The Synthesis and Selection of Alternative Separation Flowsheets

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For Claire,

who didn't realise she was marrying a thesis...

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This thesis was composed by me and all the work described in it was done by me,

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Abstract

The work described here covers some aspects of generating or otherwise choosing good separation processes. First is a review of the literature dealing with the synthesis of both separation schemes and heat exchange networks. The latter is included because it illustrates the problems encountered and reveals that the most recent and effective methods for heat exchange network synthesis have no parallel in the separations literature. The next section deals with a method of using novel criteria for selectively generating flowsheets for general separation processes. The intention is that promising designs produced by this preliminary effort will be subjected to further, more stringent screening. The results of this exercise led to a consideration of the numbers of both separator types and separation units required to fulfill a given separation task. Manual methods are presented for determining the minimum values for each of these quantities. Examples from the literature reveal that the number of separation units in a flowsheet is not a good guide to the cost of a process, unlike the analogous measure in heat exchange networks. This discovery led to a comprehensive investigation of ternary distillation schemes. This work shows, perhaps unsurprisingly, that cost is closely correlated with energy use, but no simple evaluation function which can be used instead of cost was found. Analysis of results for 4- and 5-component distillation systems reveals that energy use in turn is governed largely by the quantity of material taken as overhead product. This observation leads to a simple screening method for distillation systems. The method is especially effective for systems where one component is in large excess and where that component is not the least or next to least volatile. Other features of distillation systems are also considered but without any immediate benefit. Costs of ternary distillation sequences generated in the course of this work are compared with the results of other workers' studies in an appendix.

Contents

1 Introduction

1 Introduction

Chemical plants, like many other complex artefacts, are often designed by a mixture of rule of thumb methods developed over the years and the incremental modification and improvement of existing designs. This type of approach is usually forced on the engineer because of the lack of precise design tools for networks of processing units compared with the relative abundance of design methods for the individual unit operations themselves. The discipline of process synthesis concerns attempts to understand and systematise the design of complex processing networks at the stages of both the initial flowsheet generation and the evaluation and comparison of alternative designs.

A chemical engineer's education generally provides him with tools for designing and optimising relatively small systems of continuous variables such as temperature, pressure or reflux ratio, and with an understanding of unit operations in isolation. He may also have gained some understanding of how unit operations interact when connected together. On the other hand, the generation of process flowsheets from scratch will usually require the manipulation of discrete variables, quantities reflecting the very presence or absence of items of plant. For an example, consider the choice of a reactor: for a particular duty an engineer may have to decide whether to use a tubular or a fluidised bed reactor, a discrete choice. Similar choices arise in deciding how to interconnect equipment, whether the reaction might be done in a single reactor or in several reactors in parallel or in series.

They may occur again when the choices to be made in one part of a plant interact with the design of another part. Consider the example of the choice of reactors given above. Each of the two reactor types might give a different spectrum of products which will have to be processed further for recycle, product finishing and so on. The separation equipment required in each case may thus be different, perhaps simply at the level of changes in the continuous design variables, but possibly to the extent of needing an entirely different type of separation system. The choice of reactor type therefore becomes part of a larger problem involving both discrete and continuous variables. Mathematical techniques for systems of discrete variables are readily available, but methods suitable for mixed continuous and discrete systems are still being researched.

The reason for the delay in developing suitable synthesis techniques for the engineer lies with the large number of possible designs to be considered, the so-called combinatorial problem, coupled with the sometimes heavy computation required in evaluating each one for comparison with its competitors. Thus the analysis and evaluation of designs is another important ingredient in the study of process synthesis, and not only the analysis of completed designs. Work on the synthesis of heat exchange networks has shown that analysis of the presented data prior to any attempt at process generation can be a vital component in the synthesis of good designs, since the information so derived can be used to shrink drastically the space of likely good solutions. This allows an optimal flowsheet to be found quickly since large numbers of flowsheets which would otherwise be needlessly evaluated can be excluded from consideration.

Process synthesis then is the study of the systematic generation and evaluation of process flowsheets, concerned with the discrete choices between different items of equipment and different schemes for their interconnection. It may also concern the analysis of problem data prior to design. Even though the synthesis of complete chemical processes has been attempted, most research has fallen into distinct fields in the attempt to decompose the task into manageable pieces. The fields tackled include reaction path synthesis, the first step in the definition of any new process; heat exchange network synthesis, including a small amount of work on general energy recovery; separation scheme synthesis, the subject of this work; and control scheme synthesis, the generation of optimal control schemes for processes.

One of the points arising from heat exchange network synthesis is an enhanced understanding of the way that networks, as opposed to single processing units, behave in design. For instance in a single heat exchanger the quantity of heat or cold recovered from a process stream can be maximised only at the expense of infinite heat exchange area. Conventional wisdom will apply the same argument to networks of heat exchangers, namely that the degree of heat recovery is dependent on the capital cost of the network. This has been found not to be so: networks which recover all the available heat or cold are usually cheapest since they require the minimum quantity of external heating or cooling service.

That this sort of understanding has yet to be developed for separation networks has largely determined the course of the work described here. It begins with an attempt to solve a very general separation problem and ends with a look at a very specific separation system, trying to understand that better. Chapter 3 presents a very general method of separation scheme synthesis based on an intuitive method for the evaluation of alternative flowsheets. The method allows candidate flowsheets to contain separation units with unknown properties, details of which are expected to be filled in by further research, either at the laboratory bench or among the archives. Chapter 4 deals with some targets which might be useful in the design of separation schemes using only a known set of candidate separator types. More particularly it provides simpler and more comprehensive methods for finding out some of the information given by the method of Chapter 3. Chapter 5 is concerned with a more specific problem still, namely the costing of simple ternary distillation schemes and the comparison of cost with other evaluation functions. The aim is twofold, firstly to discover if there is any function which gives a good approximation to cost, and secondly, and perhaps more importantly, to discover what factors mainly influence the cost of distillation. Chapter 6 continues this thrust and considers the distillation of systems of more than three components.

2 Literature Review

2 Literature Review

The general difficulty with process synthesis is the size of the problem to be tackled. The current work is concerned with separation scheme synthesis, a much larger problem than heat exchange network synthesis and yet even in that field the size of the problem has seemed a major difficulty. This review of the literature is in two parts. The first deals with heat exchange network synthesis and is included for two reasons: firstly it illustrates the range of solution methods, common to both fields, which are employed to try to make the problem manageable; and secondly the latest developments in the field of heat exchange network synthesis are only just beginning to have parallels in separation scheme synthesis. This second point suggests possible areas for development, some of which are pursued in this work.

2.1 Heat Exchange Network Synthesis Literature

The heat exchange network synthesis problem is often formulated along the following lines:

"Given a set of process streams of known heat capacities and flowrates, each to be heated or cooled from an initial to a target temperature, and unlimited supplies of auxiliary heating and cooling media at known temperatures, find the network of heat exchangers which brings each stream to its target temperature at minimum overall cost."

It has been pointed out in earlier reviews [1-5] that the methods used to tackle this problem are many and exploit every possible feature. This review will therefore centre on the methods used, and for two reasons. The first is to familiarise the reader with these methods since the same types of approach are used in separation scheme synthesis. That subject is more complex and familiarity with the solution methods allows the review of it to concentrate on the variations in the problem. The heat exchange network synthesis problem is simpler and shows few variations in the literature. The second reason is that the more successful methods employed in heat exchange network synthesis have no parallels in the field of separations as yet, and so an understanding of the reasons for their success might suggest new approaches to the separations problem.

2.1.1 Discussion of Terms

Firstly terms applicable to heat exchange networks themselves are discussed and then terms describing design methods. As described above the heat exchange network consists of hot and cold process streams. The characteristic of a HOT stream is not particularly that it is at a high temperature but that it is to be cooled. A COLD stream similarly is to be heated. HOT and COLD utilities, for instance steam and cooling water, provide auxiliary heating and cooling. When streams are paired in a heat exchanger they are said to be MATCHED. A CYCLIC network is one where the same pair of streams is matched more than once. An ACYCLIC network is obviously one where any pair of streams is matched once and once only. A network containing a LOOP may be acyclic, but it is possible to trace a continuous path from one exchanger through the pipework back to that exchanger. An example is where hot stream A is matched with cold stream B, which is also matched with hot stream C, and C is matched with cold stream D which is also matched with stream A. A cyclic network contains the smallest possible loop. Finally PARALLEL STREAM SPLITTING, or just stream splitting is the technique of dividing a stream into two streams of smaller flowrate which may then be matched in parallel with other streams.

Two important terms occurring in any description of synthesis procedures are ALGORITHM and HEURISTIC. An algorithm is a rule or set of rules guaranteed to produce a given result. Thus an algorithmic procedure for solving the problem defined above would be guaranteed to find the cheapest possible network to do the job. Unfortunately, algorithmic methods often suffer from the disadvantage that the amount of work they require is governed loosely by the number of possible solutions to the problem. This number grows very quickly with the size of the initial problem and thus for realistic problems an algorithmic method may require an excessive amount of computer time. Algorithmic methods are therefore often modified by using heuristics to cut down the space of solutions which need to be searched. This heuristic trimming may be entirely arbitrary and governed by, for instance, the particular representation being used by the researcher concerned. Alternatively it may reflect an understanding of the characteristics of networks which are likely to be far from optimal and exclude them accordingly. There are also methods which are entirely heuristic, and in both uses heuristics can give extremely good results. Their drawback is that there is generally no method of telling how good the results are. A heuristic or heuristically limited method may therefore fail quite drastically with no warning.

Algorithms, then, guarantee to find a given result, possibly in a limited space or set of solutions. They may require excessive effort to solve large problems. Heuristics incorporate experience and learned lessons to suggest how the problem may be simplified. Care must be taken that they are applied appropriately and that the lessons are learned correctly. They may fail without warning.

2.1.2 The Literature

2.1.2.1 Embedding and Structural Parameters

Perhaps the most obvious approach to the synthesis of networks is to extend the classical optimisation procedures applied to systems of continuous variables. The resulting problem contains both discrete and continuous variables and its objective function, the overall cost of the system, is usually non-linear. It is therefore a mixed integer non-linear programming (MINLP) problem, the most difficult of the optimisation problems. This MINLP formulation of the heat exchange problem has been attacked (or circumvented) in several ways.

The first published work in the field of heat exchange network synthesis was by Hwa in 1965 [6]. He used the method of separable programming, an extension of linear programming, to optimise both structure and heat loads. To avoid the difficulties of discrete variables he used a technique which has since been used mainly in sythesising whole processes. A structural parameter is a variable describing how a stream is divided between two alternative processing paths. A large flowsheet is constructed including all the likely processing options in parallel with structural parameters describing how flow is divided between them. It is then optimised with the structural parameters constrained to lie between zero and unity. Any equipment which has no flow through it in the optimal flowsheet is removed and the discrete choice is avoided. Shah and Westerberg [7] expose a problem using this approach.

The second approach is that taken by Grossmann and Sargent [8] who begin by expressing the problem as a mixed integer non-linear program but then note the difficulty of solving such a problem directly, particularly the difficulty of avoiding local minima. The method they opt for is to decompose the problem, solving the integer problem first and then to use a standard non-linear programming technique for the continuous variables. Kelahan and Gaddy [9] on the other hand use a random search method to optimise on both discrete and continuous variables simultaneously.

Drawbacks to the method of embedding and optimisation other than those already mentioned are concerned with the initial large, combined flowsheet required. One difficulty is its size and the resulting size of the optimisation problem which can become unmanageable for even relatively small problems. Grossmann and Sargent give the amount of computer time required to generate their optimal configurations. Interestingly, it seems that for their method the number of streams in the problem is not so significant in determining the CPU time required as are factors governing the flatness of the optimal region - the number of solutions with near optimal costs is more important than the number of solutions per se, but there is no guidance given as to how such problems might be detected.

A second and related problem is the requirement for the engineer to use his imagination to include all the necessary items of equipment and the appropriate connections in the initial flowsheet. Presumably for heat exchanger systems where the range of equipment and possible connections is relatively small the initial flowsheet might be generated automatically, though for more complex problems this might not be so. If the engineer were called upon to generate the initial network himself it is possible that the optimal network would be left out simply by oversight.

2.1.2.2 Integer Programming Approaches

Although Grossmann and Sargent used an integer non-linear programming method for the continuous variables in their study the three papers described next apply a very different approach based on the optimal assignment problem of operations research and integer programming. In 1969 Kesler and Parker [10] used a modified linear programming form coupled with the optimal assignment algorithm to tackle the heat exchange network synthesis problem. Their approach was to split each stream into "packets" of equal heat content at different temperature levels and then to use the method to assign packets of hot and cold streams to each other in "exchangelets". Modifications have to be made to the linear programming algorithm to allow for the non-linear objective function, to keep the synthesised process feasible and ensure that hot and cold utilities are not matched with each other. Exchangelets between the same pairs of streams are eventually lumped together to form full size exchangers. It has been suggested that this method only works with a linearised objective function, though Kesler and Parker's paper would seem to say otherwise. The difficulty they do mention is a combinatorial one: they solve a six stream problem with 20 equations in 175 unknowns but suggest that industrial problems would need several hundred equations in several thousand unknowns. The problem formulation does not allow cyclic networks or parallel stream splitting.

In 1971 Kobayashi, Umeda and Ichikawa [11] described a broadly similar technique used in a rather different way. They first of all assume each stream to transfer the same quantity of heat, which allows the number of equations handled by the assignment algorithm to be reduced to the number of either the hot or cold streams. In most cases this approximation is not sufficient and so they resort to parallel stream splitting to define heat packets with the same supply and target temperatures as the original streams but with smaller flowrates. A two-level, iterative approach is adopted, first generating the optimal structure for a given quantity of heat exchanged between streams and then recalculating this total heat load in a continuous optimisation. The process is continued until the structure stabilises.

The "assignment problem" approach seems to have languished until 1977 when Cena, Mustacchi and Natali [12] approached the problem very simply. They divided all the streams into sequential packets of heat, as Kesler and Parker, and then used the assignment algorithm to produce a network with a large number of exchangelets. These are then lumped together by hand, the linearised costs associated with each lumped unit recalculated and the algorithm repeated, a la Kobayashi et al. They report that cases where the optimal structure changed after the first iteration were few. During the hand lumping phase parallel splits and cyclic arrangements may be produced at the engineer's whim.

These three papers adopt rather different methods of adapting a linear programming method to a non-linear objective function. Kesler and Parker use a novel method of adapting the standard linear programming method to non-linear systems, which does not seem to have been taken up elsewhere. Kobayashi et al use the standard optimal assignment approach and avoid the problem by assuming that each exchanger is the same size, and thus effectively linearising the cost. Cena et al linearise cost explicitly, solve the problem, and then iterate, re-linearising the cost in the region of the solution.

2.1.2.3 Heuristic Programming

The next approach considered is that of Rudd and his co-workers. In 1969 they began a series developing a theory of process synthesis. The first paper [13] discusses problem decomposition, the technique of breaking a currently insoluble problem into pieces that can be solved, and then integrating the solutions. The technique is illustrated by means of a very simple heat exchange problem. The second paper [14] tackles a more complex heat exchange problem by heuristic methods, but the technique is developed further into a general process synthesis method, eventually appearing as the AIDES package [15,16]. The approach is to start with a set of heuristic rules and let the computer 'learn' which rules are most important for cheap design as the design process progresses. Thompson and King [17,18] use a similar method for the synthesis of separation processes.

2.1.2.4 Infeasible Branch and Bound

Although this method was also developed by Rudd and co-workers (Lee, Masso and Rudd [19]) it is sufficiently different from the work described above to warrant separate treatment. It is still based on the notion of problem decomposition, and the feature of the heat exchange network synthesis problem that the authors regard as rendering it insoluble is the constraint that a stream may only be used in one place at a time. They argue that if the designer did not have to worry about which match was best for any stream but could use it as many times as he liked then he would have little difficulty in producing a good network. The drawback is of course that such a design might well be impossible to build! Lee, Masso and Rudd's solution to this dilemma is to list all the good networks in increasing order of cost, the first feasible network in the list being the optimal 'real' design. Unfortunately it is shown later [20] that the method for excluding infeasible solutions also excludes some feasible ones, though this is not a fundamental criticism of the method. Menzies and Johnson [21] use the same technique in their method for the synthesis of general energy recovery systems, considering pressure as well as heat recovery. Ponton and Donaldson [22] use it as a basis for comparison with their own method.

2.1.2.5 Tree Searching Procedures

The first workers to apply an algorithmic method based on a search of all possible designs were Pho and Lapidus [20]. They present a blind search, total enumeration method whereby all possible acyclic designs without parallel stream splitting are considered. The optimal sequence is found by direct comparison of all alternatives. For any but the smallest problems this method requires an unrealistically large amount of computation and so for larger problems the authors recommend an alternative method. For problems with more than a handful of streams they present a partial enumeration method using a look-ahead strategy similar to techniques used in chess playing programs. Here only the most promising alternatives at any stage of design are considered and so the workload is reduced. The disadvantage with this procedure is that the optimal design may be overlooked if it contains one unpromising stage offset by many good ones.

Rathore and Powers [23] develop a similar method but use a bounding strategy to reduce the number of alternatives which must be considered. The bounding criterion is based on the degree of energy recovery, a feature which, with the authors' discussion of the desirability of low numbers of heat exchangers and also of methods of deciding how much heat to transfer in any one match, relates this work to the TC method of Flower and Linnhoff [24], to be discussed in the next section.

2.1.2.6 Preanalysis, Target Setting and Related Procedures

Finally we discuss a large family of loosely related procedures based on thermodynamic analysis of the heat exchange problem prior to design, and on related heuristic rules which can be used to restrict the number of solutions which need be considered.

The first work of this kind was by Nishida, Kobayashi and Ichikawa [25] and appeared in 1971. They present various theorems applying to networks having the minimum heat transfer area in the interior subsystem. (The interior subsystem is the network of matches between hot and cold process streams; the exterior subsystem is that of matches between process streams and utilities. This partitioning and other features are shared with the contemporary work of Kobayashi et al [11] described above.) These rules are then used to synthesise a network for the interior subsystem, assuming a quantity of heat transferred. Since this quantity affects the design and cost of the exterior subsystem an iterative approach is used, optimising the heat loads in the system and resynthesising the interior subsystem until the design is stable.

Also in 1971 Hohmann [26] presented methods for determining the minimum number of heat exchange units required in any network, the minimum requirement for hot and cold utilities and the minimum heat transfer area. These quantities, particularly the first two, are important factors influencing the cost of a heat exchange network, and in company with Lockhart [27] Hohmann develops an assessment method for heat transfer networks using the above findings.

One of the theorems derived by Nishida et al above states that in the minimum area network the streams will be matched such that the hottest hot stream will exchange heat with the cold stream with the highest target temperature. This rule is used by two sets of workers as a heuristic for generating nearly optimal networks. First Ponton and Donaldson [22] present a method whereby this hottest/highest (H/H) rule is used repeatedly to synthesise a network: after each match the rule is used on the streams which have not yet reached their target temperatures. The networks generated by this method are often cyclic, with more than one match between any pair of streams. Though the final network is often very good, sometimes better than the best found by other methods, the technique sometimes fails and alternative heuristics are presented for use in such cases. As with all totally heuristic methods however it is impossible to tell if the heuristic gives poor results without using a different technique for comparison.

Shah and Westerberg [28] use Ponton and Donaldson's method in their procedure. They use it to generate an initial network which is then the subject of an evolutionary procedure aimed at improving the design. Finally they optimise the whole network. Compared to the method of Ponton and Donaldson this procedure obviously reduces the chance of generating a very bad design.

A third entirely heuristic method using elements of the work of both Nishida et al and of Hohmann is that of Wells and Hodgkinson [29]. They present a large selection of heuristic rules, some applicable to other areas of process synthesis, not to heat exchange alone, and explain the use of the heat content diagram, a representation of heat exchange networks also used by Nishida et al. These tools enable the designer to create and manipulate networks by hand and give him some guidance as to which alternative designs might be improvements over the current one. No attempt is made to achieve or even define optimality.

The method of Nishida, Kobayashi and Ichikawa described above was extended and improved by Nishida, Liu and Lapidus in 1977 [30]. They use the same basic approach as the earlier work but instead of optimising the total heat duty they calculate a maximum degree of heat recovery before the minimum area flowsheet is generated. This minimum area, nearly minimum cost flowsheet is then subjected to an evolutionary procedure in an attempt to lower the cost still further. The authors comment on the need to minimise the number of heat exchangers in order to make the best use of the effect of economy of scale on the network.

Essentially the same approach is taken by Linnhoff and Flower [31] but they use different targets. Instead of heat exchange area they identify utilities costs as dominant and so Hohmann's utilities target is used to guide the synthesis procedure. Their temperature interval (TI) method arises naturally from the technique used to derive the utilities target, but tends to result in networks comprising a large number of small exchangers. The complementary evolutionary development (ED) method can be used to reduce the number of exchangers toward the predicted minimum number required. A particular advantage claimed or the method is that since energy costs, and thus utilities costs, dominate the cost of a network, then any network that meets the minimum utilities target is likely to be nearly optimal. The TI/ED method can also be used to generate a range of networks with maximum energy recovery and near optimal cost which may then be further screened on the basis of start-up and control characteristics, plant layout constraints or any other criterion.

The last method to be considered here is the thermodynamic combinatorial (TC) method of Flower and Linnhoff [24]. It is based on similar premises to the TI/ED method, namely that any network with the minimum number of heat exchange units and featuring minimum utilities usage is likely to be near optimal. However it also recognises that there may be many other thermodynamic constraints placed on the design of networks if these targets are to be met without parallel stream splitting. In fact in some cases the problem is so constrained that it is possible to develop all possible networks achieving these three goals, minimum units, minimum utilities and no stream splitting, by hand. If Flower and Linnhoff's premise that the globally optimum network will achieve both the units and utilities targets is accepted then the TC method will find that network if it does not require stream splitting. In extreme cases where this is not so then the method may fail completely and generate no networks at all, however the authors claim that in such cases the TI/ED method works particularly well. A general overview of the full approach is given in [32]. It is reported that these latest methods have been applied to industrial problems with considerable success [33,34].

2.1.3 Heat Exchange Networks: Conclusion

The study of heat exchange network synthesis has progressed in two particular though often intertwined directions. The first is the development of mathematical methods which aim to make the potentially very large and general problem of selecting the best network from among many equally promising alternatives manageable. This approach has been successfully used in a number of ways, from the application of standard optimisation procedures to the development of numerical methods apparently unique to this problem. The fundamental difficulty underlying these methods is that they attempt to treat the task in such a general manner, assuming that the optimal solution might lie anywhere and making little or no attempt to locate likely areas of near optimality before beginning the search procedure. The consequence for the engineer is that no matter how powerful his computer he can have little interaction with the design tool due to the vastness of the search task. Thus the application of such tools is limited since factors such as plant layout and control considerations, which are not at the moment readily amenable to simple evaluation, cannot be included in the design procedure.

The second strand of development has been a deepening understanding of the basic features of heat exchange networks [32]. This progress has allowed the improvement of heuristic based techniques so that the latest methods provide good results with less risk of failure than earlier approaches. Heuristics have been used to restrict the number of solutions which need to be considered and the latest techniques use both heuristic rules based on methods used for the evaluation of networks (for instance that good networks have low utilities usage) and analytical methods based on the data of particular problems (the minimum utilities target for example). The virtue of such an approach is not only that it makes better use of the available data than earlier methods, but also that the reduction of the search space allows the engineer to become involved in the design process. The method therefore remains a tool for the engineer to use rather than attempting to replace him. Being heuristic in nature however, these methods cannot guarantee optimality, although the departure from it is likely to be small.

2.2 Separation Scheme Synthesis

Compared to separation scheme synthesis the heat exchange network synthesis problem is very simple and straightforward. Even so it has been necessary to resort to various techniques aimed at relieving the combinatorial difficulties. Such approaches, for instance branch and bound or heuristic/evolutionary schemes, reduce the space of possible solutions, and have been aided by the development of targetting procedures - heuristic rules which describe attributes expected of optimal and near optimal solutions. Nonetheless, because of the relative simplicity of the problem, in the foregoing review the work was classified in terms of the solution methods employed. The solution methods employed in separation scheme synthesis are for the most part identical in approach, save that the target setting approaches are largely absent, and therefore in the review of the literature on this subject it is intended to concentrate on the different facets of the problem and mention the solution methods only in passing, except in cases where the solution method puts limits on the problems which may be treated.

2.2.1 The Problem and Its Simplification

The separation scheme synthesis problem has been stated by Thompson and King [17] as

> "given a feed stream of known conditions (i.e. composition, flowrate, temperature and pressure), systematically synthesise a process that can isolate the specified products from the feed at minimum cost".

This is a restricted version of the more general problem of isolating a set of product streams from a set of feed streams, a problem which is in its turn a subproblem of the general process synthesis problem tackled by Rudd and co-workers [13-16,19] and Motard and co-workers [35,36]. The intermediate problem has not been tackled as such, possibly because it rarely crops up in the industrial environment, and for the most part even the problem as stated by Thompson and King is simplified more or less radically. The simplifications usually applied are reviewed briefly below.

a) Restricted Set of Separator Types: The problem statement as given puts no restriction on the methods used for isolating the specified products. In almost every case studied the problem is recast to select the appropriate separator types from a predefined set. In some cases this set is reduced to contain only distillation. Part of the work reported in this thesis is aimed at identifying separator types it would be useful to include in the set, an approach suggested by Johns

[37].

b) Two-Product Separators: Virtually all separator types in common use produce only two product streams. Examples are standard distillation processes, solvent extraction and so on. Separator types with multiple product streams are possible, such as chromatographic techniques or complex distillation designs, but these have not been investigated except by a few workers (Petlyuk et al [38], Tedder and Rudd [39,40]), and then not as part of a synthesis method.

c) Sharp Separations (High Recoveries): For some solution methods, most notably dynamic programming, it is necessary to assume that all separations are perfect, that is that all of every feed component appears in a single product stream. The corollary of this assumption is that no component may appear in more than one product of the synthesised process. For the purposes of design this is a fair assumption, since small quantities of foreign components generally have little effect on the design of a separator. However most workers using methods which do not require this assumption nevertheless make use of it as it simplifies the synthesis problem considerably. Exceptions are Tedder and Rudd [39,40], Nath [41] and Motard and Westerberg [42] (reported by Nishida, Stephanopoulos and Westerberg [5]). The work of Tedder and Rudd also relates to the previous point, since their low purity products are produced in a three-product-stream distillation column.

d) Immediate recovery of mass separating agents (MSAs): again a restriction imposed by dynamic programming, where each separator type must be a self-contained unit. Most other solution methods can regard an MSA, once added, as simply another product to be isolated. In fact only Hendry and Hughes [43] actually apply dynamic programming to choosing between separator types, so only they apply this restriction. However many other workers make a point of mentioning that they do not apply it.

e) No Heat Integration: A very important point. The majority of work in this field has considered systems where all heating and cooling is performed by utilities: steam, cooling water etc.. There is however a small but growing body of work covering the synthesis of distillation systems with heat integration: systems where the reboiler load of one column is met by the condensing duty of another. There is as yet no work considering a range of separator types with heat integration, though some of the lessons learned from distillation systems will no doubt be applicable to more general situations.

f) Optimisation of Pressure, Reflux Ratio etc.: This is more of an observation of practice rather than a restriction on design. In distillation networks without heat integration the pressures inside individual columns do not interact strongly, pumping costs forming a fairly small part of the total network cost. There are adequate heuristic methods for fixing ranges of pressure, reflux ratio and so on where cost will be nearly optimal (see Tedder and Rudd [39,40], Bakhshi and Gaddy [44], King [45] and discussion in chapter 5) and this can be done for each column independently. However when heat integration is introduced the heat transfer possible between streams varies with the temperatures of the streams involved, and thus with the column pressures. These then interact strongly. Many workers not studying heat integration, especially the later ones, do not bother to optimise individual column designs explicitly but rely on heuristics for near optimal conditions. Workers on heat integration must take such variables, especially pressure, into account.

Having noted the variations on the problem the literature breaks down into three main areas, namely work covering distillation without heat integration, distillation with heat integration, and general separation schemes without heat integration. There is also a body of work concerned with the analysis of the problem and such things as alternative evaluation functions. This work will be dealt with under the most appropriate of the above headings.

2.2.2 Distillation Without Heat Integration

The synthesis of an optimal distillation scheme is a relatively simple task nowadays, the problem being to pick one design from a finite, indeed rather small set of alternatives (see

Freshwater and Henry [46]). Most of the work in this field has therefore concentrated on two topics. The first of these is the derivation of heuristic design rules which will enable the designer to look at his problem and almost immediately be able to pick an optimal or near optimal equipment configuration. The second topic is that of suggesting alternatives to cost as an evaluation function, with the aim of screening alternatives quickly. This is also a heuristic based approach, the heuristic being that minimising some other evaluation function will tend to minimise cost. There are, of course, papers covering problem analysis and support which will also be mentioned.

2.2.2.1 Heuristics

Lockhart, 1947 [47], published what is almost certainly the first paper on separation scheme synthesis. He produced heuristic rules for the design of systems for the removal of light components from natural gasoline. The heuristics were based on the composition of the feed, and were derived for 2-column, 3-product sequences. Other workers covering similar fields have been Petlyuk et al [38], Freshwater and Henry [46], Bakhshi and Gaddy [44] and Tedder and Rudd [39,40]. Freshwater and Henry and Bakhshi and Gaddy looked at simple distillation schemes for three, four and five component feeds over a range of volatilities and compositions. Petlyuk et al and Tedder and Rudd looked at different column designs in attempts to discover which, if any, would provide cheaper networks than those given by conventional designs.

2.2.2.2 Evaluation Functions

The first worker in this field was Harbert, 1957 [48], who worked on the premise that since the cost of energy was the largest part of the cost of distillation, then it might as well be treated as the only cost (and this in the USA in 1957!). On this premise he developed two heuristic rules, and to supplement them he provided a simple semi-graphical method of estimating the energy requirement of distillation. This he used as an alternative to costing to indicate the optimal sequence or near optimal sequences. He was thus the first to advocate the use of an alternative evaluation function. Rod and Marek [49] followed closely with a broadly similar method applied to 3- and 4-component systems, based on the assumption that cost is proportional to the total amount of material vapourised. They report fairly large errors in some cases and, as may be seen in the appendix, there is only rough agreement with the results of other workers.

The two papers mentioned above are the only two to use energy requirements as a substitute for cost in evaluating separation schemes, though the area is explored further in this current work. Another approach, though still based on an appreciation that the important variable is energy consumption, is to use the theoretical minimum separative work (see King [45] for a full explanation). This approach was adopted by Maikov and co-workers [50,51] who also noted the similarity of the separative work function to the rather simpler entropy of choice function from information theory. Maikov [51] gives several examples, both real and imaginary, and points out that though the function favours 50:50 splits (where the flowrates of top and bottom products are equal), the whole process must be considered, not the individual separators. Thus a system with moderately asymmetrical splits may be favoured over one in which a highly asymmetrical split is forced by a previous 50:50 one. There are two obvious difficulties with this approach. One is that no account is taken of the effect of physical properties such as relative volatility or latent heat on the ease of separation. The other is that the theoretical minimum work of separation is never closely approached in distillation. These two features are inherent in the methods described, but even so the reported results for 3- and 4-component systems seem quite acceptable.

2.2.2.3 Summary

In conclusion, workers in the synthesis of systems employing only distillation have not attempted to solve the problem by any algorithmic method, but instead have concentrated on two aspects:

i) discovering heuristic rules which will enable an engineer to design good but not necessarily optimal systems (compare the work of Ponton and Donaldson [22]);

ii) discovering evaluation functions which, though still heuristic in nature, allow some objective comparison between different configurations.

The problem is, even for quite complex mixtures, amenable to relatively easy solution using simple methods. It is therefore rather surprising that no workers have considered algorithmic solutions to this common problem.

2.2.3 Distillation with Heat Integration

Allowing reboiling and condensing streams to exchange heat with one another in distillation systems destroys the simplicity of the synthesis problem. Apart from the combinatorial problem of which streams to match with which there are also the problems raised by allowing the reflux ratio and pressure to vary, thus varying the amount of heat required or available and the temperatures at which it is supplied or accepted. Not only then is there a combinatorial problem, but the design of the heat exchange system interacts strongly with the designs of the individual columns.

The first workers in the field were Rathore et al [52,53]. In their first paper they tackled the simplified problem where all columns are run at the same pressure. They use the infeasible branch and bound method of Lee, Masso and Rudd [19] and dynamic programming to produce the optimal solution. They also present five rules for checking the feasibility of the heat exchange matches. In the second paper they extend their method to to cover systems where pressure is not fixed. In each case the solution method requires sharp splits. Freshwater and Ziogou [54] performed a series of case studies similar to the earlier work of Freshwater and Henry [46], but this time for isobaric distillation systems with energy recovery. They question some of the rules proposed in Rathore et al's first paper, and come to the conclusion that they are a useful but not exact guide. In passing they also question some of the assumptions made in the second paper. Muraki and Hayakawa [55] extend the method of Rathore et al to include a full optimisation of each two-column subproblem (a pair of columns exchanging heat). They admit that their method requires more computational effort than the earlier work, but they claim that the fuller optimisation produces much cheaper networks.

Faith and Morari (and vice versa) [56,57] meanwhile used a branch and bound strategy with upper and lower bounds derived from the Lagrangian theory. Their major conclusion, apart from the fact that they present a viable solution method, is that total energy recovery -- recovering all the heat that it is technically feasible to -- is not always economically attractive.

The latest work is by Linnhoff and co-workers. First Dunford and Linnhoff [58] apply one of the results of heat exchange network analysis to distillation schemes. This result is that in any heat exchange problem, heat integrated distillation systems included, there exists a temperature level known as the 'pinch'. If maximum heat recovery is to be achieved then heat must not be transferred from streams at temperatures above the pinch temperature to streams at temperatures below. (For further details see Linnhoff, Mason and Wardle [32].) The authors use this phenomenon to determine the appropriate and inappropriate application of heat integration schemes, including the more unusual distillation schemes and those employing heat engines and heat pumps. Their main point is that simple distillation schemes should be analysed in this way before heat integration techniques are applied.

Sophos, Stephanopoulos and Linnhoff [59] (reported by Nishida et al [5]) suggest a synthesis approach similar in concept to the earlier work in heat exchange network synthesis of Linnhoff and colleagues. The technique is a two level one, first to identify a small set of very good unintegrated flowsheets, chosen for their small heat loads and temperature differences. It is shown that these systems provide the best opportunities for heat integration and will include the optimal system when heat integration is applied. The second stage is to use targetting procedures to allow good systems to be designed and selected. As with Linnhoff and co-workers' earlier approach to heat exchange network synthesis further screening is possible but will probably not be justified, the final choice of design being left to the engineer's judgement.

In conclusion then there have been two algorithmic solution methods proposed for the synthesis of distillation systems with heat integration. One of these uses dynamic programming and the other a branch and bound method. Other more recent work is similar in style to earlier work in heat exchange network synthesis in that it applies heuristics not to the design method, but to the final designs which are produced, and then leaves to the engineer himself the finding of an appropriate design from a pool of good ones.

2.2.4 General Separation Scheme Synthesis

As remarked before, the synthesis of distillation schemes without heat integration is a fairly simple matter. However, when heat exchange between reboiling and condensing streams is allowed the complexity of the problem increases manyfold. A similar increase takes place when separator types other than distillation are considered for the case without heat integration. In fact no worker has yet tackled the problem of separation scheme synthesis with a range of separator types and heat integration. In the work that has been done there is the usual blend of algorithmic and heuristic approaches, as with the other two fields covered, together with some papers covering the background and techniques.

2.2.4.1 Early Developments

The first paper in the field (Souders [60]) is not strictly about synthesis as it has come to be understood, but is about counter-current separation processes in general. The author presents simple graphical methods for deciding which of the various techniques would be cheapest in a given situation, the decision being based on the relative separation factor of the key components (for distillation the relative volatility) for each different separator type.

