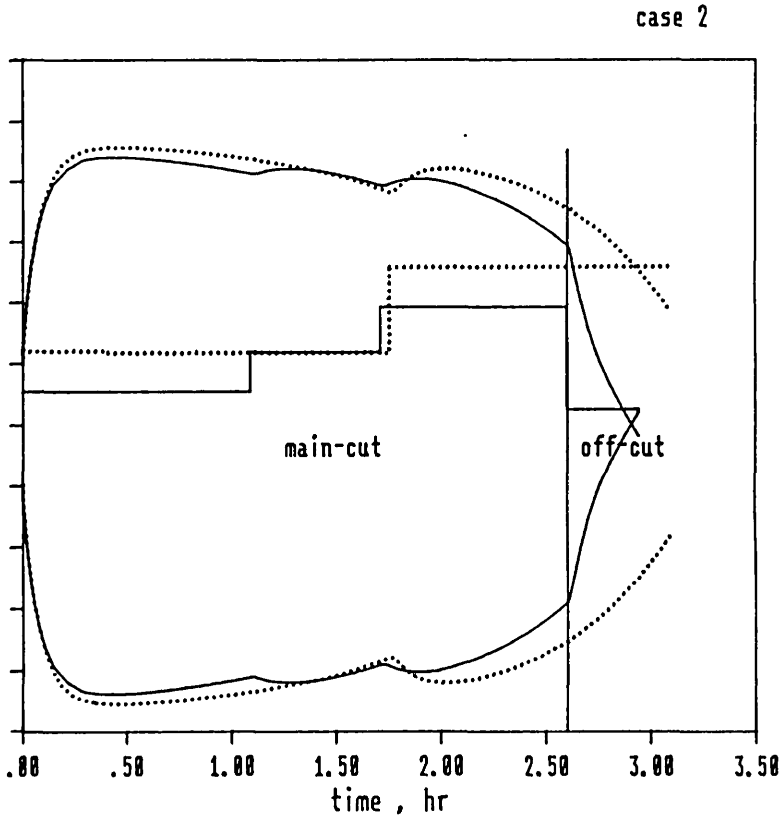


Figure 4.6a. Accumulated Distillate Composition

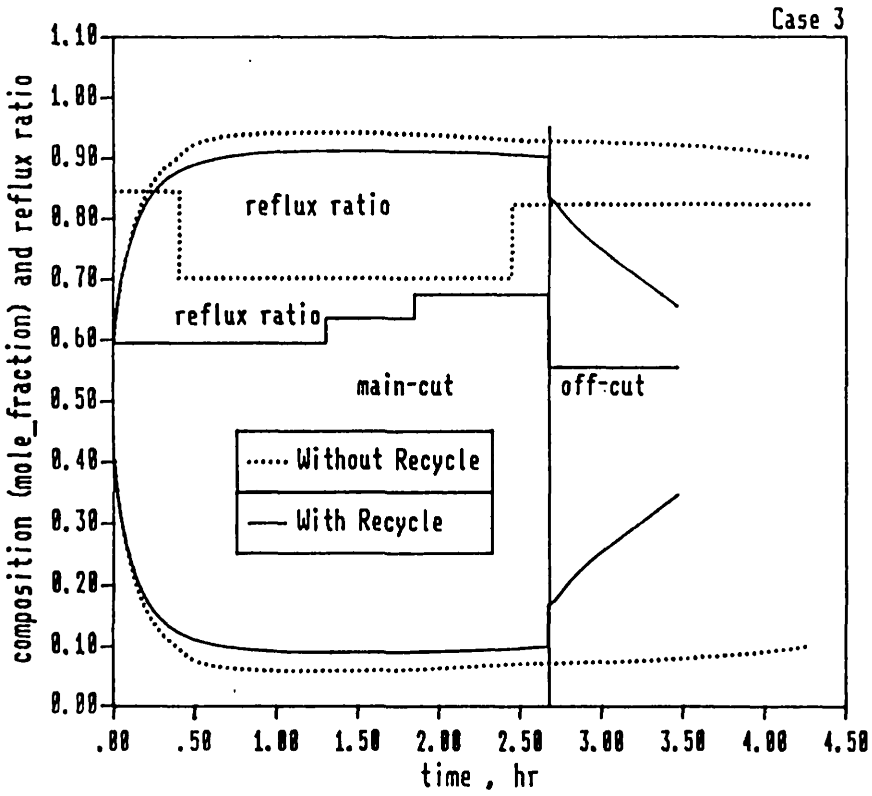


Figure 4.6b Instant Distillate Composition

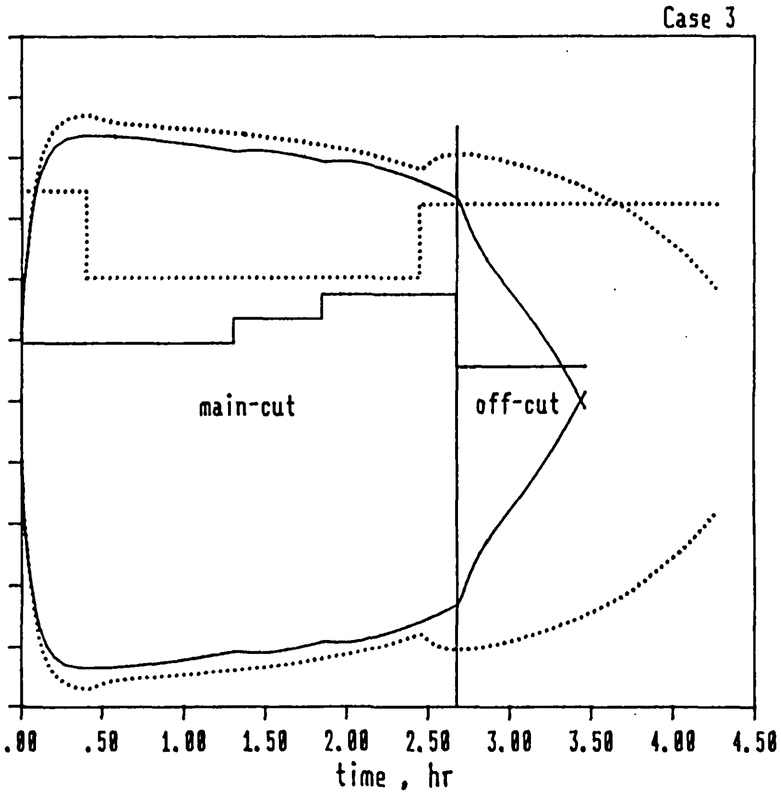


Figure 4.7a Accumulated Distillate Composition

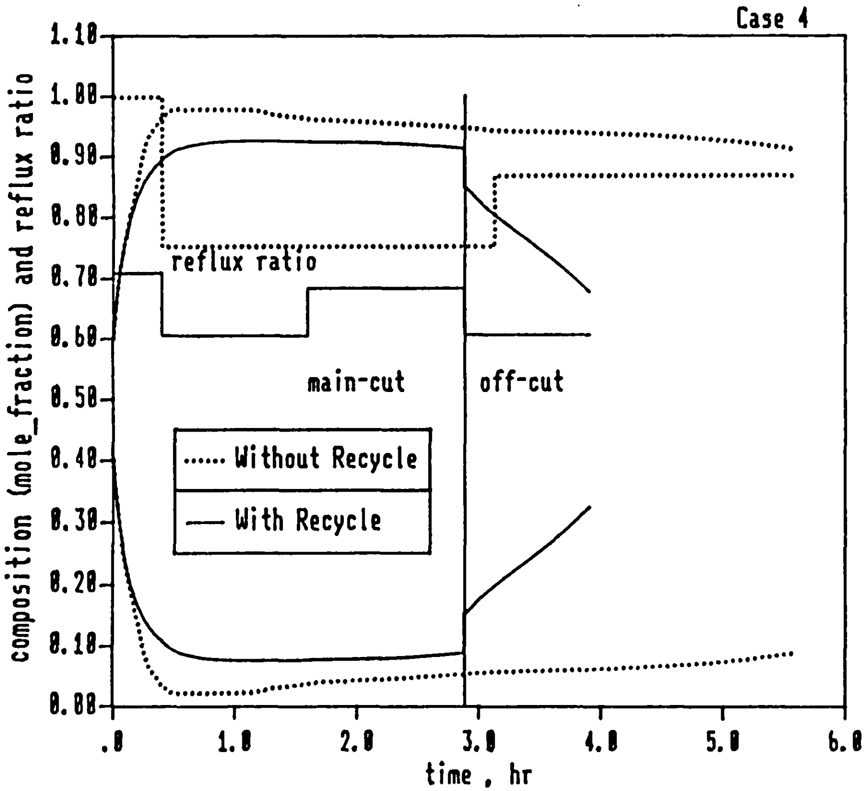


Figure 4.7b Instant Distillate Composition

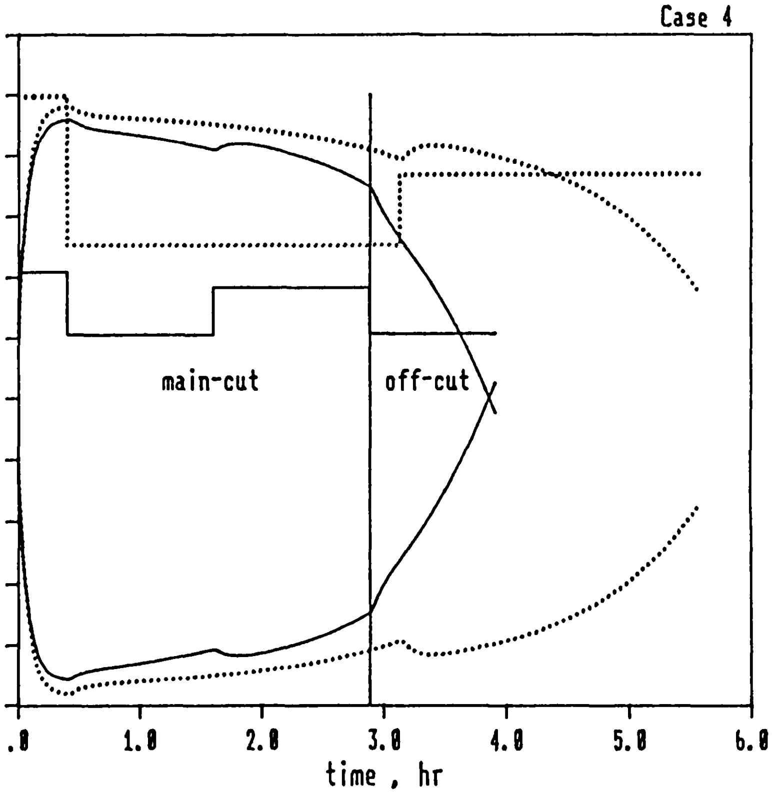


Figure 4.8a Accumulated Distillate Composition

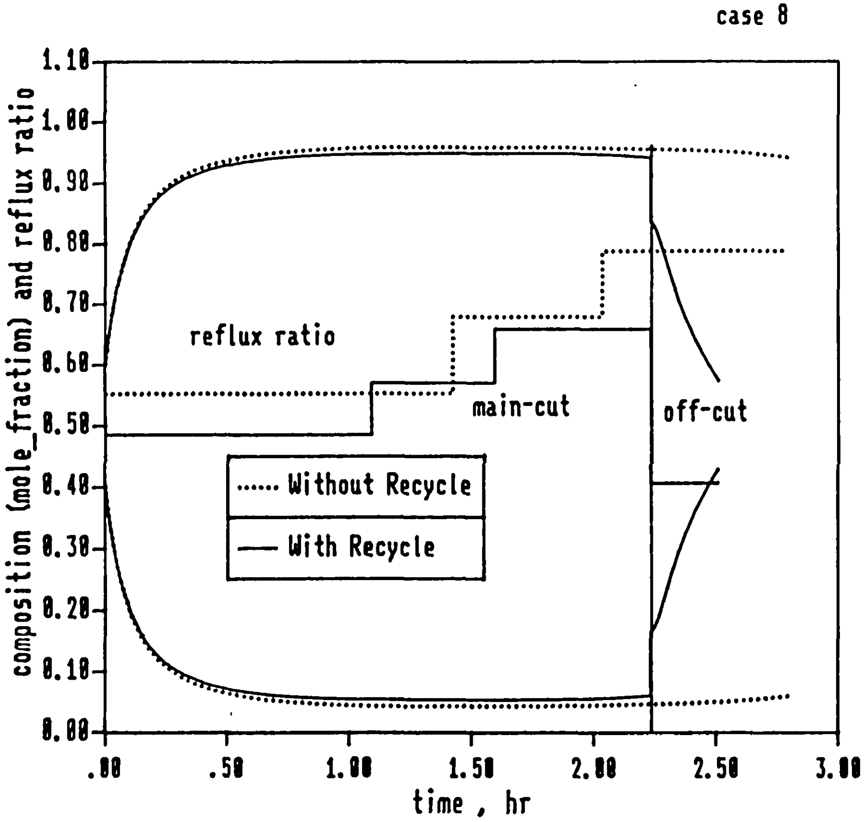


Figure 4.8b Instant Distillate Composition

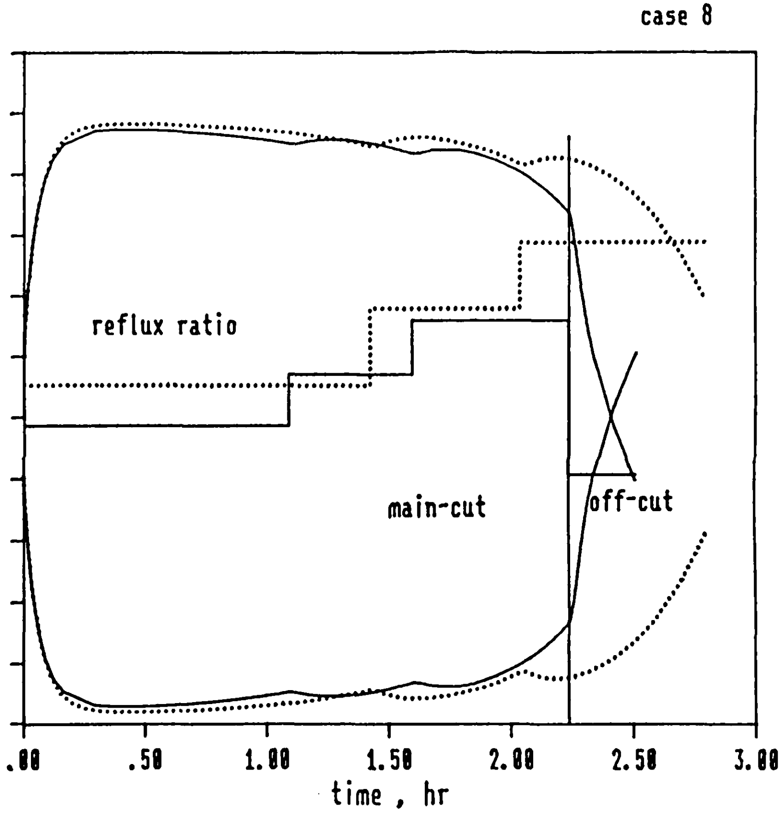


Figure 4.9a Accumulated Distillate Composition

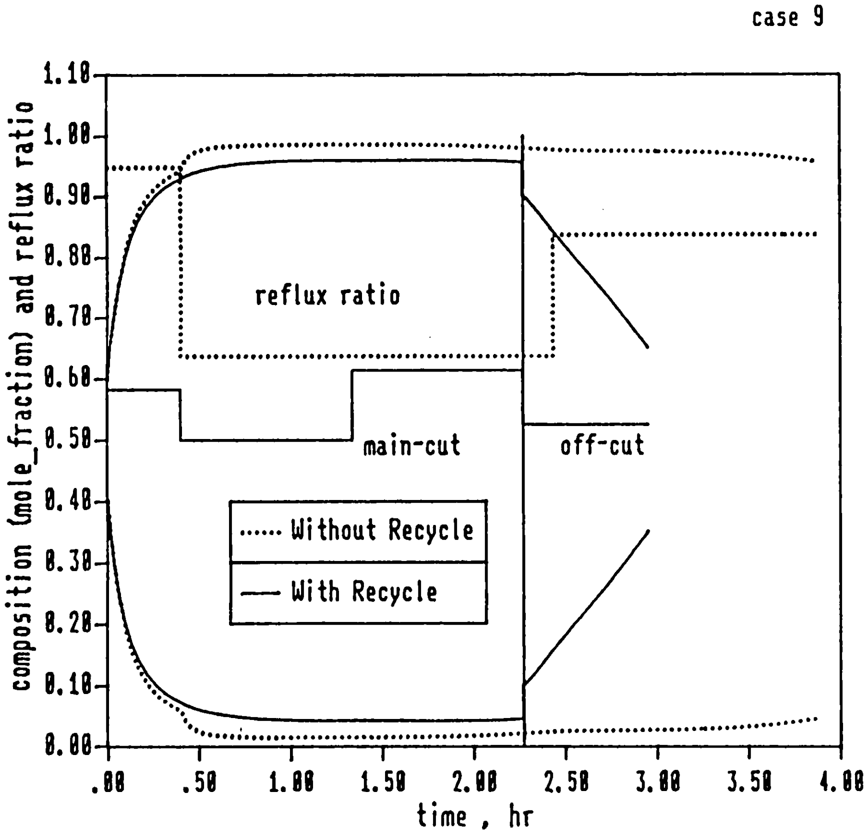
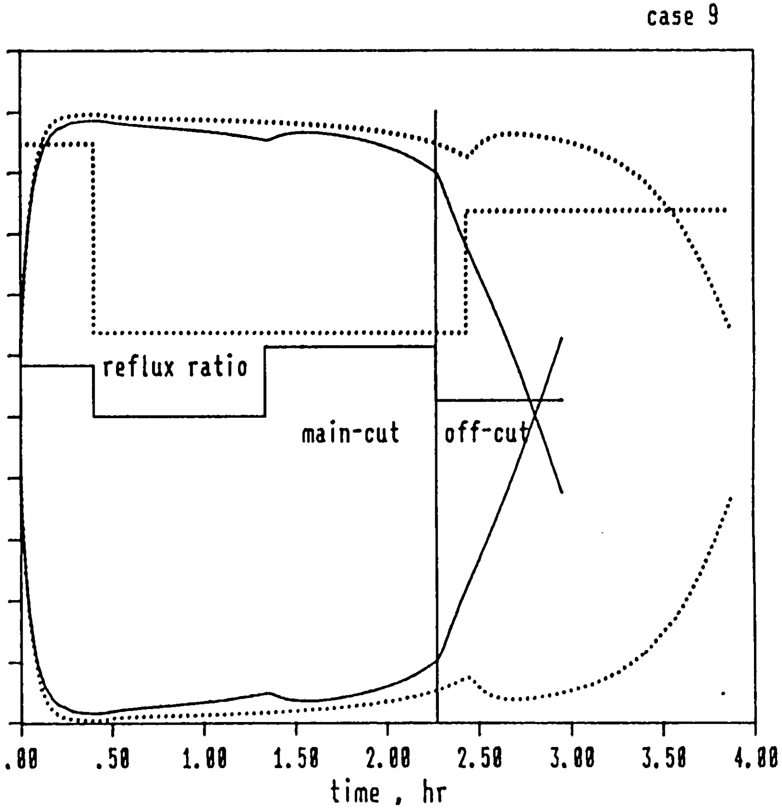


Figure 4.9b Instant Distillate Composition



4.5 One Level Optimal Control Formulation

Refering to Figure 4.4 and given a batch charge B_0, x_{B0} ,a desired amount of distillate $\underline{D}_1$ of specified purity x^*_{D1} and final bottom product $\underline{B}_2$ of specified purity x^*_{B2} we wish to determine the amount and composition of the off-cut (R^*_1, x^*_{R1}) and the reflux rate policy r(t) which minimizes the overall distillation time. In this formulation instead of optimising R_1, x_{R1} we optimise the mixed charge to the reboiler (B_C, x_{BC}) and at the end of the solution we evaluate the optimal R^*_1, x^*_{R1} from the overall balance around the mixer in Figure 4.4. The optimal control problem is formulated as :

P4 $$\min_{B_C,\, x_{BC},\, r(t)} \quad J = t_1 + t_2 = t_F$$

subject to interior point constraints

$$\begin{aligned} D_1(t_1) &= \underline{D}_1 \\ x_{D1}(t_1) &= x^*_{D1} \end{aligned} \qquad (4.11)$$

and end point constraints

$$\begin{aligned} B_2(t_F) &= \underline{B}_2 \\ x_{B2}(t_F) &= x^*_{B2} \end{aligned}$$

where t_1 is defined by one of the switching times and t_F is the final time.