The next developments came in 1972, with two papers exemplifying different approaches to the problem. Hendry and Hughes

[43] gave details of an algorithmic method using dynamic programming which showed both the virtues and the shortcomings of this approach. The method is guaranteed to find the optimal flowsheet, given the assumptions that all the products are essentially pure, and that any mass separating agents used are recovered immediately. On the other hand Thompson and King [17,18] describe an almost completely heuristic method using the 'cheapest first' rule. The method uses estimates of the cost per stage for each type of separator to synthesise a network by picking the cheapest to be the next unit included in the process. The costs are updated as the synthesis process proceeds. When a flowsheet is completed the synthesis process is repeated using the updated costs and possibly changing the product set until the best process is found. The 'cheapest first' heuristic used here is similar to Ponton and Donaldson's [22] 'hottest/highest' heuristic for heat exchange network synthesis. Thompson and King's procedure is similar to Ponton and Donaldson's in that it will often give a very good result, but it cannot be guaranteed to be optimal. Hendry and Hughes give a single example comprising a six component feed to be separated into four products using two separator types. Thompson and King give three examples, two hydrocarbon separations with six and eight component feeds, and a ten component problem using various extraction methods for separating rare earth metals in solution. A large part of Thompson and King's work revolves around a computer subroutine for determining which products may be produced by which separators, a subject also covered in the current work.

2.2.4.2 Algorithmic Methods

Algorithmic methods form the bulk of the remaining work in general separation scheme synthesis. Unlike Hendry and Hughes' approach all use some sort of branch search or branch and bound procedure. Rodrigo and Seader [61] used a heuristically ordered depth first search with bounding to find the optimal sequence. Like Hendry and Hughes [43] they make the point that near optimal sequences must also be generated by good synthesis procedures. The reason for this is that the costs of several near optimal sequences often lie close together, and a decision will often be made not on cost but on other less readily quantifiable grounds such as safety or operability and control. Westerberg and Stephanopoulos [62] also make this point while discussing their branch and bound strategy. Their method employs upper and lower bounds in a two-level Lagrangian procedure, the few flowsheets retained at the end of the first stage being the 'nearly optimal' set. Kafarov et al [63] also used a branch and bound procedure apparently similar to that of Rodrigo and Seader. Gomez and Seader [64] used a predictor-based search, a breadth first search using the cost of the remaining binary separators in an uncompleted flowsheet as a lower bound on cost. They claim that the method has better computational characteristics than Rodrigo and Seader's method.

The remaining algorithmic method for general separator systems is very different from the ones noted above. The method described by Johns [37] and Johns and Romero [65] does not perform the detailed design and costing used by all the other methods. It is intended for use during the very early stages of design and uses only very simple models of unit operations, the costing functions usually being calculated only from the feed flowrate and the relative separation factor. No component is considered to be distributed between more than one stream and this allows a very simple stream notation to be used. Binary flags signal the presence or absence of species, an approach also adopted in some of the present work. The synthesis technique is a depth first branch and bound with some features of dynamic programming. The method is very versatile, being able to cope with reactors and, to an extent, heat exchangers as well as separation units, though it is unsuited to heat exchange network synthesis.

2.2.4.3 Heuristic Methods

As well as algorithmic approaches several workers have proposed heuristic synthesis methods. Apart from Thompson and King there have been three, Stephanopoulos and Westerberg [66], Seader and Westerberg [67] and Nath [41], all suggesting evolutionary procedures. The underlying idea is that an initial flowsheet is generated heuristically and then changes are applied in a systematic way to produce neighbouring flowsheets. If one of these is better (that is cheaper) than the original flowsheet then its neighbours are generated until the cheapest flowsheet is found. The attractions of this approach are the relatively small amount of effort required to generate good flowsheets compared to the strictly algorithmic methods, and also the fact that the search can be made interactive, allowing the engineer to apply his own experience to the choice of flowsheet. Stephanopoulos and Westerberg were the first workers to recommend this approach and they present a set of rules for generating neighbour processes and discuss strategies for applying them. Seader and Westerberg present six rules for the initial generation, use the same evolutionary rules as the above and suggest a systematic strategy for applying them, based on how the generation rules were applied originally. Nath presents an automatic method based on the similar notion of challenging the application of the original generating heuristics. The final flowsheet is then optimised with respect to component recoveries, reflux ratios and column pressures.

2.2.4.4 Summary

When attempting to include separators other than distillation whilst synthesising flowsheets the problem changes from being quite simple to very complex. The most widely used approaches have been algorithmic, based mainly on direct search procedures, dynamic programming requiring too much computational effort and placing too many restrictions on the problem formulation. Heuristic approaches are also presented. After one unusual but quite successful attempt to use the 'cheapest first' heuristic all the further work uses heuristic generation of an initial flowsheet followed by evolutionary change to examine neighbouring processes. There is as yet no work of the problem analysis/target setting type applicable to synthesis of general separation schemes.

2.2.5 Other Work and Conclusions

2.2.5.1 Other Work

All the work discussed so far has concerned sharp separations, separations where each component of the feed appears in only one product stream. However in certain cases, the processing of reactor feed streams, for instance, sharp splits are not required, and less sharp or 'sloppy' separations are generally more economical in any case. The assumption of the sharp approach is that in each separator the only components that are distributed are the key components, and of these only a very small amount appears in the 'wrong' product: essentially all the light key component and everything lighter than it appear in one product, and all the other components in the other. Nath [41] devotes a chapter to the consideration of sloppy distillation systems using semi-sharp or non-sharp splits where one or both keys are distributed appreciably between the product streams. He presents a representation and a heuristic synthesis method for sloppy systems, though his conclusion is that much more work needs to be done. The only other workers to have touched on this aspect of the synthesis problem are Tedder and Rudd [39,40] who point out that if the feed mole fractions are suitable and/or product purities are low enough then three product streams can be produced from a ternary feed in a single distillation column with a side draw-off stream.

Another area not mentioned above is the use of the second law of thermodynamics in the analysis of processes. This has mainly been applied to whole processes (Gaggioli and Petit [68]; Townsend and Linnhoff [69]; Umeda, Harada and Shiroko [70]) though two papers particularly refer to distillation. Umeda, Niida and Shiroko [71] report a method of modifying existing designs for distillation with heat integration by using a debottlenecking procedure based on the thermodynamic concept of available energy or availability. Linnhoff and Smith [72] do not present any sort of synthesis method, but instead discuss the thermodynamic efficiency of distillation. They discuss the concept of 'ideal distillation' as opposed to an ideal separation process and advocate efficiencies based on the former, which takes account of the unavoidable energy losses of distillation, rather than the latter which does not.

2.2.5.2 Separation Scheme Synthesis: Conclusions

Most of the work in the literature has been aimed at simplified versions of the general composition adjustment network synthesis problem. In particular almost all workers have considered only sharp separations. The most simplified problem is where only distillation is used without considering heat integration and using only simple equipment configurations. This is a straightforward sequencing problem and yet workers tackling it have concentrated on developing heuristic rules to aid the designers own judgement rather than on algorithmic methods. When heat integration is considered in distillation systems the problem becomes much more interesting and several algorithmic methods are presented for its solution, but there is no purely heuristic method proposed. Synthesis of systems using several separator types but no heat integration has employed both these approaches, with emphasis on branch and bound and heuristic/evolutionary procedures. As yet there has been no work specifically directed at the problem of multiple separator types with heat integration, and little at sloppy separation systems. Similarly little work has been aimed at analysing separation problems and using the results to guide synthesis procedures as has been done with heat exchange network synthesis.

2.3 Overall Conclusion

Workers in heat exchange network synthesis have used both algorithmic and heuristic approaches to the problem with varying degrees of success. Purely algorithmic methods suffer from the disdvantages that they often cannot cope with problems of realistic size and that their methodology does not readily allow for considerations which are not easily quantified. Although avoiding these two drawbacks and frequently yielding good results, purely heuristic methods can sometimes give poor results without warning. Arguably the most successful methods to date apply and extend knowledge gathered from earlier research to provide algorithmic and heuristic tools for the analysis of network problems prior to design. The design goals derived using these techniques may then be employed by the engineer either in traditional design procedures or in synthesis methods developed specifically to utilise these goals. Cases have been reported where these methods have been applied extensively and successfully to industrial problems, suggesting that this particular topic is beginning to find its way out of Universities into the industrial environment.

In the synthesis of separation schemes the same sorts of approach are found as in the earlier work on heat exchange networks, namely a selection of algorithmic and heuristic methods relying on relatively simple views of the problem. There are however relatively few attempts to analyse design methods or problem data such as has led to the more recent work in heat exchange network synthesis. This lack may simply be due to no workers having approached the problem in this way, and the recent appearance of work utilising this type of approach [59] might bear out this conclusion. However there may be another reason, namely that the separations problem is inherently more complicated than the heat exchange problem, and thus the required analysis and goals are correspondingly more complex and less obvious than those for heat exchange. One field where work of this type is being carried out however is in the analysis of chemical plant using the second law of thermodynamics.

3 S6 -- Sam's Systematic Separation Scheme Synthesis System

3 S6 -- Sam's Systematic Separation Scheme Synthesis System

S6 is a computer program based on ideas provided partly by Johns' work in the field of separation scheme synthesis [37,65] and partly on an idea of Jack Ponton's. Johns' basic idea is a method of representing the streams of a separation process using simple binary flags to signal the presence or absence of components. He then uses ordered lists of components and separation factors to do very simple evaluation of alternative network designs. The fact that components are considered to be only either present or absent allows the use of dynamic programming techniques to considerably speed up a branch and bound search for the optimal process (see discussion of 'subtrees' in Rodrigo and Seader [61]). A similar method and data structure are used in S6 to implement Jack Ponton's idea for the evaluation of alternative designs. Rather than using any sort of surrogate cost function for evaluating designs and finding the optimum as Johns does the method uses other criteria to find a set of flowsheets which are likely candidates for further evaluation.

3.1 Details of Method

3.1.1 Basic Concepts

Johns' method uses a ranked list of components for each of a set of separator types and then performs a branch and bound search of all possible flowsheets. The search covers all possible distributions of components, not only those which are feasible using the supplied set of separator types. This technique allows future expansion of the method to include such separators with the intention of prompting the engineer's inventiveness. It is intended that the inclusion of such an unknown separator in an otherwise attractive flowsheet will send him off to discover how the separation might be achieved. It should be noted however that the current implementation of Johns' method only produces results which correspond to known processes, that is they do not include unknown separator types. S6 uses a similar idea but uses criteria other than cost for bounding the search. This means that separators which are not known to be feasible, that is types which are not presented to the program, may be allowed. The resulting output from the program is a list of flowsheets which, it is hoped, are likely candidates for development into good processes, some of which may contain separators which are not feasible by any of the separation methods presented to the program to begin with. The aim of this feature is to spur the designer into looking for new separator types whose use would provide better processes.

Instead of cost S6 uses the feasibility, ease and desirability of individual separations as criteria for evaluating flowsheets. These terms have particular meanings in this case so they are explained below.

-- A FEASIBLE separation is one which is possible using one or more of the separator types presented to the program. A separation which is not feasible may be either simply not known to be feasible or may be specifically excluded. For instance this feature may be used to prevent separations which are known to have been studied for years without success from being marked simply as unknown.

- EASE is only defined for separations which are known to be feasible. A separation is easy if the relative separation factor of the key components is greater than a limit defined individually for each separator type.

- A DESIRABLE separation is defined as one which does not split a multicomponent product (MCP). This is a rather inflexible criterion for characterising a separation, but so far none more versatile has come to light.

3.1.2 Screening of Alternative Designs

S6 attempts to generate all possible flowsheets in turn, screening each as it is generated, and printing out all which survive the screening process. This screening is at two levels, firstly for the individual separations and secondly for the entire flowsheet. Each individual separator is screened as it is generated, and as much of the flowsheet as has been generated at that stage is screened at the same time. If either screening stage indicates that the process should be abandoned then it is and the next flowsheet considered. Details of the two screening stages are given below.

Each individual separation is screened using the definitions of feasibility, ease and desirability described above and the screening procedure is summarised in table 3.1. A flowsheet may either be kept or discarded depending on the feasibility, ease or desirability of the separations contained in it. The possible actions are described below.

-- If during the generation of the flowsheet a separation is encountered that is specifically not feasible the flowsheet is discarded.

-- A flowsheet containing a separator of unknown feasibility is kept if that separator is also desirable, but is discarded if it is not -- what is the point of keeping for further investigation a process which we do not particularly want to use and do not know how to build anyway?

-- If a separation is known to be feasible then what happens depends on the ease and desirability criteria -- separations which are easy, desirable or both are kept, otherwise the flowsheet is discarded.

The second stage of screening is based on the number of separations in the flowsheet which are of unknown feasibility, are not easy or are not desirable. For instance the program may generate only flowsheets which contain a single separator of unknown feasibility or which have only easy separations, or which have no undesirable separations. This is an extension of Johns' idea of

Table 3.1: Individual separator screening

FEASIBILITY (Do we have the technology?)	EASE (Is it easy?)	DESIRABILITY (Do we really want to do it?)	Keep the Process?
Y	Y	Y	Y
		N	Y
	N	Y	Y
		N	N
Don't Know	n/a	Y	Y
		N	N
Definitely not	don't care	don't care	N

Table 3.2: Whole Flowsheet Screening

Denoted by:	Ui Dj (M)	
	i	maximum number of separators of unknown feasibility allowed in flowsheet
	j	maximum number of difficult, i.e. not easy, separators allowed in flowsheet
		either or both of i and j may be unlimited, denoted by '*' instead of a number
	M	if present signifies no splitting of MCPs allowed

including a single unknown separator in a flowsheet. Thus it is possible to produce a list of flowsheets containing a single unknown separator to be investigated or a list of the flowsheets which do not split multicomponent products and therefore have the minimum number of separation units (see the next chapter for a discussion of this point). The notation used for this second level of screening is summarised in table 3.2, and is used in the following discussion of results. Each screening regime is denoted by an expression of the form UiDj(M), where i is the number of unknown separations allowed in a flowsheet, j is the number of difficult separations, that is ones which are not easy according to the above criteria, and the M, if present, denotes that MCPs may not be split. Either i or j may be replaced by '*' which signifies that no limit is placed on that particular quantity.

3.2 Results

All the results presented here were generated with the screening for individual separators set as in table 3.1, but with no separations marked as being definitely not feasible. Screening of whole flowsheets was done in various ways, each with a designation of the type shown in table 3.2. Three examples are covered. They are the one due to Hendry and Hughes [43] and the two hydrocarbon separations studied by Thompson and King [17].

3.2.1 Example 1: Hendry and Hughes' Example

Details of this example are given in table 3.3. It has a six component feed, two separator types and a three component MCP. Running S6 with flowsheet screening of U0D0 produced no flowsheets at all. U0D* produced 41, each of which had a single difficult separation and in which Hendry and Hughes' optimum, shown in figure 3.1, was number 20. Using U1D0 produced 17 flowsheets. This exercise was intended to highlight separators it might be useful to investigate further. In every case the unknown separation was used to avoid splitting of the MCP. The particular separation is the splitting of the butene isomers from an alkane mixture, a very desirable separation but by no means a new problem.

3.2.2 Example 2: Thompson and King's example 1

Details of this example are given in table 3.4. It has only two of the ten separator types presented by Thompson and King, but analysis (see the next chapter) shows that these are sufficient to isolate all the required products. Note the two different levels of difficulty set for the separators: the second set makes extractive distillation more likely to be difficult than distillation. As with example 1 U0D0 gives no flowsheets. U0D1 gives no flowsheets with the first case, but with the second gives 47, as does U0D* with both cases. The optimal process found by Thompson and King is shown in figure 3.2 and is flowsheet number 43 from the 47 generated. U1D0

Table 3.3: Hendry and Hughes' example, as modified by Johns.

Feed Components

- propane (C3)
- but-1-ene (B1)
- n-butane (NB)
- trans-but-2-ene (B2T)
- cis-but-2-ene (B2C)
- n-pentane (C5)

Multicomponent Product

- but-1-ene
- trans-but-2-ene
- cis-but-2-ene

Separation Methods

Distillation

component	rel. sepn. factor
C3	1.12
B1	1.05
NB	1.00
B2T	0.99
B2C	0.97
C5	0.90

ease limits:
0.98 - 1.02

Extractive Distillation

component	rel. sepn. factor
C3	1.05
NB	1.00
B1	0.98
B2T	0.95
B2C	0.90
C5	0.85

ease limits:
0.833 - 1.20

Table 3.4: Thompson and King's example 1

Feed Components

- ethane
- propane
- butane
- pentane
- propene
- but-1-ene

Multicomponent products

1. butane
   pentane

2. propene
   but-1-ene

Separators

Distillation

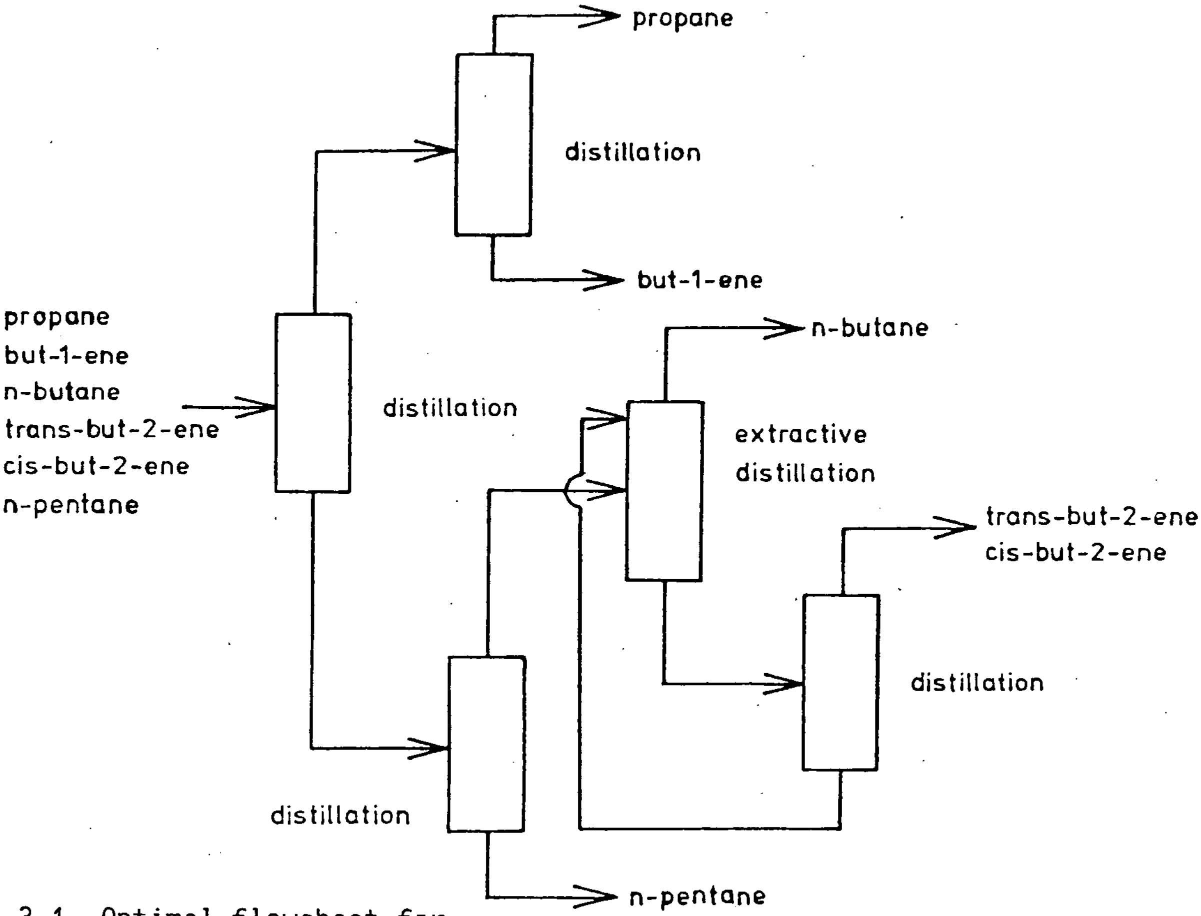
components	rel. sepn. factor
ethane	1144.9
propene	331.26
propane	276.44
but-1-ene	100.47
butane	81.130
pentane	27.421

ease limits:
(i) 0.80 - 1.25
(ii) 0.83 - 1.20

Extractive Distillation with THF

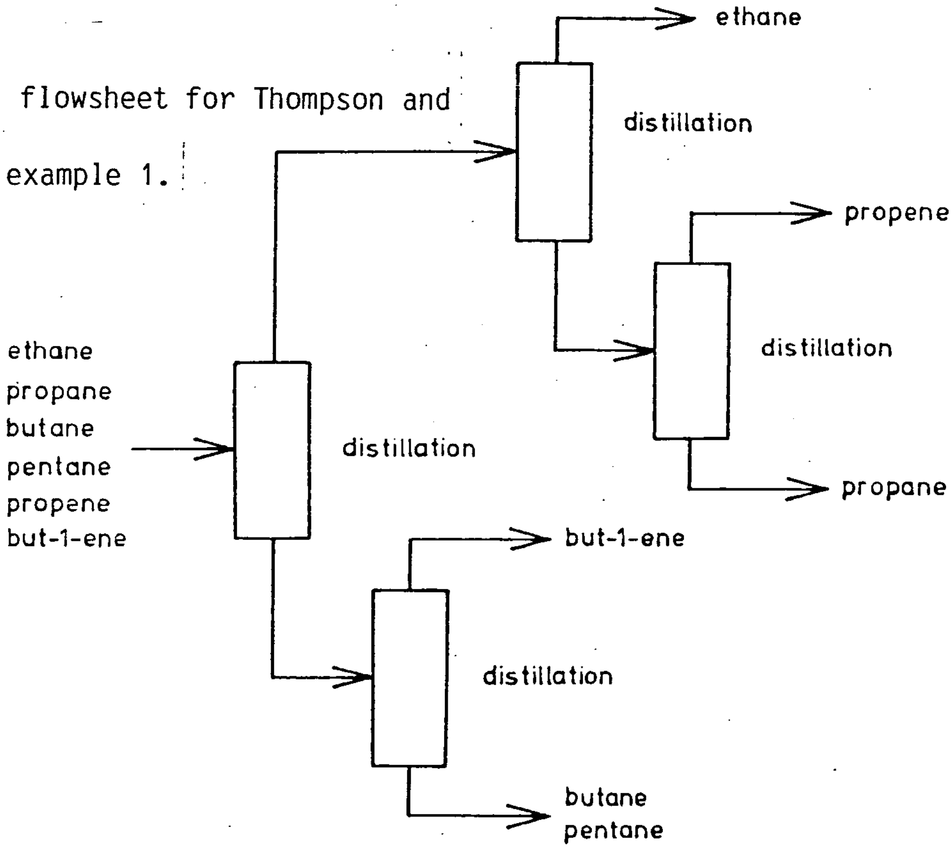
components	rel. sepn. factor
ethane	1678.2
propane	435.71
propene	407.31
butane	117.25
but-1-ene	112.54
pentane	37.383

ease limits:
(i) 0.80 - 1.25
(ii) 0.71 - 1.40



igure 3.1 Optimal flowsheet for Hendry and Hughes' example.

igure 3.2 Optimal flowsheet for Thompson and King's example 1.



in the first case gave 17 flowsheets, in the second 40. In every case the unknown separator was used to preserve one or both MCPs, suggesting that a new and useful separator type would separate alkenes from alkanes.

3.2.3 Example 3: Thompson and King's Example 2

This example is Thompson and King's second hydrocarbon example with only four of the ten original separator types -- distillation and three extractive distillation types as used by Nath [41], though ~~without~~ the product set used is different from both the original one and the corrected one used by Nath. The problem is described in table 3.5. All of U0D1, U0D* and U1D0 produce hundreds of processes -- over 300 in each case before the program failed with excess output. Inspection revealed that only a small part of the possible processes had been printed at this stage. Further results were therefore obtained specifying that no MCPs were to be split. U0D1M produced no flowsheets and U0D*M and U1D0M produced 56 each. Unfortunately Thompson and King's optimal flowsheet splits one of the MCPs and so this is not in the set produced by U0D*M. In fact the cheapest process found by Thompson and King which splits no MCP is about twice as expensive as the cheapest which splits one. Again all the unknown separators found using U1D0M are used to preserve MCPs.

Table 3.5: Thompson and King's example 2, modified product set

Feed Components

n-pentane
n-hexane
n-heptane
benzene
toluene
cyclohexane
1-hexene
1-pentene

Multicomponent Products

1. benzene
   toluene

2. n-pentane
   n-hexane

Separators

Distillation

components	rel. sepn. factor
1-pentene	42.012
n-pentane	29.231
1-hexene	11.780
n-hexane	9.8753
benzene	8.2442
cyclohexane	6.5224
n-heptane	4.8030
toluene	2.7252

ease limits:
0.83 - 1.20

Extractive Distillation with Phenol

components	rel. sepn. factor
n-pentane	95.907
1-pentene	61.219
n-hexane	33.838
1-hexene	32.840
n-heptane	18.842
cyclohexane	9.6401
benzene	8.0913
toluene	2.5991

0.71 - 1.40

Extractive Distillation with THF

components	rel. sepn. factor
n-pentane	41.861
1-pentene	38.634
1-hexene	14.737
n-hexane	14.273
benzene	7.8699
n-heptane	7.2111
cyclohexane	6.7183
toluene	2.4285

ease limits:
0.71 - 1.40

Extractive distillation with 1-hexene

components	rel. sepn. factor
1-pentene	40.636
n-pentane	27.879
1-hexene	11.300
n-hexane	9.5995
benzene	9.4848
cyclohexane	6.9195
n-heptane	4.6584
toluene	3.1451

0.71 - 1.40

3.3 Conclusions

The results produced by S6 were rather disappointing. The aims of the work were twofold: to screen the set of known feasible flowsheets to produce a list of good flowsheets for further screening; and to highlight separations which are not known to be feasible but would might merit investigation, for instance to look for a new extraction solvent. Comments on both of these follow.

In screening for feasible flowsheets S6 gave perhaps more candidates than might have been desired for medium sized problems with six components -- forty flowsheets for close evaluation is rather many. Even though some might be removed by hand this would tend to run counter to the object of the exercise.
In both six component examples studied the optimum flowsheet found by other workers was among those presented. For a larger problem the method produced an unrealistically large number of candidate flowsheets. This could be overcome only by restricting the method so that the optimum flowsheet could not be generated. These problems might be removed by fine tuning the various parameters, for instance the 'ease' level of relative separation factor for the different separator types. However considering the possibility of solving similar problems by simpler methods (see Seader and Westerberg [67]) the effort seems likely to give little return except perhaps to improve the engineer's level of understanding of his problem, a point which will be considered in later chapters.

Concerning the identification of useful new separator types the results were also disappointing since they only seemed to reveal things that were known already, for instance that it would be useful to separate alkenes from alkanes. One of the more interesting points to emerge from the development of methods for the synthesis of heat exchange networks is that much can be learned about the characteristics of 'good' processes prior to any actual design work. The work developed in the next chapter presents aids for doing this sort of thing by hand, and so, looking back, S6 can be seen as a rather expensive alternative to pencil and paper.

4 Minimum Number of Separators

4 Minimum Number of Separators

4.1 Introduction

The ideas for this section of work arose from two sources: (a) the observation that the S6 program described in the last chapter often told us things that we knew already, and (b) the work of Linnhoff et al [24,31-34] on the analysis of data in heat exchange network design. The work of Linnhoff et al brings out two particular criteria for successful heat exchange network design, namely that the network should contain the minimum number of heat exchange units and that it should use the minimum amount of heating and cooling utilities. The number of units in a network is closely related to the capital cost. This is due largely to the effect of the economy of scale (see Boland and Linnhoff [33] for further discussion). Utilities usage not only governs the operating cost of a heat exchange network (cost of steam, cooling water, etc.), but also has an effect on the capital cost: by a simple heat balance the amount of net heating or cooling required by the process streams is fixed so any extra heat supplied to the system must also be removed, which requires extra heat transfer area for both heating and cooling. Again see [33] for details. Linnhoff et al give procedures for determining the minimum number of heat transfer units and the minimum utility requirement prior to design and thus provide the engineer with useful targets to aim at.

Such analysis of problem data has not previously been attempted for separation networks. This chapter is concerned with the concept of the 'minimum number of separators'. Two meanings are considered: the first is the minimum number of separator TYPES required to produce a given set of products from a feed mixture; the second is the minimum number of separation UNITS needed in a flowsheet which produces a set of products with a given set of separator types. The number of separator types is not a particularly useful quantity to know, but the technique used for its derivation also reveals some other useful information. In many cases it will be trivial to determine each of these quantities. In particular for the situation where each product consists of a single component then only a single separator type, for instance distillation, will be required. In such a situation it is virtually impossible to imagine how an engineer designing a simple distillation scheme for such a duty could use more units than the minimum, one less than the number of products. The quantities studied here become more important however when products containing more than one component are required. In such cases it may be necessary to split a product and then remix the components. Figure 4.1 gives examples of flowsheets which produce products containing components (A,C) and (B) from a mixture of the three where the physical properties do not allow the products to be isolated directly.

During the design of any network it is important to use a clear representation of the problem which does not limit the design process. Some representations of heat exchange networks do not allow process streams to match more than once, for example. The work described here uses two complementary representations for the

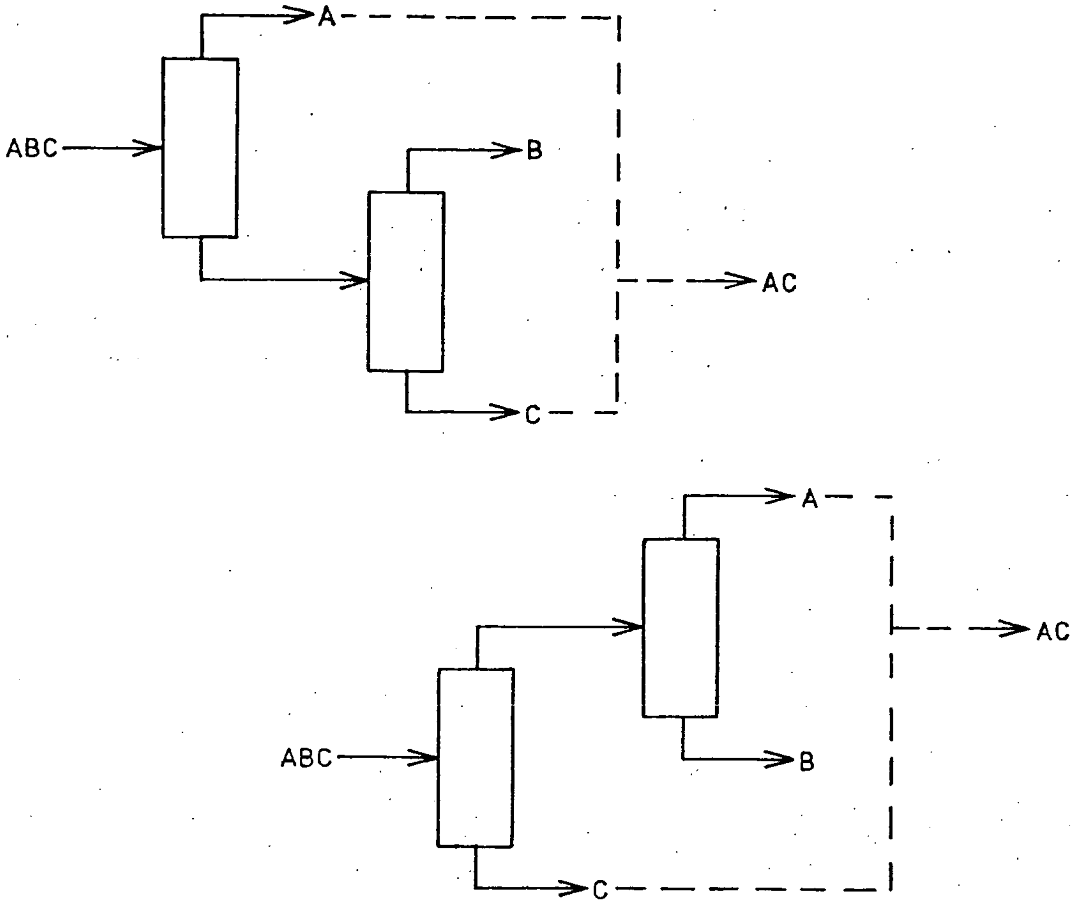


Figure 4.1 Two simple configurations producing (AC) and (B) from (ABC).

possible distribution of components in a separation unit. These are the Product-Component Matrix (PCM) and the Product-Product Matrix (PPM). They are used to determine which separator types are needed to produce a given set of products and also which products may be isolated using a given set of separator types. This information is used to determine the minimum number of separation units.

4.2 Number of Separator Types

In this section the determination of the minimum number of separator types will be shown by means of examples and the introduction of the Product-Component Matrix and the Product-Product Matrix. The more interesting matter of which products may be isolated using a given set of separator types is also discussed.

4.2.1 Product/Component Matrices

Consider example 4.1. Here a mixture of five components, numbered from 1 to 5, is to be split into four products, components 2 and 5 appearing in one product. Three types of separator are available, labelled S1 to S3, and the orderings of the components by relative separation factor for these three types are given in table 4.1.

Figure 4.2 shows the Product-Component Matrix (PCM) for separator type S1, which might be distillation. The matrix is

Table 4.1

Example 4.1

Orderings for components 1-5 in separator types S1-S3:

S1	S2	S3
1	1	1
2	3	2
3	2	3
4	4	5
5	5	4

Products required: (1), (2,5), (3), (4).

(number of components) by (number of products) and each row, corresponding to a component, contains the number of that component or some other identification. The order of components in the rows corresponds to the ordering by relative separation factor -- relative volatility or K-value in the case of distillation. Thus the most volatile component (mvc) will appear in the top row and the least volatile (lvc) in the bottom row. The products are ordered arbitrarily and which column a component appears in depends on which product it appears in.

As can be seen from the PCM in figure 4.2, the MCP (2,5) cannot be isolated intact using separator type S1 since any attempt to separate either product (3) or product (4) from it will split it. Let us now consider separator type S2 (figure 4.3). With separator type S2 we can remove (3) without breaking (2,5), but we still cannot remove (4). However, consider type S3, figure 4.4. Using separator type S3 we can separate (2,5) intact from (4). Now, though in none of the separator types available to us can we isolate (2,5) whole, with judicious use of separator types S2 and S3, and possibly S1, (2,5) can be removed without splitting and remixing. If neither of types S2 and S3 introduces an MSA, it is easy to see that a solution using three units is possible and that at least the two types S2 and S3 are required. Otherwise four separation units would be required.

The above example covered the case where an MCP enclosed other, single component products within its volatility range. It is obvious that a single component product may always be separated

```
  Products  --->
             ---------   mvc
Components  ! 1       !
          ! !   2     !   !
          ! !     3   !   !
          V !       4 !   V
            !   5     !
             ---------   lvc
```

Figure 4.2 Product-Component Matrix for Separator type S1, example 4.1

```
 ---------
! 1       !
!     3   !
!   2     !
!       4 !
!   5     !
 ---------
```

Figure 4.3 Separator Type S2, example 4.1

```
 ---------
! 1       !
!   2     !
!     3   !
!   5     !
!       4 !
 ---------
```

Figure 4.4 Separator Type S3, example 4.1

intact, and, if it lies outside the volatility range of an MCP, may be separated leaving the MCP intact. But what about the case of two MCPs, which may be interleaved in any of several ways? Consider example 4.2, similar to the 'top' half of Thompson and King's [17] example 2, with the corrected product set given by Nath [41] (see example 4.6 in section 4.4.3 below). Here we have four components, (1,2,7,8), two products (1,2) and (7,8), and several separator types, the PCMs for two of which, S1, distillation, and S2, extractive distillation with phenol, are shown in figure 4.5.

Neither of these two separator types allows either of the products to be separated intact, though by judicious juggling we could isolate either product, splitting the other, in two separators, not counting MSA recovery. Nath, however, allows another separator type, extractive distillation with tetrahydrofuran (THF), type S3. This has the ordering shown in figure 4.6.

Here, using two units, product (7,8) may be isolated, but any operation will split (1,2). Even with all the separators, S1, S2 and S3, any operation we can perform will split one or other of the products, if not both. Let us consider another separator type to see whether its use will enable both products to be isolated: type S4, liquid-liquid extraction with Chateau Mouton Rothschild '66, figure 4.7.

It might seem that both products are now isolable intact, since (7,8) may be separated intact in S3, and (1,2) in S4. This is not the case. Even with all four separator types no split can be

```
 -----                                  -----
!   8 !  Separator type S1             ! 1   !  Separator type S2
! 1   !                                !   8 !
!   7 !  Distillation.                 ! 2   !  Extractive distillation
! 2   !                                !   7 !  with phenol.
 -----                                  -----
```

Figure 4.5 PCMs for separator types S1 and S2, example 4.2

```
 -----
! 1   !  Separator type S3
!   8 !
!   7 !  Extractive distillation
! 2   !  with THF.
 -----
```

Figure 4.6 PCM for separator type S3, example 4.2

```
 -----
!   8 !  Separator type S4
! 1   !
! 2   !  Liquid-liquid extraction
!   7 !  with CMR'66.
 -----
```

Figure 4.7 PCM for separator type S4, example 4.2

performed which does not break one of the two products. The best that can be done is still to separate one product intact whilst breaking the other. This observation leads to the following important result: in order for two MCPs to be separated from each other they may not be interleaved at all in at least one separator type. It is apposite at this point to insert a note that what is being done in splitting an MCP is the redefining of the product set, as done by Thompson and King in a rather complex computer program. It is demonstrated below that much the same thing can be done quite simply using pencil and paper. This is achieved using the Product-Product Matrix discussed next.

4.2.2 Product-Product Matrices

Although the Product-Component Matrices contain all the required information about the different separator types they represent, they are somewhat inconvenient to use to decide which separator types would be useful in a certain situation. Indeed the problem gets worse the more components and, especially, the more MCPs there are. Jack Ponton came up with the basic idea for the following representation, since christened the Product-Product Matrix (PPM). This is a square matrix, (number of products) by (number of products), for each separator type. Row i, column j is marked with a '1' if product i may be separated from product j without splitting either. The unmarked elements are filled with '0's. Figure 4.8 gives both PCMs and PPMs for example 4.1 described above. Two points are immediately obvious:

```
          ---------    ---------    ---------
         ! 1       !  ! 1       !  ! 1       !
PCM      !   2     !  !     3   !  !   2     !
         !     3   !  !   2     !  !     3   !
         !       4 !  !       4 !  !   5     !
         !   5     !  !   5     !  !       4 !
          ---------    ---------    ---------

          J No Products
          ---------    ---------    ---------
       λ !   1 1 1 !  !   1 1 1 !  !   1 1 1 !
PPM      ! 1   0 0 !  ! 1   1 0 !  ! 1   0 1 !
         ! 1 0   1 !  ! 1 1   1 !  ! 1 0   1 !
         ! 1 0 1   !  ! 1 0 1   !  ! 1 1 1   !
          ---------    ---------    ---------
             S1           S2           S3
```

Figure 4.8 PCMs and PPMs for example 4.1

```
      S1            S2         S1+S2 (=S2)
  ---------     ---------     ---------
 !   1 1 1 !   !   1 1 1 !   !   1 1 1 !
 !     0 0 !   !     1 0 !   !     1 0 !
 !       1 !   !       1 !   !       1 !
 !         !   !         !   !         !
  ---------     ---------     ---------

      S3       S1+S3 (=S3)   S1+S2+S3 (=S2+S3)
  ---------     ---------     ---------
 !   1 1 1 !   !   1 1 1 !   !   1 1 1 !
 !     0 1 !   !     0 1 !   !     1 1 !
 !       1 !   !       1 !   !       1 !
 !         !   !         !   !         !
  ---------     ---------     ---------
```

Figure 4.9 Combining PPMs for example 4.1

- -- The elements on the main diagonal are not used -- a product cannot be separated from itself.

- -- the matrices are symmetrical about the main diagonal -- if product i can be separated from product j then product j can be separated from product i. For clarity the redundant lower left hand half of the PPM will be omitted in the remainder of the diagrams.

What is not immediately obvious is that the matrices can be combined, element by element, to give a matrix representing any combination of separator types. The method of combination is a logical OR, treating the '1's as logical TRUE. The result is a matrix which represents a set of separator types and shows which products may be isolated using the separator types in the set. Any remaining zeros show which products may still not be separated from each other. It allows the designer to spot what split or splits cannot be done with a given set of separator types and which it might therefore be useful to investigate. This last was, of course, one of the aims of the S6 effort. Figure 4.9 shows the production of combined matrices for example 4.1 from those in figure 4.8.

From the combined PPMs it is immediately clear that with only separator types S1 and S2 we still cannot separate products (2,5) and (4) from each other, and with just S1 and S3 we cannot separate (2,5) and (3). However, with S1, S2 and S3, in fact with just S2 and S3, each product can be isolated intact.

The PPMs for example 4.2 are given in figure 4.10. The example is trivial but worth noting. Suffice to say the products obviously cannot be separated at all using any combination of the four separator types given.

The PCMs and PPMs shown in figure 4.11 are for yet another example with interleaved products, example 4.3 based on Thompson and King's example 1. In separator type S1 product (3,5) cannot be separated from (4,6), though (1,2) can be separated from both, and in S2 (4,6) can be separated from the other two products, though they cannot be separated from each other. The PPMs indicate this and the combined PPM for both separator types shows that using both allows all three products to be isolated intact.

4.2.3 Use of Matrices and Discussion

The Product-Component and Product-Product Matrices described above can be used in several ways. For instance they can be used to find the minimum number of separator types required to produce a given set of products. This is done by choosing the smallest set of separators whose combined PPM has no zeroes in it. However this measure is probably not particularly useful, since it may simply tell us that all the required products may be produced using a single exotic and very expensive separation method. A more valuable use is to discover which separator types other than a single base type would be needed to isolate the product set. For instance if not all the products are isolable using, say, distillation, shown by

```
           S1       S2       S3       S4
          -----    -----    -----    -----
        !   8 !  ! 1   !  ! 1   !  !   8 !
  PCM   ! 1   !  !   8 !  !   8 !  ! 1   !
        !   7 !  ! 2   !  !   7 !  ! 2   !
        ! 2   !  !   7 !  ! 2   !  !   7 !
          -----    -----    -----    -----

          -----    -----    -----    -----
  PPM   !   0 !  !   0 !  !   0 !  !   0 !
        !     !  !     !  !     !  !     !
          -----    -----    -----    -----
```

Figure 4.10 PCMs and PPMs for example 4.2

```
              S1         S2
            -------    -------
          ! 1     !  ! 1     !
          ! 2     !  !   3   !
   PCM    !   3   !  ! 2     !
          !     4 !  !   5   !
          !   5   !  !     4 !
          !     6 !  !     6 !
            -------    -------

                                 S1+S2
            -------    -------    -------
          !   1 1 !  !   0 1 !  !   1 1 !
   PPM    !     0 !  !     1 !  !     1 !
          !       !  !       !  !       !
            -------    -------    -------
```

Figure 4.11 PCMs and PPMs for example 4.3

zeroes in the PPM for that separator type, then inspecting the other PPMs to see which have '1's where the distillation PPM has '0's will reveal which other separator types are useful.

In the situations described above are where all the products may be separated intact using the supplied separator types. In some cases this may not be so, or it may be desirable to use only a subset of the separator types available, and multicomponent products may have to be broken. In these cases zeroes in the PPMs for the combined PPM for the set or subset of separator types show which pairs of products cannot be separated, and the PCMs for the individual separator types will show which of the products must be split. This information is used in the derivation of the minimum number of separation units described below.

The information about which products cannot be isolated using some type or types of separator can also be used to stimulate a search for hitherto unconsidered types to be included in a flowsheet. This was one of the aims if S6, and the use of these matrices provides a much simpler way of discovering some of the things revealed by that program.

4.3 Minimum Number of Separation Units

The minimum number of separation units required in a flowsheet to produce a given set of products using a given set of separator types is discussed in this section. The introduction to the next chapter considers the failings of this measure as an indication of the cost of separation systems. First comes a discussion of terms and what is meant by a 'unit'.

4.3.1 Terms and Simple Case

Some workers treat separators which require mass separating agents (MSA separators) and the associated MSA recovery equipment as a single unit. Most of these workers use the example originated by Hendry and Hughes [43], where immediate solvent recovery was forced by the use of dynamic programming to solve the synthesis problem. Other workers, using approaches similar to that of Thompson and King [17] treat MSAs like any other component in the system, to be isolated in exactly the same way as any other product. We will refer to these two possibilities as the 'immediate recovery' convention and the 'delayed recovery' convention. If an MSA is used then the delayed recovery convention will obviously give a larger number of units than the immediate recovery convention for the same flowsheet.

For either of these cases there is one situation where the minimum number of units, U_{min}, is trivial to determine. This is the situation where each product is composed of only a single component and a separator type which does not use an MSA is available. In this situation the product ordering is unimportant and any separator type may be used for any split. U_{min} in this case is always one less than the number of products, $N_p - 1$.

4.3.2 Immediate Recovery Convention

In this view the determination of the minimum number of units is particularly simple. If all the products can be isolated intact with the separator types available, then the minimum number of units will be equal to the number of products less one,

$$U_{min,i} = N_p - 1$$

If this is not the case then it will be necessary to split one or more products to create a new product set with N_p' products. The minimum number of units will now equal one less than the new number of products,

$$U_{min,i} = N_p' - 1.$$

The method of deciding which products to split will be discussed cursorily later.

4.3.3 Delayed Recovery Convention

Using this convention the separators must be divided into two classes, those which use an energy separating agent (ESA separators) and do not introduce any extra species, and those which introduce mass separating agents which must be removed later in the process (MSA separators). The only commonly used ESA separator is distillation, but MSA separators are more numerous, including extractive distillation, solvent extraction and so on. There is one simple case, namely that where all the products can be isolated using only the set of ESA separators. (In most cases this will of course mean that all the products may be isolated using distillation.) In this case the minimum number of units is the usual, number of products less one

$$U_{min,d} = N_p - 1.$$

If not all the products can be separated by use of the ESA separators then the determination of the minimum number of units is done in two stages. First a new set of products which can be isolated using the ESA separators is found. This set has $N'_{p,esa}$ members. Now consider the MSA separators as well. If all the original products can be isolated using both the ESA and MSA separators, then the minimum number of units will either be $N'_{p,esa}$ less one, or the number of original products less one plus the number of MSAs separators needed to isolate the products. The number of MSAs used has to be added to account for the units required to recover the MSAs.

$$Umin,d = \min [N'_{p,esa} - 1, N_p - 1 + N_{msa}]$$

where N_{msa} is the number of MSA separators needed to isolate the original products.

If, in the most extreme case, the use of MSAs still does not allow the isolation of all the products then the products must again be split to provide another set of products which can be split using the MSAs available. This set will have $N'_{p,msa}$ members, and the minimum number of units will then be given by

$$U_{min,d} = \min [N'_{p,esa} - 1, N'_{p,msa} - 1 + N_{msa}]$$

There is one exception to the above rules. This occurs when a single MSA is used more than once, but need be recovered only once. Figure 4.12 shows an example of this situation where an extractive distillation entrainer is used in two columns sequentially. In this example the term N_{msa} may be decreased by one, however there are disadvantages attached to this sort of arrangement. One is that it limits the number of equipment configurations that may be used, since the two extractive distillation columns may only be used in the order shown. If the split between the keys in the second column were attempted first then the entrainer would leave in the bottom product and not be available for the second separation. Also in such cases the entrainer is introduced into the second column with the feed rather than near the top of the column as is usual, which will not be ideal. It is also clear that such a scheme of using the MSA twice but recovering it only once cannot be used for separators using immiscible phases such as solvent extraction.

4.3.4 Splitting Products

The above procedure for determining the minimum number of separation units will sometimes depend heavily on being able

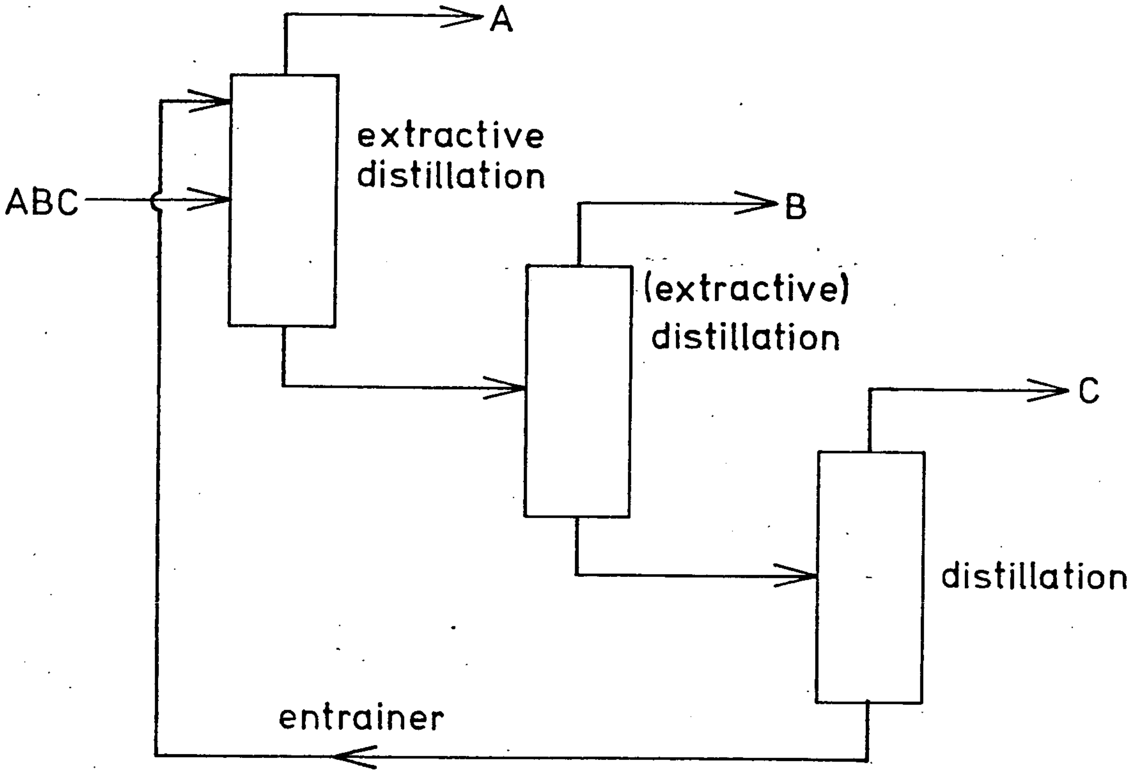


Figure 4.12 Example of an extractive distillation entrainer used twice before recovery

notionally to split multi-component products which cannot be isolated intact. Since the minimum number of units is so dependent on the number of products to be isolated then we obviously need to create the minimum number of new products when deciding which products to split and into which new products. Little will be said about this subject, except to reiterate what was said in the discussion on the number of separator types: it will generally be obvious from the Product-Product Matrices which product or products must be split since a zero entry indicates that two products are inseparable with a given separator type or combined set of separator types. The Product-Component Matrices then tell whether one or both of the products need be split and into what further products.
It is conceivable that in very complicated (and rare) cases the best way of splitting the products will not be clear. in those cases it is left to the engineer to investigate the possibilities.

4.3.5 Minimum Number of Units: Summary

In this section a method has been described for finding the minimum number of separation units required in a flowsheet to produce a set of products from a feed stream using a set of separator types. Two different results are obtained depending on what convention is adopted regarding the recovery of MSAs: MSA recovery may be regarded as part of a single unit, the immediate recovery convention, or the MSA may be thought of as being recovered in a separate unit, the delayed recovery convention. Using either convention, but particularly the delayed recovery, the desired product set may have to be notionally redefined for the determination of the minimum number by splitting multicomponent products. In such cases which products must or may be split will usually be obvious from the PCMs and PPMs, but in complicated cases it may be necessary for the engineer to exercise his own judgement.

4.4 Further Examples

The following are some examples which illustrate the determination of the number of separator types required and also the minimum number of units.

4.4.1 Example 4.4

This is the example used by Thompson and King [17] and simplified by Nath [41] as his example no.4. Its features are given in table 4.2 and the PCMs and PPMs in figure 4.13.

4.4.1.1 Number of Separator Types

Figure 4.13 shows that both of the individual separator PPMs contain zeroes but that the combined PPM has none. Therefore the minimal set of required separator types contains both S1 and S2, and the minimum number of separator types required is two. Similarly, if distillation, S1, is taken as a base type to be used by default

Table 4.2

Example 4.4

Nath's example 4, [41]

Component		required in product number:
1 ethane	(C2)	1
2 propane	(C3)	2
3 butane	(C4)	3
4 pentane	(C5)	3
5 propene	(C3=)	4
6 but-1-ene	(C4=)	4

Separator types available:

S1 distillation
S2 extractive distillation with tetrahydrofuran (THF)

Component orderings at 54.4 degrees Centigrade:

S1	S2
C2	C2
C3=	C3
C3	C3=
C4=	C4
C4	C4=
C5	C5

```
                S1                        S2
          --------------            --------------
        ! C2             !        ! C2             !
  PCM   !           C3=  !        !    C3          !
        !    C3          !        !          C3=   !
        !           C4=  !        !       C4       !
        !       C4       !        !          C4=   !
        !       C5       !        !       C5       !
          --------------            --------------

                                                         S1 + S2
            ---------           ---------            ---------
          !   1 1 1 !         !   1 1 1 !          !   1 1 1 !
    PPM   !     1 0 !         !     1 1 !          !     1 1 !
          !       1 !         !       0 !          !       1 !
          !         !         !         !          !         !
            ---------           ---------            ---------
```

Figure 4.13 PCMs and PPMs for example 4.4

```
                    S1
          ------------------
        ! C2                 !
  PCM   !          C3=       !
        !    C3              !
        !               C4=  !
        !       C4           !
        !       C5           !
          ------------------

            ----------
          !   1 1 1 1 !
  PPM     !     1 1 1 !
          !       1 1 !
          !         1 !
          !           !
            ----------
```

Figure 4.14 PCM and PPM for example 4.4, distillation, modified product set then one split, between products 2 and 4, (C3) and (C3=,C4=), must be done using extractive distillation with tetrahydrofuran. Alternatively, if distillation alone is to be used then the PCM reveals that (C3=,C4=) must be split. This is shown in figure 4.14.

4.4.1.2 Minimum Number of Units

Using the immediate recovery convention the minimum number of units is clearly 3, since all 4 products can be isolated using the given separator types, thus

$$N_p = 4$$

$$U_{min,i} = 4 - 1 = 3$$

For the delayed recovery convention it is clear that with just ESA separators product number 4, (C3=, C4=), needs to be split. This situation is shown in figure 4.14 which now shows 5 products. $N'_{p,esa}$ is thus five. Now using all the available separator types all the products are isolable so there is no need to split products and determine a value for $N'_{p,msa}$. It has been determined above that with the original product set S2 must be used once for separating product 2, (C3), from product 4, (C3=, C4=). N_{msa} is thus one. We therefore have the following:

$$N'_{p,esa} - 1 = 5 - 1 = 4$$

$$N_p - 1 + N_{msa} \quad = \quad 4 - 1 + 1 \quad = \quad 4$$

$$\text{whence} \quad U_{min,d} \quad = \quad 4$$

4.4.1.3 Other Workers' Results

In fact the optimum flowsheet is found by splitting the (C3=, C4=) product and using just distillation. The best flowsheet found ~~by Thompson and King using the original product set, and~~ by both Thompson and King and Nath with the modified product set is shown in figure 4.15.(p85) It uses 4 units.

4.4.2 Example 4.5

This is the example originally studied by Hendry and Hughes [43] and by Nath [41] as his example 5. Like the previous example it has six components and two separator types -- distillation and extractive distillation. It also has four products, but, unlike the example above has only one 3-component MCP. Details are given in table 4.3. The PCMs and PPMs are given in figure 4.16.

4.4.2.1 Number of Separator Types

Here the PPM for S2, extractive distillation with furfural, shows that all the products are isolable using this one separator

Table 4.3

Example 4.5

Hendry and Hughes' example, [43].

Components		required in product number
1 propane	(C3)	1
2 n-butane	(NB)	2
3 but-1-ene	(B1)	3
4 trans-but-2-ene	(B2T)	3
5 cis-but-2-ene	(B2C)	3
6 n-pentane	(C5)	4

Separator types available:

S1 distillation
S2 extractive distillation with furfural

component orderings at 54.4 degrees centigrade:

S1	S2
C3	C3
B1	NB
NB	B1
B2T	B2T
B2C	B2C
C5	C5

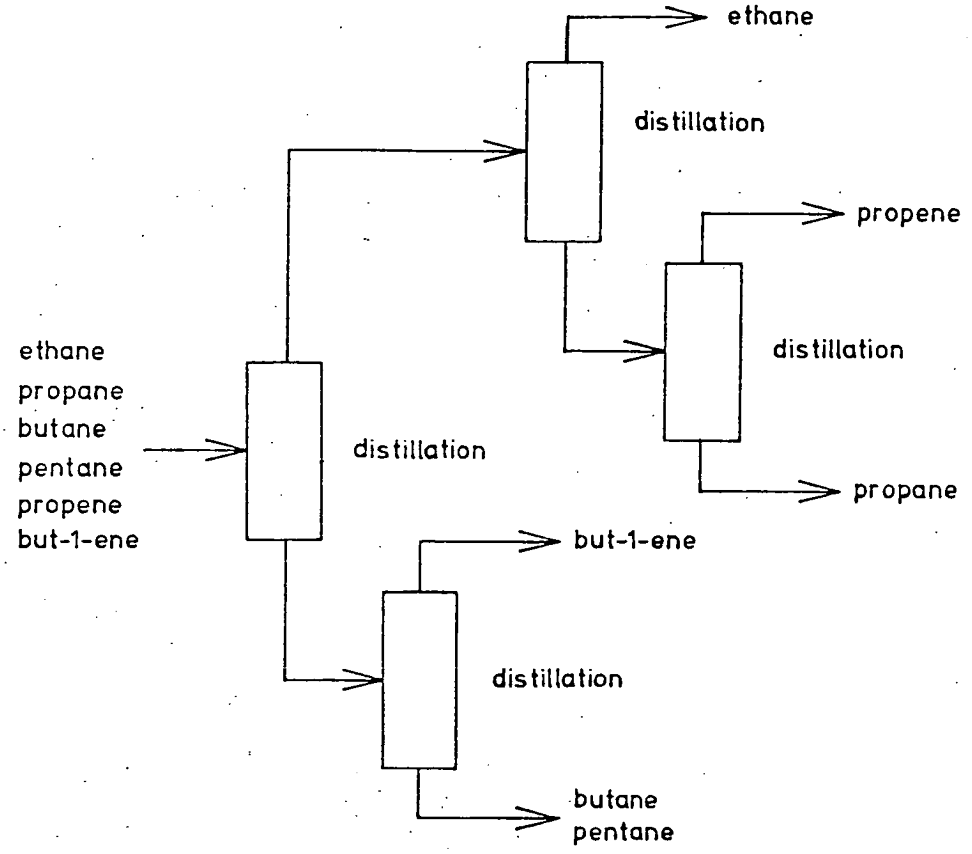


Figure 4.15 Optimal flowsheet for example 4.4, Thompson and King's example 1.

```
              S1                   S2
        --------------       --------------
       ! C3           !     ! C3            !
  PCM  !       B1     !     !    NB         !
       !    NB        !     !       B1      !
       !       B2T    !     !       B2T     !
       !       B2C    !     !       B2C     !
       !           C5 !     !           C5  !
        --------------       --------------

                                                      S1 + S2
            ---------           ---------            ---------
           !  1 1 1 !          !  1 1 1 !           !  1 1 1 !
  PPM      !    0 1 !          !    1 1 !           !    1 1 !
           !      1 !          !      1 !           !      1 !
           !        !          !        !           !        !
            ---------           ---------            ---------
```

Figure 4.16 PCMs and PPMs for example 4.5

```
                  S1
          ----------------
         ! C3             !
  PCM    !       B1       !
         !    NB          !
         !          B2T   !
         !          B2C   !
         !             C5 !
          ----------------
```

Figure 4.17 PCM for example 4.5, distillation, modified product set type. The minimum number of separator types required is therefore one. If distillation, S1, is taken as the base separation method then either S2 must be used to separate (NB) from (B1,B2T,B2C) or the 3-component product must be split into (B1) and (B2T,B2C), as shown in figure 4.17.

4.4.2.2 Minimum Number of Units

Since all four products are separable then the immediate recovery convention gives the minimum number of units as three,

$$N_p = 4$$

$$U_{min,i} = 4 - 1 = 3$$

Taking the delayed recovery view product 3, (B1, B2T, B2C), must be split into (B1) and (B2T, B2C) if only ESA separators are used, thus $N'_{p,esa}$ is five. All four products can be separated using the supplied separator types, and only one MSA need be used to separate (NB) from (B1, B2T, B2C). Thus we have

$$N'_{p,esa} - 1 = 5 - 1 = 4$$

$$N_p - 1 + N_{msa} = 4 - 1 + 1 = 4$$

whence $U_{min,d} = 4$

4.4.2.3 Other Workers' Results

Results of other workers investigations are shown in figure 4.18. (p90) Note that using the minimum number of units in the final flowsheet results in a cost some 2.5 times greater than the optimal. In fact the optimal flowsheet not only splits a product so that all the products could be isolated with distillation only, but then uses extractive distillation. Westerberg and Stephanopoulos [62] give the cost of the extractive unit with MSA recovery in the optimal flowsheet as \$520 000, the cost of the competing distillation unit for the same duty being more than \$3 million.

4.4.3 Example 4.6

The third example is the second hydrocarbons separation example of Thompson and King [17], as corrected and simplified by Nath [41], being his example 6. The details are in table 4.4. This is an eight component problem with six products and 4 candidate separator types, one distillation and three extractive distillation. The initial PCMs and PPMs are given in figure 4.19.

4.4.3.1 Number of Separator Types

Here the products are all separable using only one of the available separator types, namely extractive distillation with tetrahydrofuran, S3. The minimum number of separator types required

Table 4.4

Example 4.6

Thompson and King's 2nd example [17], Nath's example 6 [41].

Components		required in product number
1 pentane	(C5)	1
2 hexane	(C6)	2
3 heptane	(C7)	3
4 benzene	(B)	4
5 toluene	(T)	3
6 cyclohexane	(CH)	5
7 hexene	(C6=)	6
8 1-pentene	(C5=)	6

Separator types available:

S1 distillation
S2 extractive distillation with phenol
S3 extractive distillation with tetrahydrofuran
S4 extractive distillation with 1-hexene

Component orderings at 54.4 degrees Centigrade:

S1	S2	S3	S4
C5=	C5	C5	C5=
C5	C5=	C5=	C5
C6=	C6	C6=	C6=
C6	C6=	C6	B
B	CH	B	C6
CH	C7	CH	CH
C7	B	C7	C7
T	T	T	T

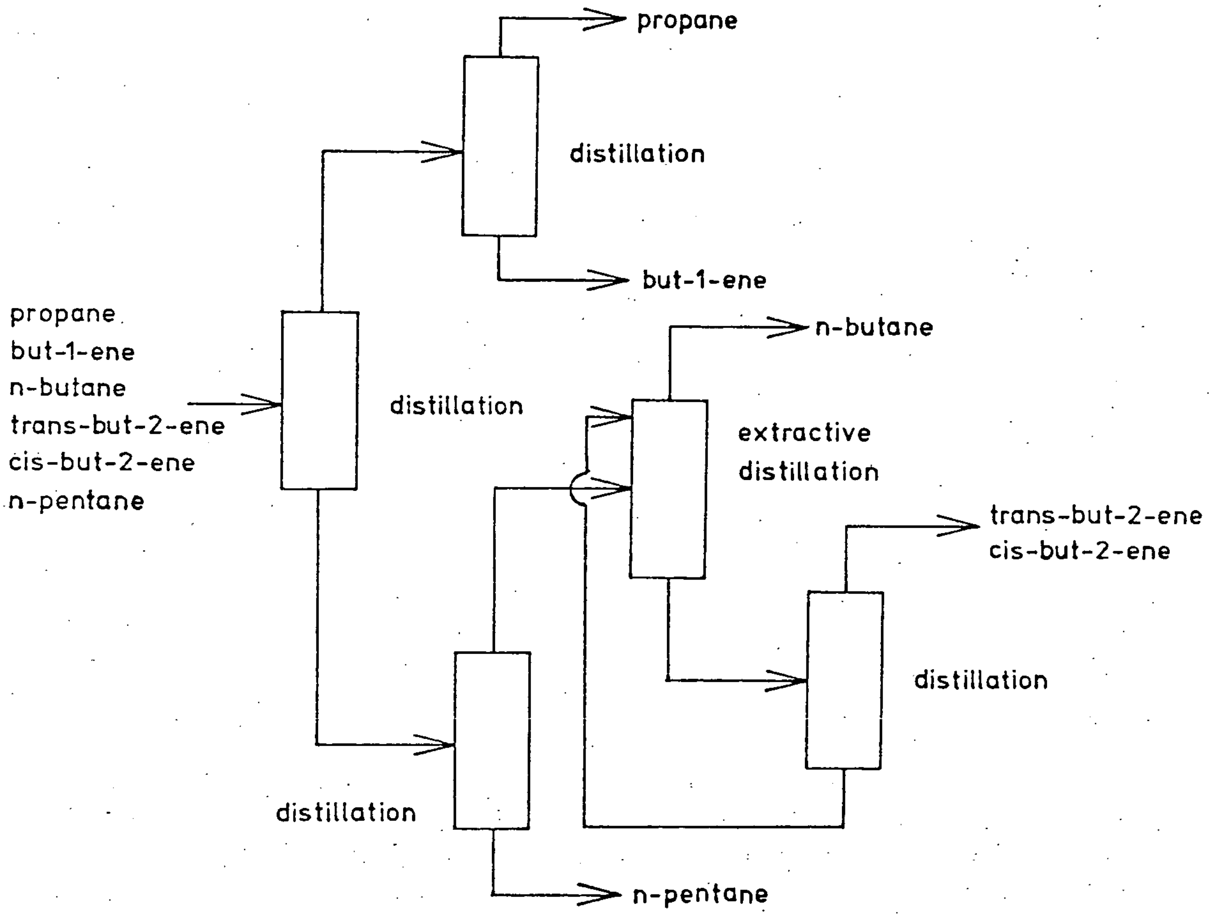


Figure 4.18 Optimal flowsheet for example 4.5, Hendry and Hughes' example.

```
               S1                        S2
      -------------------       -------------------
     !               C5= !     ! C5                !
PCM  ! C5                !     !               C5= !
     !               C6= !     !    C6             !
     !    C6             !     !               C6= !
     !          B        !     !            CH     !
     !            CH     !     !       C7          !
     !       C7          !     !          B        !
     !       T           !     !       T           !
      -------------------       -------------------

               S3                        S4
      -------------------       -------------------
     ! C5                !     !               C5= !
PCM  !               C5= !     ! C5                !
     !               C6= !     !               C6= !
     !    C6             !     !          B        !
     !          B        !     !    C6             !
     !            CH     !     !            CH     !
     !       C7          !     !       C7          !
     !       T           !     !       T           !
      -------------------       -------------------


               S1                        S2
         -------------             -------------
        !   1 1 1 1 0 !           !   1 1 1 1 1 !
   PPM  !     1 1 1 1 !           !     1 1 1 0 !
        !       1 1 1 !           !       0 1 1 !
        !         1 1 !           !         1 1 !
        !           1 !           !           1 !
        !             !           !             !
         -------------             -------------

               S3                        S4
         -------------             -------------
        !   1 1 1 1 1 !           !   1 1 1 1 0 !
   PPM  !     1 1 1 1 !           !     1 1 1 1 !
        !       1 1 1 !           !       1 1 1 !
        !         1 1 !           !         1 1 !
        !           1 !           !           1 !
        !             !           !             !
         -------------             -------------
```

Figure 4.19 PCMs and PPMs for example 4.6

is thus one. If distillation, S1 is used as the base type then either product 6, (C5=,C6=), must be split to separate it from product 1, (C5), the situation shown in figure 4.20, or one of S2 and S3, extractive distillation with phenol and tetrahydrofuran respectively, must be used.

4.4.3.2 Minimum Number of Units

All the products are isolable with the given separator types, therefore using the immediate recovery convention the minimum number of units is one less than the number of products,

$$N_p = 6$$

$$U_{min,i} = 6 - 1 = 5$$

Using the delayed recovery convention the products are not isolable using distillation alone, but the (C5=, C6=) product must be split. This gives seven products in the modified product set. $N'_{p,esa}$ is thus seven. Now all the original products are isolable with the separator types available, and only one MSA is needed, thus N_{msa} is one.

$$N'_{p,esa} - 1 = 7 - 1 = 6$$

$$N_p - 1 - N_{msa} = 6 - 1 + 1 = 6$$

```
          S1
 -----------------------
!               C5=     !
! C5                    !
!                   C6= !
!    C6                 !
!          B            !
!            CH         !
!       C7              !
!       T               !
 -----------------------
```

Figure 4.20 PCM for example 4.6, distillation, modified product set whence $U_{min,d} = 6$

4.4.3.3 Other Workers' Results

The result of Nath's using the modified product set which allows distillation alone to be used is shown in figure 4.21. There are seven products and we therefore expect six units. That the flowsheet should be optimal or near optimal is remarkable since there are in fact eight units. Extractive distillation with phenol is used twice, or perhaps three times since in one instance it is used in two separators before being recovered.

4.4.4 Example 4.7

This is an imaginary example to show what happens when all the products cannot be isolated using the supplied separator types. The example is based on example 4.1, shown in in figures 4.8 and 4.9, but has only two separator types and three products, two of them MCPs. The two separator types are distillation, S1, and liquid-liquid extraction, S2. The PCMs and PPMs are shown in figure 4.22.

4.4.4.1 Number of Separator Types

The combined PPM shows that product 2, (2,4), cannot be separated from product 3, (3,5) using the available separator types.