In addition, all differential and algebraic model equations act as equality constraints in problem P4 with suitable boundary conditions as mentioned in the two formulation.

The formulation presented in P4 automatically establishes the quasi-steady state operation of Figure 4.4 where the recycle from the previous batch is identical in amount and composition to those of current batch.

4.6. Comparison of the Two Level and the One Level Formulations

One example problem is presented here to compare the results (optimal policy, computation time) using the two level and the one level optimal control formulations.

4.6.1 Problem Description

Benzene and Toluene is considered as the binary mixture for the purpose. The column consists of 3 internal plates, reboiler and a total condenser. The reboiler is charged with a fresh feed of 5 kmol with Benzene mole fraction 0.6. The total column holdup is four percent of the charge. Half the holdup is in the condenser and the rest is distributed over the plates. The vapor load to the condenser is 3 kmol/hr. The required purities are $x^*_{D1} = 0.90$ and $x^*_{B2} = 0.15$. The solution of equations (4.1-4.4) therefore gives $\underline{D}_1 = 3.0$ kmol and $\underline{B}_2 = 2$ kmol. This problem is same as case 3 in Table 4.1. We use three control intervals to achieve ($\underline{D}_1$, x^*_{D1}) and one control interval to achieve ($\underline{B}_2$, x^*_{B2}).

4.6.2 Results and Discussions

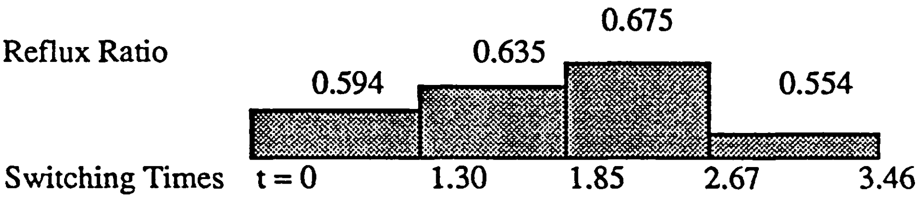
The reflux ratio policies together with switching times and optimal amount and composition of the recycle obtained by two methods are presented in Table 4.2 and the solution statistics are given in Table 4.3.

The results presented in Table 4.2 are in good agreement. The small differences between the results might be due to the different accuracy set for the optimisation (see Table 4.3). Since the gradients in the two level formulation were solved by finite difference the inner loop problems (P1 and P2) had to be solved very tightly (accuracy for the optimiser = 1.0E-4). Whereas, the outer loop problem (P0) of the two level formulation and the one level problem (P4) were solved using the optimiser accuracy = 1.0E-2.

The interesting part of the results by two methods is the difference in computation time. As presented in Table 4.3 , the one level control was about 5 times faster than that of the classical one. In fact the total time for the one level solution was only 1.5 times the time required to evaluate just one function (or gradient) values for the two level formulation.

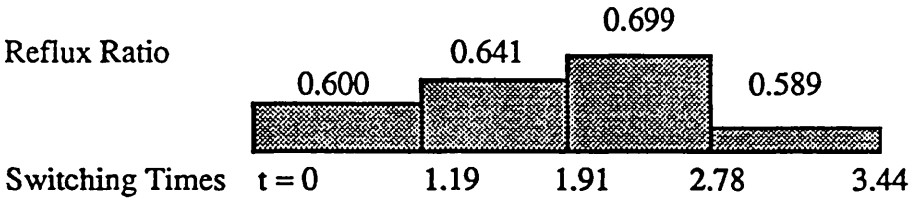
Table 4.2 Comparison of the Results Obtained by Two Methods

Two Level Fomulation :



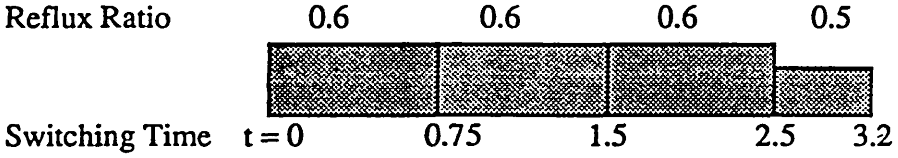
Optimum : R_1 = 1.07 kmol x_{R1} = 0.653

One Level Fomulation :



Optimum : R_1 = 0.986 kmol x_{R1} = 0.630

Initial Reflux Profile



Initial choice of R_1 = 0.8 and x_{R1} = 0.6

Table 4.3 Solution Statistics Using Two Methods

Two Level Formulation :

No. of Function Evaluations (outer loop) = 5
No. of Gradient Evaluations (outer loop) = 3

Optimiser Accuracy

Inner Loop = 1.0E-4
Outer Loop = 1.0E-2

Time for each Function or Gradient Evaluation (approx) = 10 min (CPU)
Total Time Spent (including translator and optimiser time) = 81 min

% of total time spent in Physical Properties = 30

One Level Formulation :

No. of Function Evaluations = 15
No. of Gradient Evaluations = 12

Optimiser Accuracy = 1.0E-2

Time for each Function Evaluation = 0.4 min
Time for Each Gradient Evaluation = 0.7 min

Total Time Spent = 15.5 min

% of total time spent in Physical Properties = 30

4.7 More Examples using the One Level Optimal Control Formulation

4.7.1 Problem Description

A typical nonideal binary mixture and a variety of separation specifications are used to demonstrate how optimal recycle policies can be obtained and to assess the validity of the "degree of difficulty" measure. The mixture is Acetone - Ethanol. The SRK equation of state is used to evaluate vapor liquid equilibria. More accurate physical property models (UNIQUAC, Wilson) can also be used for such mixture. However, the use of SRK in this problem will not hamper seriously the conclusions drawn from this example. Column configuration, initial charge to the column and separation requirements for several cases are presented in Table 4.4. For all cases the initial amount of the fresh feed is 5 kmol with acetone concentration of 0.55 (mole fraction). In all the cases 4% of the fresh feed was used as total column holdup, half of it in the condenser, the rest equally distributed on the plates. The amount of distillate required (D_1) and the vapor load to the condenser are 2.5 kmol and 2.5 kmol/hr respectively in all cases. As before the minimum time required for a given separation (D_1, x^*_{D1}) is reported in Table 4.4 as t_r for the case with off cut recycle and as t_{nr} for the case with no recycle. In both cases the reflux ratio was discretised into 3 control intervals during the main cut period and a single interval during off cut production.

4.7.2 Results and Discussions

Results are summarised in Table 4.4. Table 4.5 shows the optimal reflux ratio profiles for cases with recycle and without recycle. Typical solution statistics for the cases studied are given in Table 4.6.

Table 4.4 Input Data and Results (nonideal case)

Case	N_T	x^*_{D1}	q	t_{nr} hr	t_r hr	% Time Saved	Optimal R_1(kmol)	x_{R1}
1	5	0.900	0.607	3.77	3.70	1.86	0.50	0.558
2	5	0.910	0.638	4.11	3.98	3.11	0.70	0.608
3	4	0.900	0.728	4.72	4.40	6.68	0:73	0.662
4	3	0.900	0.910	11.32	5.56	50.86	1.91	0.673

Key: % time saved is based on t_{nr}

The benefits of recycling are again quite clear and the discussion presented in section 4.4.2 equally applies for the cases studied. Table 4.4 shows that recycling is again beneficial for **q** values greater than approximately 0.60. The solution statistics presented in

Table 4.5 Optimal Reflux Ratio Policies for Different Cases in Table 4.4

Case 1:						
No Recycle :						
Reflux Ratio		0.709	0.725	0.744		
Switching Times	t = 0	1.45	2.45	3.77		
Recycle :						
Reflux Ratio		0.670	0.690	0.719	0.479	
Switching Times	t = 0	0.92	1.99	3.29	3.70	
Case 2:						
No Recycle :						
Reflux Ratio		0.906	0.704	0.781		
Switching Times	t = 0	0.40	3.31	4.11		
Recycle :						
Reflux Ratio		0.808	0.674	0.735	0.479	
Switching Times	t = 0	0.40	1.67	3.58	3.98	
Case 3:						
No Recycle :						
Reflux Ratio		0.998	0.749	0.802		
Switching Times	t = 0	0.40	3.09	4.72		
Recycle :						
Reflux Ratio		0.749	0.721	0.763	0.563	
Switching Times	t = 0	1.14	2.45	3.89	4.40	
Case 4:						
No Recycle :						
Reflux Ratio		0.999	0.899	0.927		
Switching Times	t = 0	0.40	7.67	11.32		
Recycle :						
Reflux Ratio		0.999	0.718	0.761	0.376	
Switching Times	t = 0	0.4	1.67	4.33	5.56	

Table 4.6 shows that computation time with nonideal mixtures is about 3 times greater than with ideal mixtures (see Table 4.3 for one level formulation case). This can be explained from the fact that most of the time is consumed in evaluating physical properties. In ideal mixture cases (Table 4.3) the time spent in physical properties calculation is about 28-30% of the total time compared to 65-70% in nonideal cases (Table 4.6).

Table 4.6 Typical Solution Statistics for the Cases in Table 4.4

No. of Function Evaluation = 15
No. of Gradient Evaluation = 12
Time for each Function Evaluation (approx) = 1.7 min (CPU)
Time for each Gradient Evaluation = 2.1 min
Total Time (approx) = 52 min
% of Total Time spent in Physical Properties = 65 - 70

4.8 Conclusions

The proposed one level optimal control formulation is found to be much faster than the classical two level formulation to obtain optimal recycle policies in binary batch distillation. In addition, our experience is that the one level formulation is also much more robust. The reason for the robustness is that for every function evaluation of the outer loop problem, the two level method requires to reinitialize the reflux ratio profile for each new value of (R_1, x_{R1}). This was done automatically using the reflux ratio profile calculated at the previous function evaluation in the outer loop so that the inner loop problems (specially problem P2) could be solved in a small number of iterations. However, experience has shown that even after this reinitialization of the reflux profile sometimes no solutions (even sub-optimal) were obtained. This is because of failure to convergence within a maximum limit of function evaluations for the inner loop problems. On the other hand the one level formulation does not require such reinitialization. The reflux profile was set only at the beginning and a solution was always found within the prescribed number of function evaluations.

Recycling of an off-cut is found to be beneficial in most cases for q values greater than about 0.60. On the basis of these results this measure can qualitatively predict the benefit of using off cut recycle and therefore can be used as a decision variable. Optimal values for the amount and composition of the recycle and the reflux policies can then be obtained using the methods presented in this work. The methods are capable of showing when and how long an initial total reflux operation is needed for a particular separation. In some cases more than 70% savings of batch time was found when recycle was used. Further improvements on the column performance may be expected from finer control of the reflux ratio, obtained using a larger number of control intervals.

In all the case studies presented in this chapter it was assumed that the amount of fresh feed to be processed in the long production campaign is fixed for every batch cycle, but the reboiler was oversized to some extent so that it could accomodate the extra charge from the off cut recycle. The optimal amount of recycle was obtained within that bound (40% oversized) so that maximum benefit could be achieved out of the given column.

However, in practice it is always desirable to charge the reboiler to its full capacity and in that case, it might be required to determine the optimal amount of fresh feed, the optimal amount of recycle and its composition so as to maximize the amount of fresh feed processed in unit time (productivity). Although a proper optimal control solution was not produced for this problem, we scaled the results (amount of fresh feed and recycle) presented in Table 4.1 to a fixed reboiler capacity of 5 kmol in order to assess whether the reductions in operation times do indeed translate into higher productivity. Using the same recycle compositions as those mentioned in Table 4.1 we calculated the minimum operation time to separate the mixed charge into distillate and bottom products of the same purity (x^*_{D1}, x^*_{B2}). This will automatically produce an off-cut identical in amount and composition to that recycled (scaled) and therefore satisfies the quasi steady-state requirement. The productivity thus obtained [fresh feed(scaled) processed in unit time] is then compared with the productivity obtained by processing a fresh fixed reboiler charge (5 kmol) for the same given separation (x^*_{D1}, x^*_{B2}).

This study was done for case 2 and case 4 of Table 4.1 (for a low and high value of q). The results are summarized in Table 4.7. The purpose of this study is to show that recycling is beneficial even on a fixed reboiler capacity basis. Although these scaled results will in general not be strictly optimal, the benefits of recycling in terms of productivity are quite clear from Table 4.7. Also the benefit increases with q as before.

Table 4.7 Benefits of recycle in terms of Productivity (based on fixed reboiler charge)

Case	B^*_C kmol	B^S_0 kmol	R^S_1 kmol	x^*_{D1}	x^*_{B2}	$\underline{D}^S_1$ kmol	q	t_{nr} hr	t_{rS} hr	P_{nr} kmol/hr	P_r kmol/hr	% Increase in Productivity
2	5.0	4.545	0.455	0.900	0.150	2.727	0.597	3.10	2.62	1.613	1.733	7.44
4	5.0	4.019	0.981	0.912	0.132	2.411	0.799	5.57	3.09	0.897	1.300	44.93

Key : B^*_C = Fixed Reboiler Charge   Scale Factor (SF) = $B^*_C / (B_0 + R_1)$ (of Table 4.1)

$B^S_0 = B_0 \times SF$ (fresh feed) $R^S_1 = R_1 \times SF$ (recycle)

$B^*_C = B^S_0 + R^S_1$

x^*_{D1} x^*_{B2} are the specifications on the top main product and final bottom product and are same as those set in Table 4.1. (note: these specifications yielded $\underline{D}_1$ = 3.0 kmol in Table 4.1)

$\underline{D}_{1S}$ is the solution of equations (4.1-4.4) given B^S_0, x^*_D, and x^*_{B2}

t_{nr} is the batch time without recycle using 5 kmol (fixed reboiler charge) of fresh feed for the given separation x^*_{D1} and x^*_{B2} (this time is same as those reported in Table 4.1 as t_{nr})

t_{rS} is batch time with recycle using 5 kmol of mixed charge (B^*_C) for the same separation x^*_{D1} and x^*_{B2}

P_{nr} = Productivity without recycle = B^*_C / t_{nr}, kmol/hr

P_r = Productivity with recycle = B^S_0 / t_r, kmol/hr

% Productivity increase = $(P_r - P_{nr}) \times 100 / P_{nr}$

Note: In all cases x_{B0}, V_C and x_{R1} are the same as those used in Table 4.1

Chapter 5

OFF-CUT RECYCLE IN MULTICOMPONENT BATCH DISTILLATION

5.1 Introduction

This chapter addresses the formulation of an optimal control problem to obtain optimal recycle policies in multicomponent batch distillation. Some special cases are identified where the methods used for the binary case can be applied fairly easily to multicomponent mixtures. The previously mentioned measure **q** of "the degree of difficulty" of separation is used to identify those special cases. A new operational strategy regarding the order of off-cuts recycle in a multicomponent environment is discussed. The Benefits of recycling are correlated against the measure **q**.

The work published so far (Mayur et al., 1970; Christensen and Jorgensen, 1987) on the optimal recycle policy was restricted to binary mixtures. In their work and also in the previous chapter the benefits of recycling were measured in terms of a reduction in batch time although increase in productivity could be a possible alternative. Luyben (1988) considered this productivity measure (he defined it as "capacity" which includes both batch time and a constant charging and cleaning time) in a simulation of multicomponent batch distillation with recycle. But he was only interested to show the effect of different parameters (no of plates, relative volatilities) on the productivity and did not consider the effect of off-cuts recycle on the productivity.

In the past, the mathematical formulation of the optimal control problem to obtain optimal off-cut recycle policy was done for a quasi-steady state operation using binary mixtures where there was only one main-cut and one off-cut (Mayur et al., Christensen and Jorgensen) and the off-cut was always recycled at the beginning of the batch.

Since in a multicomponent mixture there can be n_C-1 main-cuts and n_C-1 off-cuts, there could be a number of operational stragies regarding the way the off-cuts are recycled. In this work these strategies will be discussed first and then a general optimal control formulation will be presented to obtain off-cut recycle policy for one of such operational strategies. The possible difficulties for solving the general formulation are outlined. However, special situations are identified and explained with examples where the method used for binary case is applicable.

5.2 Operational Strategies for Off-cut Recycle.

Recycling of all the off-cuts to the beginning of a new batch is quite common (see Figure 5.1).

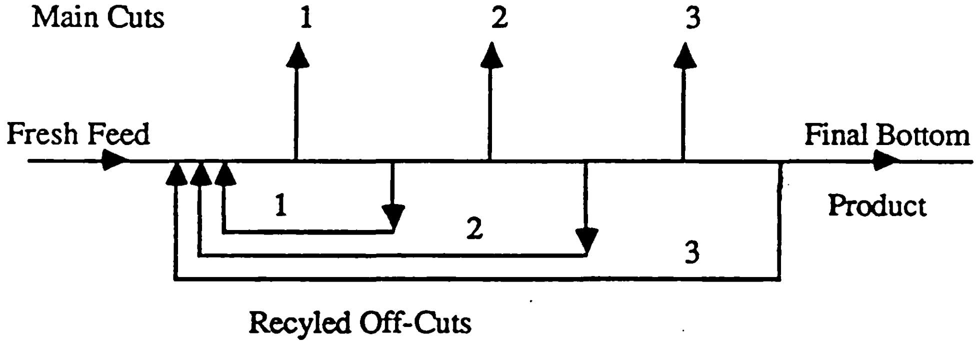


Figure 5.1 Typical Multicomponent Batch Operation with Recycled off-Cuts

This type of recycle policy has the follwing disadvantages:

a) As the number of off-cuts increases, the total amount of recycle increases, thus significantly reducing the amount of fresh feed that can be be processed for a given reboiler capacity. This consequently means a corresponding reduction in the total amount of product for a quasi-steady state operation.

Luyben (1988) considered the above operational strategy for off-cut recycle for a three component separation problem. Because of this strategy the amount of fresh feed processed at quasi-steady state was decreased by about 33% and the total product was decreased by the same amount. Although his intention was not to show the advantage or disadvantage of recycling, it was clear from his work that because of recycles, productivity was decreased by 13-16% with respect to that obtained with only fresh feed and no recycle on a fixed reboiler charge basis.

b) Recycling of heavy cuts to the initial charge will cause dilution of the lighter components in the reboiler mixture thus meaning higher energy requirement at the beginning because of a higher boiling point. This is not a thermodynamically sound policy since it involves remixing of products which had already been separated. However the main advantage of this strategy is that only one storage vessel is required for all the off-cuts.

To overcome the disadvantages of the above recycle policy, the following strategy could be a possible alternative:-

Collect and store the off-cuts in sequential order and recycle each of them to the reboiler in that order. Figure 5.2 illustrates the strategy. In practice this is obtained by adding off-cuts from the previous batch to the reboiler at suitable times.

With this recycle policy the reduction in the amount of product is only equal to the amount of the first off-cut for a quasi-steady state operation. This approach will also reduce the possibility of having a high boiling point in the reboiler mixture at the beginning of the process. In addition remixing of products already separated is minimized. However this policy will require n_C-1 distinct intermediate tanks and a more sophisticated controller to charge the off-cuts at the right time.

5.3 Optimal Off-Cut Recycle Policy Formulation for Multicomponent Batch Distillation

We consider the sequential off-cut recycle strategy (Figure 5.2) in this formulation. An optimal control formulation is presented with an objective to minimize the overall batch distillation time as mentioned in case 1 in section 5.1. The formulation uses the nomenclature shown in Figure 5.2.