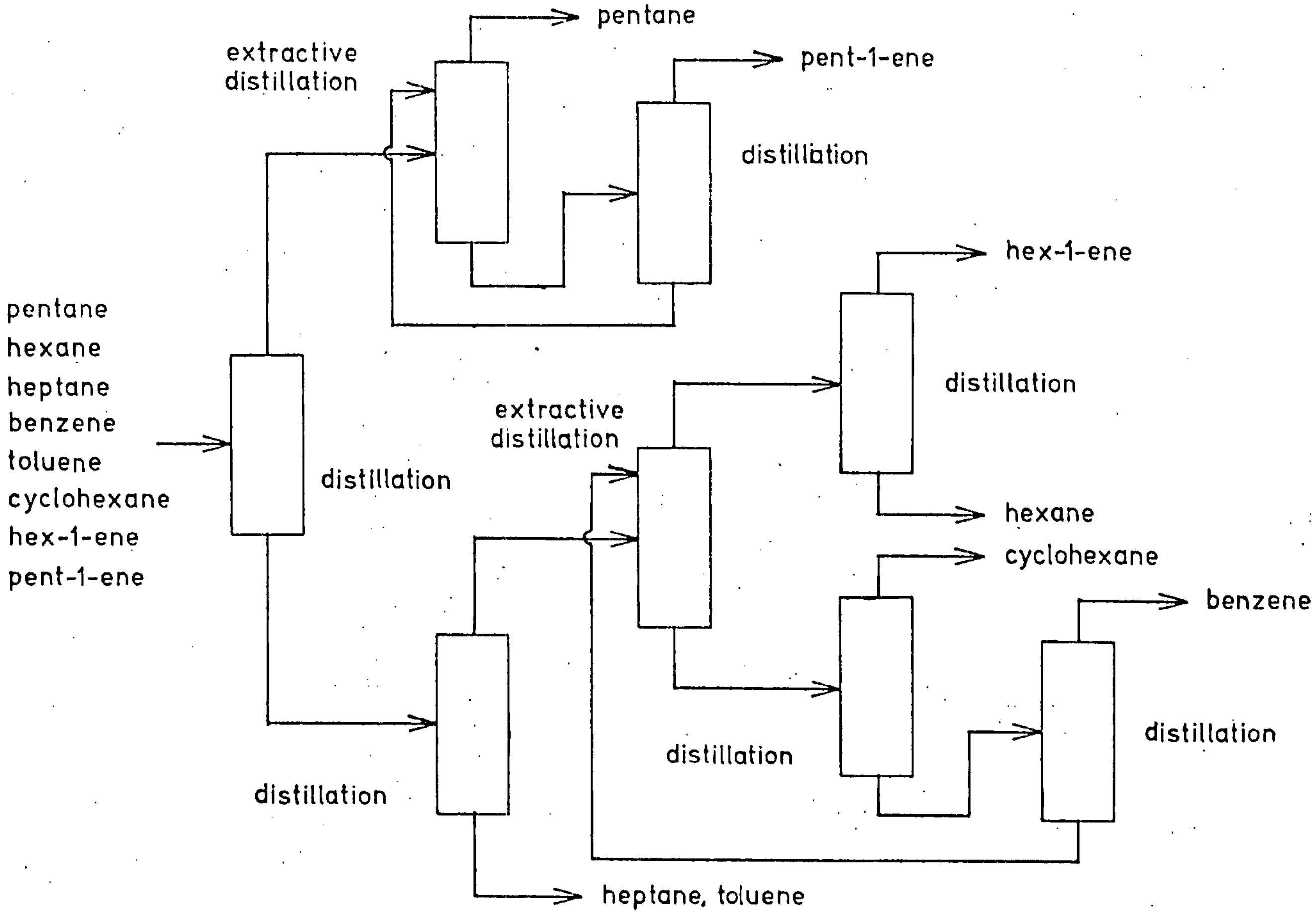


Figure 4.21 Optimal flowsheet for example 4.6, Nath's correction of Thompson and King's example 2.

```
          S1            S2

        -------       -------
       ! 1     !     ! 1     !
       !   2   !     !   2   !
PCM    !     3 !     !     3 !
       !   4   !     !     5 !
       !     5 !     !   4   !
        -------       -------

                                    S1 + S2
        -------       -------       -------
       !   1 1 !     !   1 1 !     !   1 1 !
PPM    !     0 !     !     0 !     !     0 !
       !       !     !       !     !       !
        -------       -------       -------
```

Figure 4.22 PCMs and PPMs for example 4.7, distillation and liquid-liquid extraction.

Before proceding farther the product set must be redefined by splitting one of these two products, and the PCMs suggest that product 2 be broken. The PCMs and PPMs for this new product set are given in figure 4.23. All the products may now be isolated with separator type S2 alone. If distillation, S1, is considered as the base separator type then either both it and S2 must be used or the other MCP, (3,5), must also be split.

4.4.4.2 Minimum Number of Units

Using the new product set let us now determine the minimum number of units for the immediate recovery case. N_p is three but the products cannot be separated, so the new product set gives N_p' as four, thus

$$N_p' = 4$$

$$U_{min,i} = 4 - 1 = 3$$

For the delayed recovery case the product set must be further modified to determine $N'_{p,esa}$, as shown in figure 4.24. $N'_{p,esa}$ is five. Using the MSA separator the original products are not isolable so we use the set in figure 4.23 with $N'_{p,msa}$ as four. The PPMs show that the MSA must be used once to separate (4) from (3,5). N_{msa} is therefore 1. This gives the following

$$N'_{p,esa} - 1 = 5 - 1 = 4$$

```
              S1              S2

          ---------       ---------
         ! 1       !     ! 1       !
         !   2     !     !   2     !
   PCM   !       3 !     !       3 !
         !     4   !     !       5 !
         !       5 !     !     4   !
          ---------       ---------

                                            S1 + S2
          ---------       ---------        ---------
         !   1 1 1 !     !   1 1 1 !      !   1 1 1 !
         !     1 1 !     !     1 1 !      !     1 1 !
   PPM   !       0 !     !       1 !      !       1 !
         !         !     !         !      !         !
          ---------       ---------        ---------
```

Figure 4.23 PCMs and PPMs for example 4.7, modified product set.

```
              S1

          ----------
         ! 1        !
         !   2      !
   PCM   !       3  !
         !     4    !
         !        5 !
          ----------

          ----------
         !  1 1 1 1 !
         !    1 1 1 !
   PPM   !      1 1 !
         !        1 !
         !          !
          ----------
```

Figure 4.24 PCM and PPM for example 4.7, distillation, final product set.

$$N'_{p,msa} - 1 + N_{msa} = 4 - 1 + 1 = 4$$

whence $U_{min,d} = 4$

4.4.5 Comment on Examples

The main point arising from these examples is that neither the minimum number of separator types required nor the minimum number of separation units is necessarily of value in assessing the optimality of a flowsheet. Of the three examples studied by other workers only one has the minimum number of units in the optimal flowsheet. in the other two the product set is redefined so that distillation alone may be used and then expensive distillation units are replaced by MSA separators requiring extra units. The conclusion to be drawn is that in these cases at least, MSAs are introduced not to preserve MCPs and thus minimise the number of products but to avoid expensive ESA splits. The related aims of minimising the number of products by not splitting MCPs and using the minimum number of separation units may thus be misleading targets for the engineer to aim at.

4.5 General Conclusion

The two targets available in heat exchange network synthesis, the minimum number of units and the minimum utilities required, are both useful for the designer to aim at. This chapter gives a method for determining the minimum number of separation units required in a flowsheet and a related quantity, the minimum number of separator types required. The second of these is not a particularly useful measure, but the representation introduced to find it also reveals clearly which products may or may not be isolated using particular separator types. This information can be used in the search for previously unconsidered separator types which might be usefully included in a flowsheet. The examples considered in this chapter reveal that the minimum number of units and the minimum number of separator types are not particularly good guides to process optimality. In the next chapter functions are considered which might give a good correlation with cost in simple distillation sequences.

5 Evaluation Functions in Ternary Distillation

5 Evaluation Functions in Ternary Distillation

5.1 Introduction

5.1.1 Targets in Heat Exchange Networks

To recap, work by Linnhoff et al [24,31] (based on earlier work by Hohmann [26] and Nishida et al [25,30]) gives two targets for the designer of heat exchange networks to aim at:-

-- the minimum number of heat exchange units that a network need comprise;

-- the minimum consumption of hot and cold utilities that a network need use.

Synthesis of networks can be done either by traditional methods using these targets as design aids, or by using systematic design methods based specifically on the targets. The essence of the method is the heuristic rule that any network which meets these goals will be 'better', that is cheaper and simpler than one which does not. This view is justified because the quantities which the targets reflect correlate closely with cost. Because of economy of scale a network of a given heat exchange area will be cheaper if that area is spread across fewer rather than more heat exchange units, and thus networks with the minimum number of exchangers tend to be cheaper. More importantly a network which uses more than the minimum amounts of hot and cold utilities not only incurs higher heating and cooling costs but also has to convey larger quantities of heat throughout. This will require larger than minimum heat transfer areas if temperatures are to be kept constant. The two targets are also useful because they are simple to calculate and in the vast majority of cases can both be achieved for real networks.

5.1.2 Targets in Separation Schemes

In separation scheme synthesis there are as yet no corresponding targets for a designer to aim for. It is shown in the preceding chapter that the number of separation units does not correlate well with cost. This is because separation processes are in some aspects simpler and in others more complex than heat exchange processes. Thus for systems using only distillation and separating reasonably ideal mixtures it is difficult to conceive flowsheets with other than a fixed number of simple separation units, and yet the different configurations may exhibit a wide range of costs. At the other end of the scale if several separator types are used then it may be that an arrangement using several stages of MSA addition and recovery may be significantly cheaper than a single distillation column designed for the same duty. The simple distillation problem is simpler than the heat exchange problem in that a smaller range of equipment configurations is possible, and yet each 'unit' is much more complex, leading to a wider range of costs. The situation with multiple separator types is even more complex since each unit may utilise any one of a range of phenomena to drive the separation, as against only one driving force for heat exchange or distillation, namely temperature difference or relative volatility respectively.

5.1.3 Aims of This Work

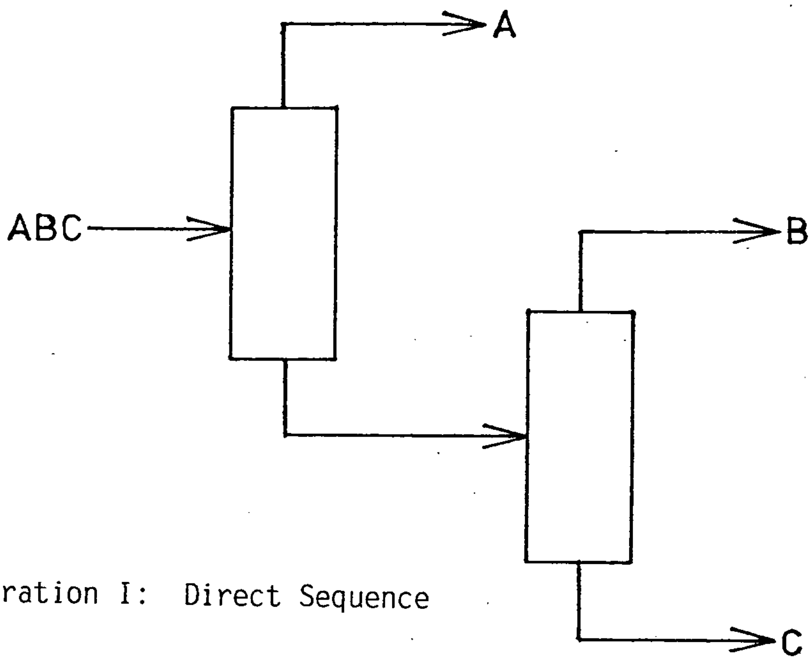
This particular section of work was begun with rather hazy ideas of discovering functions which could be used as alternatives to cost in the design of distillation systems. As it progressed it became apparent that a less empirical approach might be more useful. The final results are therefore valued more for revealing what features cost depends on rather than for any close approximation to the costing function in the situations studied. This is just as well for two reasons. The first is that those functions which best approximate cost are almost as complex to calculate as cost itself and therefore will give little saving of effort in an evaluation procedure. The second reason is that a cost estimation procedure is an approximation to the real cost of a project only in a given set of economic circumstances. An alternative evaluation function is therefore an approximation to this approximation and the chances of its maintainng a good correlation with cost as economic circumstances change may therefore be rather low. To design good distillation systems consistently requires a more fundamental understanding of the problem. Perhaps inevitably this chapter comes to focus on energy use as the significant factor in distillation design and the next chapter considers some further aspects of this finding.

5.1.4 Method

The tactic used in this work was to plot contours of the difference in cost of the two simple ternary schemes, the 'direct' and the 'inverted' sequences (figure 5.1) on a ternary diagram, the coordinates being the mole fractions of feed components to the system, and to compare the results with those given by using functions other than cost. Some of these were simple functions either used by other workers or devised for this work. Various other functions based on reflux ratio or reboil rate were also tested. The idea of plotting "regions of optimality" for different separation schemes for differing feeds on a ternary diagram came from Tedder and Rudd [39,40]. However, this study differs in several ways from theirs - they studied eight different column configurations, this study uses only two; they considered cost estimates and attempted to derive a list of heuristic design rules, this work studies several different evaluation functions to see which might shed some light on the design process, or might even be used for evaluation as an alternative to costing.

The reason why ternary systems using only two simple column configurations were studied were these.

-- To allow comparison of these results with those of Tedder and Rudd, and possibly earlier workers, see the Appendix on Costs of Ternary Distillation.



Configuration I: Direct Sequence

Configuration II: Inverted or Indirect Sequence

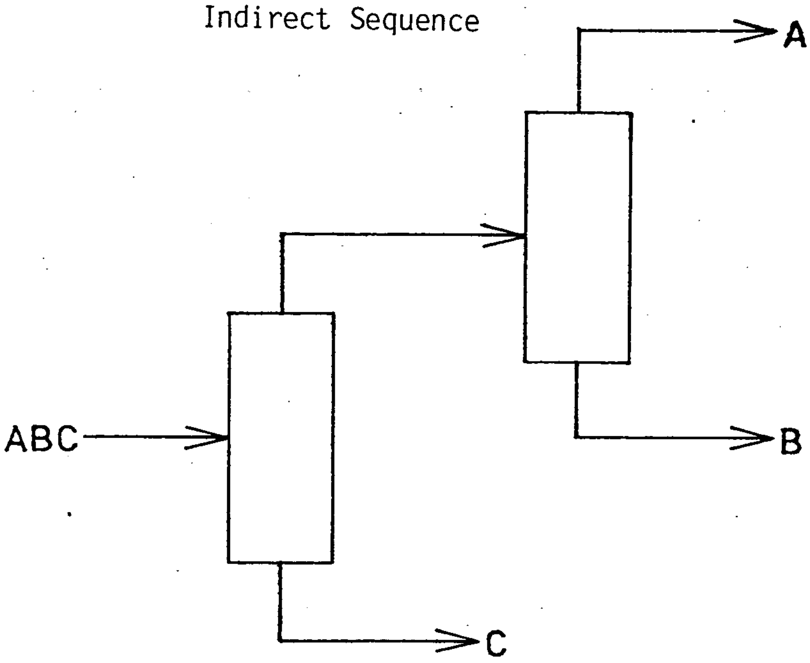


Figure 5.1 Simple ternary distillation sequences.

- With a ternary system the results may be correlated with the feed composition and presented graphically on a triangular diagram. With more than three components this is not be possible, making the interpretation of the results more difficult.

- With only two column configurations (and a computer program which draws contours) it is possible to see which is the cheaper for any given feed simply by drawing the zero contour of the cost on the ternary diagram "map". A larger number of configurations would have required a more complex method with which it would not have been easy to compare different functions.

5.2 Results

This discussion of results is divided into four sections. First are some brief general notes followed by a short section on the cost results, covered more fully in the Appendix, second a section on functions related to external flows, the entropy of choice and those based on mass flow, and finally a section on the reboil related functions.

5.2.1 Notes on Results and Presentation

5.2.1.1 Feed Mixtures, ESI and MESI

The ternary mixtures used as feeds in this study are four of the six hydrocarbon mixtures used by Tedder and Rudd (number 1 and numbers 4 to 6 in this study) together with others using the same set of components in different combinations. Details of the feed mixtures are given in table 5.1. The indeces used to characterise the feeds are the Ease of Separation Index (ESI) and the Modified ESI (MESI). ESI is used by Tedder and Rudd and is defined as the ratio of the relative volatility of the lighter pair of components to that of the heavier pair, thus

$$\mathrm{ESI} = \frac{\alpha_{AB}}{\alpha_{BC}} = \frac{K_A/K_B}{K_B/K_C} = \frac{K_A \cdot K_C}{K_B^2}$$

where A is the lightest component, C the heaviest. It is presumably intended as a measure of the relative difficulty of the two splits. However, as noted below in section 5.2.3.4, the relative volatility does not provide an intuitively satisfactory measure of the difficulty of a separation. This is because as the relative volatility tends to unity a separation becomes infinitely difficult. The relative volatility therefore does not match the trend of cost. A possible alternative is to use the excess relative volatility, $(\alpha - 1)$, which tends to zero as α tends to 1. If this quantity is used to characterise the difficulty of separations the ratio of difficulties in ternary separations analogous to the ESI is the MESI defined as -

Table 5.1

Components Present	Feed number 1	2	3	4	5	6	7	8
propane						✓		
i-butane			✓		✓	✓	✓	✓
n-butane		✓	✓		✓	✓		
i-pentane		✓		✓	✓			✓
n-pentane	✓	✓		✓				
n-hexane	✓		✓	✓			✓	✓
n-heptane	✓						✓	

Characterisation Indices	Feed number 1	2	3	4	5	6	7	8
MESI	1.07	4.93	0.06	0.15	0.28	3.63	6.41	1.01
ESI	1.04	1.86	0.18	0.47	0.59	1.72	4.22	1.01
HI	0.27	0.14	0.71	0.71	0.60	0.17	0.15	0.25
a_{AB}	2.57	2.35	1.36	1.25	1.38	2.38	10.42	3.24
a_{BC}	2.47	1.28	7.52	2.65	2.35	1.38	2.47	3.31
a_{AC}	6.35	3.05	10.23	3.31	3.24	3.28	25.73	10.74

$$\text{MESI} = \frac{\alpha_{AB} - 1}{\alpha_{BC} - 1}$$

Feeds numbers 7 and 8 were chosen to replace Tedder and Rudd's numbers 2 and 3 because they gave a better spread of MESI.

5.2.1.2 Column Design and Product Specification

The column design and costing method used was that of Rathore et al [52,53], details of which are given in table 5.2. The feature of the designs not covered there is the relationship of the product streams to the feed stream in each column. Two distinct conventions were used. The results marked 'recoveries', figures 5.8(b)-13(b), were obtained by specifying 95% recovery of key components and 100% recovery of non-keys. Thus the tops stream from a column will contain 95% of the light key fed to the column, 5% of the heavy key and any light non-key which might be present. The rest of the results were obtained by specifying the concentration of the key components in the 'wrong' product: light key in the heavy product and heavy key in the light product. The top product in these cases will consist of any light non-key, a maximum of 1% mole fraction heavy key and the remainder being that light key not forming a 1% impurity in the bottom product.

5.2.1.3 Presentation of Results

The ternary diagrams shown here are right angled and are

Minimum number of stages

$$\left(\frac{\text{kg moles of component } i \text{ in distillate}}{\text{kg moles of component } i \text{ in bottoms}}\right) = (\alpha_{i\text{-}r})^{S_m} \left(\frac{\text{kg moles of component } r \text{ in distillate}}{\text{kg moles of component } r \text{ in bottoms}}\right)$$

$\alpha_{i\text{-}r}$ value is geometric mean of the values at the column top and bottom.

Minimum reflux ratio

$$\sum_{J=1}^{N} \frac{\alpha_i Z_{i,F}}{\alpha_i - \theta} = 1 - \phi$$

$$R_m = \sum_{J=1}^{N} \frac{\alpha_i X_{i,D}}{\alpha_i - \theta} - 1$$

Number of stages

For $\left[\frac{(R - R_m)}{(R+1)}\right] < 0.125;\ \left[\frac{(S - S_m)}{(S+1)}\right] = 0.5039 - 0.5968\left[\frac{(R - R_m)}{(R+1)}\right] - 0.0908 \log\left[\frac{(R - R_m)}{(R+1)}\right]$

and for $\left[\frac{(R - R_m)}{(R+1)}\right] > 0.125;\ \left[\frac{(S - S_m)}{(S+1)}\right] = 0.6257 - 0.9868\left[\frac{(R - R_m)}{(R+1)}\right] + 0.5160\left[\frac{(R - R_m)}{(R+1)}\right]^2$

$- 0.1738\left[\frac{(R - R_m)}{(R+1)}\right]^3$

Diameter of the column

$$D_c = \left[\left(\frac{4}{\pi V}\right)(D)\,(R+1)\,(22.2)\left(\frac{T_{DV}}{273}\right)\left(\frac{1}{P}\right)\left(\frac{1}{3600}\right)\right]^{1/2}$$

where

$$V = .761\left(\frac{1}{P}\right)^{1/2}$$

Height of the column

$$H_c = 0.61\left(\frac{S}{\eta}\right) + 4.27$$

Cost basis of optimization:

$$\text{Total annual cost} = \text{annual operating cost} + \frac{\text{total installed equipment cost}}{\text{Project life}}$$

Column cost:

$$\text{Installed cost of the column} = 4.34\left[762\,D_c\left(\frac{H_c}{12.2}\right)^{.68}\right]$$

If the column pressure is more than 3.4 atm. a correction factor

$[1 + 0.0147\,(P - 3.4)]$ is applied.

Cost of trays

$$\text{Installed cost of trays} = 70\left(\frac{S}{\eta}\right)\left(\frac{D_c}{1.22}\right)^{1.8}$$

Instrumentation cost:
column instrumentation cost = 4,000.00
Maintenance cost of the column
2% of the total installed cost of the column
Heat exchanger cost:

$$\text{Total installed cost} = 3.39\left[9000\left(\frac{A}{92.1}\right)^{.65}\right]$$

If the pressure is more than 10.2 atm. a correction factor
of $[1 + 0.0147\,(P - 10.2)]$ is applied.
Heat exchanger operating cost = 8500 $(C_u Q)$ + 2% of the total installed cost.
Material of construction: carbon steel

Assumed values:

η = 80% in all the columns
$R = 1.2 \times R_m$

Feed condition = saturated liquid feed for all columns
Operating hours in a year = 8500
Project life = 10 years

Table 5.2 : Design equations from Rathore et al [53]

always presented with 100% most volatile component (referred to as A) in the top corner, 100% middle component (B) at the right angle and 100% least volatile component (C) at the bottom right. These points will be referred to as the A, B and C corners respectively. The edges joining them are the AB, AC and BC edges, AC being the hypotenuse. This format results in some stretching of scale along the AC edge.

5.2.1.4 Evaluation Functions Considered

Following is a summary of the evaluation functions used in the study. In each case the value of the function for the two column ternary system is the sum of the values for each column.

i) Cost: The design and costing method used by Rathore et al [52,53] as discussed above. Cost is in 1969 US dollars.

ii) Mass Load or Total Mass Load: The sum of the feed flowrates to the individual separators in a network. It has been used as an evaluation function by Mahalec and Motard [35,36].

iii) Entropy of Choice: A function similar in form to the theoretical minimum separative work required for a separation, and used as an evaluation function by Maikov [51]. Minimum separative work is discussed by King [45]. It is based solely on the distribution of mass through the system, not on any physical properties.

iv) Mass Load / Relative Volatility: The column feed flow rate divided by the relative volatility of the key components. This was used by Johns and Romero [37,65] as an evaluation function in their simple synthesis method.

v) Mass Load / Excess Relative Volatility: As iv) but modified due to the suspicion that the difficulty of a separation depends not on the relative volatility of the keys but on its difference from unity. 'Excess relative volatility' is the quantity (α - 1) where α is the relative volatility, see above.

vi) Reboil Related Functions: Two functions connected with internal vapour rate: the actual vapour rate in kmol per second and the reboiler duty in kilowatts.

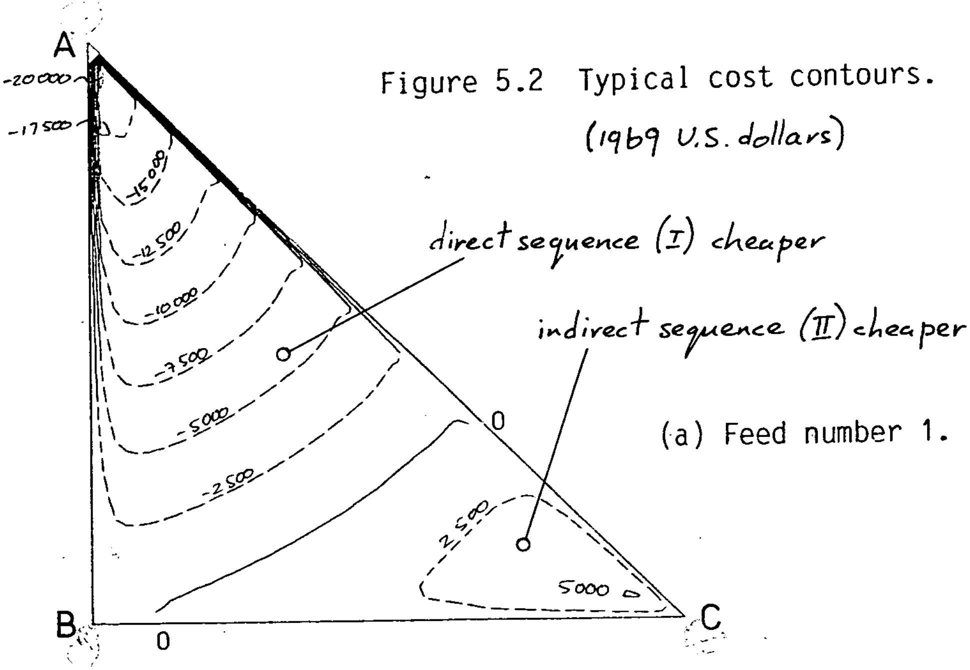
Two other functions were considered, but were rejected at an early stage. These were: minimum reflux ratio and reboil rate in units of the feed flowrate of key components to the column. The reasons for their inclusion and rejection are considered under 'Reboil Related Functions' below.

5.2.2 Cost Results

Figures 5.2(a)-(c) show typical results for cost difference between the two distillation designs, that is, the cost of the 'direct sequence' minus the cost of the 'inverted sequence', shown

Figure 5.2 Typical cost contours.

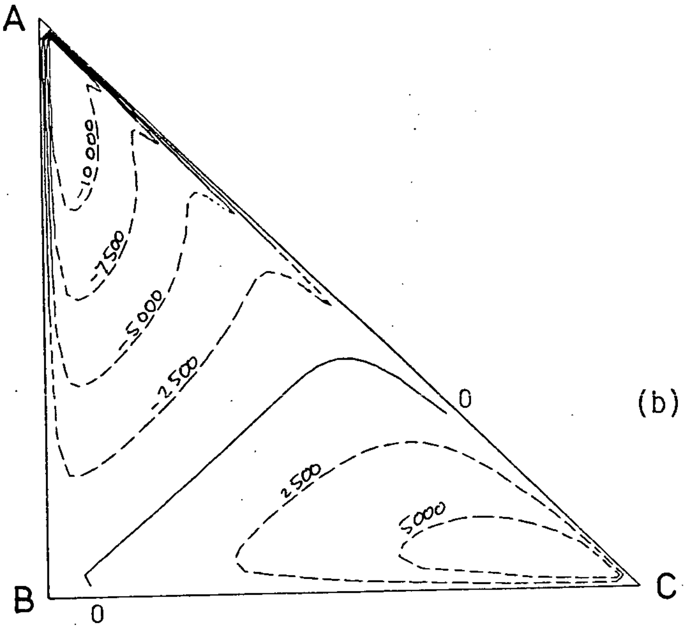
(1969 U.S. dollars)



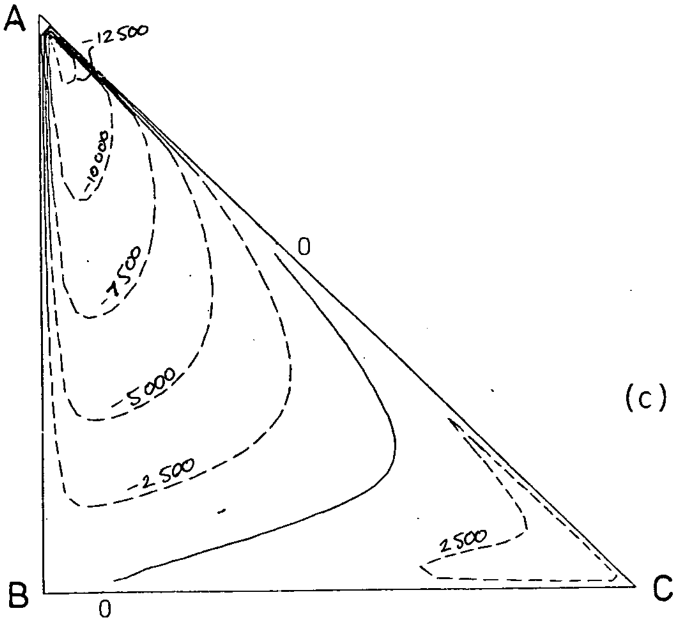
direct sequence (I) cheaper

indirect sequence (II) cheaper

(a) Feed number 1.



(b) Feed number 5.



(c) Feed number 6.

in figure 5.1. The zero contour is marked with a solid line and the surface is depressed towards the A corner, showing an advantage for configuration I, the direct sequence in that region, and raised toward the C corner showing an advantage for configuration II, the inverted sequence. This is to be expected as the direct sequence is favoured when there is a larger amount of light component in the feed. The results shown are for feeds no.1, no.5 and no.6, with ESIs of 1.04, 0.59 and 1.72 respectively, and MESIs of 1.07, 0.28 and 3.63. The designs are for systems where the reboiler steam was assumed to be available at any desired temperature rather than at a set of fixed levels (for further discussion of this point see the Appendix). They are included here as examples of the general form of the cost contours which an evaluation function must be expected to match. An important point to note is that even for ESI or MESI of approximately one the direct sequence is favoured over more of the diagram. This suggests that it is more expensive to take material overhead, since the main difference in such cases is in the amount of overhead product from the first column in the sequence.

5.2.3 External Mass Flow Based Functions

These functions are referred to as 'external mass flow based' because they are calculated from the external flows to and from a column, the feed and product stream flowrates. More particularly they are not related to any internal vapour or liquid flows in a column.

5.2.3.1 Mass Load

Simply the sum of the feed flowrates to the two columns, this function is completely linear over the whole ternary diagram. As shown in figure 5.3 the zero contour bisects the diagram from the B corner to the BC edge. It is at best only a rough guide to the choice of configuration since it is not affected by any change of species in the feed.

5.2.3.2 Entropy of Choice

This function, used by Maikov [51], is calculated from the fraction of the feed which appears in the overhead product, ϵ, and is defined as

$$H = -\epsilon \log \epsilon - (1 - \epsilon) \log (1 - \epsilon)$$

where the logarithms are to the base 2. (For comparison purposes the logs may be taken to any convenient base.) The basis for its use is that it is similar in form to the expression for the minimum theoretical separative work (discussed by King [45], p661ff, as noted above). Unlike the other functions studied here H should be maximised, so a preference for the direct sequence in figure 5.4 is shown as a high area. This area lies toward the A corner. The zero contour bisects the triangular diagram so, at least for ternary systems, this function shows no superiority to mass load, despite the claims made for it by Maikov. One

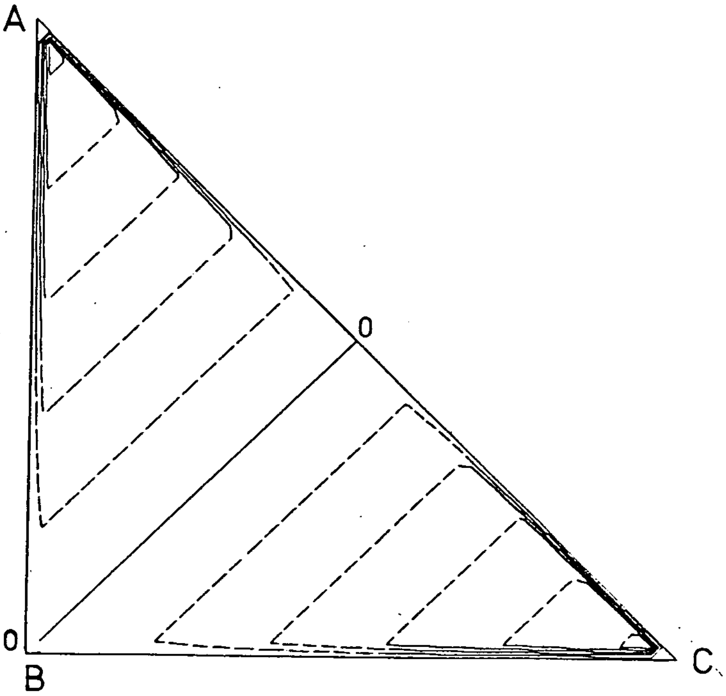


Figure 5.3 Mass load.

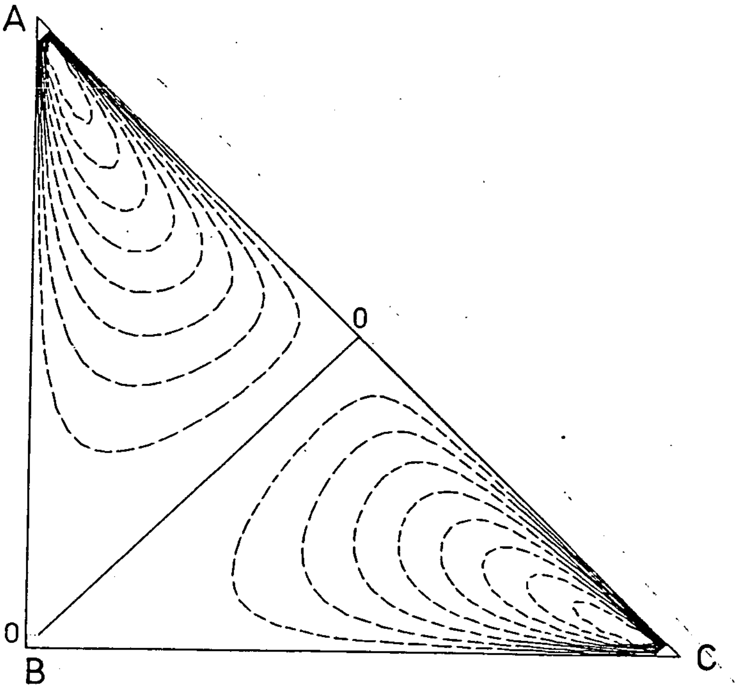


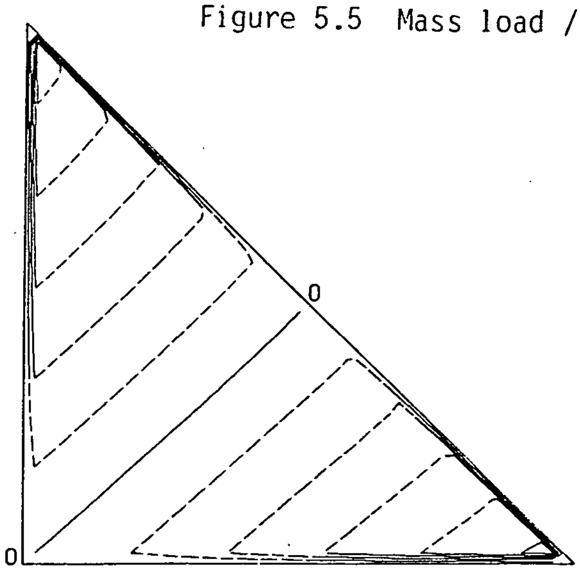
Figure 5.4 Entropy of choice.

possibility which has not been explored is that the entropy of choice might be used as the basis of a more complex evaluation function. The fact that its contours are curved rather like those of cost suggests that it might be quite good in this role.

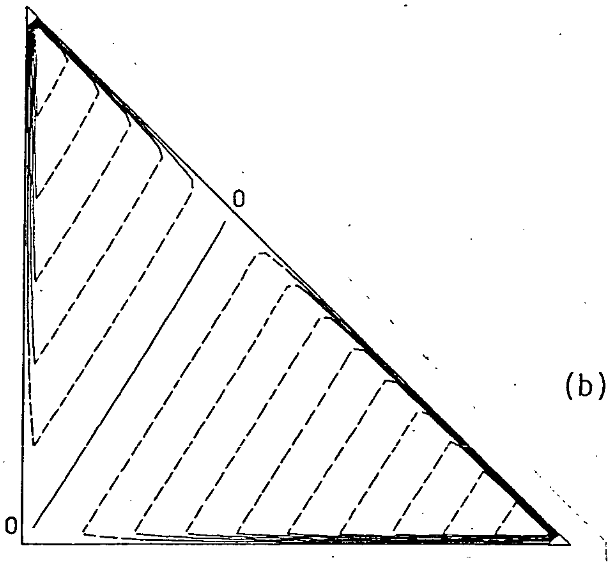
5.2.3.3 Mass Load / Relative Volatility

This function represents an attempt to modify the mass load function to allow for the different difficulties of the separations involved. Feed flowrate divided by the relative volatility of the key components is summed over all the columns in the flowsheet. Johns and Romero [37,65] used variations on this theme, using other relative separation factors for other separator types, in a quite powerful synthesis program. The function has two main drawbacks, however. Firstly it does not show the curvature of the cost function -- compare figure 5.5(a)-(c) with figure 5.2(a)-(c). The reason for this is that the relative volatilities of the components stay almost constant as the design conditions vary with feed composition so the function remains almost linear. Secondly it does not reflect the apparent extra cost of taking a product overhead which results in the zero contours for cost difference being pushed towards the BC edge. Thus for a case with ESI near to unity the zero contour for mass load / α is near the centre of the diagram whereas the zero contour of cost difference is not: compare figures 5.2(a) and 5.5(a).