Given a fresh feed (B_0, x_{B0}) and the desired purity specification of the main-cuts $(x^1_{D1}, x^2_{D2}, x^3_{D3}, ...)$ determine the amount and composition of the off-cuts $[(R_1, x_{R1}), (R_2, x_{R2}), (R_3, x_{R3}), ...]$ and the reflux ratio policy r(t) which minimize the overall distillation time. The optimisation problem can be written as:

$$\min_{(R_1, x_{R1}), (R_2, x_{R2}), (R_3, x_{R3}), ..., r(t)} \quad J = t_1 + t_2 + t_3 + t_4 + ... = t_F$$

subject to : (5.1)

1. bounds on the amount of off-cuts
2. bounds on the purity of off-cuts

and interior point constraints

1. purity requirements of the main-cuts
2. requirements of off-cuts to establish quasi-steady state operation

where t_1, t_2, t_3 ... are the batch times for the production of individual cuts (main-cuts and off-cuts). In addition to the constraints mentioned above the model equations act as equality constraint to the optimisation problem.

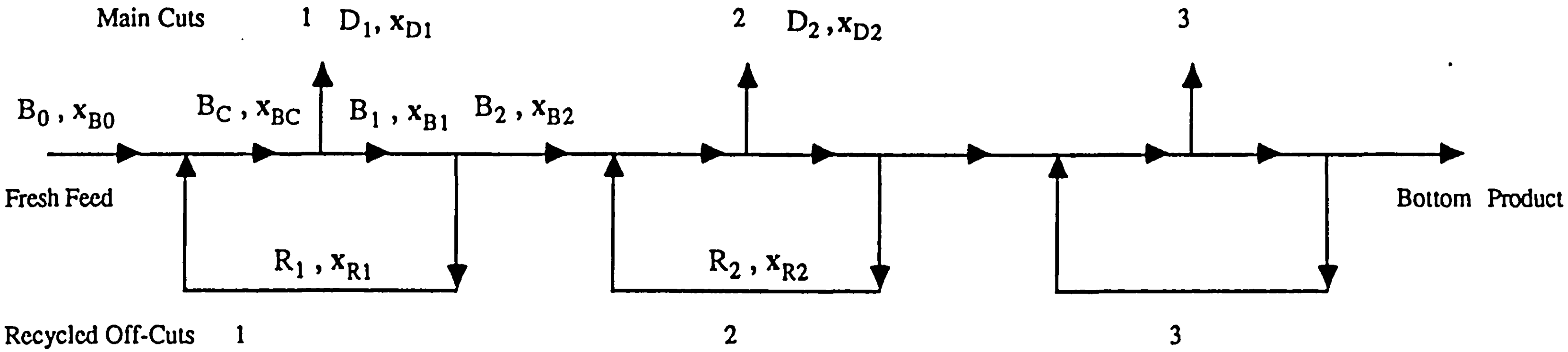


Figure 5.2 Multicomponent Batch Operation with Sequential Off-Cut Recycle

5.3.1 Solution of the Optimal Control Problem

The general formulation presented in section 5.3 results in a very complex optimisation problem and the solution of the problem using currently used methods is very difficult. Also as the number of components increase, the above formulation will result in a very big optimisation problem with a large number of optimisation variables. Even for a 3 component mixture, this will result in six decision variables (R_1, x^1_{R1}, x^2_{R1}, R_2, x^1_{R2}, x^2_{R2}) in addition to the variables arising from the number of control intervals and switching times [r(t)] in each of the four cuts (2 main-cuts and 2 off-cuts).

In addition to the above mentioned problem, numerical difficulties may arise using Morison's (1984) optimal control algorithm used in the earlier chapters. The system (model equations) describing the multicomponent off-cut recycle operation needs to be reinitialized at the end of each main-cut and off-cut to accomodate the next off-cut to the reboiler. To optimise these initial conditions (new mixed reboiler charge and its composition) it is essential to obtain the objective function gradients with respect to these initial conditions. This cannot be done in the optimal control code of Morison.

However the operational strategy shown in Figure 5.2 can be considered as a sequence of processes and an optimal control algorithm also given by Morison (1984) could be used for such sequential processes. This algorithm provides for the initialization of the sequence of processes and provides all the required gradients information. However an implementation of this algorithm was not available, so its theoretical and practical aspects could not be assessed.

Although a general solution to generate optimal recycle policies in multicomponent batch distillation would be desirable, because of the above mentioned difficulties we examined a decomposition strategy in which the whole optimal control problem is divided into a series of independent optimal control problems (see Figure 5.3). This is presented in the next section.

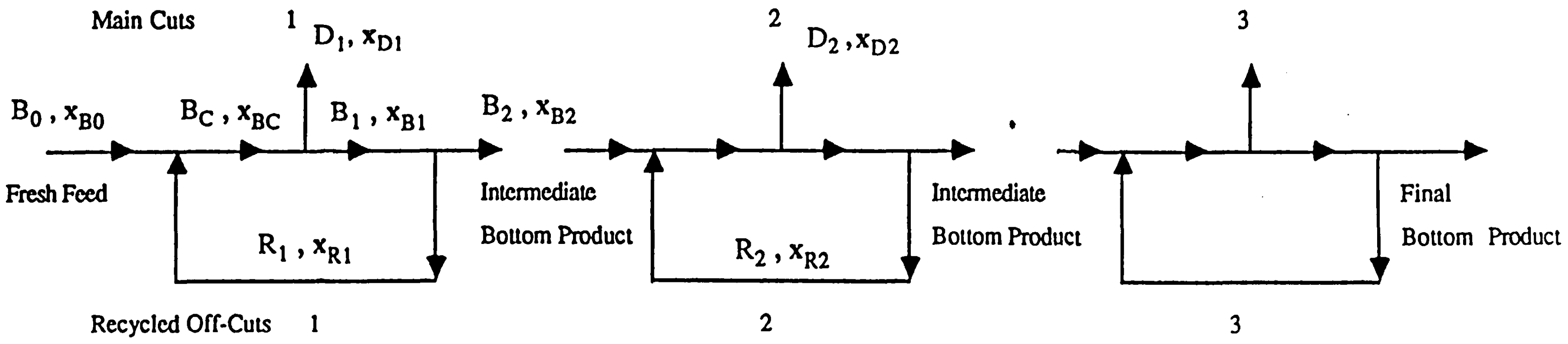


Figure 5.3 Decomposed Multicomponent Batch Operation with Off-Cut Recycle

5.4 Decomposed Optimal Control Formulation for Multicomponent Batch Distillation with Off-Cut Recycle

Before discussing how the general optimal control problem mentioned in the previous section is decomposed into a series of independent optimal control problem and before presenting the mathematical formulation for such problem the following discussion is worthwhile:

We consider a three component mixture for convenience of discussion. Consider the overall and component mass balance around loop 1 shown in Figure 5.4 (the decomposed optimal control problem for the first cut). The balance equations are the same as those presented in chapter 4 (equations 4.1-4.4). For a given charge (B_0, x_{B0}) and for a 3 component mixture there will be 5 equations with 8 unknown variables (D_1,x^1_{D1}, x^2_{D1}, x^3_{D1}, B_2, x^1_{B2}, x^2_{B2}, x^3_{B2}). Therefore, the degree of freedom (DF) of the steady state mass balances is 3. Several choices of consistent specifications are to specify (x^1_{D1}, x^2_{D1}, x^1_{B2}) or (x^1_{D1}, x^1_{B2}, x^2_{B2}) etc. In all cases, however we need to specify at least 2 compositions in either the main-cut (D_1) or the bottom product (B_2). From the mathematical point of view any one of these specifications can be used to solve the mass balance equations, but it is very difficult to achieve a separation when specifying two component puritiesin a single product (top or bottom). This is because these purities depend on various other factors (relative volatilities, number of plates, etc.).

Usually separation specifications in multicomponent mixtures are imposed on a particular component of the cut, e.g. on component 1 in main-cut 1, on component 2 in main-cut 2 and likewise. Of course, it is possible to set intuitively some of the heavier component compositions to zero during lighter cuts and some of the lighter component compositions to zero during heavier cuts. But it is really difficult to specify independently the compositions of the preceeding and few successive component compositions in a particular cut.

The question that immediately comes into mind in such cases is why not specify only one component either on the top or on the bottom product and solve the optimal control problem. The answer is that with such specifications it is very easy to formulate and solve an optimal control problem for multicomponent case without recycle (several examples were presented in chapter 3 and also see Diwekar et al., 1987), but such a problem with off-cut recycles is very difficult. This is because, it is very difficult to satisfy the conditions for the quasi-steady state operation. However, the problem can always be formulated and solved with great ease if the only concern is to produce cuts (main-cut and off-cut) satisfying the required amount and purity on one of the components only (see the results section) and which does not require to consider a quasi-steady state operation.

To determine the optimal quasi-steady state off-cut recycle policies we restrict ourselves to the special case considered by Luyben (1988), that is, to the case of sharp separation between all components (either using wide range of relative volatilities or a large number of plates). The main assumptions are further illustrated with reference to Figure 5.4.

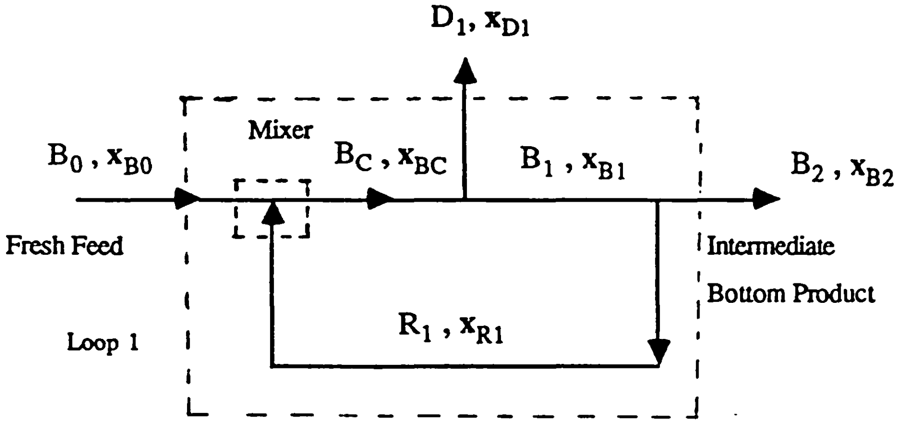


Figure 5.4 Off-Cut Recycle after the First Cut

The assumptions are as follows:

1) No 3rd component with respect to a particular main-cut component ("cut component" was defined in chapter 3) would appear in the main cut. e.g. $x^3_{D1} = 0$ for main-cut 1, $x^4_{D2} = 0$ for main-cut 2 etc.

2) Any amount left of a particular cut component will be removed totally in the next main-cut. For example the amount left of component 1 after main-cut 1 will be totally removed in main-cut 2 and therefore component 1 will not appear in subsequent cuts.

3) No 3rd component with respect to a particular off-cut component would appear in the off-cut.

e.g. for off-cut 1 $x^3_{R1} = 0$, for off-cut 2 $x^4_{R2} = 0$ and likewise. This assumption together with assunption no. 2 imply that each off-cut would only comprise of the main-cut component and the next component in the mixture.

Now with these assumptions and for given specifications (x^1_{D1}, x^1_{B2}) on component 1 in main-cut 1 and in the intermediate final bottom product (bottom product after 1 main-cut and 1 off-cut), solving the mass balance around the loop 1 (Figure 5.4) will give the rest of the variables (D_1, B_2,.....etc.). Let the solution of D_1 be given by $D_1 = \underline{D}_1$, and that of B_2 be given by $B_2 = \underline{B}_2$.

Also with these assumptions and specifications the whole optimal control problem shown in Figure 5.2 is now decomposed into a series of independent optimal control problems (Figure 5.3). Referring to Figure 5.4 each optimal control problem may now be described as follows:

Given a batch charge B_0, x_{B0}, a desired amount of distillate $\underline{D}_1$ of specified purity x^1_{D1} and final bottom product $\underline{B}_2$ of specified purity x^1_{B2} determine the amount and composition of the off-cut (R_1, x^1_{R1}) and the reflux rate policy r(t) which minimizes the overall distillation time.The optimisation problem is now the same as P4 (equation 4.11 in chapter 4) for binary mixtures. For convenience it is reproduced here.

P5
$$\min_{B_C,\, x^1_{BC},\, r(t)} \quad J = t_1 + t_2 = t_F$$

subject to interior point constraints

$$D_1(t_1) = \underline{D}_1$$
$$x_{D1}(t_1) = x^1_{D1} \qquad (5.2)$$

and end point constraints

$$B_2(t_F) = \underline{B}_2$$

$$x_{B2}(t_F) = x^1_{B2}$$

where t_1 is the time required to produce the main-cut and is defined by one of the switching times, t_2 is the time required to produce the off-cut and t_F is the final time. In this formulation we optimise B_C, x^1_{BC} (the mixed reboiler charge) instead of R_1, x^1_{R1}. At the solution R_1, x^1_{R1} are obtained from a material balance.

Also because of the assumptions made earlier, the formulation presented above automatically accounts for the quasi-steady state operation.

The above optimal control problem is solved using the codes used in the previous chapter and the benefits of recycling is correlated against the measure of "the degree of difficulty" of separation, **q**. The same measure is also used to identify the situations when the assumptions mentioned above are valid.

The formulation presented above is equally applicable to subsequent cuts.

5.5 Measure of "the degree of difficulty" of Separation, q for Multicomponent Mixtures

Referring to Figure 5.4, the same definition of "the degree of difficulty" of separation for binary mixtures is applied for multicomponent mixtures.

$$q = \int_{x^1_{B2}}^{x^1_{B0}} \frac{(N_{min} + 1)\, dx^1_B}{(x^1_{B0} - x^1_{B2})\,(N_T + 1)} \qquad (5.3)$$

N_{min} is again evaluated using the Fenske's equation :-

$$N_{min} + 1 = \frac{\ln\left(\frac{x^1_{D1}}{x^r_{D1}}\right)\left(\frac{x^r_B}{x^1_B}\right)}{\alpha_{1-r}} \tag{5.4}$$

where r is any reference component (not to be confused with the reflux ratio). x^1_B and x^r_B are intermediate compositions of components 1 and r as the reboiler composition changes from x^1_{B0} to x^1_{B2}. x^1_{D1} is the specified composition of the cut-component in main-cut 1 and x^r_{D1} is the composition of component r in main-cut 1.

In order to evaluate equation (5.4) we must obtain the values of all x^i_B ($i=1,2,....n_c$) at intermediate points as the reboiler composition changes from x^1_{B0} to x^1_{B2}. This is because we need to obtain the values of x^r_B in equation (5.3) and need to calculate a geometric average value of α_{1-r} at each intermediate point. The average relative volatility α_{1-r} is obtained based on the relative volatilities on the top and bottom of the column which are evaluated using bubble point calculations. Diwekar and Madhavan (1986) in their multicomponent batch distillation simulation work obtained these x^i_B with respect to the change in composition of a reference component. In our case, say for the first cut, we can use component 1 as a reference component for this purpose (note: this reference component must not be confused with the reference component r mentioned previously). The following equation given by these authors can be used to evaluate x^i_B:

$$x^i_B = x^i_{D1} - \frac{x^i_{D1} - x^i_{B0}}{x^1_{D1} - x^1_{B0}}\left(x^1_{D1} - x^1_B\right) \tag{5.5}$$

The derivation of this equation was simply done by doing a material balance over the entire column ignoring column holdup. Detail of this derivation was given by the original authors. The same equations (5.3-5) are used to evaluate **q** values for subsequent cuts. In evaluating equation (5.3) for a particular cut we use the reference component r as the component next to the "cut component".

5.6 Example Problems

5.6.1 Example 1

5.6.1.1 Problem Description: We consider a typical ternary mixture 1. Butane, 2.Pentane and 3. Hexane. For convenience and to reduce computational time we consider only the optimal operation for the first main cut and the first off-cut. Therefore only one main-cut and one off-cut is involved . Table 5.1 lists a variety of separation specification (on CUT 1) and column configurations in each case. A fresh feed of 6 kmol at a composition of <0.15,0.35,0.50> (mole fraction) is used in all cases. Also in each case a constant condenser vapor load of 3 kmol/hr is used. For convenience ideal phase equilibrium is assumed.

The column compositions are initialized to the composition of the mixed reboiler charge and a total of 2% of the fresh feed is used as column holdup. Half of the column holdup is assumed to be in the condenser and the rest is distributed equally over the plates. Reflux ratio is used as control variable of the process with 3 piecewise constant control intervals used for the main-cut separation and 1 control interval used for the off-cut.

The measure **q** is used to identify the range where a recycle policy should be applied. Also the measure is used to correlate the extent of benefit resulting from the recycle of the off-cut.

5.6.1.2 Results and Discussions: The value of **q**, the optimal batch time with and without recycle, the percentage of time savings due to recycle and the optimal amount and composition of the recycle are presented in Table 5.1 for cases wherever it is applicable. The accumulated and instant distillate compositon curves with and without recycle cases are shown in Figures 5.5-5.10. These figures also show the optimal reflux ratio profiles for each cases.

As can be seen from Table 5.1 the batch time for operation without recycle is not reported for case 3 and 4. A true quasi-steady mode of operation was not obtained for case 4 because of the violation of the sharp separation assumption mentioned in section 5.4. It is clear from Figure 5.10 that a substantial amount of the 3rd component is going to appear in the recycle cut, although the main-cut was free from that component. Therefore, the optimal control method used was not suitable for this case. However this case is an example where the off-cut recycle policies are generated by satisfying the amount and composition of the off-cut component only (i.e by violating quasi-steady state operation mode which requires other component compositions of the off-cut to be identical with those of recycled off-cut). For case 3 it was not possible to obtain the separation in the column considered without recycle. This situation clearly shows the benefit of recycling of off-cut.

Table 5.1 Input Data and Results for Example 1

Case	N_T	x^1_{D1}	x^1_{B2}	$\underline{D}_1$	q	t_{nr} hr	t_r hr	% Time Saved	Optimal R_1(kmol)	x_{R1}
1	4	0.850	0.010	1.0	0.533	3.074	1.988	35.33	0.54	0.273
2	5	0.935	0.011	0.9	0.561	3.803	2.052	46.04	0.64	0.276
3	5	0.950	0.009	0.9	0.601	------	2.273	------	0.75	0.278
4	3	0.850	0.010	1.0	0.667	------	2.290	------	1.18	0.281

Key : N_T = no. of plates t_{nr} = batch time without recycle t_r = batch time recycle

% Time saved is based on t_{nr}

The results presented in Table 5.1 and in the Figures 5.5, 5.7, 5.9 and 5.10 clearly show that there is an upper limit of q, beyond which the assumptions mentioned in section 6.3 are not valid and the present method can not be applied. Present investigation shows that up to q equal to about 0.6, the present method is adequately applicable.

Figure 5.5a Accumulated Distillate Composition

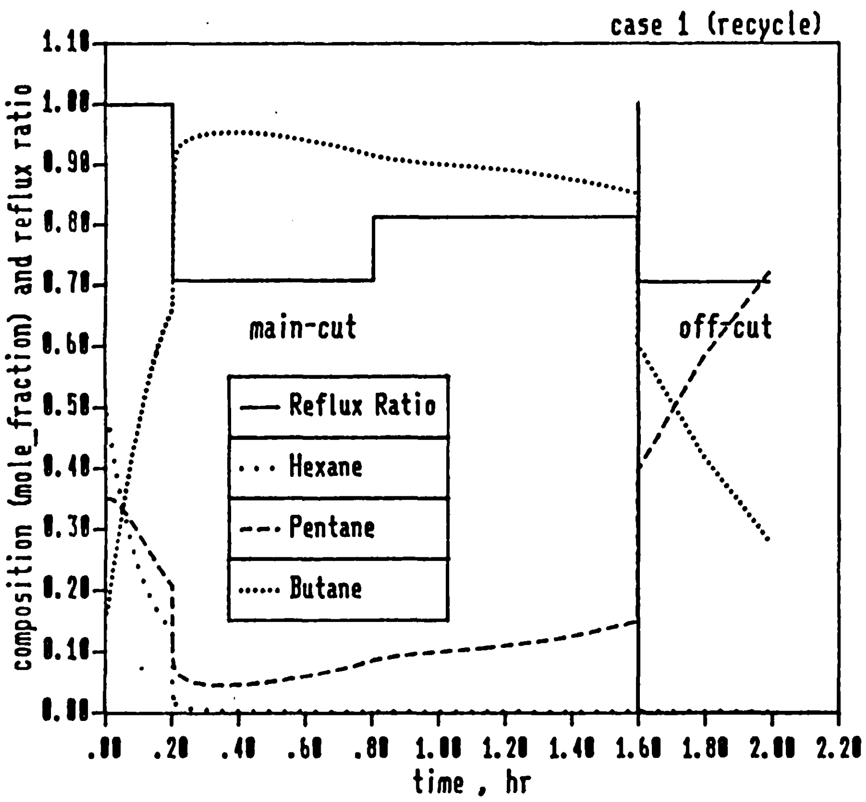


Figure 5.5b Instant Distillate Composition

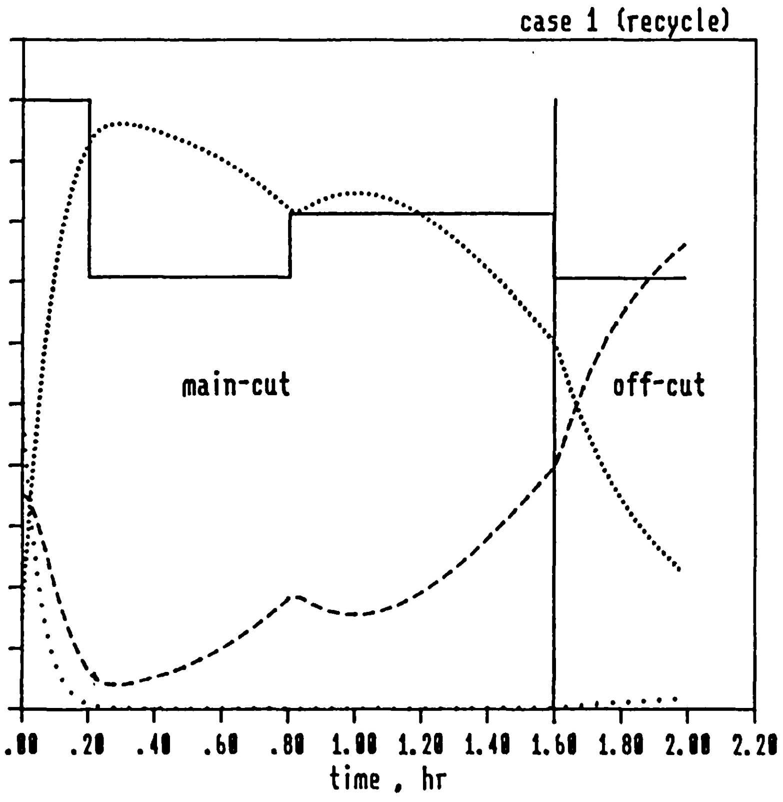


Figure 5.6a Accumulated Distillate Composition

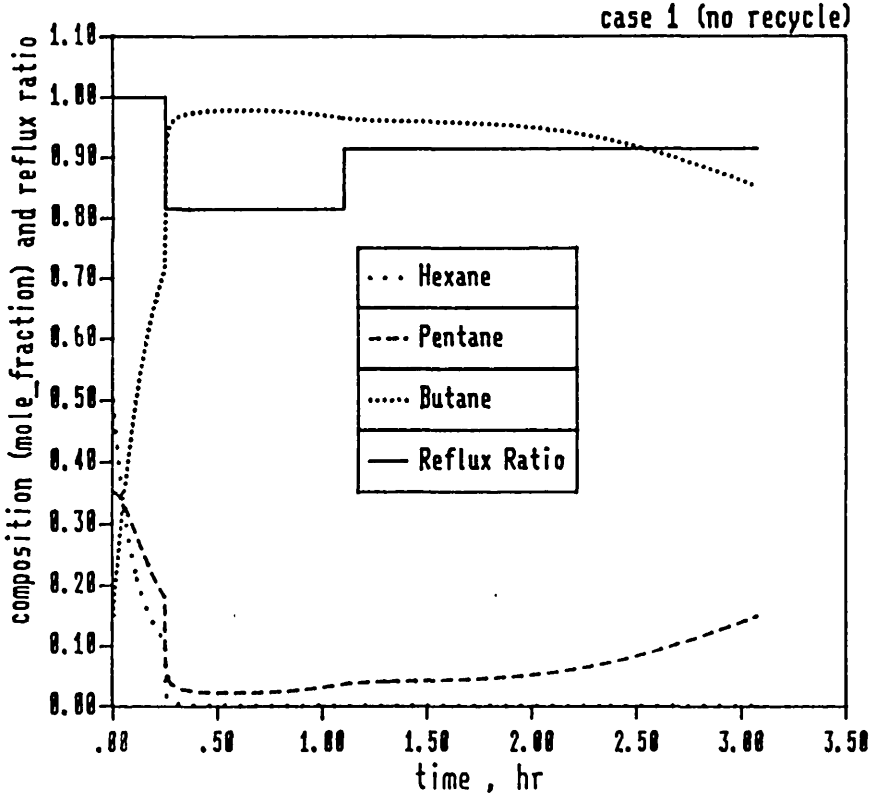


Figure 5.6b Instant Distillate Composition

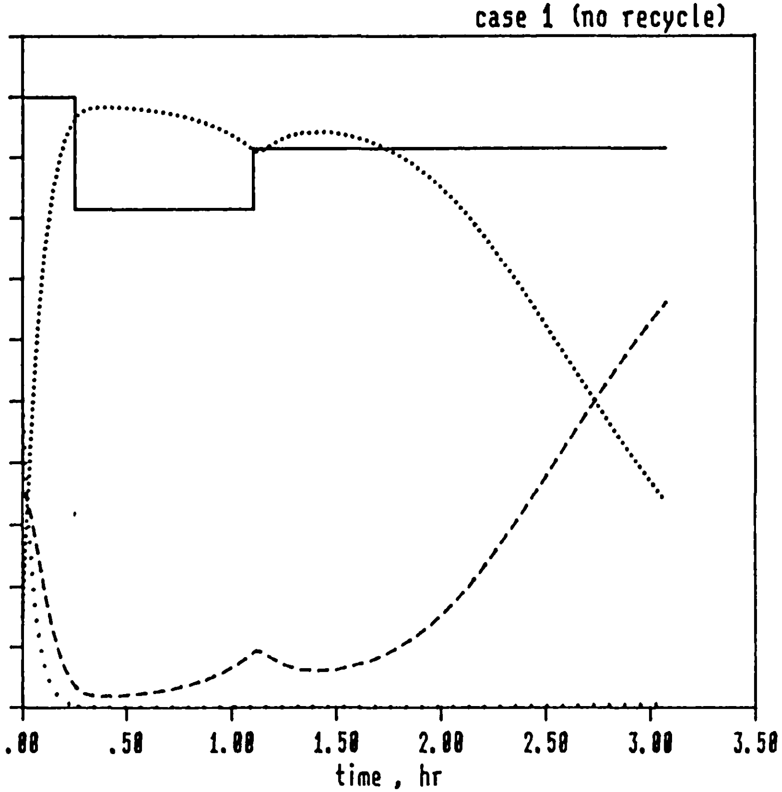


Figure 5.7a Accumulated Distillate Composition

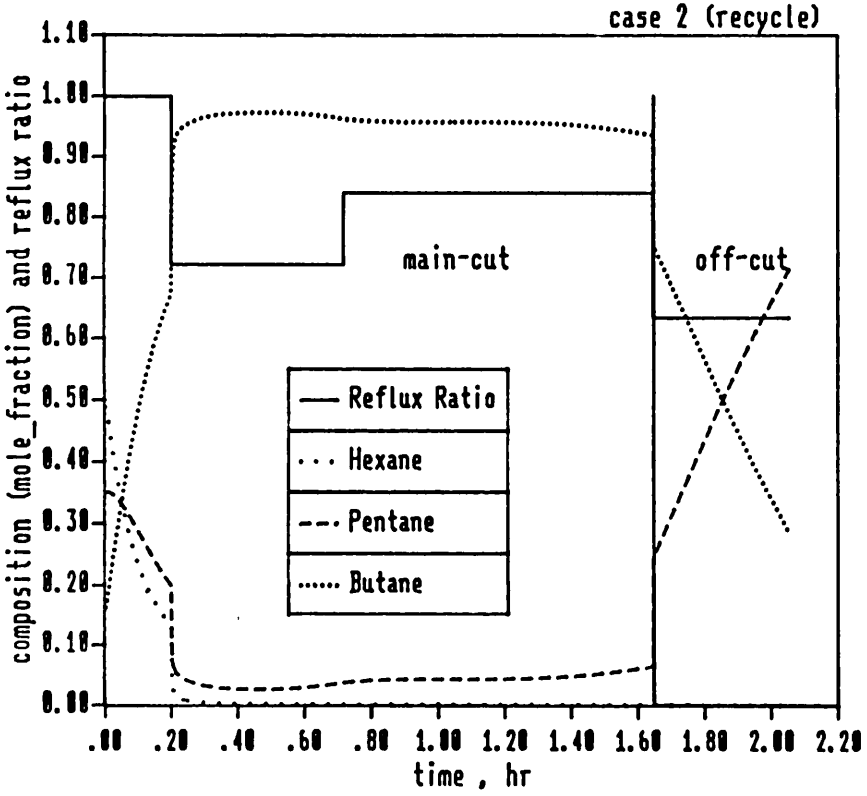


Figure 5.7b Instant Distillate Composition

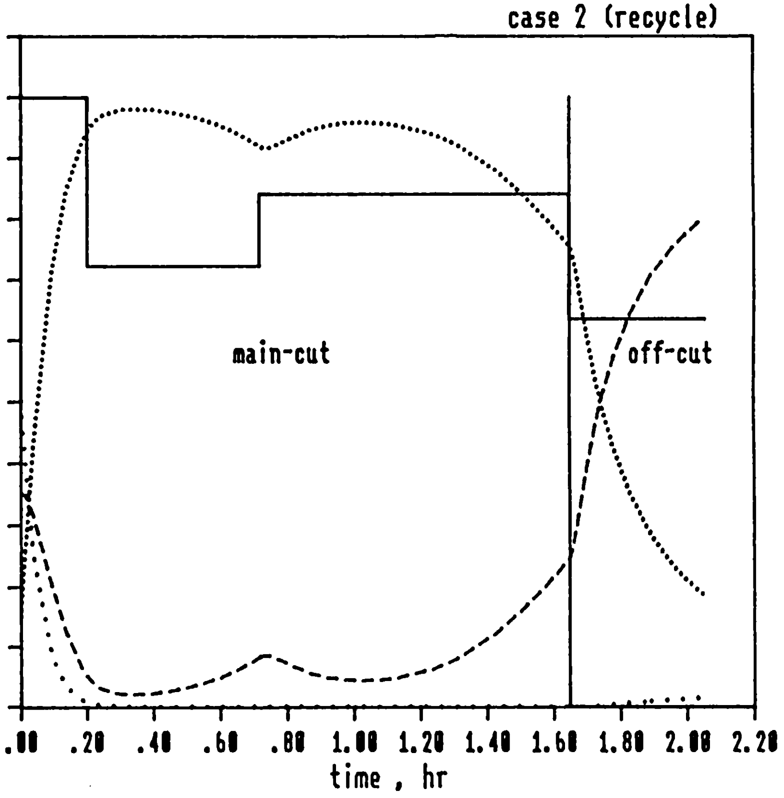


Figure 5.8a Accumulated Distillate Composition

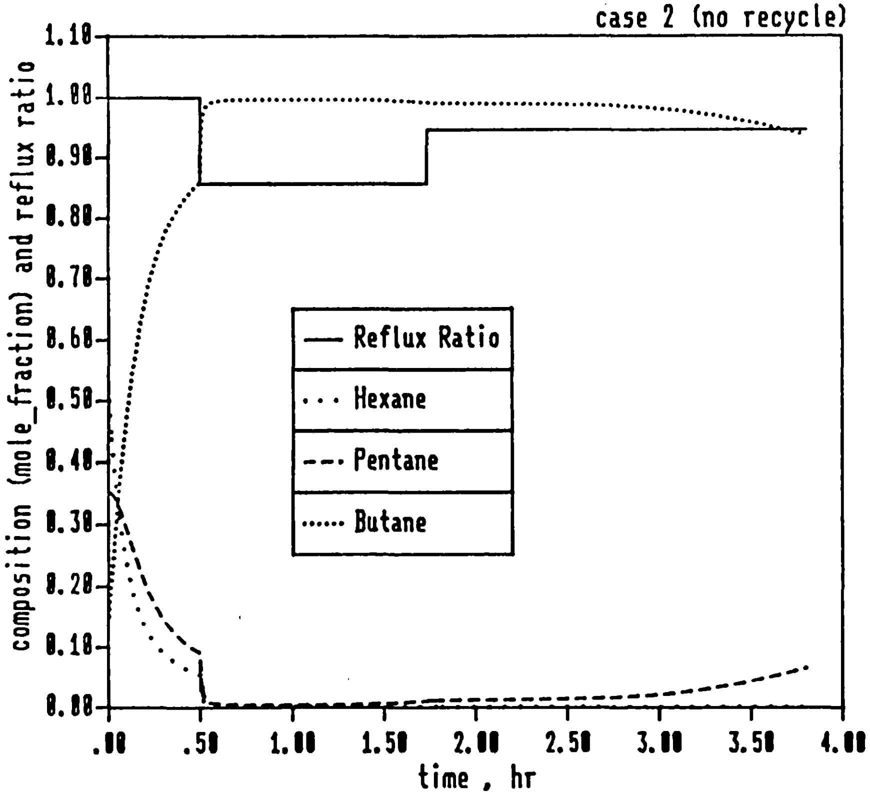


Figure 5.8b Instant Distillate Composition

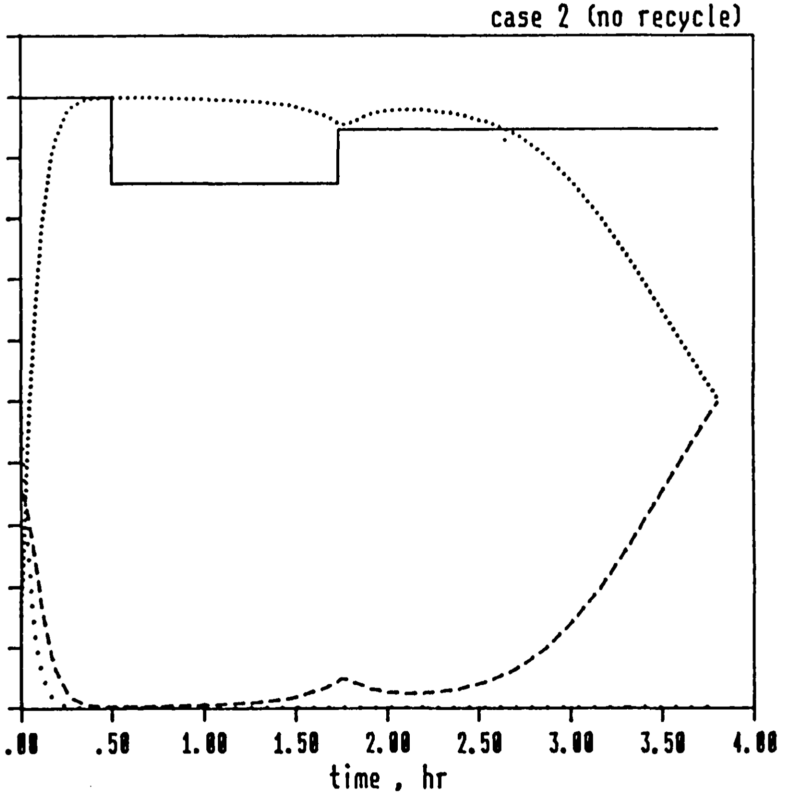


Figure 5.9a Accumulated Distillate Composition

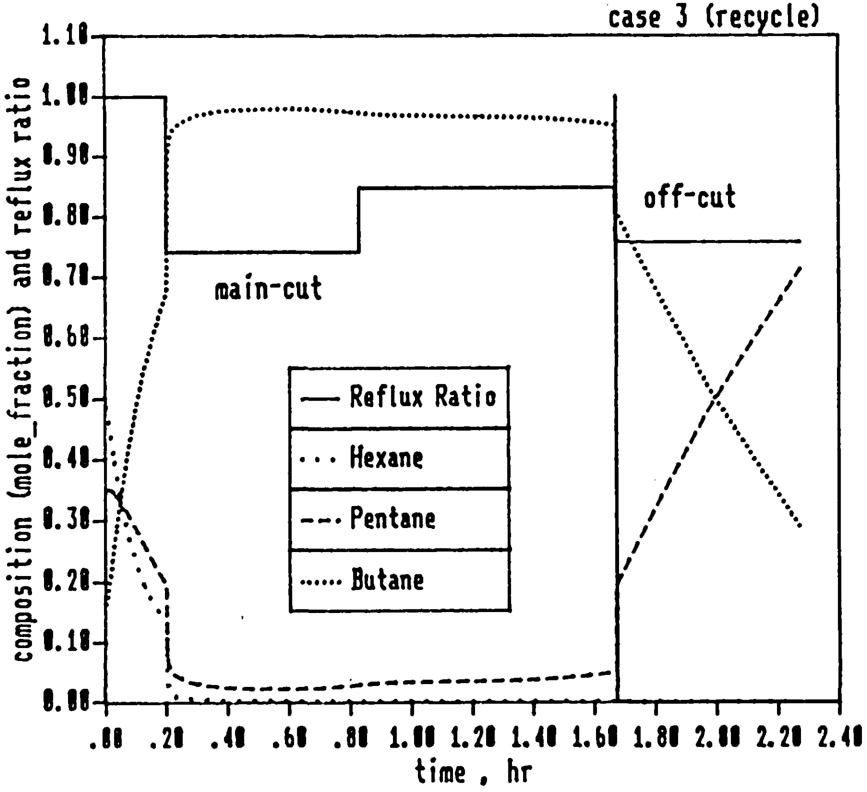


Figure 5.9b Instant Distillate Composition

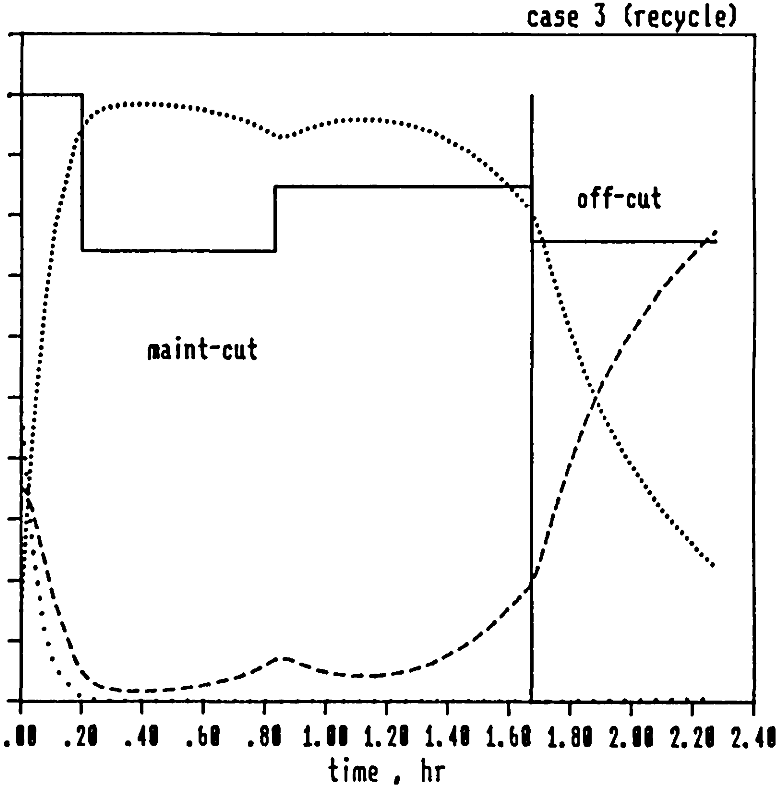


Figure 5.10a Accumulated Distillate Composition

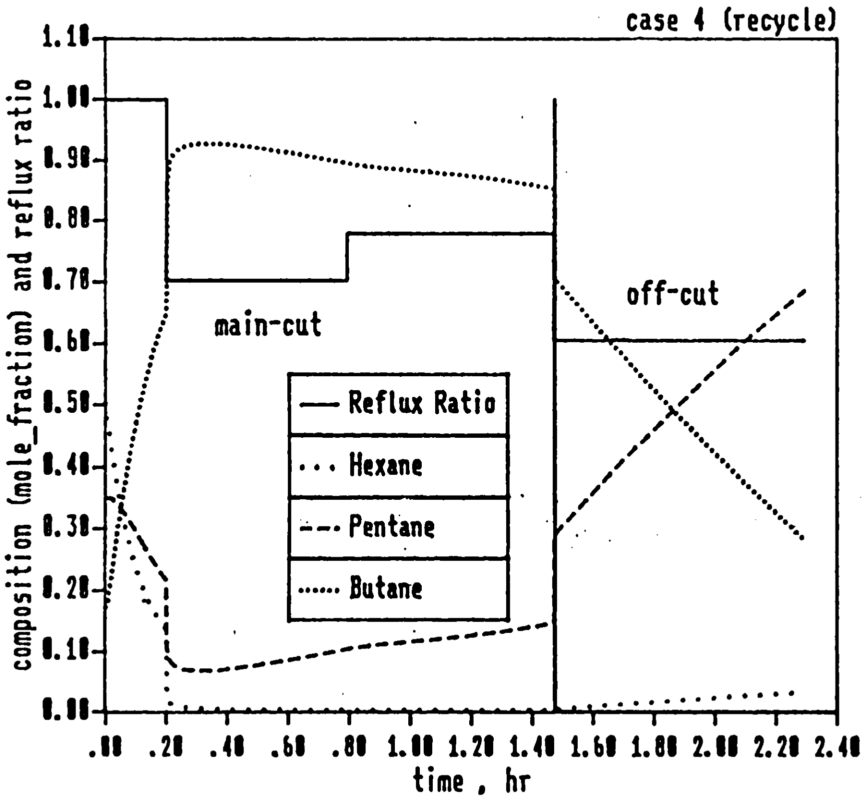
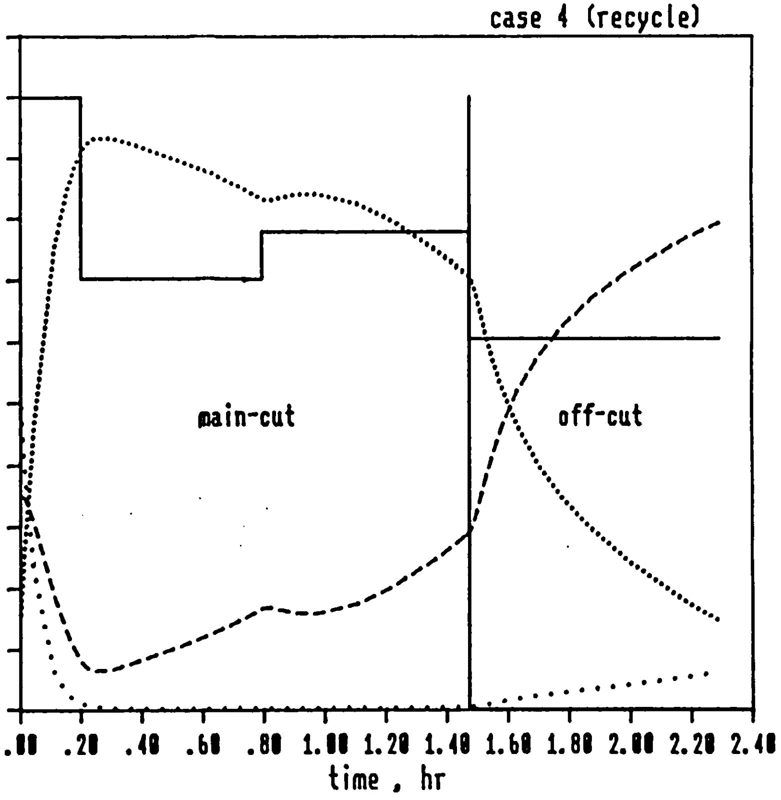


Figure 5.10b Instant Distillate Composition



As in binary cases, recycling of off-cuts offers a possibility of significant reduction in batch time. In some cases it is found to be more than 45%. The results also shows that the measure **q** can be used to predict qualitatively whether recycle is beneficial or not. As the value of **q** increases the benefit of recycle increases and so do the amount of and composition of the lighter component in the cut.

An interesting part of the results mentioned so far is that in all cases, an initial total reflux operation was found to be necessary. This was to eliminate the third component from the overheads. The length of that period again depends on the ease of separation. For no recycle cases, the times at total reflux are slightly larger than those for recycle cases. This is because the recycle of off-cut eases the separation of the main-cut. After the total reflux period of operation an increasing reflux ratio profile was obtained for the main-cut in all cases. The off-cut was always obtained at low reflux ratio as was the case with binaries.

5.6.2 Example 2

5.6.2.1 Problem Description: We consider the same ternary mixture in this example but considered the whole operation which includes 2 main-cuts and 2 intermediate off-cuts. The column consits of 5 (N_T) intermediate plates, a total condenser and a reboiler. The column is charged with the same amount and composition of the fresh feed as was in example 1. Column initialization, holdup distribution and condenser vapor load are also same as in example 1.

The specifications for the first cut are same as those presented in Table 5.1 for case 2 which is $x^{1}_{D1} = 0.935$ and $x^{1}_{B2} = 0.011$. These results in $\underline{D}_1 = 0.9$ kmol and $\mathbf{q}_1 = 0.561$. For cut 2 we specify $x^{2}_{D2} = 0.82$ and $x^{2}_{B2} = 0.13$ which results in $\underline{D}_2 = 2.0$ kmol and $\mathbf{q}_2=0.415$.

5.6.2.2 Results and Discussions: The minimum batch times for the individual cuts and for the whole operation are presented in Table 5.2 together with the optimal amount of recycle and its composition for each cut. The percentage time savings using recycle policies are also shown for the individual cuts and also for the whole operation. Figure 5.11 and 5.12

show the accumulated distillate and composition profile without and with recycle case for the whole operation. These also show the optimal reflux ratio profiles. Solution statistics for the problem are presented in Table 5.3 and 5.4 for no recycle and recycle cases.