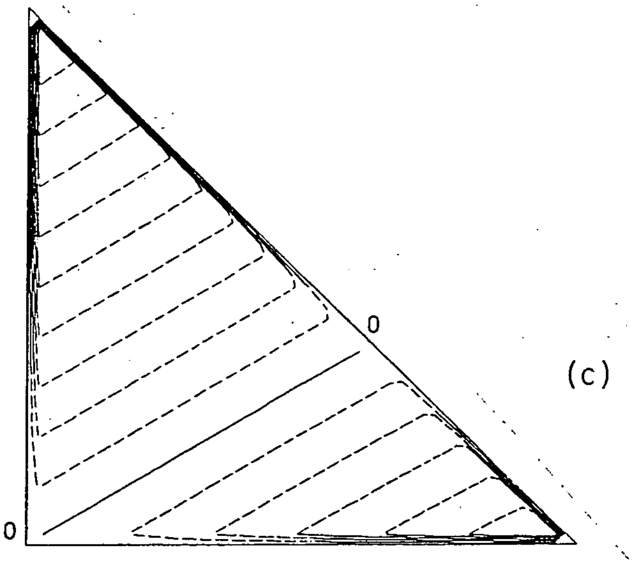
Figure 5.5 Mass load / relative volatility.



(a) Feed number 1.



(b) Feed number 5.



(c) Feed number 6.

5.2.3.4 Mass Load / Excess Relative Volatility

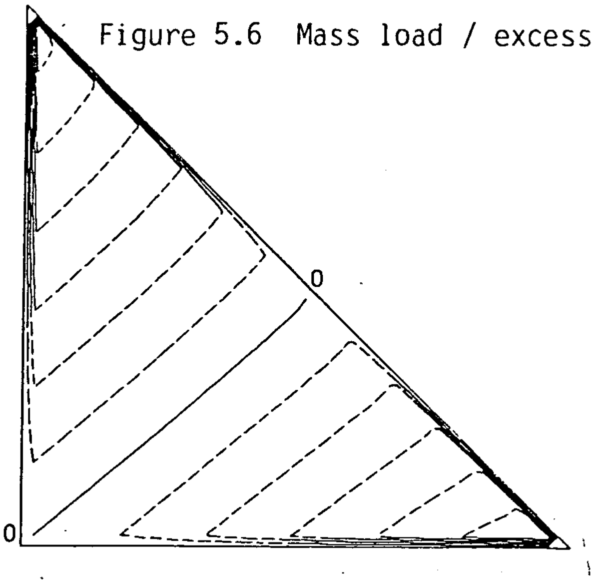
Figures 5.6(a)-(c) show mass load / (α - 1) for feeds number 1, 5 and 6. The use of this function was prompted by the observation that the difficulty of a split ought to be determined by the excess relative volatility, (α - 1), rather than α, the relative volatility, since as α tends to the all too finite value of 1 the split becomes infinitely difficult. The main points to note about these results are however

-- that the added cost of taking product overhead is still not allowed for: the zero contours for the different feeds are still centred on the B to AC bisector;

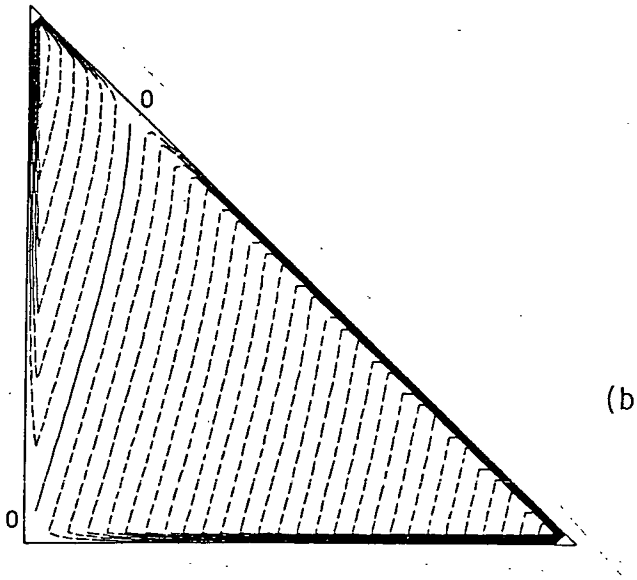
-- the effect of using (α - 1) rather than α is too extreme: comparison of the relative positions of the cost, mass load / α and mass load / (α - 1) zero contours shows that this function pushes them too far;

-- the contours show a little more curvature than the ones for mass load / α, presumably due to any change of α with temperature being exaggerated in (α - 1). In any case any such curvature would only occur in detailed design calculations since change of relative volatility with temperature is rarely considered in simple calculation.

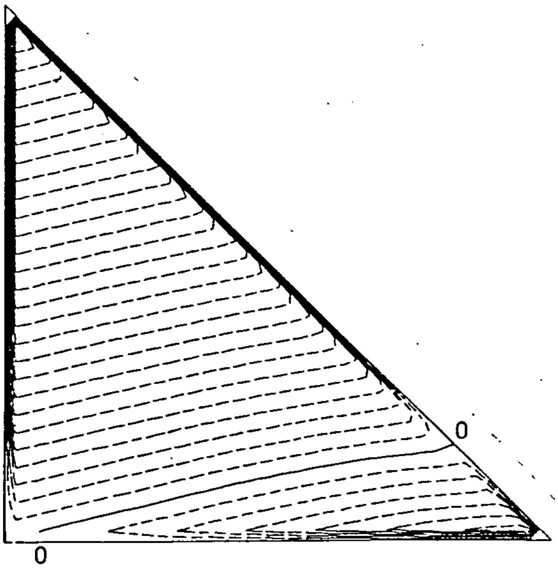
Figure 5.6 Mass load / excess relative volatility.



(a) Feed number 1.



(b) Feed number 5.



(c) Feed number 6.

5.2.4 Reboil Related Functions

5.2.4.1 Introductory Notes

Several workers, notably Harbert [48], Rod and Marek [49], Bakhshi and Gaddy [44] and King [45], point to energy use being responsible for a large part of the cost of a distillation system. Rod and Marek (1958) say that from 83% to 86.5% of the total cost of the systems they studied was due to the cost of reboiler steam; King (1981) suggests about 70%; Harbert in 1958 makes the assumption that since the cost of heat forms such a large part of the cost of distillation then it might as well be the only cost! These observations lead to the suggestion that an evaluation function based on the reboil rate might correlate well with cost. Further yet, what about using the reflux ratio, even the minimum reflux ratio (see later notes on the optimisation of designs), since vapour rate in the column depends closely on this? In the event using the minimum reflux ratio proved a dismal failure, compare figure 5.7(a) with figures 5.2(a)-(c). The other function considered and rejected was the internal vapour rate expressed in units of the feed rate of the key components. This function is used in Chapter 6, but as figure 5.7(b) shows it gives very poor results here. The other reboil related functions investigated were the internal vapour rate in kmol/s and reboiler duty in kW. Details of these results and their comparison with cost are given below.

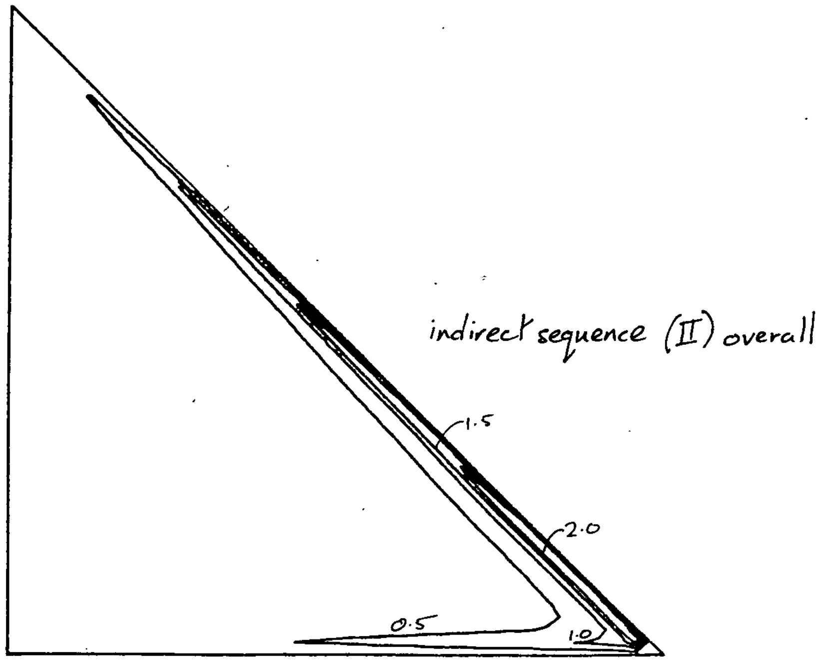


Figure 5.7(a) Minimum reflux ratio.

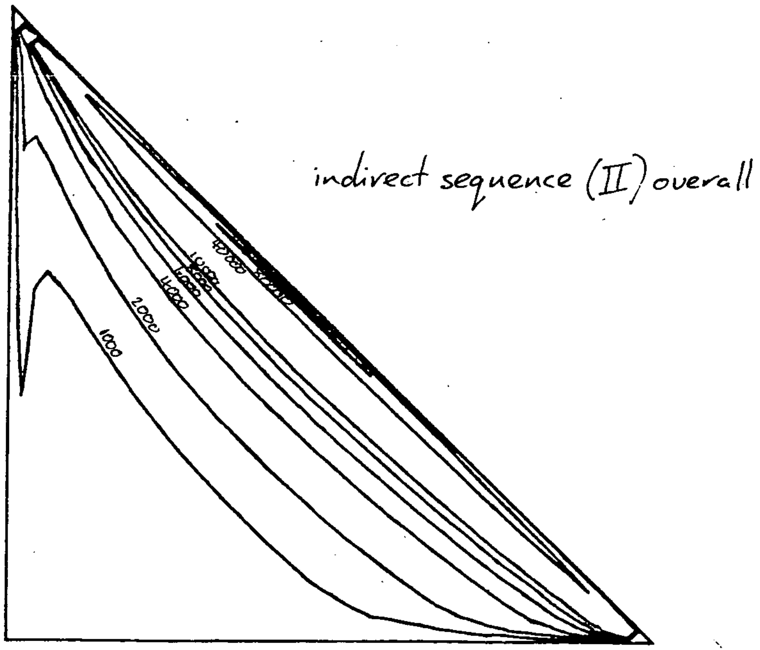


Figure 5.7(b) Internal vapour rate in units of feed rate of key components.

5.2.4.2 Reboil Related Function Results

Figures 5.8 to 5.13 show the zero contours of cost, vapour rate and reboiler duty for feeds number 1, 4, 5, 6, 7, and 8. Number 7 was chosen instead of Tedder and Rudd's number 2 since it gave a better spread of MESI in the results, and number 8 was used to try to provide a close neighbour to feed number 1 in terms of MESI. Figures 5.14 to 5.16 show comparisons for each function across the set of feeds. Each figure has two versions, (a) and (b). Version (a) gives results where the columns are designed for a 1% mole fraction of either key in the 'wrong' product. It was felt that this might give rise to undesirable edge effects as the feed concentration of a key component fell towards the edge of the diagram where there would not be enough of the component present to provide 1% in the wrong product. The diagrams labelled (b) are the 'recoveries' versions as described above, generated from designs requiring 95% of the key components to be recovered in the 'correct' product. This was expected to give smoother composition changes across the diagram.

The effects of this change of design are twofold. Firstly, as expected, various squiggles in the contours towards the edges of the diagram are smoothed out, for instance compare the cost contours in figures 5.9(a) and (b), and also most of the others. Secondly, and possibly connected, some of the squiggles observed in the (a) diagrams are where the contour heads towards the B corner and then turns sharply away: almost all of the contours seem to head toward the B corner. In the recoveries versions, (b), the contours,

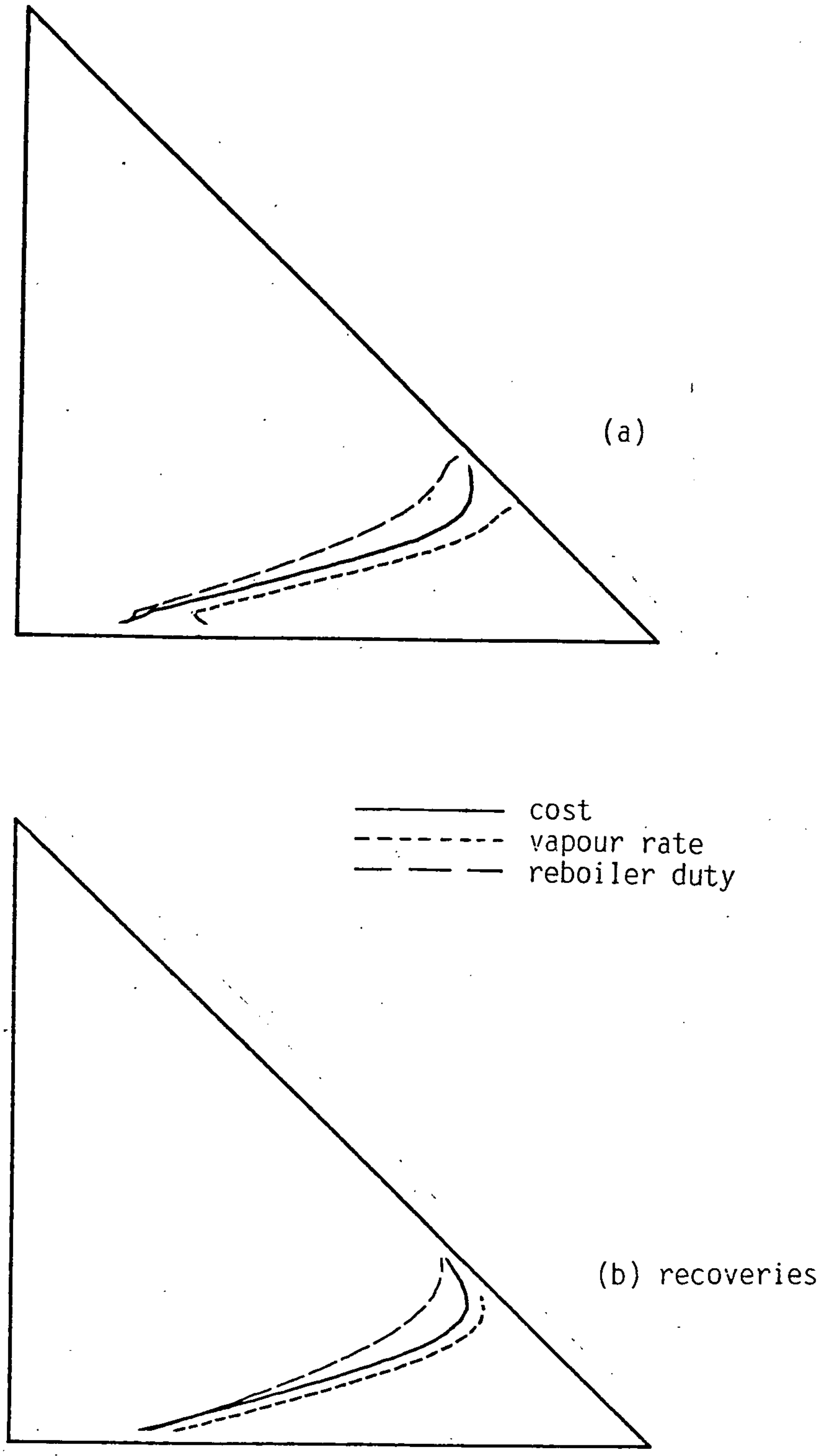


Figure 5.8 Comparison of zero contours for feed number 1.

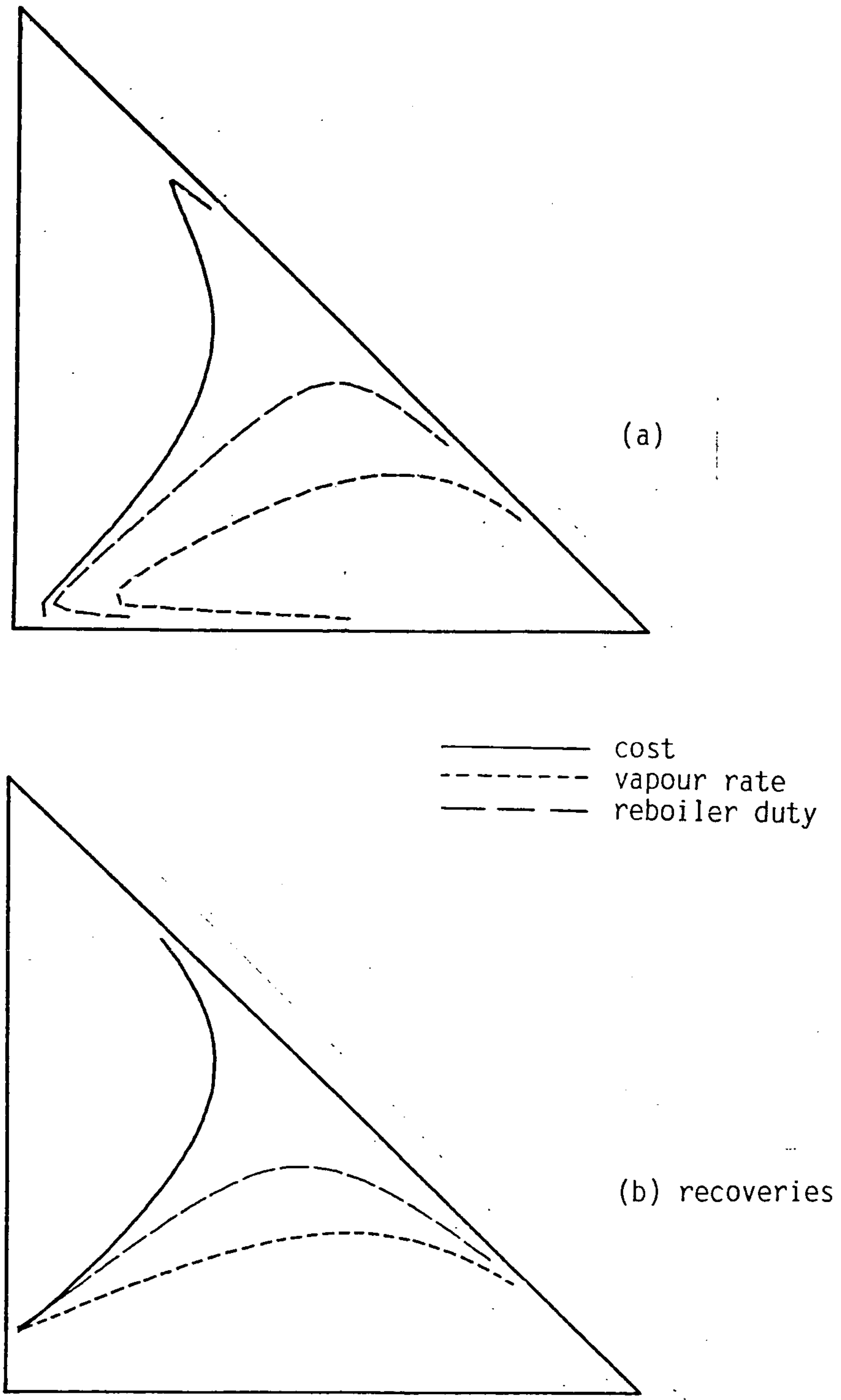


Figure 5.9 Comparison of zero contours for feed number 4.

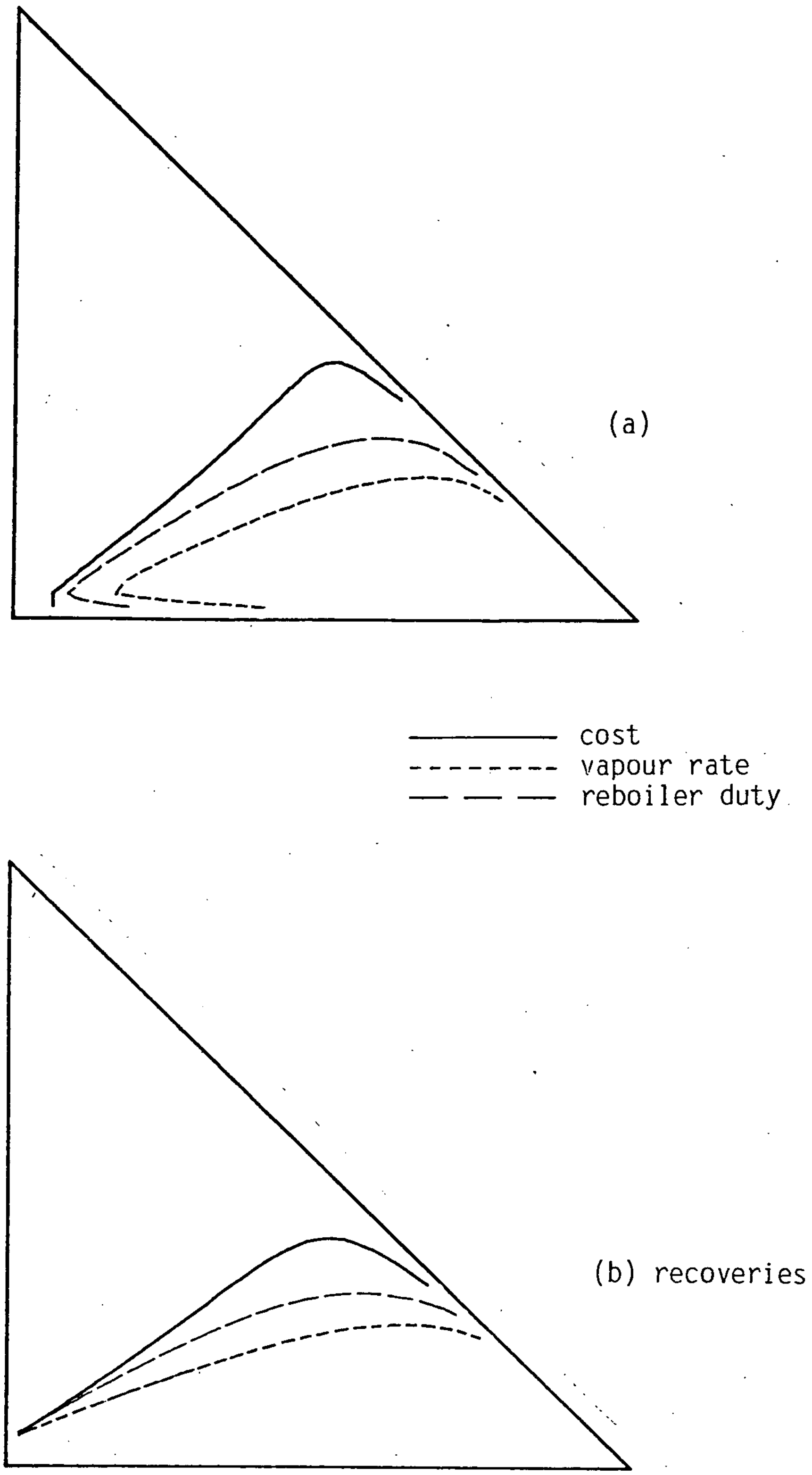


Figure 5.10 Comparison of zero contours for feed number 5.

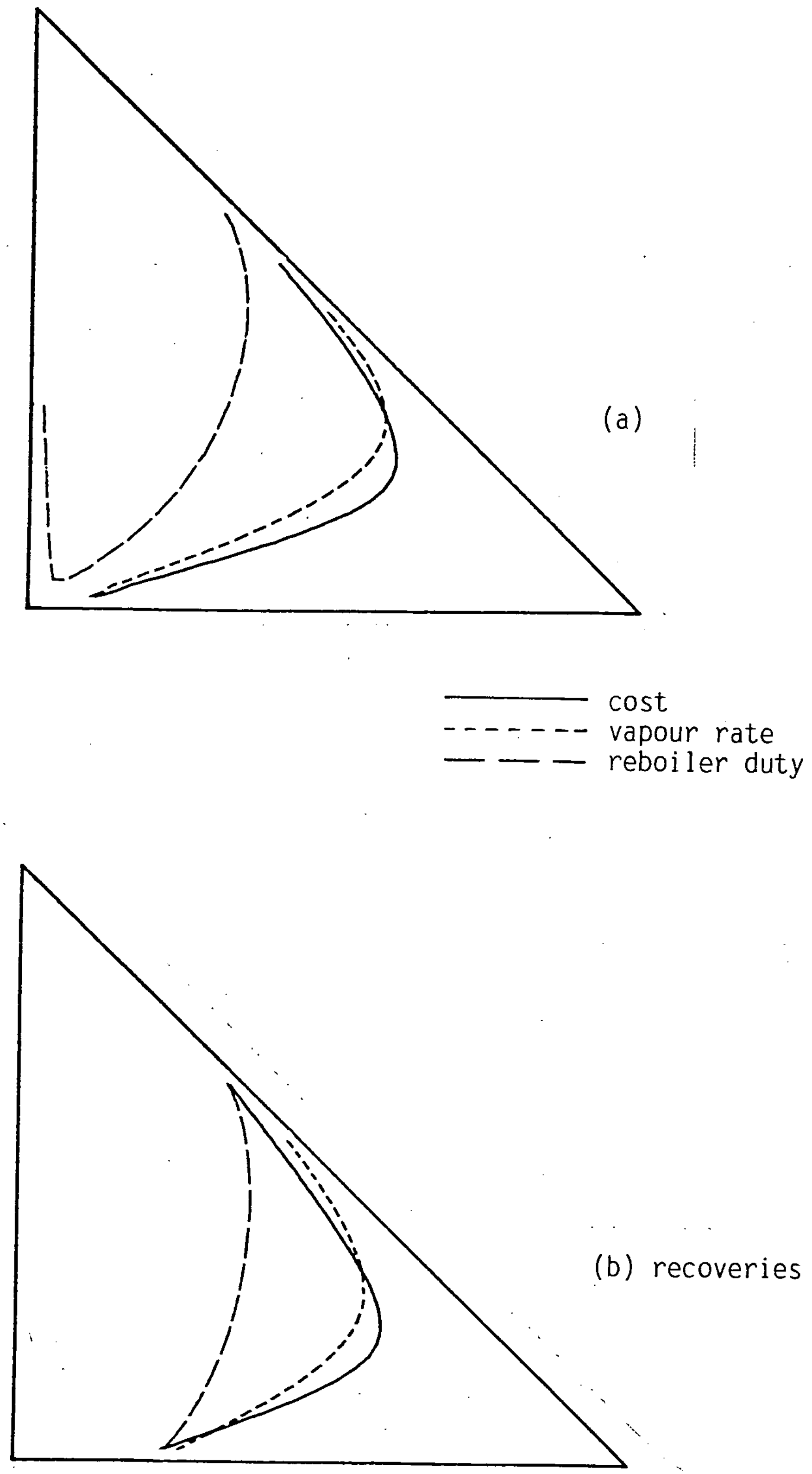


Figure 5.11 Comparison of zero contours for feed number 6.

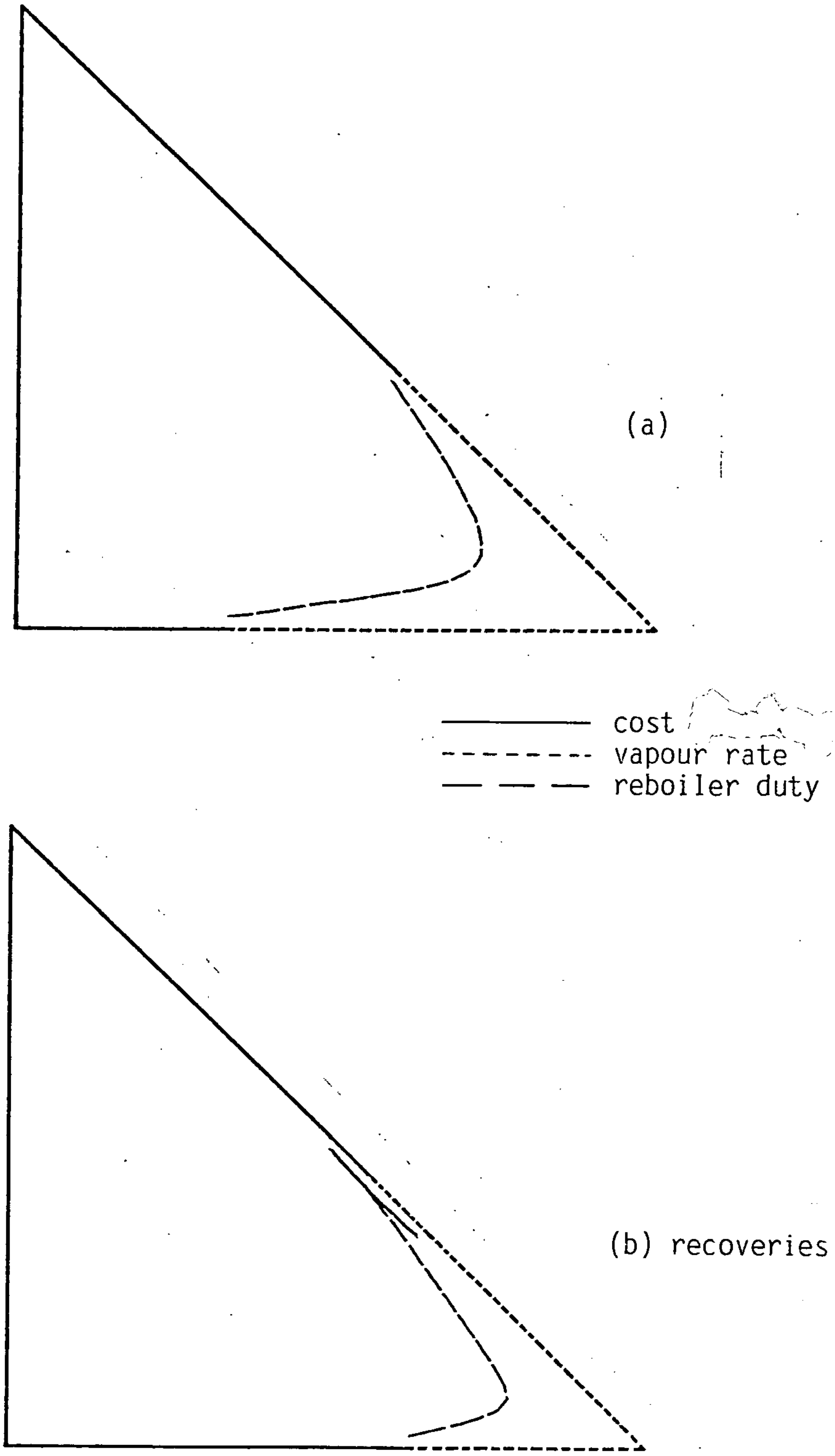


Figure 5.12 Comparison of zero contours for feed number 7.

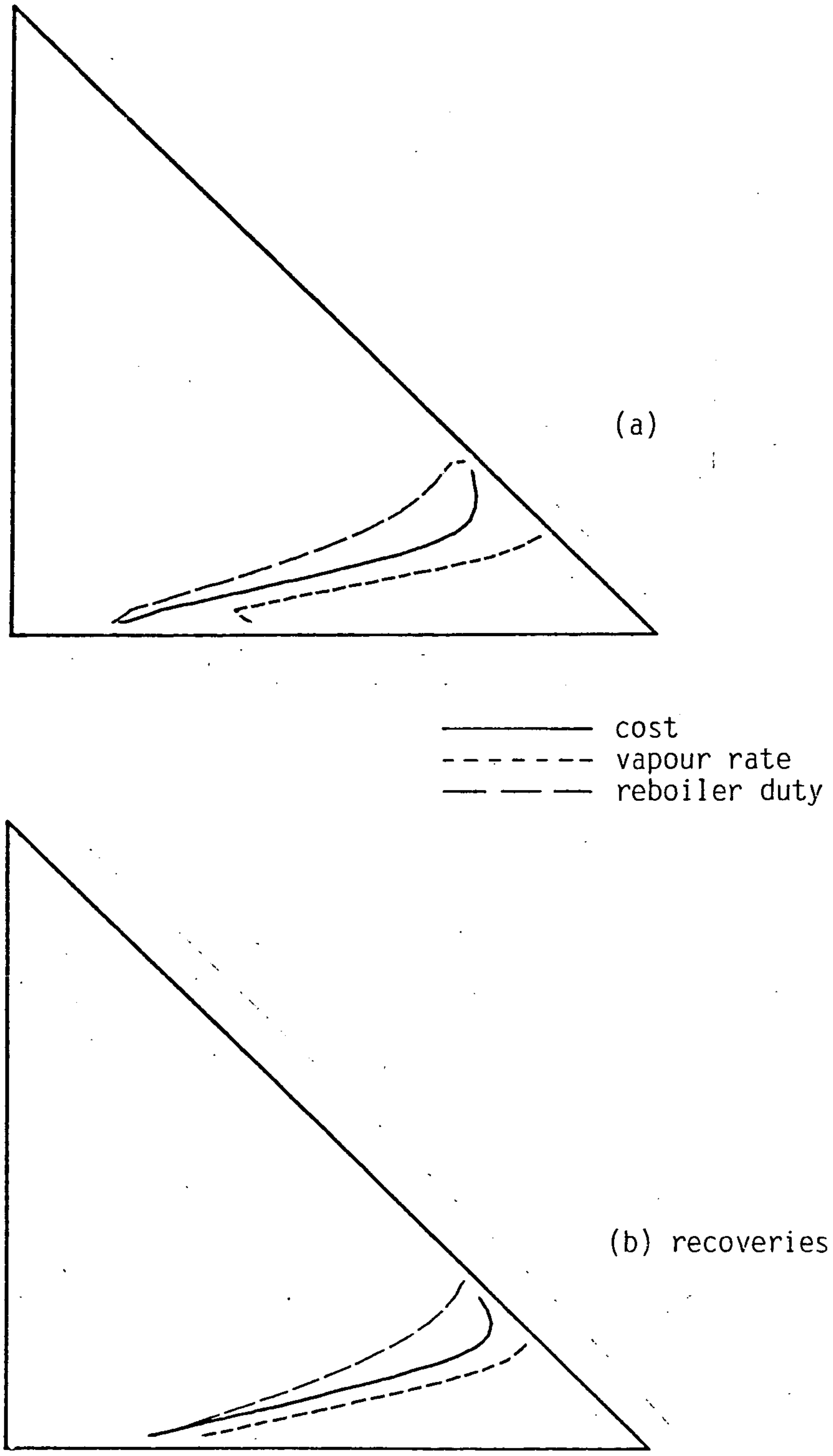
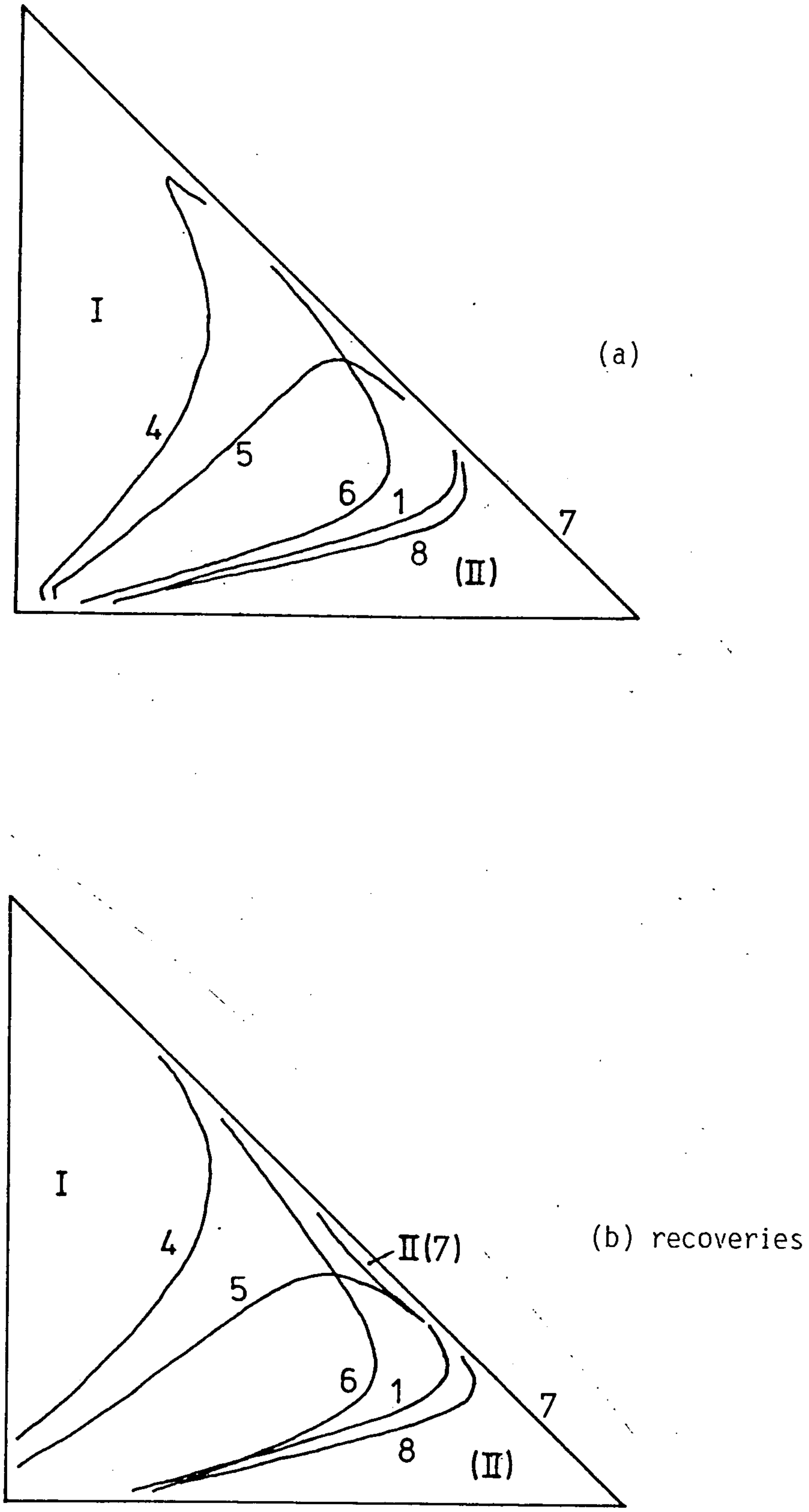


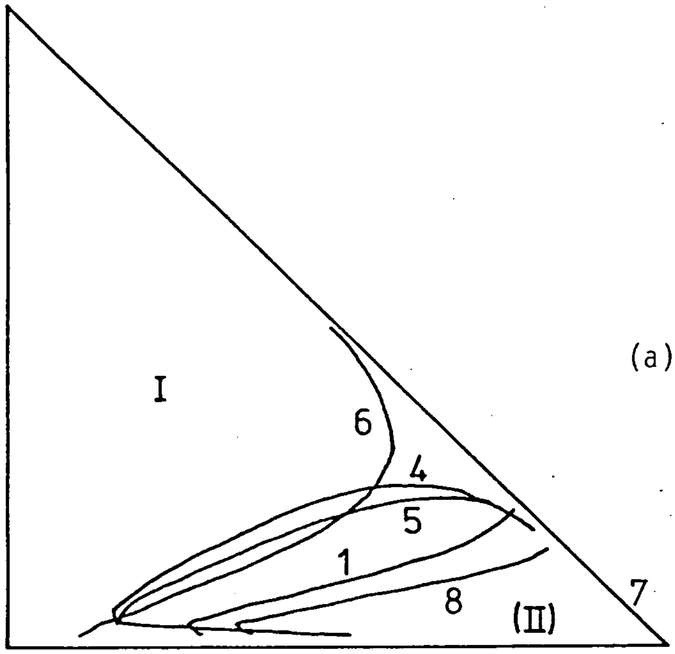
Figure 5.13 Comparison of zero contours for feed number 8.