It is clear from the Table 5.2 that although recycling of off-cut was beneficial for the first cut it was not at all beneficial for the second cut. This was quite expected because the **q** value for the second cut was quite small. However based on the total operation time recycling is still beneficial compared to no recycle case.

Table 5.2 Input data and Results for Example 2

t^1_{nr}	t^2_{nr}	t^1_r	t^2_r	Optimal				T_{nr}	T_r	T^1_S	T^2_S	T_S
hr	hr	hr	hr	R_1	x^1_{R1}	R_2	x^2_{R2}	hr	hr			
3.80	1.71	2.05	1.69	0.64	0.276	0.41	0.400	5.51	3.74	46.05	1.17	32.12

Key : t^1_{nr} = batch time without recycle for cut 1. t^2_{nr} = batch time without recycle for cut 2

t^1_r = batch time with recycle for cut 1. t^2_r = batch time with recycle for cut 2.

T_{nr} = total batch time for the whole operation without recycle.

T_r = total batch time for the whole operation with recycle.

T^1_S = % time saving based on t^1_{nr} for cut 1.

T^2_S = % time saving based on t^2_{nr} for cut 2.

T_S = % time saving based on T_{nr} for the whole operation.

It is also clear from the Figures 5.11 and 5.12 that the assumptions mentioned in section 5.4 were not violated in all cuts. Although the cut 1 operation required initial total reflux period to remove the 3rd component from the top of the column, initial total reflux period was not required for cut 2 operation because no 3rd component with respect to the "cut component"

Figure 5.11a Accumulated Distillate Composition
(Example 2, no recycle)

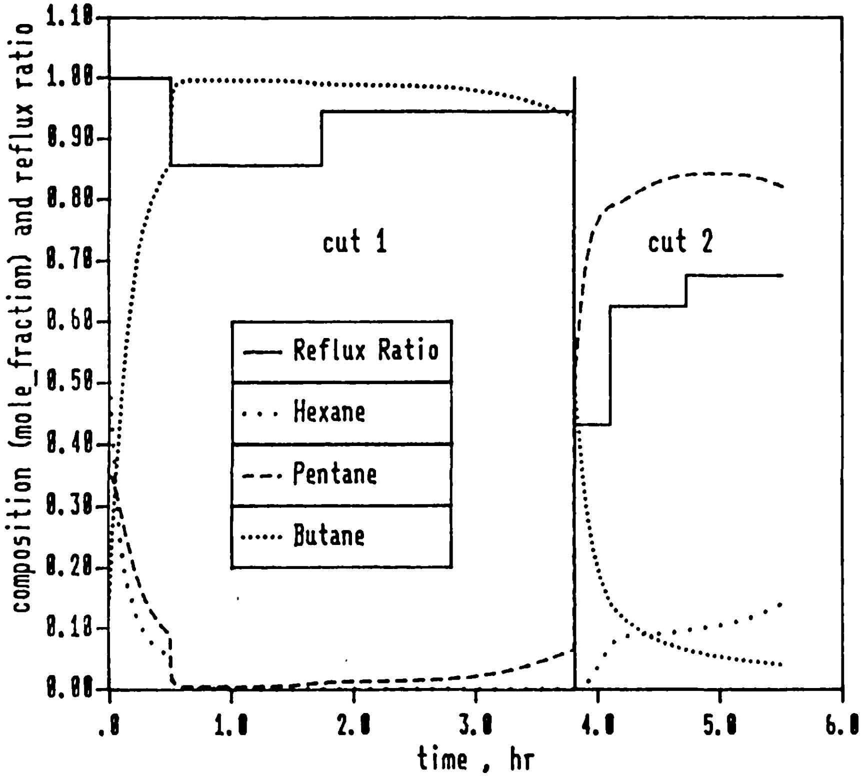


Figure 5.11b Instant Distillate Composition
(Example 2, no recycle)

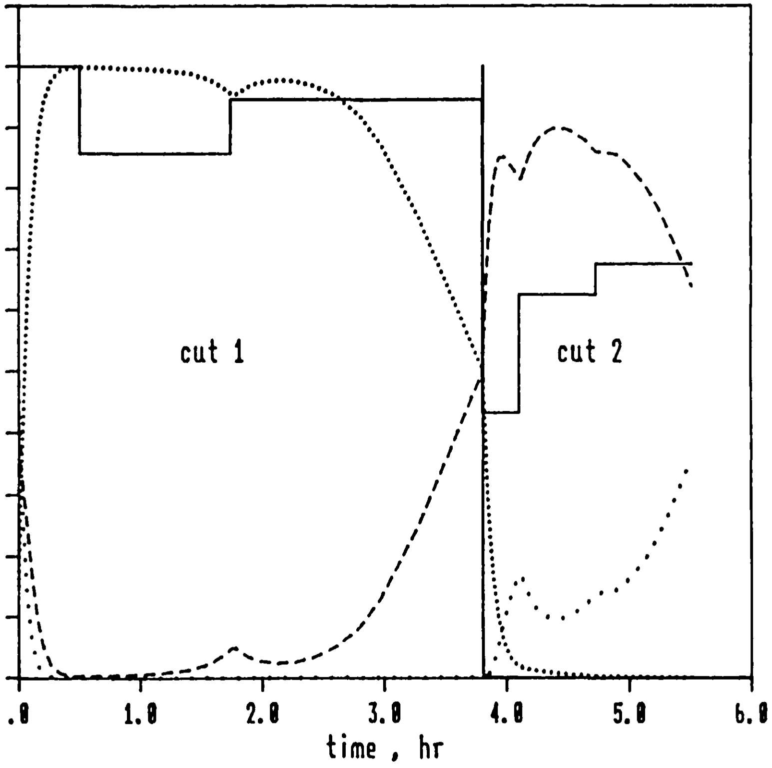


Figure 5.12a Accumulated Distillate Composition
(Example 2, recycle)

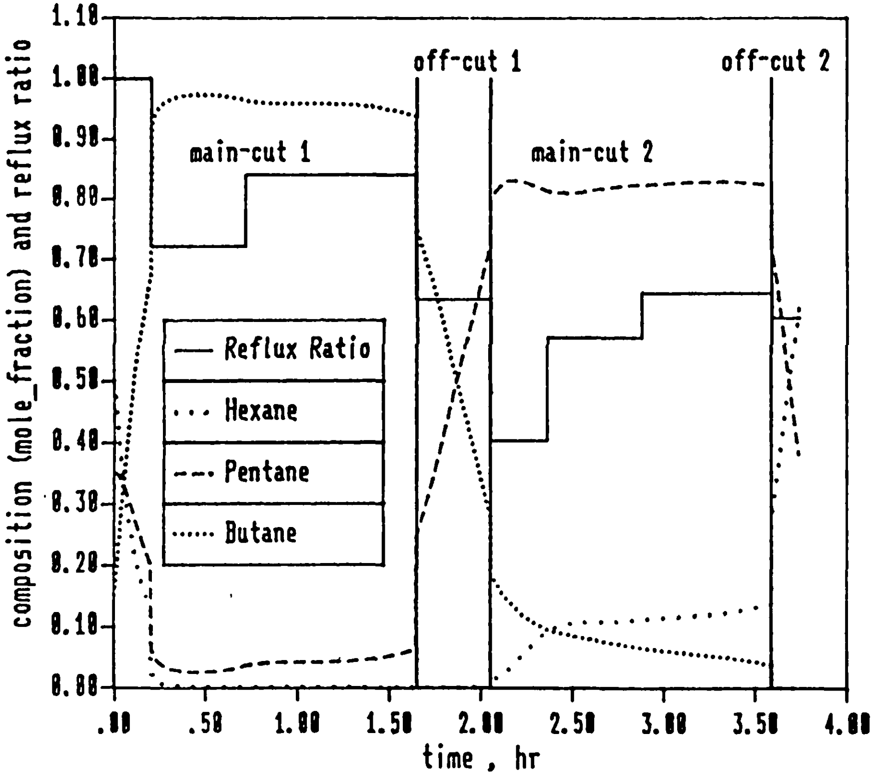
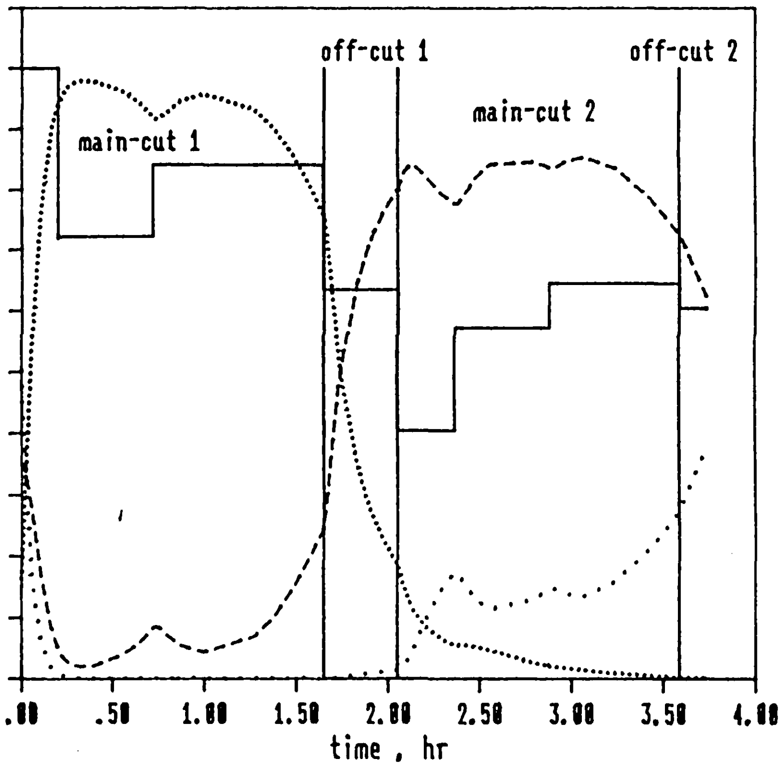


Figure 5.12b Instant Distillate Composition
(Example 2, recycle)



was present in cut 2.

Solution statistics presented in Table 5.3 for no recycle cases shows that the number of function and gradient evaluations for cut 1 is almost double of that required for cut 2 which means a larger amount of computation time. The reason is that for cut 1 the initial reflux

Table 5.3 Solution Statistics of Example 2 (no recycle case)

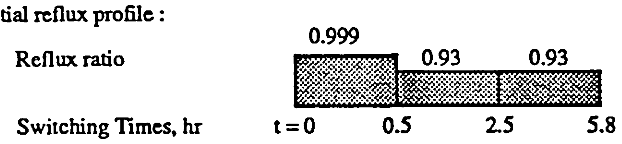
CUT 1

No. of Function Evaluations = 20
No. of Gradient Evaluations = 14

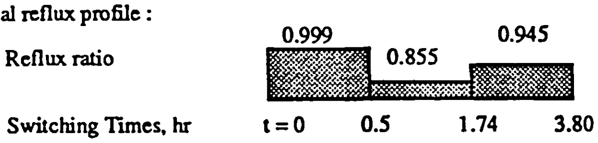
Time for each Function Evaluation (approx) = 0.7 min
Time for each Gradient Evaluation = 1.1 min

Total CPU time (including translator and optimiser) = 31 min
% of total time spent in physical properties = 29

Initial reflux profile :



Optimal reflux profile :



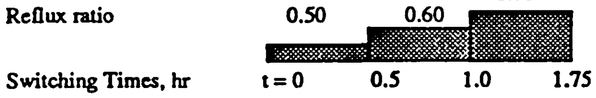
CUT 2

No. of Function Evaluations = 10
No. of Gradient Evaluations = 8

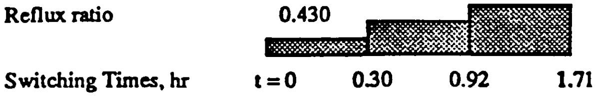
Time for each Function Evaluation (approx) = 0.5 min
Time for each Gradient Evaluation = 0.8 min

Total CPU time (including translator and optimiser) = 12 min
% of total time spent in physical properties = 28

Initial reflux profile :



Optimal reflux profile :



profile was far from the optimal one, but in cut 2 the initial reflux profile was close to that obtained optimally. This clearly shows that computation time can be saved largely by good initial guesses of the control profile. For recycle case, Table 5.4 shows that for both cuts the initial reflux profiles were close to those obtained optimally. Thus these minimized the number of function and gradient evaluation. In all cases the time spent in physical properties was in the order of 30% of the total CPU time.

Table 5.4 Solution Statistics of Example 2 (recycle case)

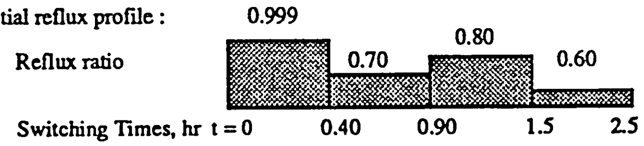
CUT 1

No. of Function Evaluations = 10
No. of Gradient Evaluations = 7

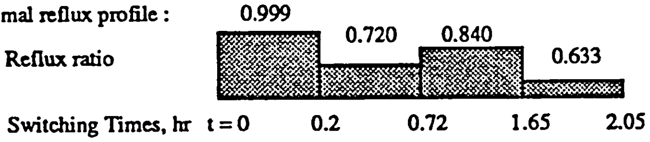
Time for each Function Evaluation (approx) = 0.6 min
Time for each Gradient Evaluation = 0.9 min

Total CPU time (including translator and optimiser) = 13 min
% of total time spent in physical properties = 28

Initial reflux profile :



Optimal reflux profile :



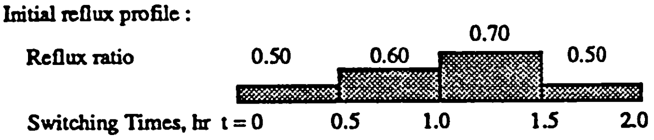
CUT 2

No. of Function Evaluations = 8
No. of Gradient Evaluations = 6

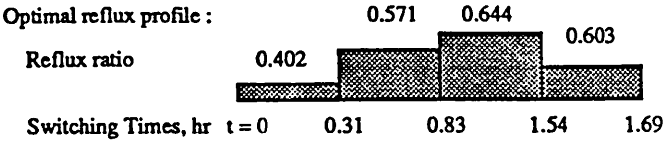
Time for each Function Evaluation (approx) = 0.5 min
Time for each Gradient Evaluation = 0.8 min

Total CPU time (including translator and optimiser) = 10.5 min
% of total time spent in physical properties = 30

Initial reflux profile :



Optimal reflux profile :



5.7 Conclusions

A general optimal control formulation is considered for establishing optimal off-cut recycle policies in multicomponent batch distillation. Several difficulties in solving such problem are discussed. A sequential strategy for off-cut recycle is proposed to overcome the disadvantages of recycling all the off-cuts at the beginning of a new batch. The suggested strategy should increase the total amount of throughput produced in a quasi-steady state operation mode and might reduce the possibility of excessive energy requirement at the beginning of the batch because of recycle of heavy off-cuts. However, the actual benefit of using this strategy could only be realised if the increase in productivity outweigh the capital costs required for storing the off-cuts in separate storage tanks.

Special cases are mentioned and identified with examples, where a quasi-steady-state mode of operation is established and where the optimal control technique used for binary mixture can be applied to multicomponent mixtures. The measure **q** of "the degree of difficulty" of separation is again found to be useful in identifying these situations where the present method can be easily applied.

The example used in this chapter shows that for **q** up to about 0.6, the assumption of sharp separation was quite valid and the present method could be safely used. Benefits of recycling for some cases were correlated against **q**. It was found that in some cases recyclying of off-cuts reduced the batch time by 45% of that required to obtain the same separation without recycle. Also the use of recycle of off-cuts made it possible in some cases to achieve a separation which was not possible within a finite time without recycle.

Chapter 6

THE ROLE OF COLUMN HOLDUP ON THE PERFORMANCE OF BATCH DISTILLATION

6.1 Introduction

The role of column holdup has been the subject of some controversy in the past and and its effect on the performance of batch distillation is not very clear. This chapter examines the role of column holdup for binary mixtures in a systematic way. It is shown that increasing holdup in some cases improves column performance and that the effect is in other cases detrimental. Both effects are explained on physical grounds and correlated in terms of a measure q of "the degree of difficulty" of the separation. This allows identifying an optimum holdup required to achieve the best performance of a column, for a given separation.

Unless there is an upset to a steady state continuous distillation operation which results in a dynamic operation, one of the major differences between batch and continuous distillation concerns the effect of liquid holdup on the system, both in the column itself and around the condenser system (Rose,1985). For a steady state continuous distillation with the assumptions of well mixed plates, the holdup has no role in the analysis (modelling of such columns does not typically include column holdup) and any quantity of liquid holdup in the system has no effect on the mass flows in the system (of course this is not completely correct, for example, if pressure drops are to be calculated from a hydraulic model). But since batch distillation is inherently an unsteady state process the liquid holdups in the system become sinks (accumulators) of material which affect the rate of change of compositions in the system and thereby affects the mass flows of the system. Therefore, modelling of batch distillation ignoring column holdup is always risky and may in some cases lead to significant error (Robinson, 1970).

Rose et al. (1950,1952) studied the effect of holdup for binary and ternary mixtures in a laboratory batch column. They defined "sharpness of separation" as the sharpness in the break (measured in mole fractions) between successive components in the graph of instantaneous distillate composition against percentage distilled. They showed that an increase in column holdup increased the sharpness of separation between components at low reflux ratio but did not have any effect at a very high reflux ratio. However, their work suffers from the limitations that they did not mention how this effect is related to the difficulty of separation, mixture properties and column configuration. Also there was no mention of how small a reflux ratio and how large a holdup one should consider for a given separation in a given column to get a significant benefit from the process.

The opposite conclusions were arrived at by Converse and Huber (1965) who studied the effect of holdup on batch distillation optimisation by including column holdup in their model. They maximized the amount of distillate of a given purity and found that in all cases, column holdup causes a decrease of the amount of maximum distillate obtained for a fixed time of operation. In another way, for a fixed amount of distillate and fixed purity higher column holdup increases the batch time. The authors did not discuss the reasons for this type of behaviour as well but concluded that the holdup was bad anyway.

Mayur and Jackson (1971) simulated the effect of holdup in a three plate column for a binary mixture, having about 13% of initial charge distributed as plate holdup with no condenser holdup. They considered a time optimal control problem for a given separation, and found that for both constant reflux and optimal reflux operation, the batch time was about 15-20% higher for the holdup case compared to the no holdup case.

Rose (1985) also drew similar conclusion about column holdup but mentioned that the adverse effects of column holdup depends entirely on the system, on the performance required (amount of product, purity), and on the amount of holdup.

These findings are therefore clearly in contradiction and the role of column holdup is not properly understood neither qualitatively nor quantitatively. Hence a systematic study of the effects of holdup is necessary to correlate the unexplained facts observed in the past.

Here the performance of a column is defined in terms of minimum batch time and the difficulty to achieve a given separation is defined in terms of the measure **q**. The effects of column holdup are then correlated in terms of **q** and of the minimum batch time required to produce a given separation.

6.2 Perfomance of the Column in terms of Minimum Batch Time

Before examining the effect of holdup on the performance of the column it is necessary to define a measure for the performance. These measures could be in terms of maximum profit, maximum product or minimum time. In the present investigation it was found that the minimum time measure was most suitable because in this case the separation requirements (e.g.product purities) are fixed and the measure, **q**, of "the degree of difficulty" of separation can be easily used. The minimum time problem was defined in detail in chapter 3.

6.3 Evaluation of the Effect of Column Holdup

Having defined the separation requirements, **q** gives an estimate of "the degree of difficulty" of the separation. Defining minimum time as the performance of the column, a series of time optimal control problem defined in section 3.3.1 of chapter 3 is solved with different values of column holdup (as a percentage of the total feed to the column) and the effects are examined. For simplicity binary mixtures are considered and a single constant reflux ratio is used throughout, with its level optimised to give minimum batch time for each case. The optimal control problem is solved using the procedure outlined in chapter 3. For a particular binary mixture the procedure is repeated for different values of **q**. The present work considers three (3) typical binary mixtures for the purpose. From the results it is shown that a correlation exists between the column holdup and "the degree of difficulty" of separation. This determines a value of the column holdup for a given separation which gives the best performance.

6.4 Example Problems

The following investigations are carried out so as to set more clear evidence on the role of column holdup:

1. Effect of increasing plate holdup on the column performance at a fixed condenser holdup.
2. Effect of increasing condenser holdup on the column performance.

We consider three binary mixtures for this purpose The mixtures are: 1. Benzene-Toluene, 2. Cyclohexane-Toluene and 3. Butane-Pentane.

6.4.1 Problem description

Column configuration and separation requirements (x^*_D) for several cases (for investigation 1) are presented in Table 6.1. For all cases the initial charge to the column is 5 kmols (B_0) with a lighter component mole fraction of 0.6 (x_{B0}) and in all cases the first component is the lighter one. The amount of distillate product required (D^*) is set to 3 kmol and a condenser vapor load of 3 kmols/hr is used for all cases.

For investigation 1, the condenser holdup was fixed to 2% of the total initial charge and the column holdup is varied as a percentage of the total initial charge to the column.

Investigation 2 is carried out only for binary mixture 1 with a **q** value of 0.332. The role of condenser holdup is examined for three different levels of condenser holdup values, 0%, 2% and 5% of the total initial charge and the plate holdup is varied in all cases as a percentage of the total initial charge to the column. There are a total of 8 internal plates with a separation required of 3 kmols (D^*) benzene at 90% purity (x^*_D). The condenser vapor load and the initial charge composition are the same as in investigation 1.

Liquid compositions of trays, condenser holdup tank and accumulator (differential variables) at time t=0 are set equal to the fresh charge composition (x_{B0}) to the reboiler. The differential and algebraic equation system is solved at time t=0 to initialize all other variables. For binary mixture 1 and 3 we use a simple Antoine's vapor pressure equation to calculate the vapor liquid equilibria. The SRK equation of states are used for binary mixture 2. Enthalpies are calculated using the same procedure mentioned previously.

In all cases the optimal control problems are solved within tolerance ε_1= 0.0001 and ε_2=0.001 (see chapter 3 for the definition).

6.4.2 Results and Discussions

6.4.2.1 Investigation 1: A series of optimal control problems were solved at different value of **q** with increasing holdup for each case. Figure 6.1 shows the minimum time solution vs percent total holdup of the column for different binaries at different **q**, and Figure 6.2 shows the corresponding optimum reflux ratio (required to get the separation in minimum time) vs percent total holdup of the column. Results are summarized in Table 6.1 which shows for each given separation the optimum value of holdup to achieve the best performance out of the given column. The corresponding best minimum batch time and the optimum reflux ratio to achieve this are also presented in the Table for each case studied.

Figure 6.1a Effect of Column Holdup at Different q

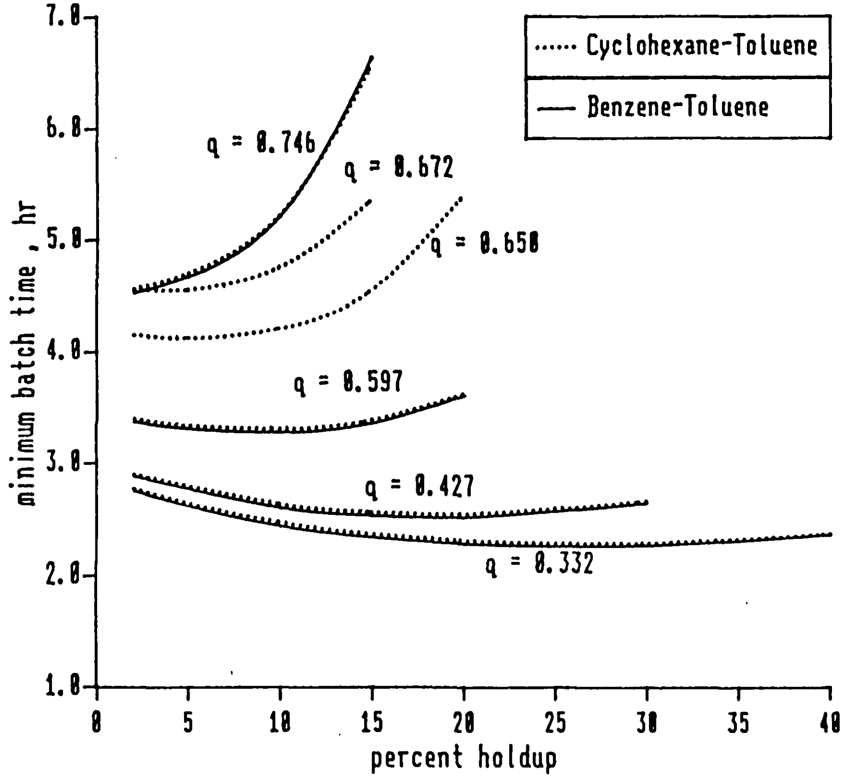


Figure 6.1b Effect of Column Holdup at Different q

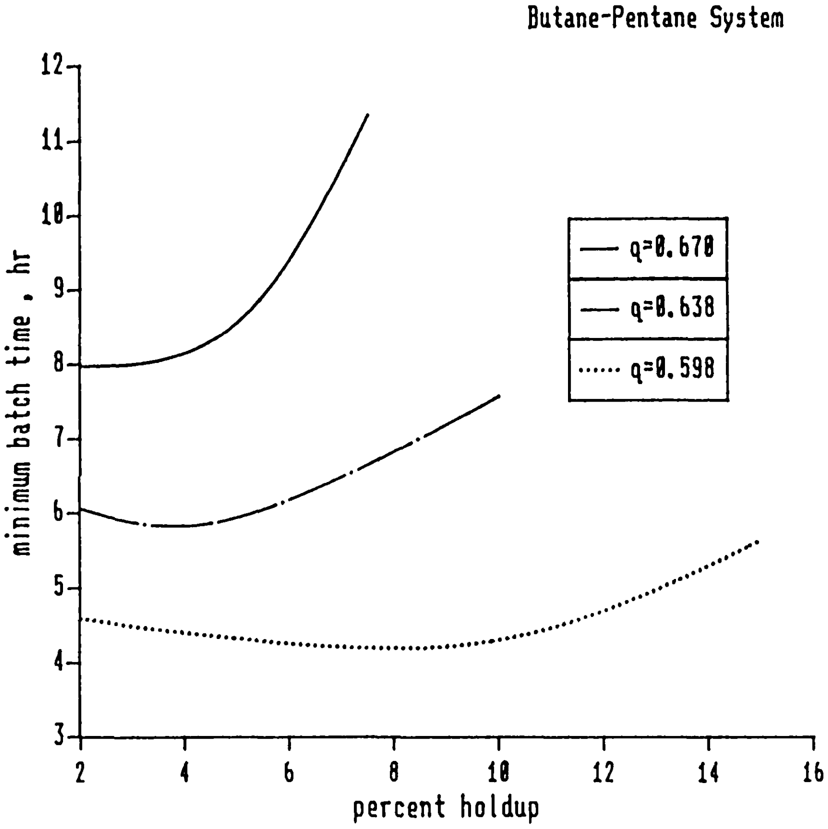


Figure 6.2a Reflux Ratio vs Column Holdup at Different q

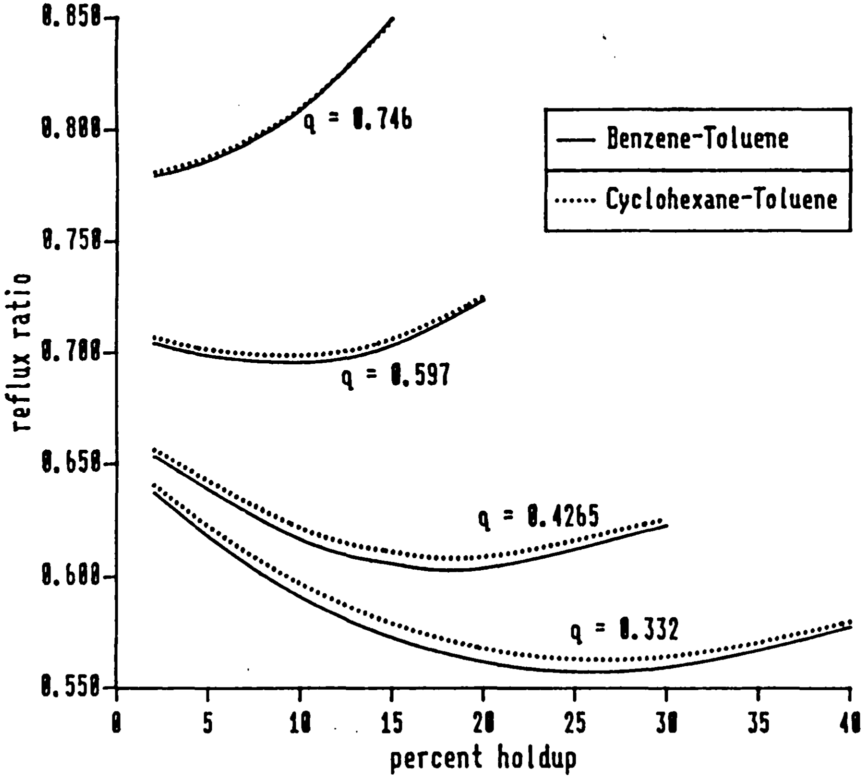


Figure 6.2b Reflux Ratio vs Column Holdup at Different q

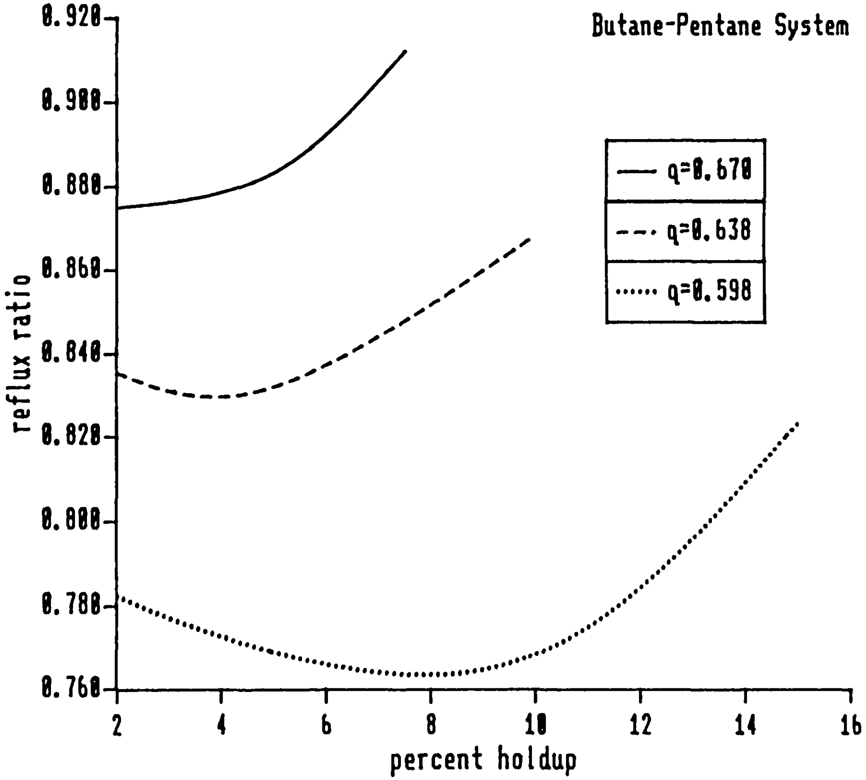


Table 6.1 Input Data and Results of Investigation 1

Case	Binary Mixture	N	X	q	Optimum Holdup %	Optimum Reflux R.	T1 hr	T2 hr	% Time Saved
1	1	8	0.900	0.332	26.0	0.557	2.69	2.26	15.98
2	1	6	0.900	0.427	18.0	0.601	2.83	2.51	11.30
3	1	4	0.900	0.597	10.0	0.695	3.38	3.28	2.95
4	1	3	0.900	0.746	2.0	0.779	4.52	4.52	-----
5	2	8	0.895	0.332	28.0	0.563	2.72	2.29	15.80
6	2	6	0.895	0.426	19.0	0.610	2.89	2.56	11.40
7	2	4	0.895	0.597	10.0	0.699	3.42	3.32	2.92
8	2	4	0.916	0.672	2.0	0.781	4.56	4.56	-----
9	2	3	0.895	0.746	2.0	0.782	4.56	4.56	-----
10	3	6	0.963	0.428	15.0	0.638	3.58	2.75	23.18
11	3	4	0.963	0.598	8.0	0.764	4.52	4.24	6.19
12	3	4	0.970	0.638	4.0	0.828	6.11	5.81	4.90
13	3	4	0.975	0.670	2.0	0.875	7.97	7.97	-----

Key: N_T = no of trays

T1 = minimum batch time for the separation using lowest plate holdup (zero %)

T2 = minimum batch time for the separation using optimal plate holdup

% time Saved = (T1 - T2) x 100 / T1

Percent reduction in batch time when using an optimum plate holdup compared to the batch time using zero plate holdup are shown as percent time saved in Table 6.1. This clearly shows the effect of using optimum column holdup. The effect of using any other plate holdup (not the optimum) are also clear from Figures 6.1-2.