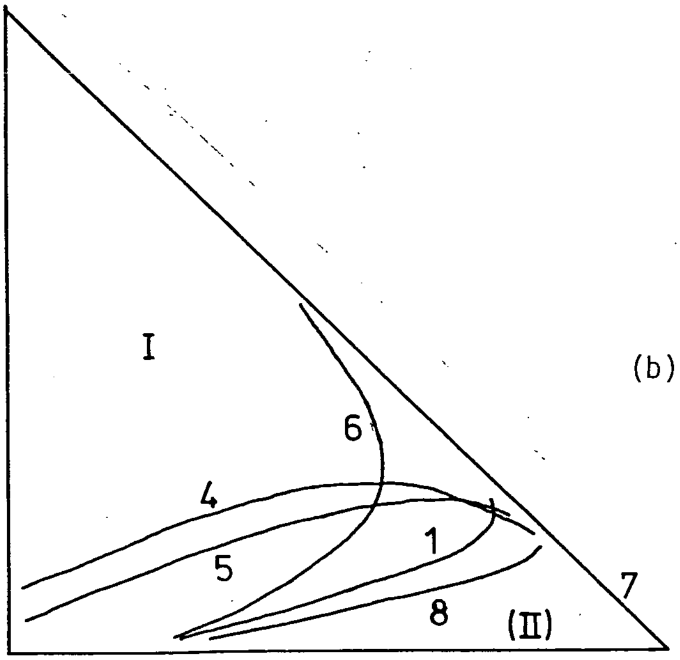
(a)

(b) recoveries

Figure 5.14 Comparison of zero contours of cost.

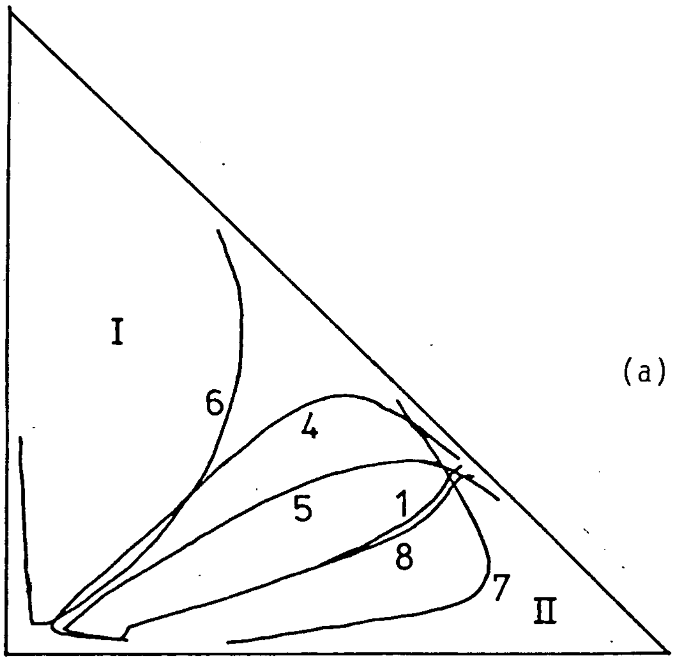


(a)

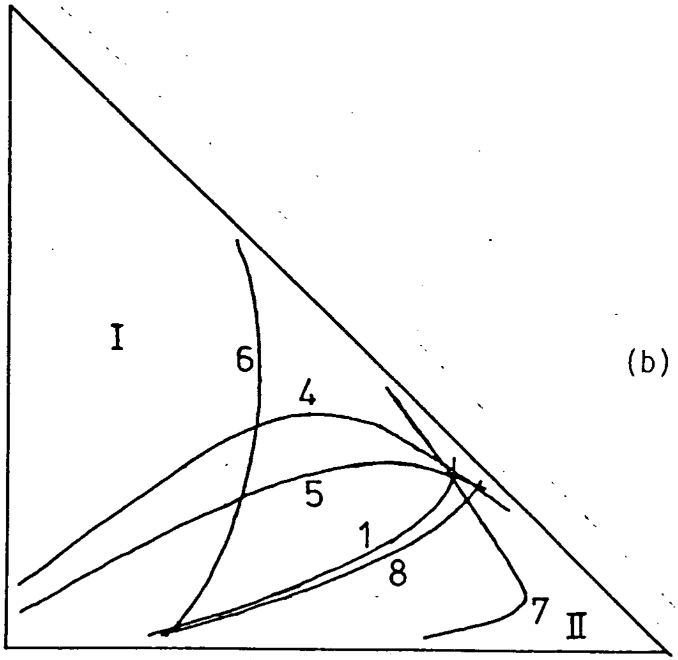


(b) recoveries

Figure 5.15 Comparison of zero contours of vapour rate.



(a)



(b) recoveries

Figure 5.16 Comparison of zero contours of reboiler duty.

although smooth, do not pass through B but instead aim for points on the AB or BC edges.

Moving on to consider the comparison of the functions under review, it will be seen that the agreement between the zero contours of vapour rate and of cost is generally quite good at high MESI (figures 5.11, 12: MESIs of 3.63 and 6.41) but worse at low (figures 5.9, 10: MESIs of 0.15, 0.28). In contrast the results for reboiler duty agree better with cost at low MESI and worse at high. One general feature of the results is the better agreement towards the B corner than towards the AC edge, though this will in part be an illusion due to the stretching of scale along the AC edge mentioned earlier. In general though neither is a good cost correlate in all circumstances.

What the diagrams presented in figures 5.8 to 5.16 fail to show is a startling correlation in the case of feed number 6 between the reboiler duty and the cost of systems designed using different conventions. The distillation systems used for cost comparisons here are designed and costed assuming steam to be available at any desired temperature and a corresponding cost (the 'smooth steam levels' case -- see Appendix), and also allowing the pressure in the columns, set by the cooling water temperature in the condenser, to fall below atmospheric pressure. The situation which the reboiler duty most closely mimics is that where steam is available only at a fixed pressure and temperature, and the agreement of reboiler duty for feed number 6 is with the cost of a system where steam is available at several fixed temperatures (the 'discrete steam levels' case). That this agreement was noticed at all is entirely fortuitous. All the results collected for systems using discrete steam levels were also for systems designed not to allow towers to run at vacuum pressures, but these were increased to atmospheric pressure if necessary. It happens that of the results for eleven feed mixtures studied (eight reported here and three others) only those for feed number 6 were unaffected by this restriction and so the reboiler duty results for designs allowing vacuum towers are comparable with the 'smooth' cost designs not allowing vacuum towers in this one case only. Unfortunately, considering the effect on the position of the cost zero contour of using discrete steam levels discussed in the appendix this agreement must also be considered fortuitous, though it would be interesting to look at other examples comparing the results for reboiler duty with cost using discrete steam supply levels.

5.2.5 Summary of Results

The correlations based only on external flows, mass load and its variations and entropy of choice, do not agree at all well with cost, though the entropy of choice does exhibit contours whose curvature is similar to that of cost and might be used in a more complex evaluation function. Two functions related to internal flows are also studied, namely the internal vapour rate and the reboiler duty. There is a reasonable but not altogether reliable agreement between either of these two functions and the cost of a 'smooth steam levels' design, with the edge perhaps going to the simpler correlation, the internal vapour rate.

5.3 Discussion

Various objections may be made which question the applicability and value of the observations reported above. Three areas are treated here.

5.3.1 Optimisation

None of the designs in the above study was optimised on either reflux ratio or pressure, and it may thus be argued that that any conclusions drawn are likely to be wrong, since what are being compared are arbitrarily poor column designs, not ones which would be used in real plant. This objection questions the applicability of the observations, however it can be argued that near optimal designs, at least of single columns if not of whole flowsheets, can be produced without resorting to optimisation procedures. Reflux ratio is dealt with first.

5.3.1.1 Reflux Ratio

The computer program used to generate all the results used here, sets reflux ratio to 1.2 times the minimum determined from the

Underwood equations. King [45] (p801) suggests that the optimum will probably be less than 1.1 times the minimum. Bakhshi and Gaddy [44] synthesise designs and optimise on both pressure and reflux ratio. The optimum value for reflux ratio always turns out to be 1.05 times the minimum, the lowest they allow. Tedder and Rudd [39,40] optimised a single column and found the optimum value dropped from 1.11 to 1.03 times the minimum as utility costs were increased tenfold. Bearing these results in mind, a value of 1.2 times the minimum seems high, but there is justification. King's worked example shows an optimum at 1.1 times the minimum, but with costs expected to increase by only 2 to 6% for reflux ratios of 1.2 to 1.3 times the minimum. Tedder and Rudd suggest a 1% increase in cost for a reflux ratio of 25% above the optimum at their standard utility rate, though at the higher rate a 23% increase is indicated. Since the cost rises only slowly at reflux ratios above the optimum, but must rise sharply towards infinity as the minimum is approached it is obviously better to err on the high side, and 1.2 times the minimum would be expected to give costs well within 5% of the optimum in most cases.

5.3.1.2 Pressure

Both Tedder and Rudd [39,40] and Bakhshi and Gaddy [44] studied cases where the cost of refrigeration is significantly higher than the cost of steam. Both pairs came to the conclusion, Bakhshi and Gaddy explicitly, Tedder and Rudd more tentatively, that the optimum pressure is close to that at which refrigeration is just avoided, that is the minimum possible to allow the condenser to be cooled by cooling water rather than refrigerant. King makes similar suggestions but makes the point that vacuum operation is usually to be avoided. In this work pressures are set by the temperature of the cooling water and, if the option of not allowing vacuum operation is invoked, then increases the pressure to atmospheric if necessary. Bakhshi and Gaddy point out that more work must be done to discover if this 'no refrigeration' principle is universally applicable, but since the species in the mixtures investigated by them are similar to the those used in this study, the procedure should give designs close to the optimum. This point does not seem to be as clear cut as the reflux ratio argument however, and is more likely to lead to significant departures from optimality.

5.3.1.3 Summary of Optimisation Discussion

To summarise then, although in designing the distillation systems for the above work no optimisation was performed, even so the design procedures followed are likely to give results whose cost is only a few percent greater than the optimal cost.

5.3.2 Computing Effort

A large part of the computational effort involved in calculating each of the reboil related functions may be expected to lie with the iterative procedures, the Underwood and bubble- and dewpoint calculations, though it might be sensible to check this supposition. The cost calculation requires the Underwood procedure, two or three bubble- or dewpoint calculations and many fiddly but trivial design calculations besides. Calculation of the reboiler duty does not need the detailed design calculations, but does need the same iterative ones. Vapour rate requires little more than the Underwood calculation. If it were planned to use reboiler duty as an alternative to doing a full costing there would probably not be a great saving in effort, even though under some circumstances it seems to be a good cost correlate. Using the vapour rate there may be a considerable saving in effort and it does seems to be a slightly better cost correlate. Even so it still involves a considerable amount of calculation.

Let us consider what hypothetical situations might correspond to using these two functions in place of cost. Using the reboiler duty as an evaluation function corresponds to using the cost of steam alone when that cost is dependent only on the quantity of heat required and not on the temperature at which it must be supplied. A real situation in which this is the case is where steam is available at only one fixed pressure. If we take this kind of simplification of the cost function one step further and assume that the latent heats of vapourisation of components in the reboiler are are all equal then the steam cost would correlate exactly with the column internal vapour rate. In this situation energy use is determined only by the quantity of material evaporated, not by its composition, and in fact corresponds quite closely to a mixture of close members of a homologous series. Some of the example feeds used here, for instance feed number 1, are indeed such mixtures. Notwithstanding this closeness to real situations, the work involved in calculating these two functions will tend to rule them out as alternatives to costing procedures, since there will be little gain in convenience. However the main gain in this study lies in discovering that cost really is well correlated with energy use.

5.3.3 Energy Integration

A serious objection to the value of this work is that the use of energy in a distillation network may be reduced by use of appropriate energy integration techniques. In fact in favourable circumstances the net bill for energy in a distillation scheme may be reduced to zero by appropriate integration of the reboiler heating and condenser cooling loads into the rest of the plant (Dunford and Linnhoff [58]). In such a situation the balance between energy and capital cost in the distillation design will change, making capital the more important component. Indeed it may be difficult to assign any energy cost at all solely to the distillation sequence: it may only be possible to view the plant as an integrated whole. In such a case not only will the basis of this work -- that energy is the major cost in distillation -- be undermined, but the only costs attibutable directly to distillation may well be due to capital. However things are not as gloomy as they might seem. There will still be incentives to produce designs with low energy usage, partly to reduce the amount of heat required by the plant in general, and also to minimise the capital cost since smaller energy flows mean smaller heat exchange equipment, other things being equal. In distillation particularly smaller energy flows are associated with smaller material flows.

5.4 Overall Summary, Conclusions and Further Work

5.4.1 Overall Summary

This study set out to devise a function which would be a good correlation with cost in simple ternary distillation systems, and looked at six possible functions. Two of these, mass load and entropy of choice, have been shown not to be good candidates since they give the same results for every choice of feed species. Two further functions, mass load divided by relative volatility and mass load divided by excess relative volatility, gave slightly closer agreement with cost, but have two drawbacks. The first is that neither introduces enough curvature into the linear mass load function to match it to the highly curved contours of the cost function, and the second is that neither recognises the higher cost of taking material overhead in distillation. The fifth function, the internal vapour rate, requires greater computational effort than the previous ones, gives much better agreement with cost in many cases but is not entirely reliable. The last function, reboiler duty, appears to correlate slightly less well with cost than vapour rate in the cases studied, and requires somewhat more effort to calculate.

The applicability of these results may be questioned on the grounds that the individual designs were not optimised, however the design procedures used are recommended in the literature to give near optimal results in most cases. An objection which affects the value of the findings concerns the computational effort required for the calculation of each function, since the best fitting ones require a significant fraction of the effort required for a full shortcut design and costing procedure. This objection is valid if it is planned to use the functions as alternatives to costing in a synthesis procedure, but this is not the only aim of the exercise. Perhaps the major objection is that since the good fit of the functions is due to the relatively high cost of the energy required in the process, (indeed he idea of using the reboil related functions came from just this observation) their value, and the value of any findings based on them, is diminished when energy costs form a smaller part of the total cost, for instance in systems with energy integration. In reply it can be argued that even in such cases systems using less energy would be desirable, due to the smaller capital cost of plant required for smaller energy and material flows.

5.4.2 Conclusions

Although the aim of this work was to discover a good cost correlate the underlying aim was not to discover an alternative to cost to use in design procedures. Instead it was to find out what cost mainly depended on. That the cost is highly correlated with energy use is, with the indispensable benefit of hindsight, a fairly obvious conclusion: both operating and capital cost in distillation systems are to a large extent dependent on the internal flows, which govern the size of the equipment and the consumption of utilities, both heating and cooling. The conclusion is therefore a potentially useful one in designing a distillation system either as a 'stand alone' unit or as part of an integrated chemical process. In a nutshell, cheap, even elegant distillation systems will have low energy flows through them.

5.4.3 Further Work

There are two obvious areas for further work. The first is to rectify one of the omissions from this study. This is to investigate some of functions intermediate in complexity between the rather simple mass flow related functions and the more complex reboil related ones. Possible candidates would include the Coefficient of Difficulty of Separation (CDS) proposed by Nath [41], or mass load / (entropy of choice x relative volatility).

The other area of further work concerns synthesis methods. The work of Hohmann [26], Nishida et al [25,30] and Linnhoff and Flower [24,31] provides targets which the engineer may aim at, and indeed achieve in his designs. In chapter 4 it was shown that a target based on the number of units is unlikely to be useful in the field of separation scheme synthesis. A comparable minimum energy target for distillation systems might be useful but there is no obvious way of calculating a value achievable in real life which does not require arbitrary assumptions -- an energy target based on minimum reflux ratios would not be achievable in real life and would also not be independent of the column configuration chosen. The next chapter takes another approach to shrinking the size of the search space by considering what factors influence energy use in distillation systems.

6 Further Investigation of Vapour Rate

6 Further Investigation of Vapour Rate

In the previous chapter the fair agreement between cost and vapour rate or reboiler load in distillation systems was noted. This chapter looks further at the use of vapour rate as a cost analogue. The work divides into two. First is a comparison of vapour rate with cost for some systems studied by Thompson and King [17] and Freshwater and Henry [46]. The study suggests a possible heuristic synthesis method for certain classes of distillation system. The second part considers the variation of minimum vapour rate with non-key component concentration, notes what seems to be a superficially simple relationship suggesting another synthesis approach, but finds that complications render this straightforward view impractical.

6.1 Comparison of Vapour Rate with Cost

This section deals with the comparison of the internal vapour rate in distillation (either the minimum vapour rate, that at the minimum reflux ratio R_{min}, or that at a fixed multiple of R_{min}) with cost, using results reported by Freshwater and Henry [46]. Patterns which may be of use in synthesising distillation systems are noted. But first comparison is made between cost, vapour rates and reboiler load for some results given by Thompson and King [17].

6.1.1 Thompsons and King's Results

Following the observation made in the previous chapter that V_{min}, the reboiler vapour rate at the minimum reflux ratio, often seemed to be a closer approximation to cost than reboiler energy load, it was decided to look at some cost results reported by other workers and compare them with the reboil related functions being studied. Results are shown in figure 6.1 and table 6.1. Note the higher correlation coefficient for V_{min} than for reboiler heat load. The following points may provide a satisfactory explanation for this phenomenon. Two factors which will influence the correlation of reboiler energy with cost will be the latent heats of vapourisation of the reboiled components, and the temperatures at which the energy is suppied (if the design and costing procedure exploits this feature by using different heating media). In fact the latent heat correction ought to make reboiler heat load a better fit to cost than minimum vapour rate, so this does not explain the discrepancy. On the other hand having to use an expensive heating medium, say high- rather than low-pressure steam, will tend to inflate the cost of a distillation system compared to the cost of one with the same total heat load required at a lower temperature, and this will therefore tend to break the strong link between reboiler heat duty and cost. This conclusion is borne out by the results to the extent that the higher-cost systems indeed use higher temperature heating media on the whole, however not enough results are available to make any firm conclusion about this explanation.

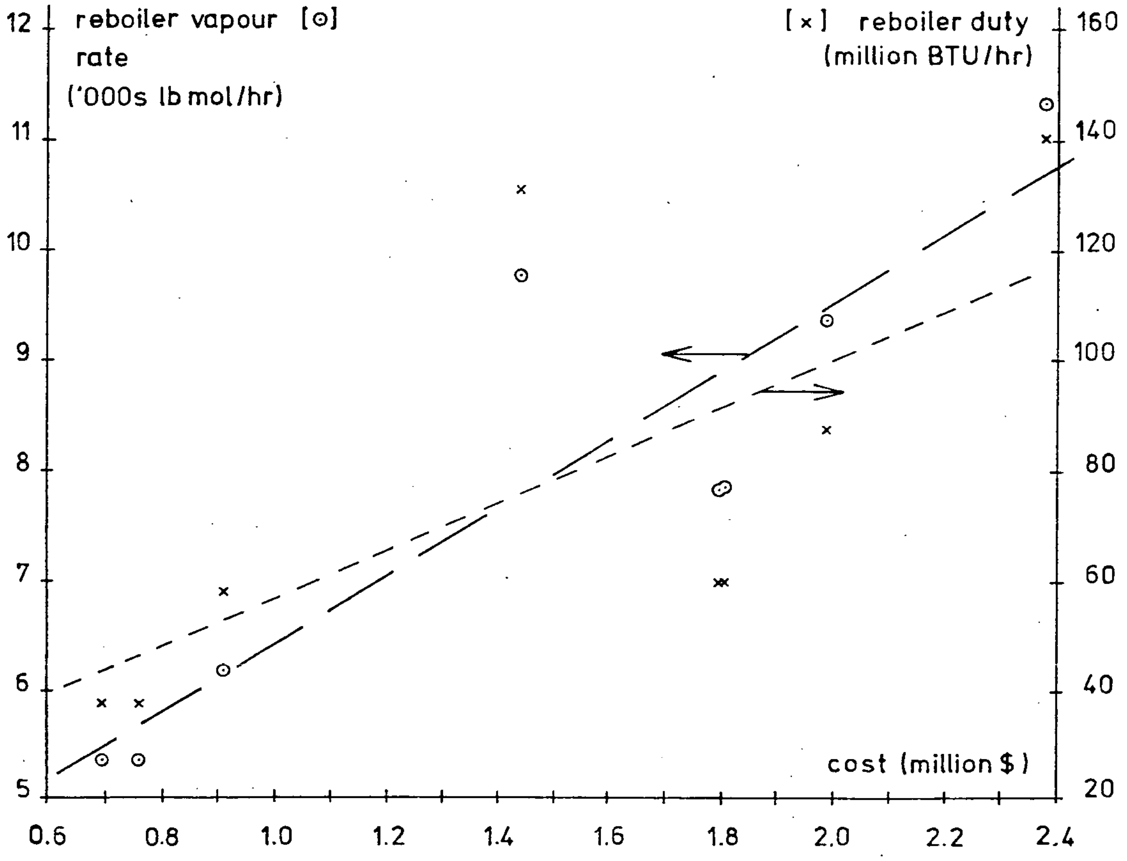


Figure 6.1 Reboiler vapour rate (V_{min}) and reboiler duty vs cost for Thompson and King's designs.

Table 6.1: Cost, vapour rate and reboiler duty figures for Thompson and King's example 1

Cost ($)	vapour rate (000 lb mol/hr)	reboiler duty (million Btu/hr)
694 536	5 357	38.00
761 312	5 359	37.96
907 513	6 192	58.55
1 440 562	9 761	131.49
1 795 429	7 828	59.80
1 804 706	7 852	60.03
1 990 944	9 360	87.47
2 378 892	11 336	140.45

correlation coefficients: vapour rate with cost -- 0.894
reboiler duty with cost -- 0.681

6.1.2 Freshwater and Henry's Systems

Since Thompson and King's method produces only a few different flowsheets their results are not ideal to use for an extensive comparison. It was therefore decided to use the work of Freshwater and Henry for investigating the relation between cost and vapour rate, since they examined all the simple configurations for 4- and 5-component distillation. They designed distillation columns with reflux ratios of 1.25 x R_{min} and at constant pressure. They use the sets of components shown in table 6.2. The physical conditions used are not specified, so for this work K-values were calculated at three sets of temperature and pressure for the 4-component mixtures, and two for the 5-component systems. These conditions and the K-values persuant thereon are also shown in the table.

6.1.2.1 Results

Freshwater and Henry's results are the costs of all the simple distillation arrangements for the feed mixtures shown in table 6.2 at the compositions given in table 6.3. The arrangements and the numbering used here are shown in figure 6.2. The numbering for the 4-component systems is different from, and I think more logical than that used by Freshwater and Henry. The results reported here are * similar but instead of cost are the values of the sum of the internal vapour rates in the columns of each configuration, each column operating at a reflux ratio of 1.25 times the minimum. This quantity will be referred to as $V_{1.25}$. Note that the presentation

Table 6.2: Feed mixtures and conditions for K-value determination

Feed number	Components	Relative volatilities		
		Conditions		
		(a) 37.8 deg C 6.9 bar	(b) 54.4 deg C 6.9 bar	(c) 37.8 deg C 1.2 bar
	i-butane			
		3.08	3.02	2.84
1a,b,c	i-pentane			
		3.69	2.68	4.56
90% recoveries	n-hexane			
		2.96	3.26	3.16
	n-heptane			
	i-butane			
		1.34	1.29	1.30
2a,b,c	n-butane			
		2.95	2.77	3.32
90% recoveries	n-pentane			
		2.92	2.50	2.96
	n-hexane			
	propane			
		2.23	2.07	
	i-butane			
3a,b		1.32	1.28	
	n-butane			
98% recoveries		2.33	2.21	
	i-pentane			
		1.37	1.27	
	n-pentane			

Table 6.3: Feed Types

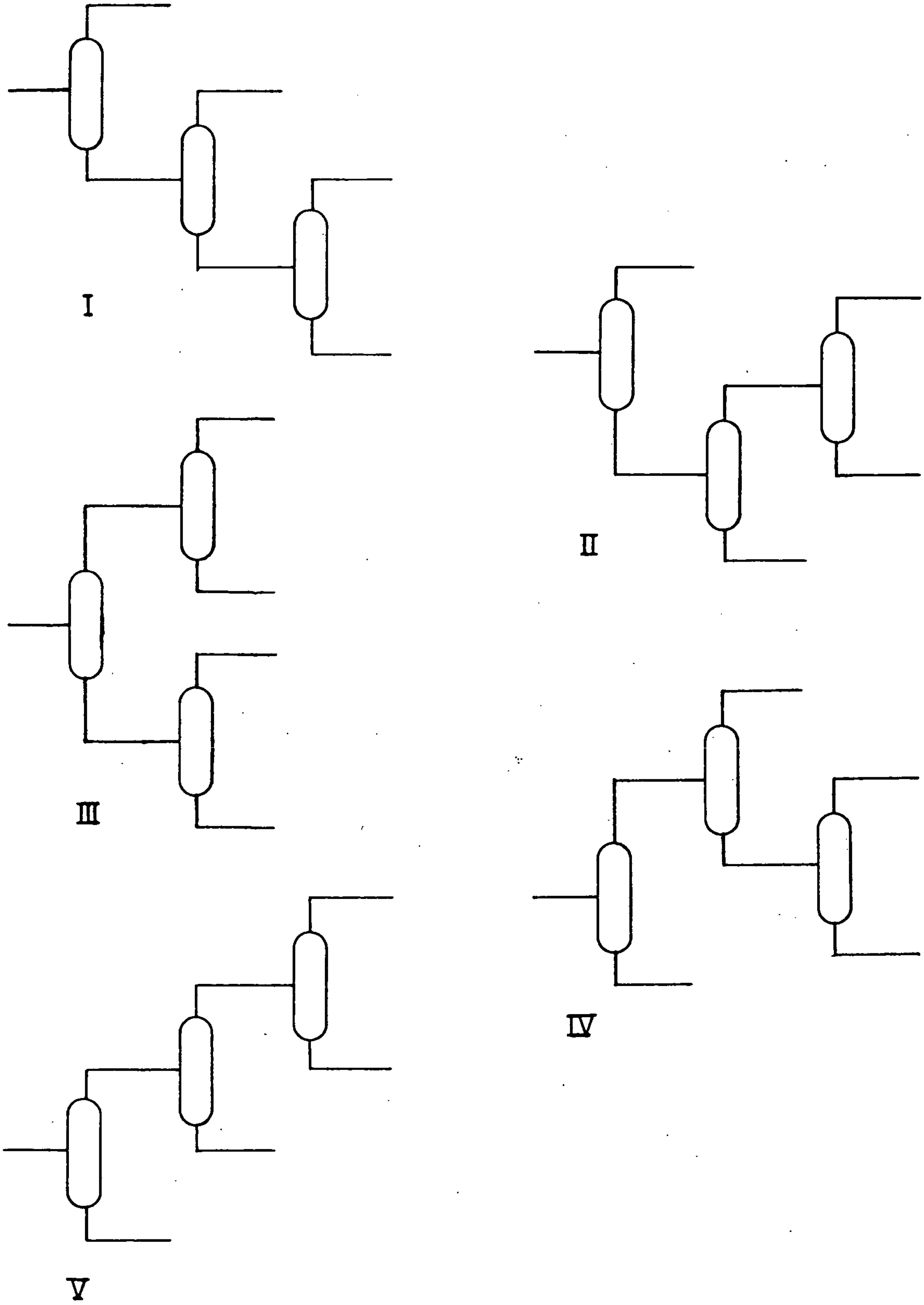
4-Component feeds

	Mole fraction of component i			
i ->	1	2	3	4
feed type				
1	0.25	0.25	0.25	0.25
2	0.7	0.1	0.1	0.1
3	0.1	0.7	0.1	0.1
4	0.1	0.1	0.7	0.1
5	0.1	0.1	0.1	0.7

5-Component Feeds

	Mole fraction of component i				
i ->	1	2	3	4	5
feed type					
1	0.2	0.2	0.2	0.2	0.2
2	0.6	0.1	0.1	0.1	0.1
3	0.1	0.6	0.1	0.1	0.1
4	0.1	0.1	0.6	0.1	0.1
5	0.1	0.1	0.1	0.6	0.1
6	0.1	0.1	0.1	0.1	0.6

Figure 6.2(a) Five simple distillation configurations for 4-component mixtures.



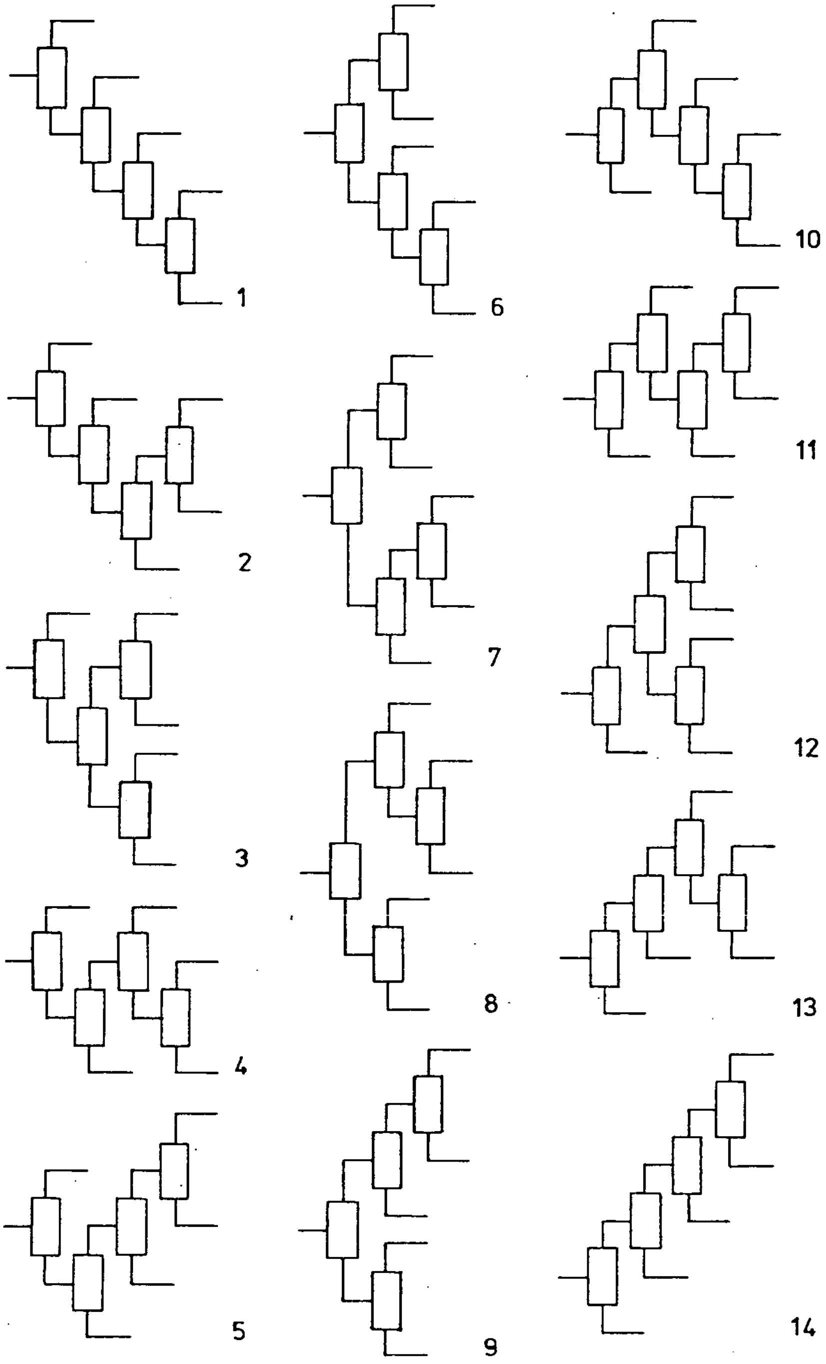


Figure 6.2(b) Fourteen simple distillation configurations for 5-component mixtures.

of results here is somewhat different from that of Freshwater and Henry. In their graphs of cost vs feed type their use of curved lines to join points for each configuration suggests a continuous functional relationship of cost with feed composition which really cannot be represented on a single axis. Their results and the new work reported here are therefore presented using straight lines which are intended to suggest nothing about variation of cost or other function between the points where the function is actually evaluated. In addition results are also presented as histograms showing the distribution of costs for each feed type, ordered by cost and grouped by feed type.