It is to be noted that **q** for a particular mixture and system measures how difficult the given separation is in the given column. But it does not guarantee that the batch times should be identical with that of other mixture for which **q** values are same. Comparison of Figure 6.1-2 shows that at **q** value of about 0.598, although optimum values of holdup for all the binary mixtures are about 10%, the batch times are different for different binary mixtures.

The liquid composition (mole fraction) of Plate 1 (top), accumulated distillate composition and reboiler composition profiles for case 3 are presented in Figures 6.3-5. Figure 6.6 presents plate 1 liquid composition profile for case 4. These are presented to show in more detail the role of increasing column holdup. This is explained in the following:

The measure "degree of difficulty" of a given separation does not only refer to how difficult the separation is for the given column but is also a measure of the extent a given column is oversized for the purpose. A **q** value of 0.3-0.4 means 30-40% of the trays available in the column are the required minimum for the given separation and therefore 60-70% of trays are in excess.

Since material holdup on the trays acts as an accumulator, larger holdup on the top trays means a larger amount of the light component is distributed on the trays at low **q** as the process moves. And since larger holdup on the trays means a slower rate of change of composition both before and after the composition reaches its maximum value, it is possible to operate the column at lower reflux with large holdup to get the specified product in a shorter time. Figure 6.3 clearly shows this and also shows that with a 10% column holdup it is possible to obtain the required separation in the shortest time. This can be seen more clearly in Figure 6.4. Although the final product compositions for different holdup cases are identical, the optimal batch time for the 10% holdup case is the shortest.

Figure 6.3 Plate 1 Liquid Composition at Different Column Holdup

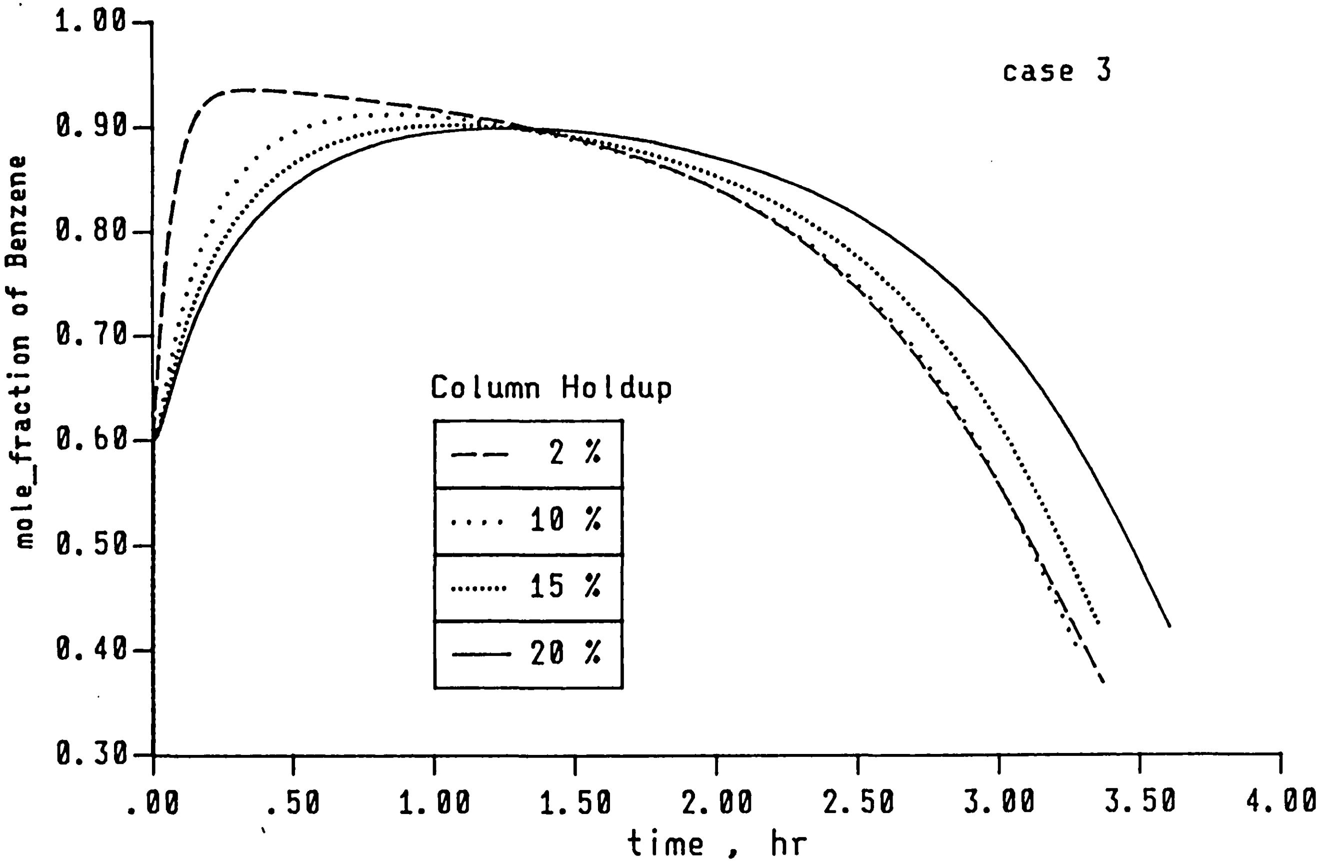


Figure 6.4 Accumulated Distillate Composition at Different Column Holdup

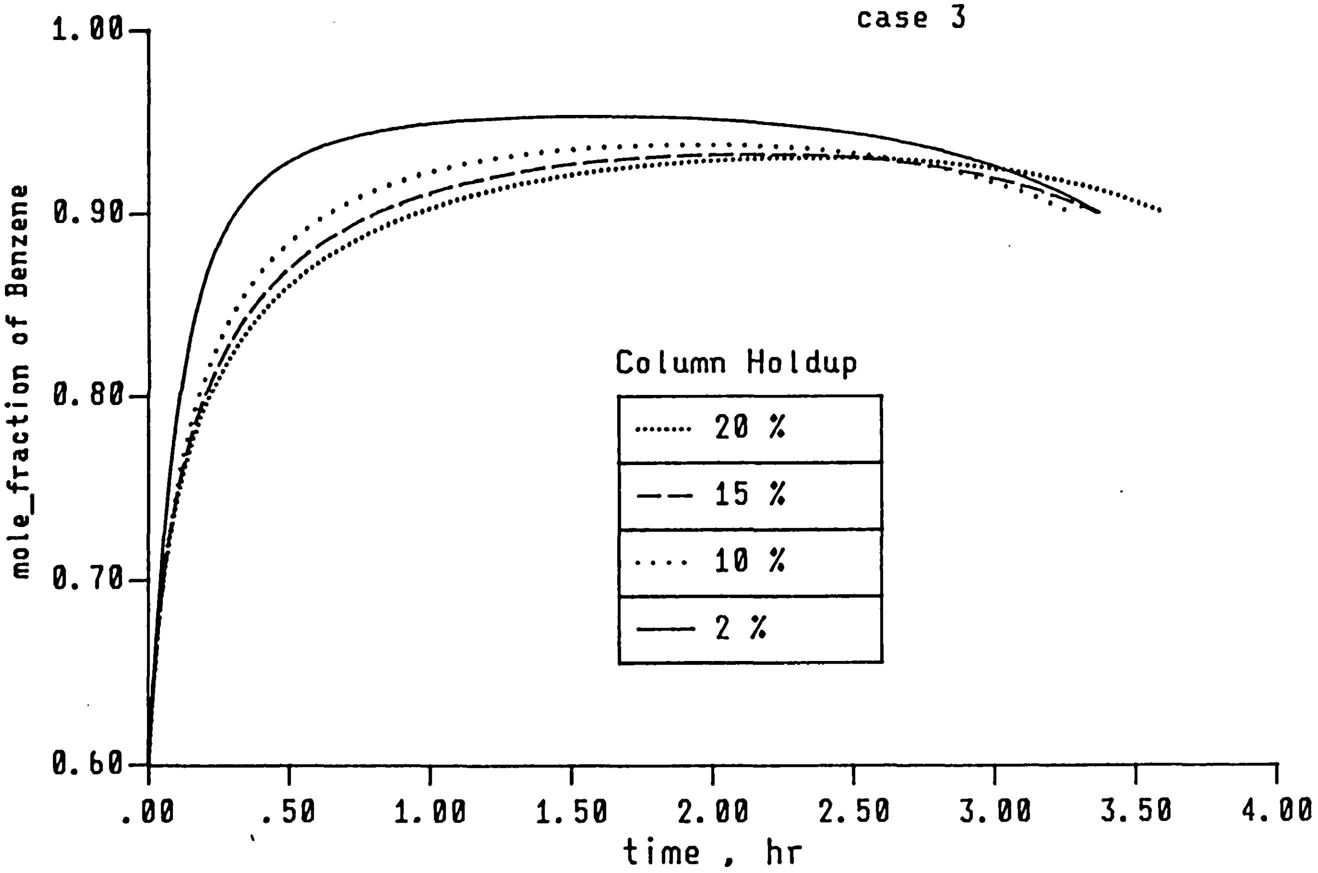


Figure 6.5 Reboiler Composition at Different Column Holdup

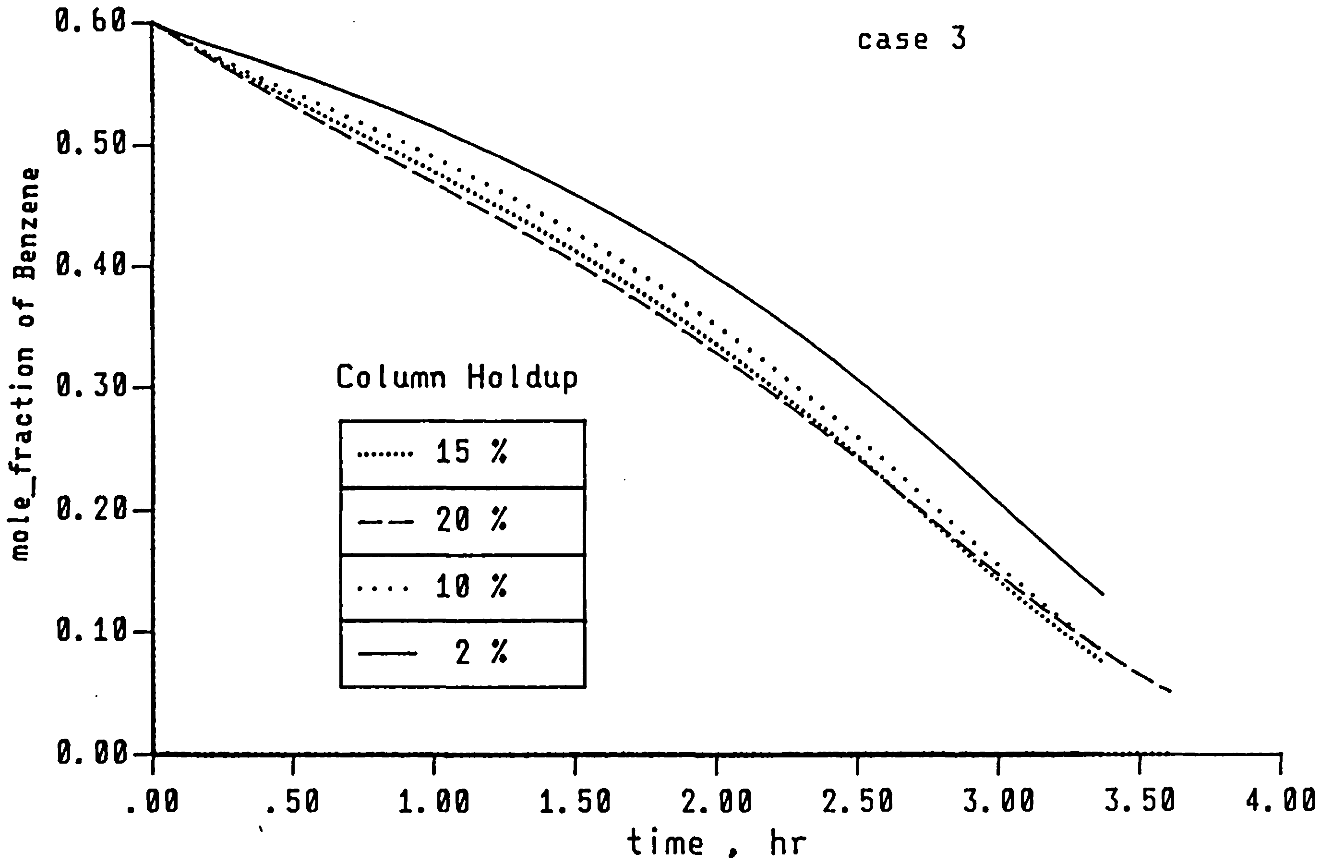
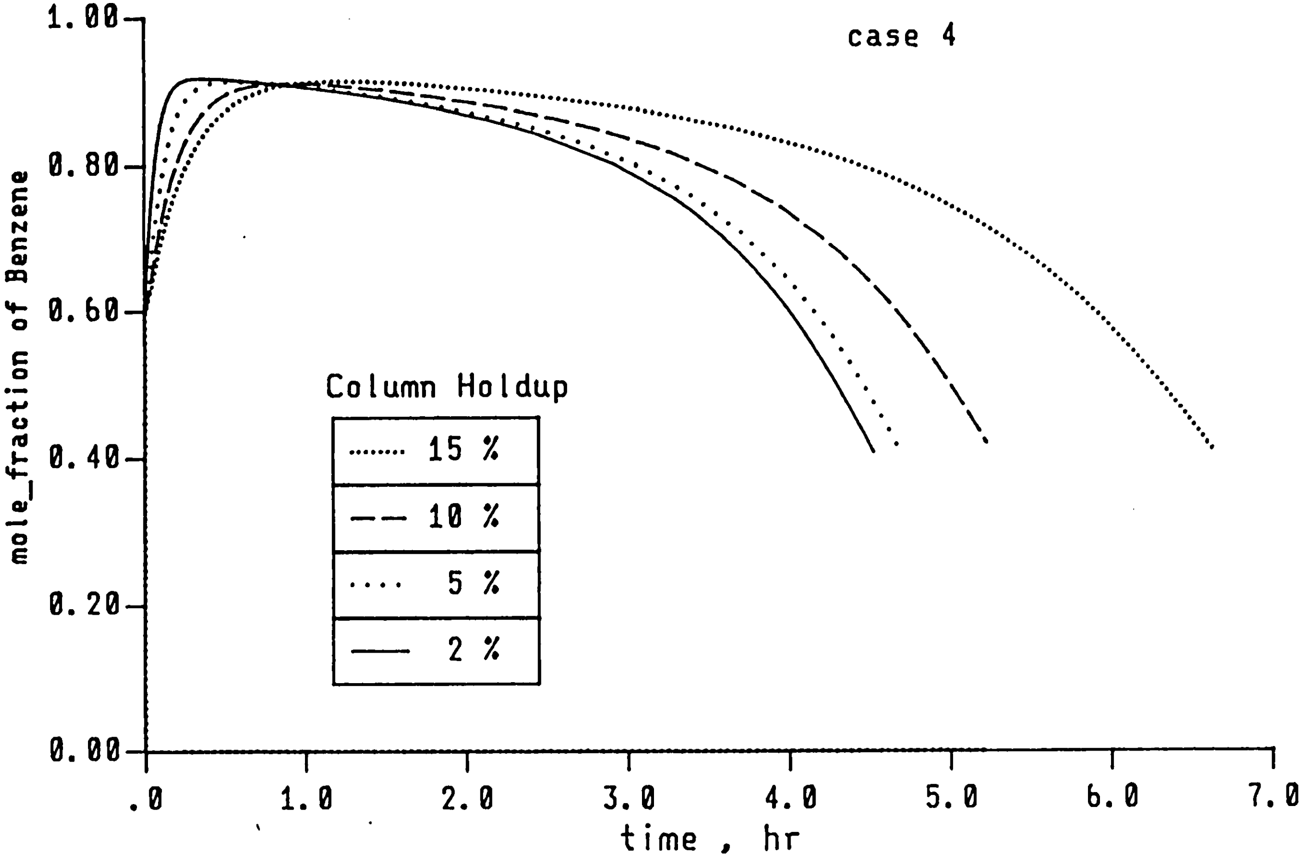


Figure 6.6 Plate 1 Liquid Composition at Different Column Holdup



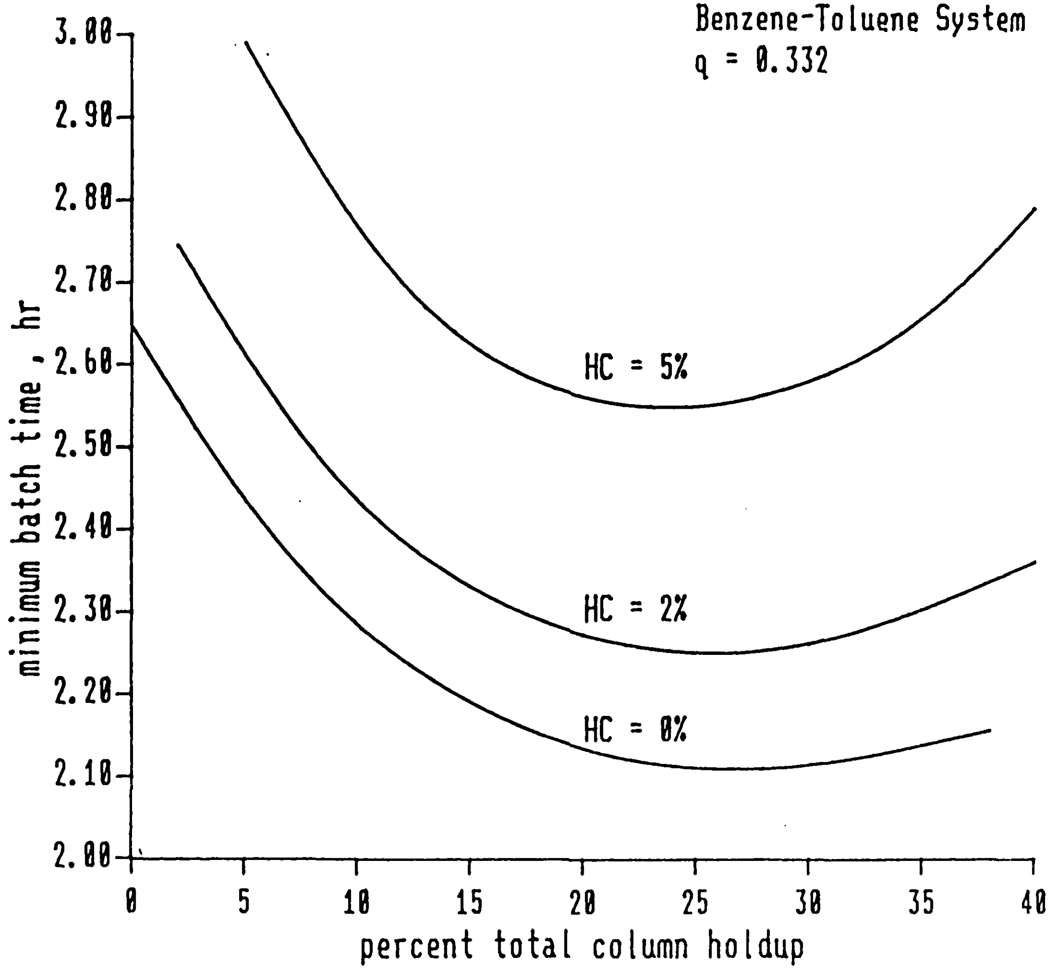
For a given separation further decrease of batch time (by operating at lower reflux values) can be obtained by increasing plate holdups until such a stage where any further holdup increases result in the reboiler being quickly depleted of the light component as the distillation progresses at which point the heavy component then begins to rise up the column. At this value of holdup and beyond it low reflux operation is no longer possible. So a high reflux is needed to maintain the product purity which consequently means longer batch time. Figure 6.5 clearly explains the above fact. For the 20% holdup case the column has to operate at a quite high reflux ratio to maintain the product quality because of the significant drop in light component in the reboiler.