6.1.2.1.1 Results for 4-Component Feeds

Freshwater and Henry's cost results for the two 4-component feeds are shown in figures 6.3 and 6.4. They also give figures for energy use for these two feeds and these are shown in figures 6.5 and 6.6. Note the distinct similarity between these results and the cost results. Note also the similarity to the new results generated for $V_{1.25}$, shown in figures 6.7 and 6.8. Various salient points emerge from these results :-

-- Considering the graphs of cost against feed type for different configurations, figures 6.3(a) - 6.8(a), the general arrangement of the points is quite characteristic.

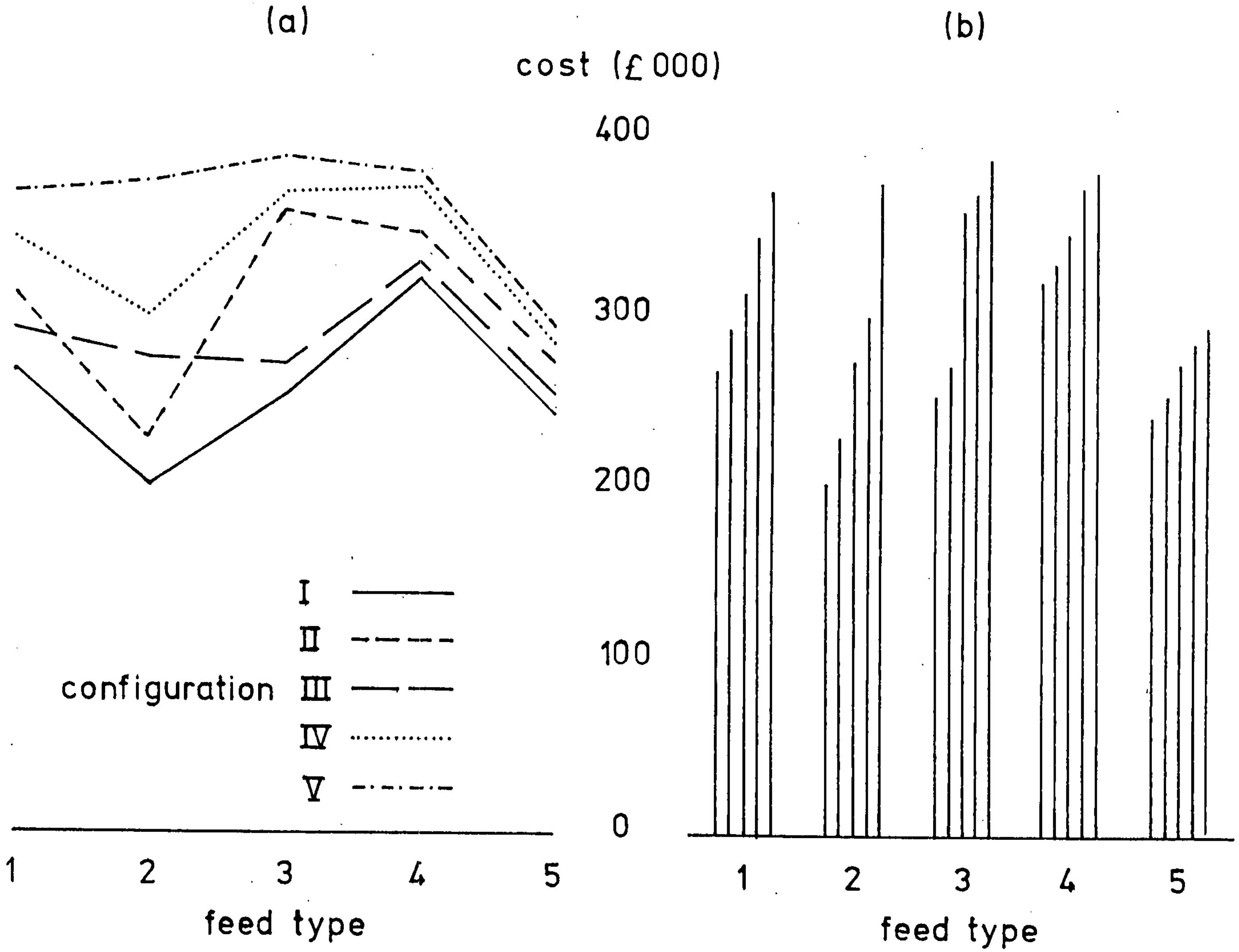


Figure 6.3 Distribution of cost with feed type for feed number 1, Freshwater and Henry's results.

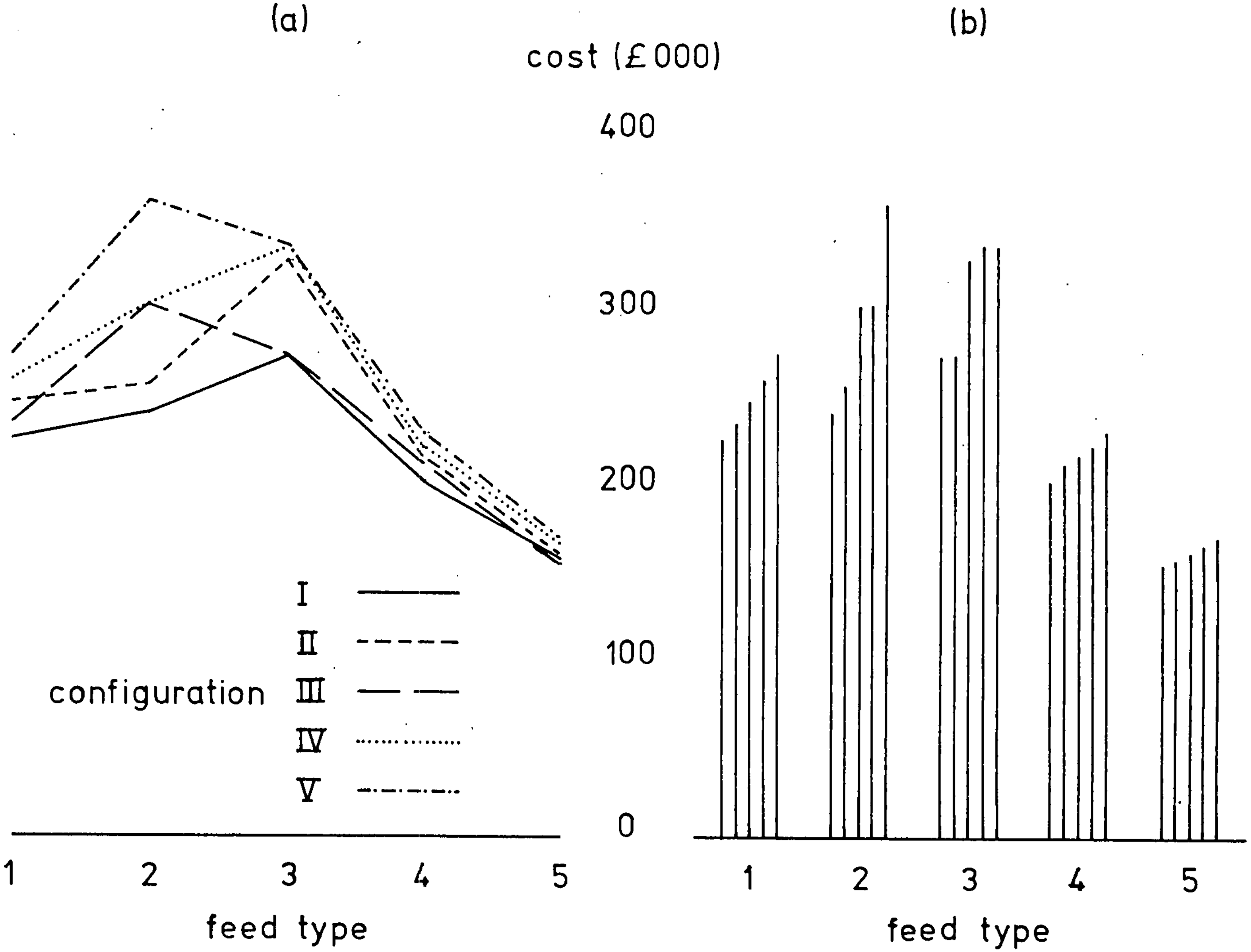


Figure 6.4 Distribution of cost with feed type for feed number 2, Freshwater and Henry's results.

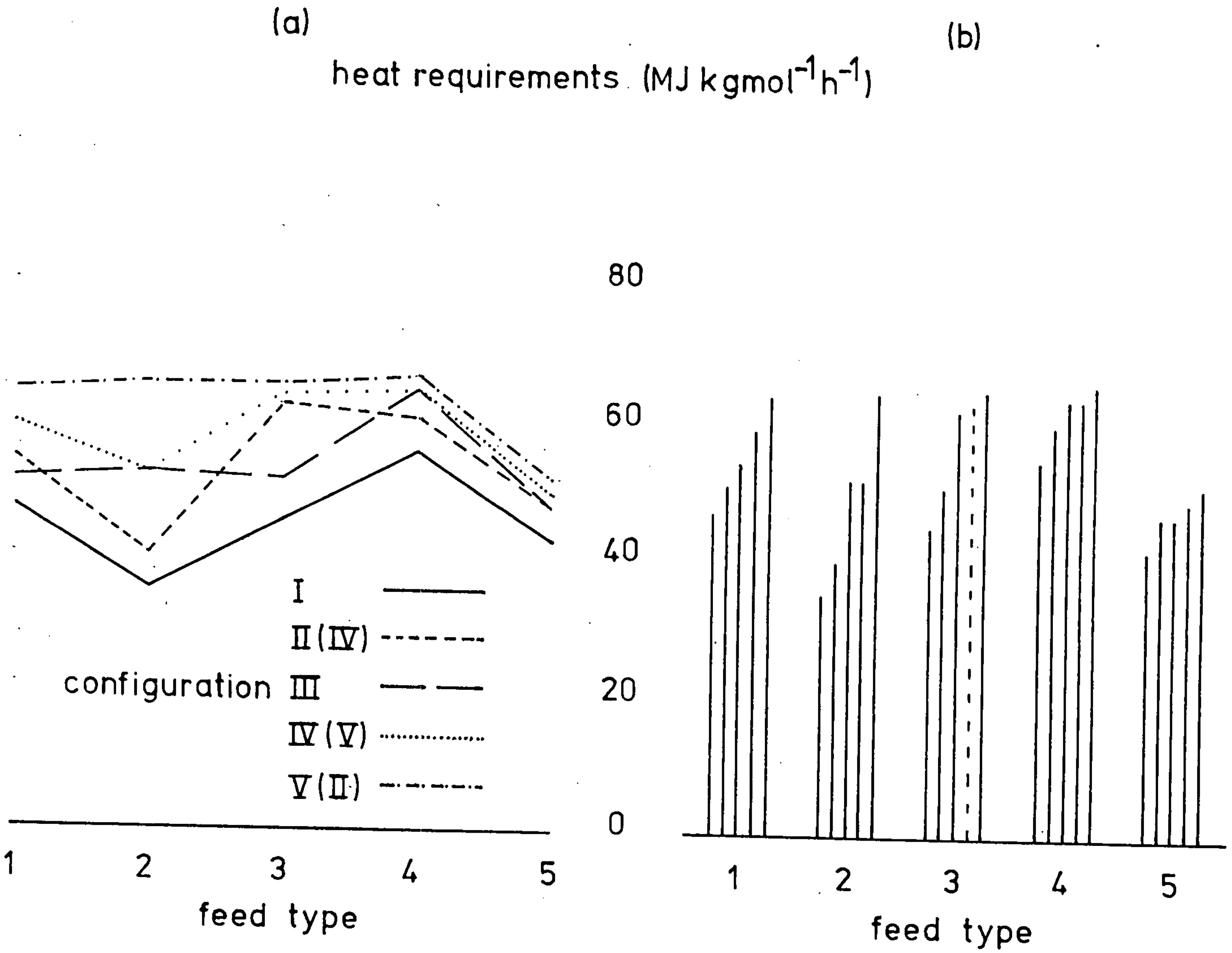


Figure 6.5 Distribution of heat requirements with feed type for feed number 1, Freshwater and Henry's results.

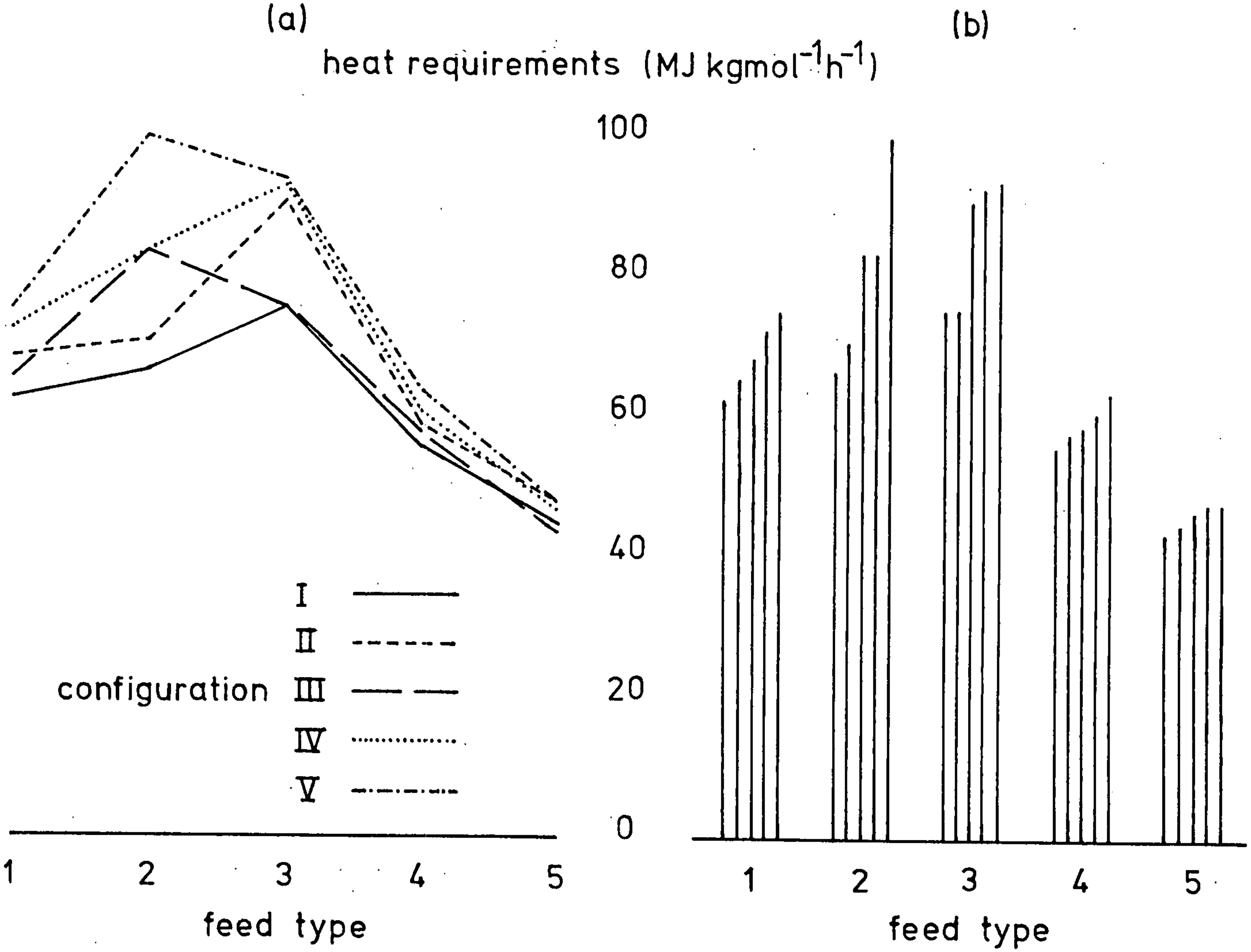


Figure 6.6 Distribution of heat requirements with feed type for feed number 2, Freshwater and Henry's results.

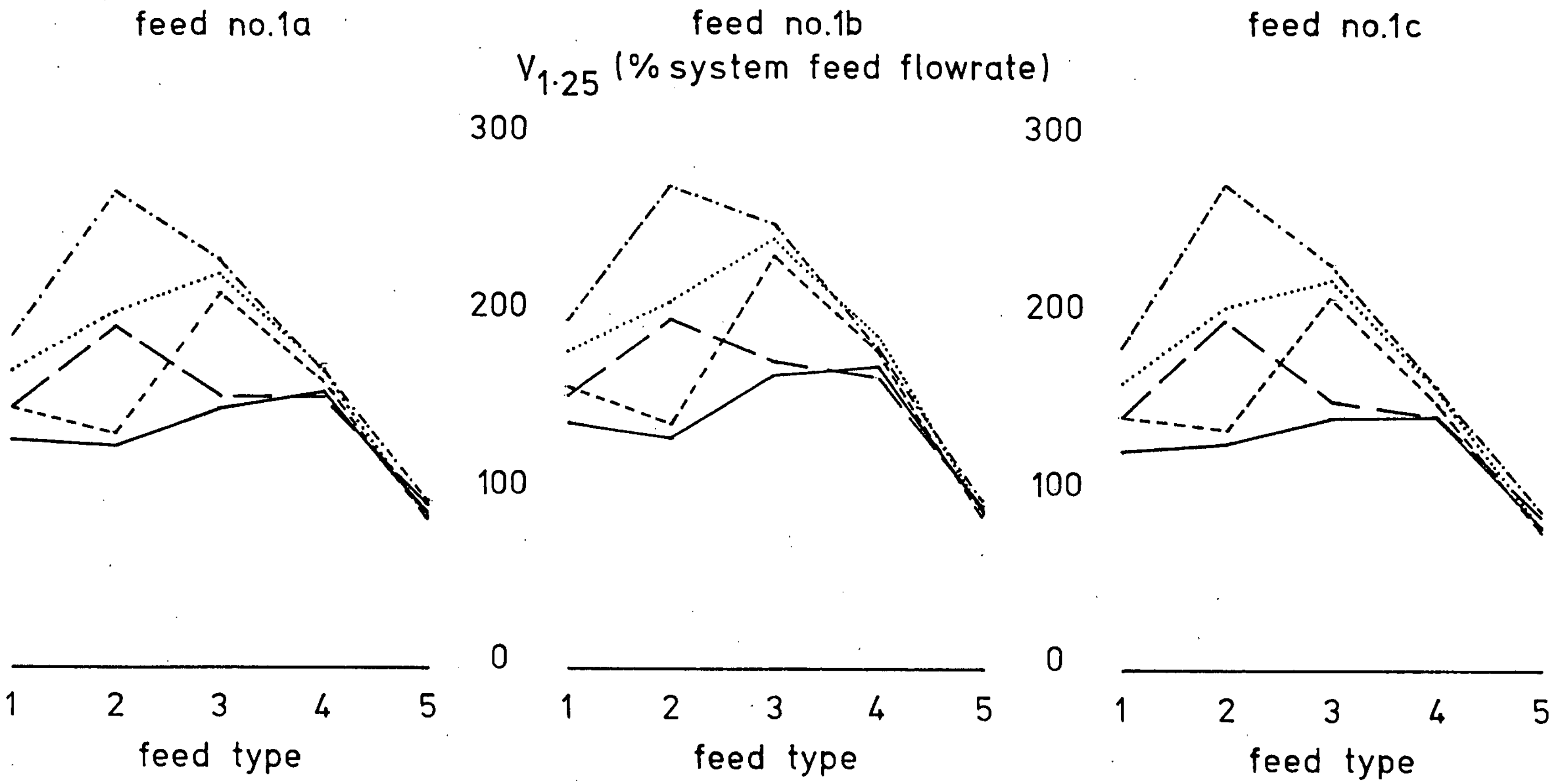


Figure 6.7(a) Distribution of reboiler vapour rate ($V_{1.25}$) with feed type for feed number 1.

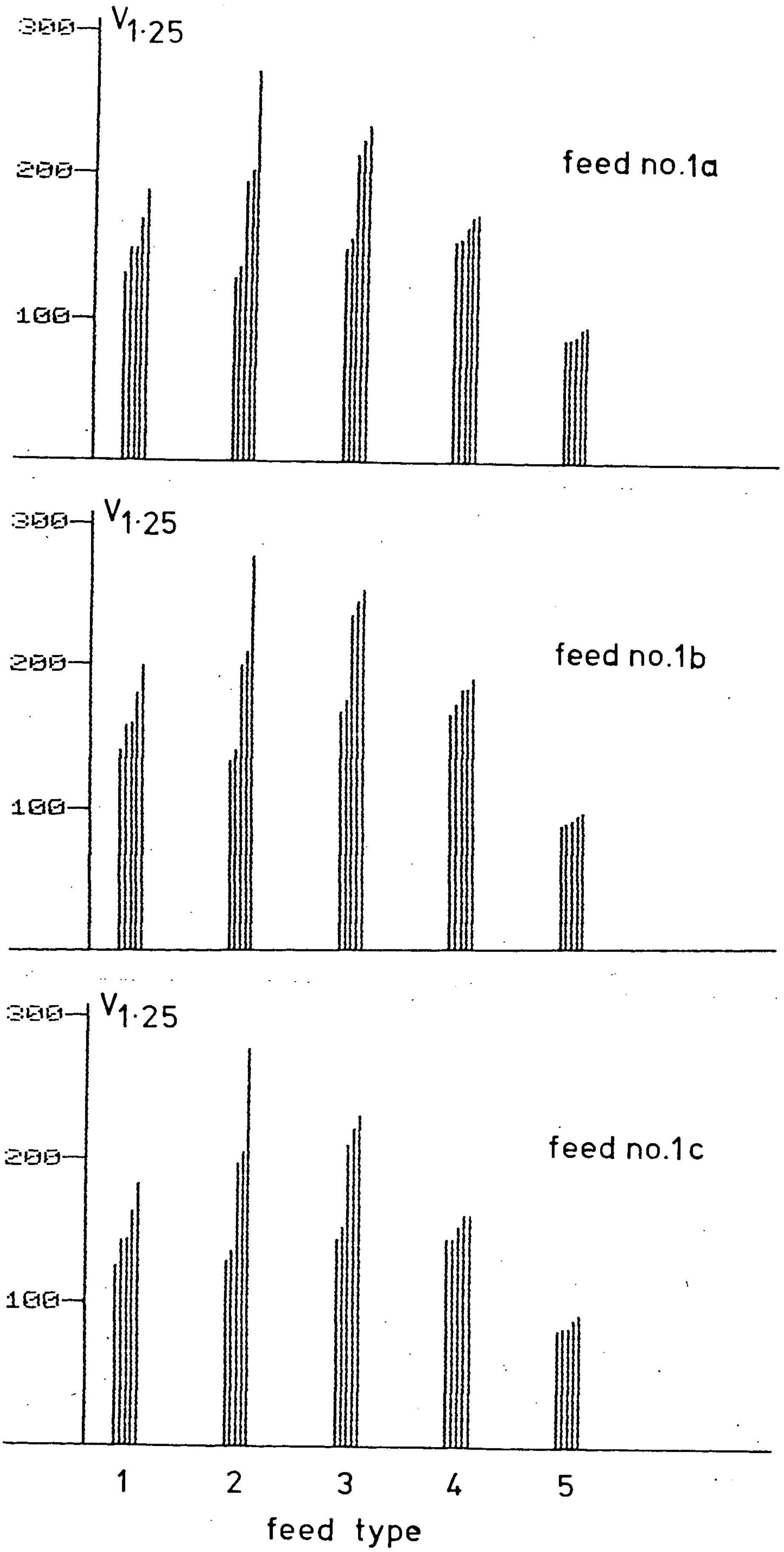


Figure 6.7(b) Distribution of reboiler vapour rate ($V_{1.25}$) with feed type for feed number 1.

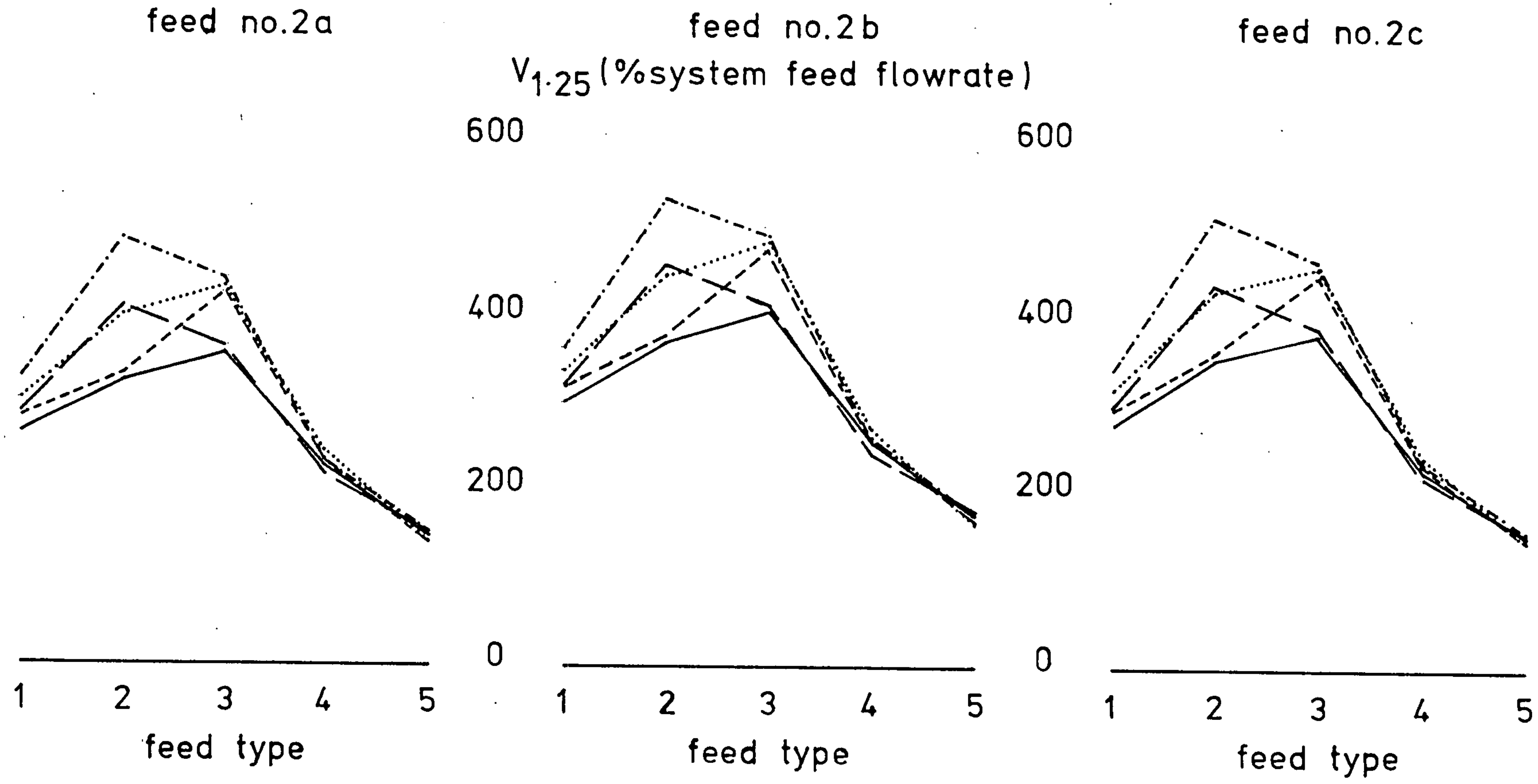


Figure 6.8(a) Distribution of reboiler vapour rate ($V_{1.25}$) with feed type for feed number 2.

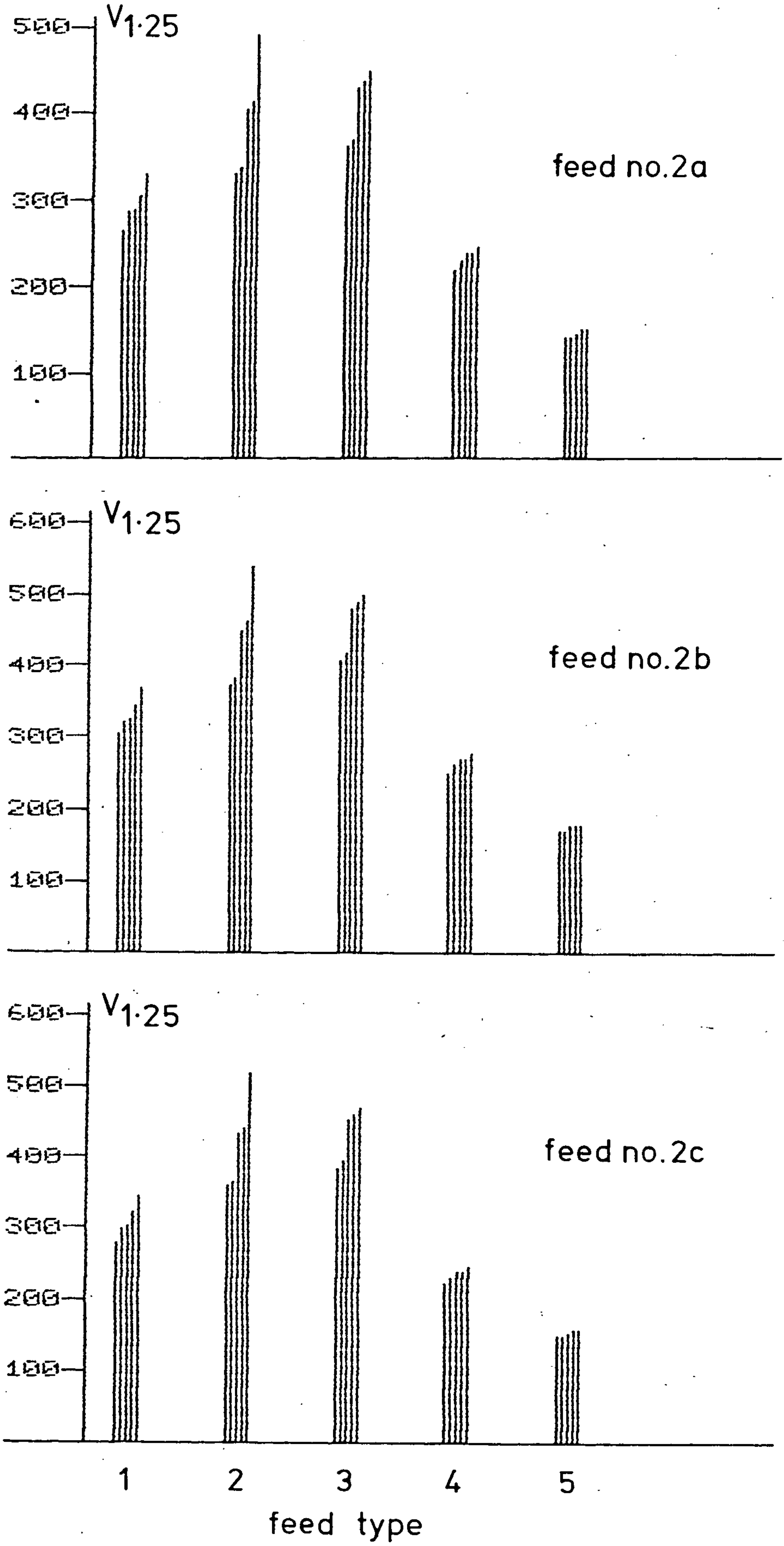


Figure 6.8(b) Distribution of reboiler vapour rate ($V_{1.25}$) with feed type for feed number 2.

-- Results for feed type 1, the equimolar feed, show a relatively smooth spread of costs or vapour rate with configuration, over a relatively large range.

-- Results for feed types 2 and 3, which have large quantities of light component, show several distinct blocks of results. This is more obvious for feed type 2 (0.7, 0.1, 0.1, 0.1) with the second example, but both examples show a (2,2,1) grouping for feed 2, and a (2,3) grouping for feed 3.

-- The histograms for feed types 4 and 5, where the major component is one of the heavier species, show no such grouping of results and are more smoothly distributed, but over a much smaller range than for the equimolar feed.

6.1.2.1.2 Results for 5-Component Feeds

Analysing the results for the 5-component systems is more difficult as Freshwater and Henry do not give any figures for energy use and their diagram of costs vs feed type is extremely confusing. However, since the results for $V_{1.25}$ show such a similarity to cost for the 4-component systems these functions will be examined here. Histograms of results for $V_{1.25}$ are shown in figures 6.9 and 6.10 and show features similar to those for 4-component feeds, viz:-

-- The equimolar feed results show a smooth and fairly wide range of values.

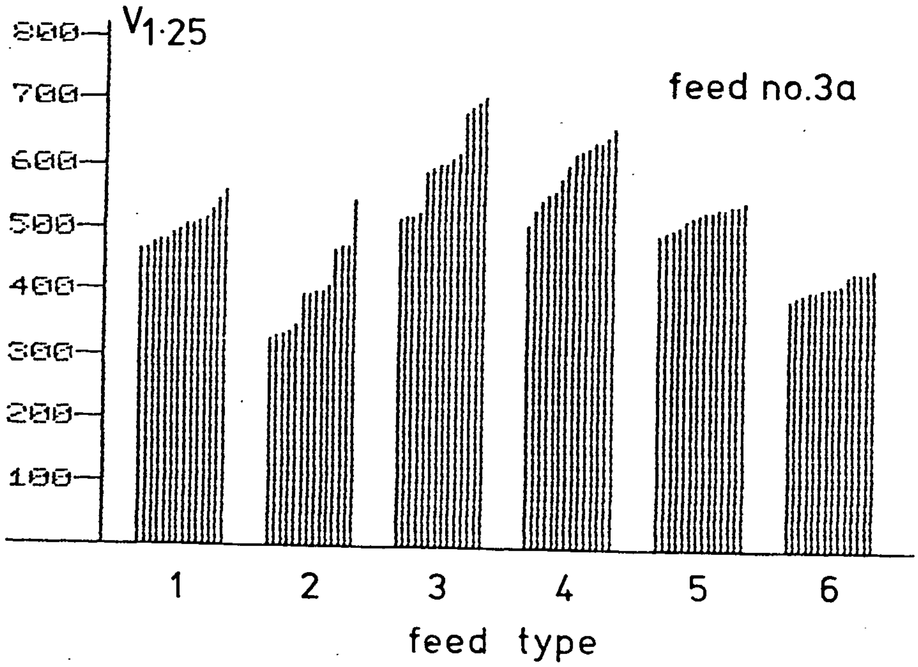


Figure 6.9 Distribution of reboiler vapour rate ($V_{1.25}$) with feed type for feed number 3a.

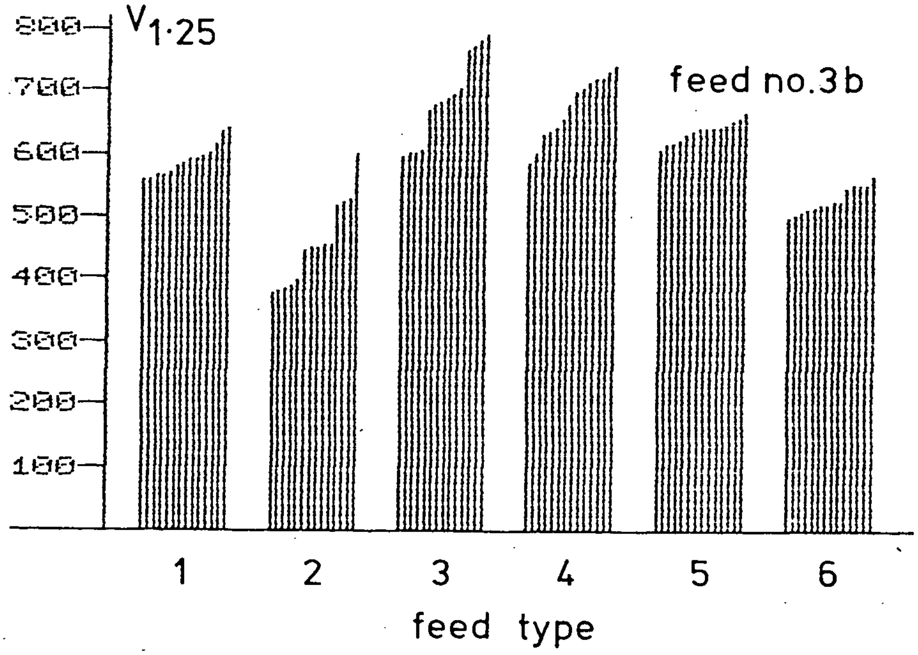


Figure 6.10 Distribution of reboiler vapour rate ($V_{1.25}$) with feed type for feed number 3b.