As the value of **q** increases (more difficult separation) the benefit of having holdup in the column decreases. This is because now the number of trays in excess of the required minimum decreases and the column should run at high reflux ratio to achieve the given separation. The effect of increasing holdup will be reversed now. Since increasing column holdup means slow rate of change of composition profile any amount of holdup on the top trays is now critical. The reason is, unless the column is run at very high reflux ratio more and more unrectified product will be withdrawn from the process and it would never be possible to achieve the given separation. For the cases studied it has been found that beyond **q**=0.67 increasing column holdup above zero is no longer beneficial. Figure 6.6 shows that at a value of **q**=0.746 batch time increases with any amount of holdup in the column.

Going back to the cases reported in the literature, the authors who found that holdup was bad for their column were operating in most cases at values of **q** greater than 0.70. Although we could not evaluate **q** for the cases reported by Rose et al. (1950, 1952) cases (product quantity and purity were not reported), we suspect that in the regions where they found holdup to be beneficial **q** would have much lower values.

6.4.2.2 Investigation 2 : Figure 6.7 shows the effect of condenser holdup on the performance of the column. Since for total condenser, holdup only plays as an accumulator of material but not as a separation stage, larger condenser holdup means longer batch time to achieve a given separation. This is quite clear from Figure 6.7. In practice there must always be

Figure 6.7 Effect of Condenser holdup (HC) on the Column Performance



a certain amount of condenser holdup, to insure a neat reflux operation, however this should be kept to a minimum.

6.5 Conclusions

The investigations carried out in this chapter clearly show how important a role the column holdup plays on the performance of the column. In practice there is always some holdup on the plates and in the condenser system (how much is involved depends on the design of the column). Therefore any analysis based on reduced order model which excludes column holdup is liable to significant errors. Of course, deviation and the extent of prediction error depend on how difficult the given separation is for a particular column.

Although the investigation presented in this chapter identifies an optimum amount of holdup (as a percentage of the total initial charge), in practice, the size of the reboiler and the amount of holdup is fixed for an existing column. Since it is always desirable to charge the reboiler to its full capacity, the holdup and the charge ratio might not be optimal for the task at hand. Also it may happen that the application of the minimum time problem to this task in the existing column may be detrimental in terms of very high processing time. However, in such situations, application of the maximum profit problem (presented in chapter 3) will forecast the optimal amount of product (at the given purity) that should be obtained from the given mixture to achieve maximum benefit out of the given column.

Nevertheless, the investigations presented in this chapter show that when the behaviour is known for a wide range of separations and mixtures, it is possible to use these information to design a batch distillation column, so that maximum benefit could be achieved out of the column.

Chapter 7

CONCLUSIONS AND FUTURE WORK

7.1 Conclusions

In this thesis optimal operational policies for batch distillation were determined using a fairly detailed and realistic model and using suitable optimal control and optimisation algorithms. The model was a mixed system of differential and algebraic equations (DAEs) of index 1 and was detailed enough to include energy balance, rigorous thermodynamics, column holdups etc. At the same time it was short enough to ease the computational burden by not including extra equations like plate hydraulics, density equations. The model was checked against several simulation and experimental data from the literature and was found to be reasonably good for use in optimal control studies.

A general purpose batch distillation code, BATCHOP, was developed with several configurational and operational options. It generates automatically the column configuration and all the necessary model equations from user supplied data. The present version of BATCHOP includes options for operating either with constant condenser vapor load or with constant reboiler duty. Other features of the code are that it provides analytical derivatives of each model equation with respect to all differential, algebraic, design and control variables which are required to solve the model. The code has the characteristics required for inclusion of integration, physical properties and optimal control packages.

In this research the code DAEINT was used for integrating the model equations. The code is based on Gear's BDF method using a variable step size and variable order method as implemented by Morison (1984). It was chosen because the Gear's method is currently considered to be the best method for solving systems arising from chemical engineering processes,which are often stiff.

The problem of finding optimal reflux ratio policies for single batch distillation operations was formulated as an optimal control problem. Optimal policies were obtained for a variety of objective functions and constraints using the formulation and solution algorithms for DAE sytems of Morison (1984) as implemented in the VOPCON code. The code was chosen because of its robustness in the solution of various batch distillation operation problems. For optimising the decision variables arising from the optimal control formulation, standard SQP methods were used.

Optimal reflux ratio policies were obtained for a number of optimal control problems found in the literature, using different types of mixtures (both ideal and nonideal) and using different column configurations and operational modes (constant vapor load or constant reboiler duty). The results obtained by using optimal reflux ratio policies in all cases were compared with those obtained by using a conventional constant reflux ratio policy. In all cases it was found that the optimal policies improved the column performances, sometimes by a very great extent.

For a given fresh feed, a desired separation and a given column, the benefits of recycling of intermediate off-cuts were demonstrated in terms of a reduction in batch time. A new one level optimal control formulation was proposed for finding an optimal recycle policy in binary batch distillation. The new method is more robust and computationally much faster than the classical two level optimal control formulation. The recycle policies included optimal amount, composition and timing of the off-cut and the optimal reflux ratio values during the main-cut and off-cut operations. The policies were again obtained using the optimal control algorithm of Morison and were presented against the measure (**q**) of "the degree of difficulty" of separation. For two typical ideal mixtures and a variety of specifications on the separation, it was found that for **q** values greater than about 0.6 the recycling was beneficial over nonrecycling cases. Benefits of recycling off-cut were also found for approximately the same value of **q** for one nonideal mixture.

For a binary mixture and based on a fixed reboiler charge, suitable scaling of the optimal recycle results obtained based on fixed fresh feed showed that recycling of off-cut improved the productivity to a large extent. The productivity was increased with an increasing value of **q**.

A general optimal control formulation was discussed for calculating optimal off-cut recycle policies for multicomponent batch separation problems and the possible difficulties for solving such problems were outlined. Some special cases were identified where the methods developed for binary mixtures could be applied fairly easily to obtain the recycle policy choice for multicomponent mixtures. The previously mentioned measure **q** of "the degree of difficulty" of separation was used to identify those special cases. Application of the method to a variety of separation specifications for multicomponent mixtures revealed the benefit of recycling over nonrecycling cases. The benefits were again correlated with the same measure **q**. A new operational strategy was proposed for off-cut recycle in multicomponent batch distillation which is physically and thermodynamically sensible.

In most optimal control studies of batch distillaion in the past, the column was initialized at total reflux steady state values. In most other cases the column was initialized at the fresh feed composition. In all cases it was not quite clear when and how long a column should run under total reflux condition to obtain best performances. In this thesis we initialized the column at the composition of the reboiler charge throughout. However, by application of the optimal control approach the solution algorithm automatically determines when and how long the column should run under total reflux operation to obtain the best operational policies.

The role of column holdup was systematically explored and was explained in relation to the measure **q**. Using typical binary mixtures and considering a time optimal control problem it was shown when the column holdup was detrimental and when it was not. It was also shown that it is possible to improve column performance to a significant extent by using an optimal amount of column holdup. Also, detailed simulation studies shows that holdup, however small, should not be neglected in the model.

7.2 Future Work

The use of distillation in which chemical reactions take place is fairly common in the chemical industries. This is because removal of one of the reaction products by distillation increases the overall conversion. Therefore, there is a large incentive for operational improvements of such columns. Although chemical reaction was considered in the present work its application was restricted only to simulation studies. Nevertheless, inclusion of the option of chemical reaction in BATCHOP is fairly easy involving only minor changes to the model and translator program, but no conceptual difficulty. This should be included in the future. The simulation study with chemical reaction as presented in Chapter 2 shows that there is a wide scope of optimal control application to increase the overall conversion of the product in this type of applications.

Also further work on the batch distillation code should involve a constant volume holdup option for components with wide liquid density range and an option of constant boilup rate operation of the column.

The benefits of off-cut recycle were realised throughout this thesis and in the past in terms of a potential reduction in batch time. But other objective functions including cost functions which accounts for the cost of the products, of storage vessels and of operation are possible alternatives which should be considered in the future so that the benefits could be realised in terms of a more direct economic measure. Productivity could also be an alternative objective function.

Implementation of the optimal control algorithm of Morison for sequential processes is most desirable in the future to develop optimal off-cut recycle policies in multicomponent batch distillation with sequential strategy of off-cut recycle. Also numerical developments are required to tackle the quasi-steady state operation constraints of the off-cuts for general multicomponent mixtures.

Development of optimal off-cut recycle policies based on a fixed reboiler charge is also most desirable in the future. This is to find an optimal amount of fresh feed and the composition of the recycled off-cut on a fixed reboiler capacity basis so that productivity or profit is increased because of recycle policy. To do this some modification of the VOPCON code will be required. This is because, in case of recycle, B_C (the amount of mixed charge to the reboiler) is the differential variable in the model equations and can be easily optimised using the present version of VOPCON (as used for one level control problem in binary recycle in chapter 4). On the other hand B_0, the amount of fresh feed, which is only charged to the reboiler at time $t = 0$, is not a time dependent variable in the model equations and therefore can not be optimised directly using the present version of VOPCON. However, to find optimal off-cut composition, the mixed reboiler composition can be optimized (as was done in this work) and the off-cut composition can be obtained from the material balance.

Before using the measure **q** of "the degree of difficulty" of separation as a qualitaitve decision variable in general, to help establish whether recycling is beneficial or not, more ideal, nonideal binary and multicomponent mixtures should be used to determine more accurately the range of this measure when the recycling is beneficial. Also the measure **q** is based on using short-cut methods for continuous distillation which use average constant relative volatilities in the column. Since for a steady state column the composition does not change with time, average relative volatilities based on the relative volatilities at the top and the bottom of the column are typically fairly representative. But in batch distillation composition always changes with time. Therefore average relative volatilities calculated based on a constant distillate composition and varying reboiler composition may not be truely representative. Therefore more accurate measure of **q** is desirable specially for nonideal mixtures.

Whether column holdup is good or bad was only determined for binary mixtures and for one specific type of operation (time optimal operation). To draw more general conclusions future work could involve investigations with other types of operation (maximum distillate, maximum profit etc.) and with multicomponent mixtures as well. In fact, the addition of a simple hydraulic model could be fairly easily accomplished as well. Whether more sophisticated hydraulic models are needed, it is not clear at this point.

Future work should also involve experimental verification of the optimal results obtained in this work using the optimal control profiles in on-line optimisation.

To improve the efficiency of the methods better integrators and optimisers should be looked into more in detail in the future. Also for multicomponent multicut operation, parallel machines could be used for each cut and an arrangement could be made to converge all the cuts in one go.

Work to determine optimal operational policies for unconventional columns can also be recommended in the future.

NOMENCLATURE

B_0, x_{B0}	initial amount (kmol) and composition (mole fraction) of the reboiler charge
B_1, B_2, B_3,....	amount of bottom product in different cuts, kmol
$\underline{B}_2$	calculated amount of the bottom product
B, x^i_B	amount and composition of the bottom product
B_C, x_{BC}	mixed reboiler charge (kmol) and its composition (mole fraction)
C_1, C_2, C_3	cost functions
D_1, D_2, D_3,....	amount of top product in different cuts, kmol
$\underline{D}_1$	calculated amount of the top product
D, x^i_D	amount and composition of the top product
D^*, x^*_D	specified amount and composition of the top product
H	condenser, reboiler or plate molar holdup
H_a	accumulator molar holdup
L, V	liquid or vapor molar flowrate
L_d	molar distillate rate
N_{min}	minimum no. of plates
N_T	actual no. of plates
Q_C, Q_R	condenser or reboiler duty KJ/hr
R_1, R_2, R_2,..	amount of recycle in different cuts, kmo
T, P	temperature (K) and pressure (bar)
T_S	start-up and shut down time, hr
V_C	condenser vapor load, kmol/hr

f_{x_1}	derivatives w.r.to x_1
f_{x_2}	derivatives w.r.to x_2
h^L, h^V	liquid or vapor enthalpy KJ/kmol
k	equilibrium constant
k^r	reaction rate constant (hr^{-1})

k^r_1, k^r_2	forward and backward rate constant, 1/hr
q	degree of difficulty of separation
r	reflux ratio (internal)
r	reaction rate (hr^{-1})
r_A	rate of formation of component A, 1/hr
t^1, t^2, t^3	cut time
t	time, hr
t_P, t_F	intermediate or final time
t^*_F	fixed final time
t_{nr}	batch time without recycle
t_r	batch time with recycle
$x_1(t)$	derivatives of differential variables
$x_1(t)$	differential variables
x_{10}	initial conditions of differential variables
$x_2(t)$	algebraic variables
x(t)	set of differential and algebraic variables
x, y	liquid or vapor composition, mole fraction
x	array of liquid composition, mole fraction
x_a, x_d	accumulator or distillate composition, mole fraction
x_A, x_B, x_C, x_D	liquid composition (mole fraction) of component A, B, C and D
$x_{B1}, x_{B2}, x_{B3}, \ldots$	composition of bottom product in different cuts, mole fraction
$x_{D1}, x_{D2}, x_{D3}, \ldots$	composition of top product in different cuts, mole fraction
$x_{R1}, x_{R2}, x_{R3}, \ldots$	composition of recycle in different cuts, mole fraction
u(t), v	control and design variables
z	control parameters

superscript and subscript

i	component number (1, n_c)
j	stage number (1, N)

greek

α	relative volatilites
$\varepsilon_1, \varepsilon_2$	small positive numbers

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APPENDIX

A. Description of BATCHOP Programme

The fortran code BATCHOP developed in this thesis has the following structure:

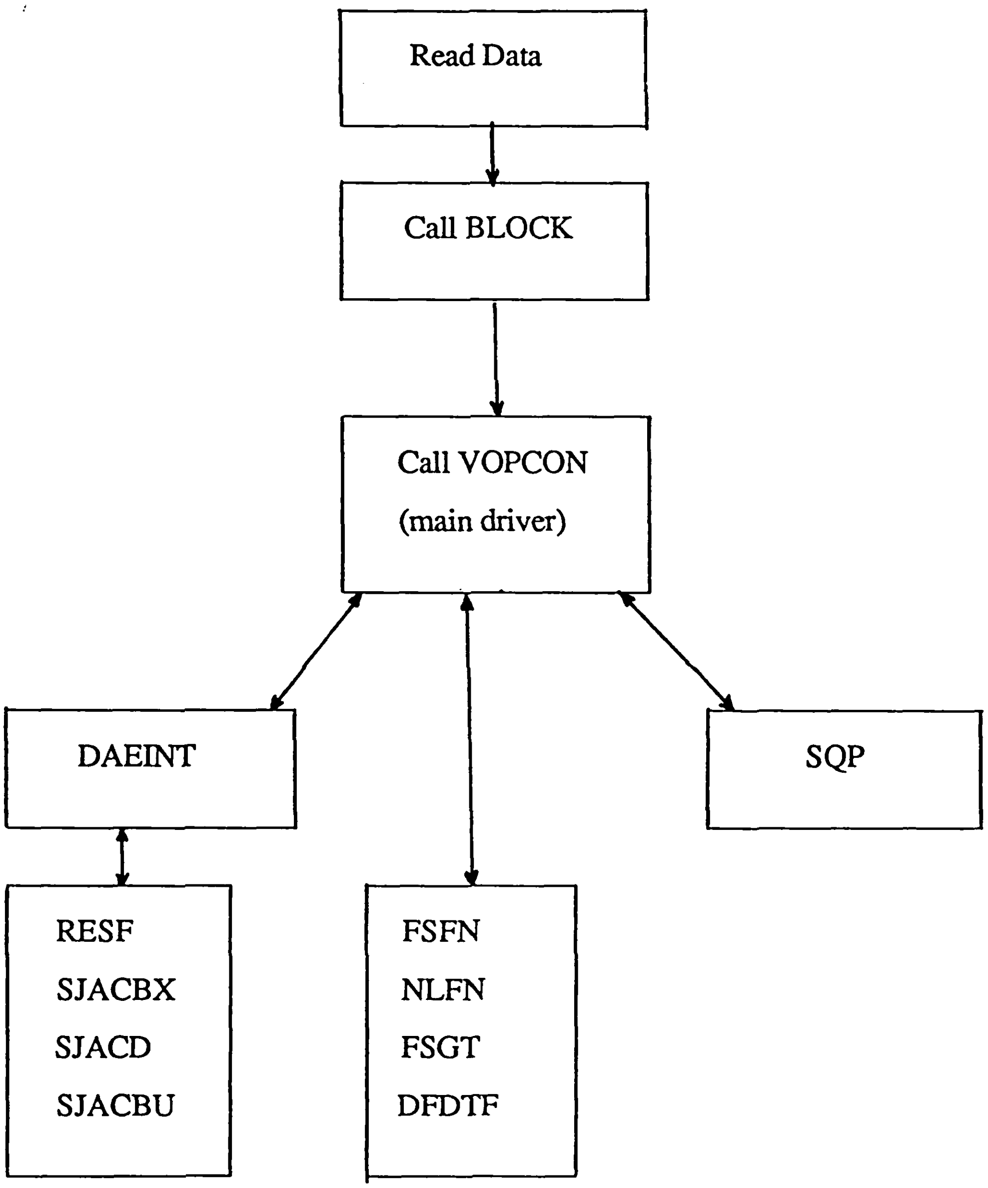

Figure A.1 Programme structure of BATCHOP

Description of the items mentioned in the above structure:-

User supplied INPUT DATA

User has to:

1. Define column configuration (no. of plates including reboiler and condenser).
2. Set OPTIONS:
   a) with or without condenser holdup tank.
   b) constant reboiler duty or constant condenser vapor load operation.
   c) ideal or nonideal physical properties.
3. Set accuracies and print levels for integrator (DAEINT), optimal control (VOPCON) and optimisation (SQP).
4. Set maximum no. of function evaluation for SQP.
5. Set initial values for differential variables.
6. Set initial guesses of algebraic variables.
7. Set design variables (plate holdups, pressure, etc.).
8 Set number of control intervals and give initial guess of control variable and switching times.
9. Set bounds on controls and on initial conditions of differential variables.

The main driver programme VOPCON calls DAEINT to integrate the DAE model equations and calls FSFN, NLFN, FSGT, DFDTF etc. to evaluate objective and constraint functions and their gradients w.r.t. decision variables. VOPCON finally calls SQP to minimize the objective function. SQP and DAEINT routines are repeatedly called by VOPCON until the solution is achieved.

Programme VOPCON : to provide gradients of objective and constraint function w.r. t. decision variables (control and its switching times etc.)

Programme DAEINT : for integration

Programme SQP : to minimize the objective function and to optimize the decision variables

Programme RESF : to generate model equations.

Programme SJACBX : to generate analytical derivatives of model equations w.r.t. differential and algebraic variables.

Programme SJACD : to generate analytical derivatives of model equations w.r. t. design variables.

Programme SJACBU : to generate analytical derivatives derivatives of model equations w.r.t. control variables.

Programme BLOCK : to generate positions of nonzero elements in sparse form for all derivatives (called only once for each new problem).

Programme FSFN : to define objective function

Programme NLFN : to define constraint functions

Programme FSGT : to provide gradients of objective and constraints functions w.r.t. differential and algebraic variables if they are explicitly dependent on them.

Programme DFDTF : to provide objective function gradient w.r.t. time if it appears explicitly in the objective function.