-- Feed types 2, 3 and 4, where the major component is not one of the two least volatile components, show blocks of results. Feed type 2 has a (5,5,3,1) grouping, feed type 3 (4,6,4) and feed type 4 (5,9).

-- The mixtures where the major component lies in the last two in decreasing order of volatility show a relatively small range of costs, with a smooth distribution.

6.1.2.2 Discussion of Results

6.1.2.2.1 Results Showing Grouping

Further work reveals a striking similarity between the results for the 4-component systems and what might be called 'tops load' or ΣD, the sum of the flowrates of overhead products from the columns in a flowsheet. This is shown in figures 6.11 and 6.12. The reason for this similarity is obvious: the vapour rate function is of the form $\Sigma(R + 1) D$, where D is the overhead product rate from the column and R is the reflux ratio. The reason for the grouping of results is also now clear: it depends on the number of times the major component in the feed is present in an overhead product from a column in any particular configuration. Thus in the 4-component systems, if the major component is the most volatile it will be taken overhead once in designs 1 and 2, twice in designs 3 and 4, and three times in design 5. If it is second in volatility ordering it will be taken overhead once in designs 1 and 3, and twice in the

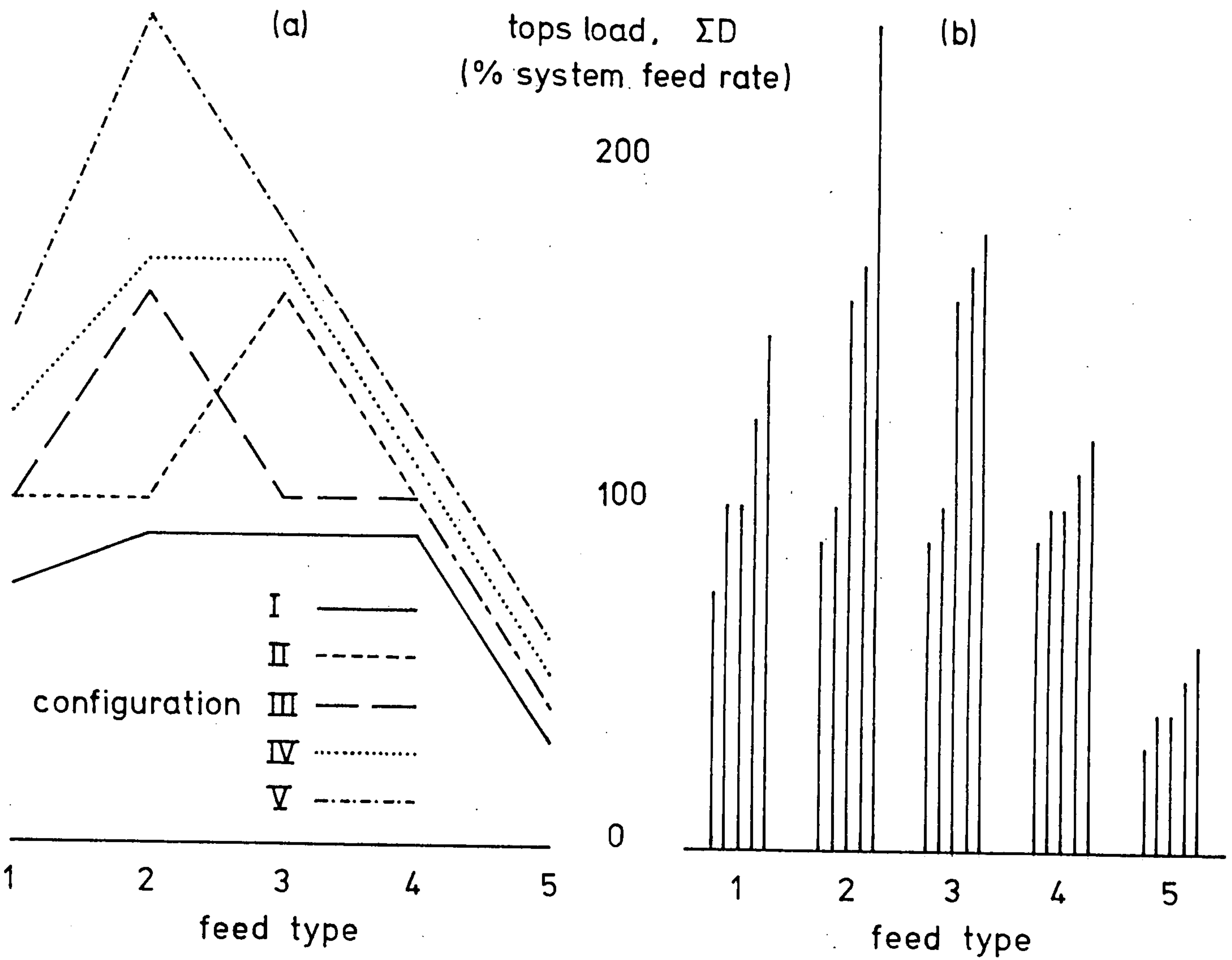


Figure 6.11 Distribution of tops load, ΣD, for 4-component feeds.

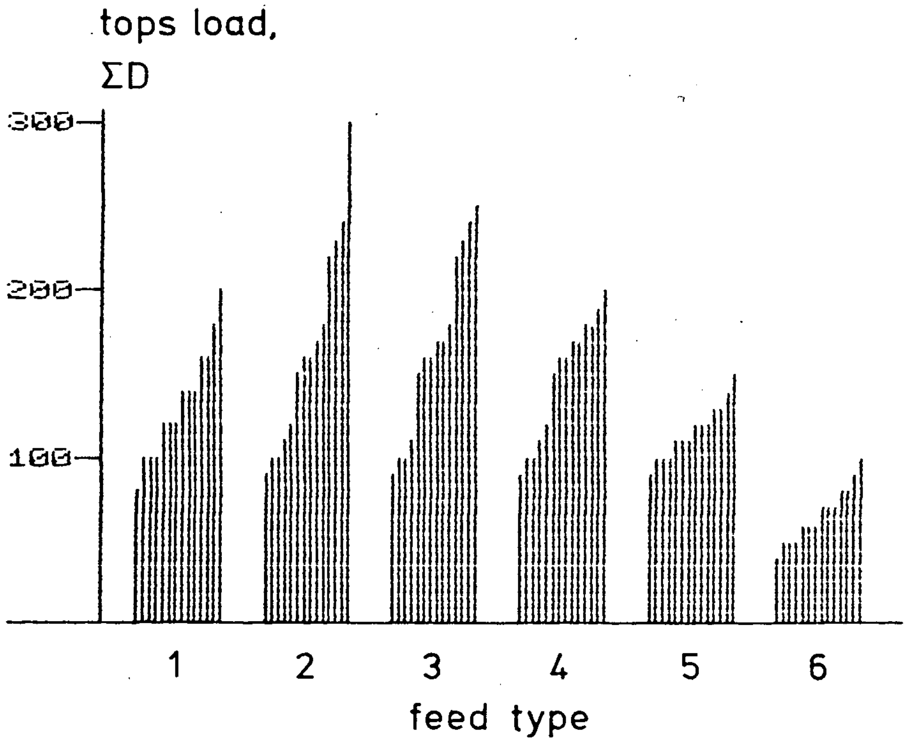


Figure 6.12 Distribution of tops load, ΣD, for 5-component feeds.

others. Inspection of the results shows that these groupings are indeed the ones found, and a similar explanation accounts for the groupings in the results for the 5-component systems.

This observation immediately suggests a simple heuristic method of screening distillation networks for further and more thorough evaluation, namely to choose that group which takes the major component overhead the smallest number of times. This heuristic is only applicable to systems which have a major component, and then only if it is not the least or next to least volatile.

6.1.2.2.2 Results Not Showing Grouping

Distillation networks processing feeds whose major component is the least volatile will never take that component overhead. Similarly where the major component is next to least volatile it will be taken overhead once in every flowsheet. It is evident that such feed types, or feeds without a major component at all such as the equimolar feeds, will not lead to the groupings of costs exhibited by the other feed types and this is borne out by the results. The simple heuristic given above is therefore not applicable. However, plotting cost for the 4-component systems or $V_{1.25}$ for the 5-component ones against ΣD, figures 6.13, 6.14 and 6.15, reveals the following points:-

-- For the equimolar feeds in each case there is a strong correlation of the evaluation function, cost or $V_{1.25}$, with Σ D.

-- For feeds with the major component low in the volatility order, the 4-component feeds show the strong correlation, but the 5-component feeds do not.

The first of these points suggests a screening method for equimolar feeds similar to the one developed above, namely to choose feeds with the lowest values of Σ D for further evaluation. This method might also be applied to 4-component feeds with heavy major components, though in such cases, with only five different configurations to chose from anyway, and these likely to exhibit only a very small range of costs, there is a case for either expressly designing each one, or for simply choosing any one of the five, for instance the direct sequence. The case of 5-component, and possibly more complex feed mixtures is rather more difficult since there is no obvious trend. However, because of the relatively small range of costs of the candidate configurations in such cases, any screening function would need to be more discriminating than the rough method of using Σ D described above.

6.1.2.2.3 Effect of Relative Volatility

Although no explicit account has been taken of the effect of relative volatility on the results reported here, some of the

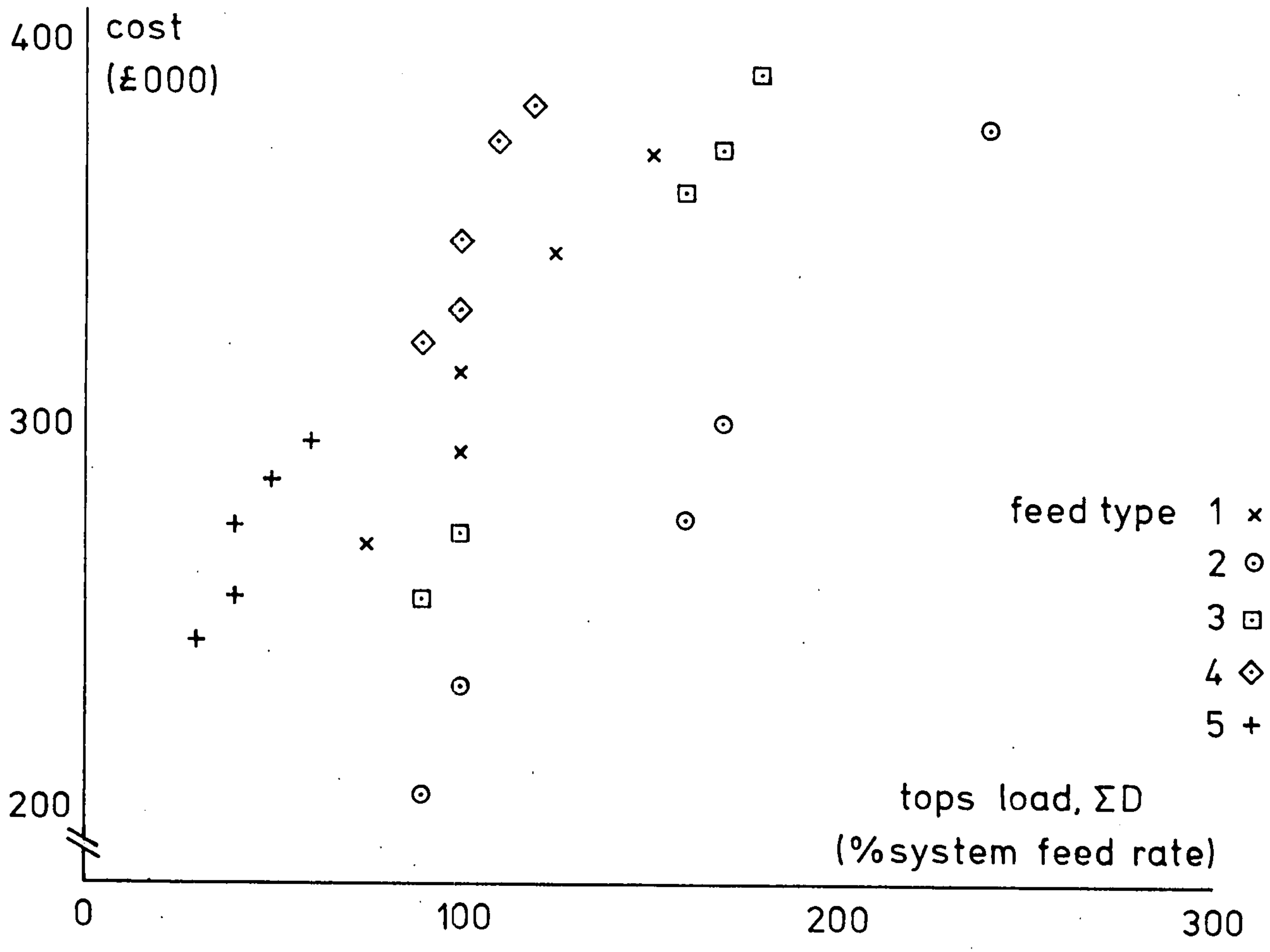


Figure 6.13 Cost vs tops load, ΣD, for feed number 1.

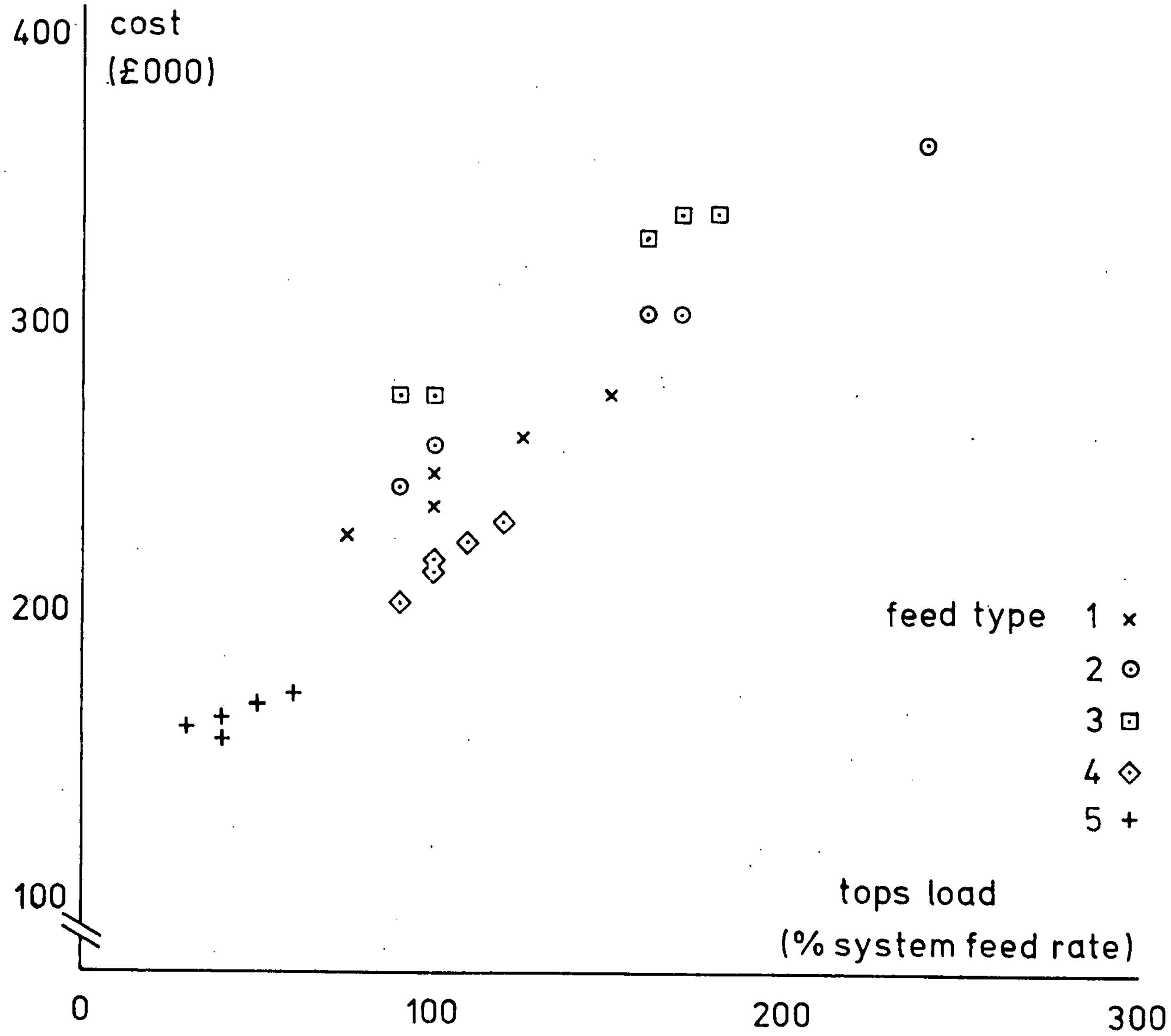


Figure 6.14 Cost *vs* tops load, ΣD, for feed number 2.

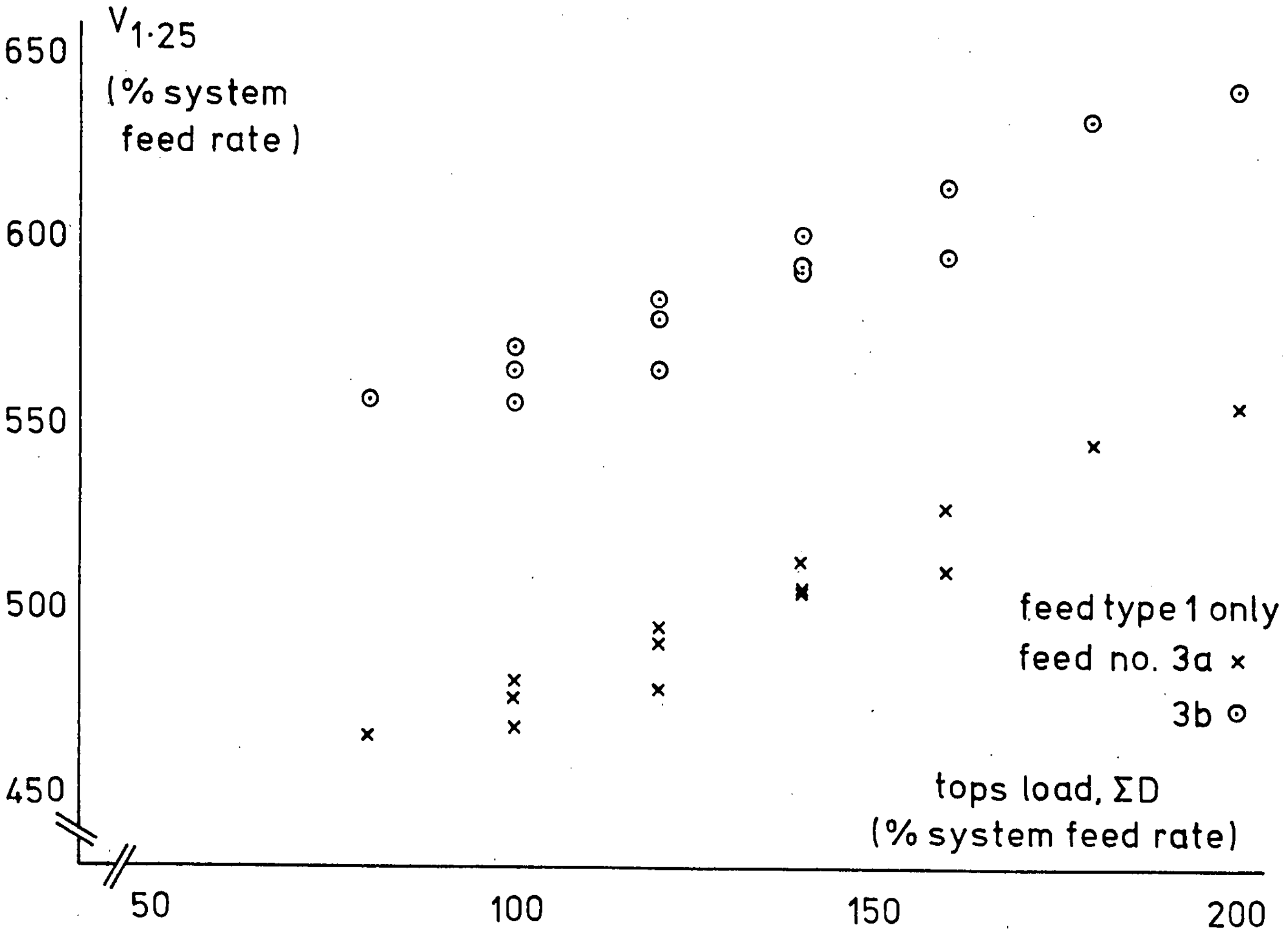


Figure 6.15 Cost vs tops load, ΣD, for feed numbers 3a,b.

physical properties of the components vary quite widely with temperature and pressure, as can be seen from table 6.2 and figures 6.16, 6.17 and 6.18. In particular the relation between the relative volatilities of the three adjacent pairs of components in the first 4-component example are very different at the three sets of conditions chosen, and are different again from the relationships in the second example. However, this variation has little effect on the general form of the results, as shown in figures 6.7 and 6.8, though it does affect the ordering of the configurations within the groups defined by the tops load, or within the equimolar or heavy major component groups. The major factor affecting the results is therefore tops load, even in cases where the ratio of adjacent excess relative volatilities (ratio of (relative volatility - 1) for adjacent pairs) is as high as 7.7 (2nd 4-component example, temperature 37.8 deg C, pressure 1.2 bar a) without affecting the grouping of results.

6.1.2.3 Conclusions

-- For distillation systems where one component is in considerable excess, and where this major component is not one of the two least volatile, the costs of the different arrangements of distillation columns form distinct groups corresponding to a similar grouping of the tops load, ΣD, of each configuration. This grouping is caused by the number of times the major component is taken in an overhead product in the sequence. The order of costs does not always correlate

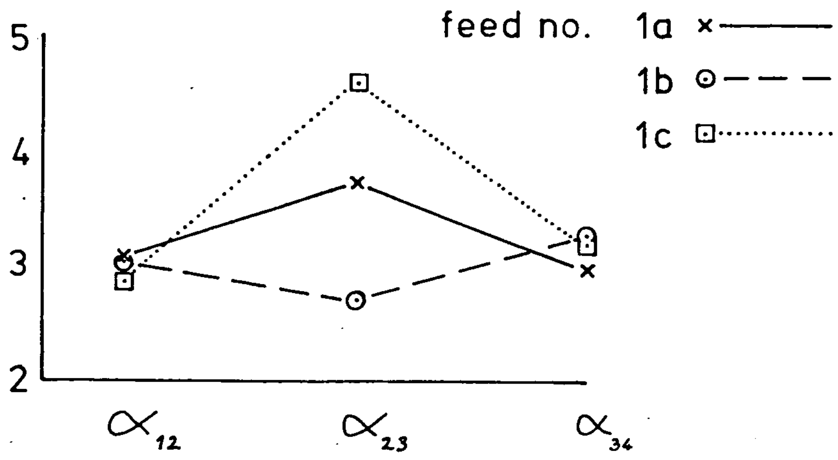


Figure 6.16 Variation of relative volatilities with feed conditions for feed number 1.

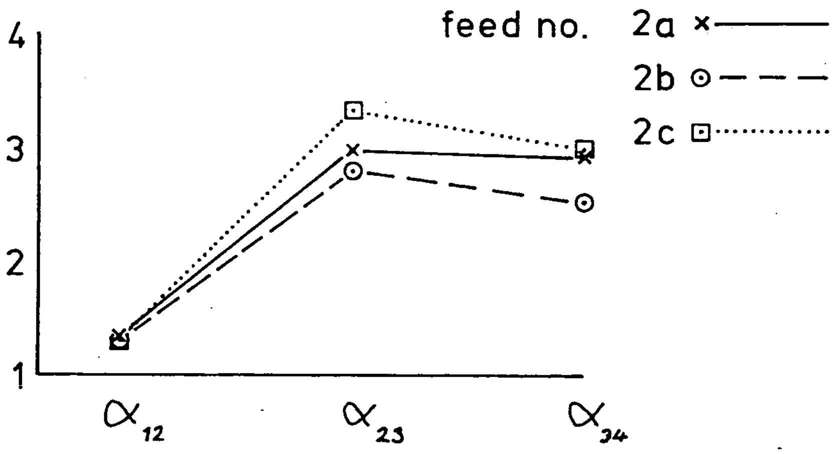


Figure 6.17 Variation of relative volatilities with feed conditions for feed number 2.

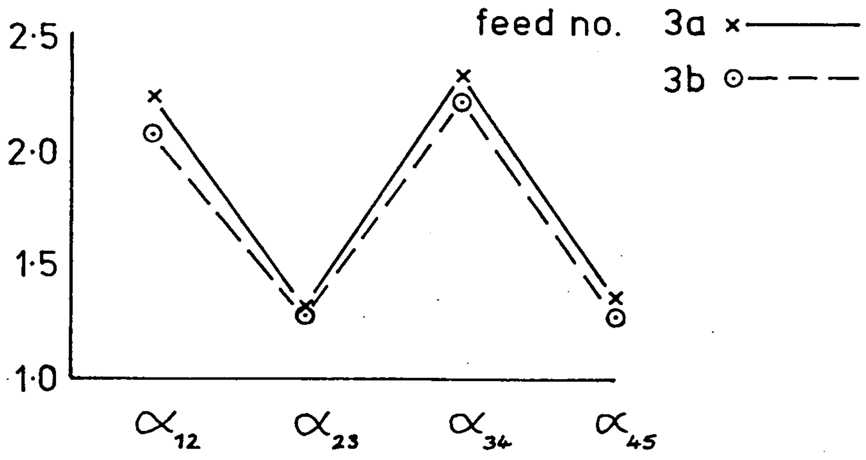


Figure 6.18 Variation of relative volatilities with feed conditions for feed number 3.

exactly with ΣD, but the grouping provides a simple way of screening out a set of configurations for more detailed design and evaluation.

-- For cases where the feed to a distillation system is equimolar this grouping will not occur, but there is still a strong correlation between ΣD and cost or vapour rate. This suggests a similar screening heuristic, namely to consider those configurations with the lowest values of ΣD. Unfortunately there is no obvious set to choose as there is with the situations covered above.

-- For feeds with a predominant component which is one of the two least volatile components no grouping of results occurs, and there appears to be no strong correlation of cost with ΣD. However the costs tend to be distributed within a narrow range and so, at least in the absence of any heat integration exercise, there is in fact little to choose between any of the configurations anyway -- the design may be chosen at the engineer's whim!

Although these conclusions have been derived for 4- and 5-component feed mixtures they are very likely to be applicable to mixtures of more components where the relative volatilities of each pair of adjacent components are not extremely different.

6.2 Use of Vapour Rate in Synthesis Methods

The work discussed here concerns one possibility for the use of the vapour rate (either V_{min} or $V_{1.25}$) in a synthesis method. It is based on two particular observations. One of these is empirical and in the event seems unlikely to lead to any further gain. The other is a fundamental feature of distillation and similar separation methods and may be of use in contexts distinct from the the use reported here. This latter observation will be discussed first.

6.2.1 First Observation -- Key Pairs

The first observation, fundamental to distillation systems, is that in any well-behaved distillation sequence, that is with only one ordering of components by separation factor, every adjacent pair of components appears as a key component pair once and once only. This observation is hinted at by Gomez and Seader [64] and Thompson and King [17,18], both of whom also make use of the assumption that a separation is cheapest in the absence of non-key components. Gomez and Seader use the assumption in a synthesis method where they take the cost of the remaining binary separations as a lower bound on the cost of completing an unfinished flowsheet. This procedure makes explicit use of the assumption that the cost of the bare binary separation is cheaper than the multicomponent separation (else the lower bound would not be lower!). It also uses the key pairs observation implicitly to construct a finite set of binary pairs still to be split.

It is clear that any of the distillation configurations for separating a given feed into given products will use precisely the same set of (n - 1) key component pairs in its (n - 1) columns, where n is the number of products to be isolated. Therefore the choice of which configuration is cheaper in any situation is governed not by which components are the keys, since the same pairs are used in every case, but by how the non-key components are distributed around the set of key component pairs. The argument holds true whether the (n - 1) products are single components or multicomponent products, but breaks down when the volatility ordering of components changes with concentration, the presence or absence of components.

Considering distillation systems in general, any system can be considered to consist of a basic skeleton of binary separations and the different practical configurations may be formed by distributing non-key components appropriately around this skeleton. The set of binary separations will allow a lower bound on the cost of the whole system to be calculated if the assumption regarding the cost of adding non-keys is true. This binary set is a fundamental feature of any mixture, and is valid for separation techniques other than distillation which have only a single ordering of components by separation factor.

The possible use of this this observation in a synthesis method lies with the recognition that the different distillation configurations are distinguished not by the key component pairs which they split but by the distribution of non-keys around the system. Thus if there were a simple method of calculating the cost of adding non-key components to a binary separation then a synthesis method could be based on the comparison of the different distributions of non-keys in the different complete processes.

6.2.2 Vapour Rate and Non-Key Concentration

6.2.2.1 Basic Observation

The second observation, which it was thought might provide a marginal costing method for the idea outlined above, is that to a fairly good approximation vapour rate is a linear function of the mole ratio of non-key component in the appropriate product, that is light non-key in the top product or heavy non-key in the bottom. To reflect the idea of adding non-keys to a binary separator vapour rate is here expressed in multiples of the feed rate of key components, hence the consideration of this quantity as a cost correlate in the previous chapter. This relationship is shown in figures 6.19 and 6.20 for simple ternary systems with a single non-key component. The parameter in this case is the ratio of the flowrates of the key components in the feed, the volatilities of all the components remaining constant. This approximate linearity also extends (figures 6.21-24) to systems of 4 and presumably more components, whether the two non-keys are either both light, both heavy or one of each. Thus it seems that in every case there may be

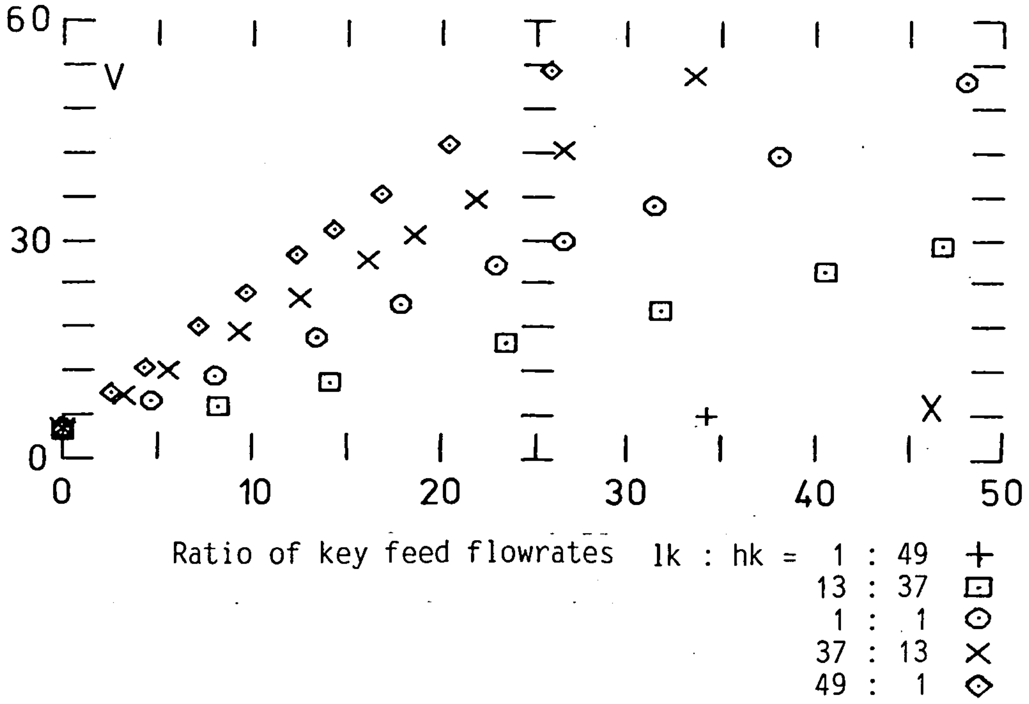


Figure 6.19 Vapour rate in units of key component feed rate vs mole ratio of light non-key in tops, ternary system.

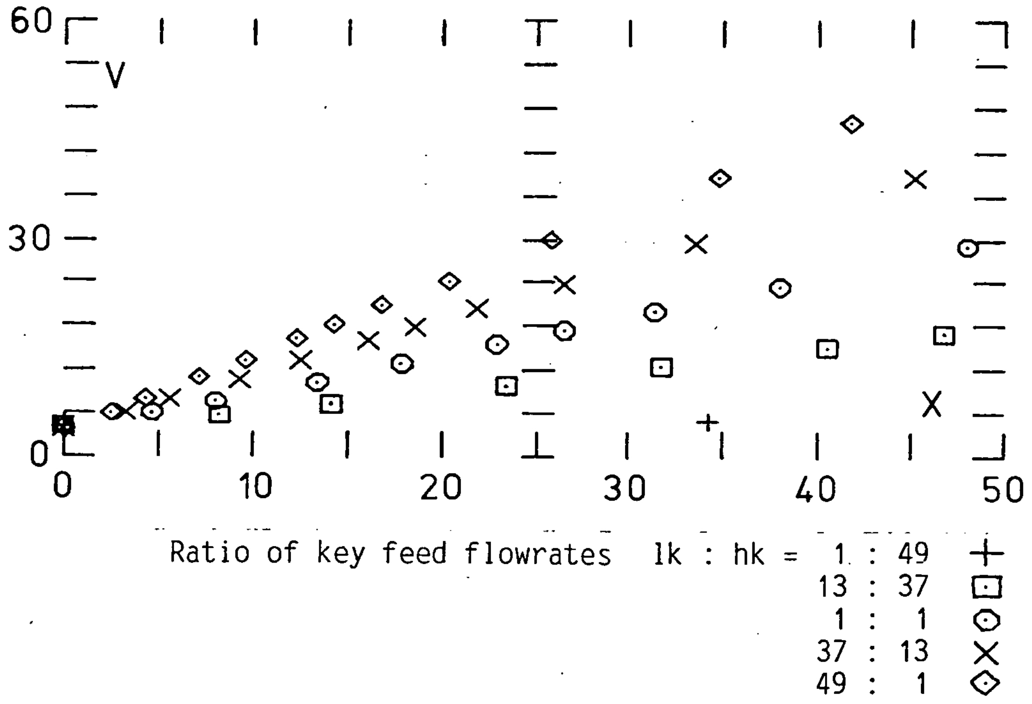


Figure 6.20 Vapour rate in units of key component feed rate vs mole ratio of heavy non-key in bottoms, ternary system.

a relatively simple relationship for the marginal cost of adding non-key components to a binary separation. for this observation to be useful we must be able to calculate the intercept and gradient of of the V vs X line for any set of components. Determining the intercept on the V axis is simple from the analytical binary form of the Underwood equations, but the gradient presents a more difficult problem.

6.2.2.2 Use

Here two fairly obvious ideas for determining the gradient of the V vs X line are discussed. The first of these is to generate another point on the line so that by simply joining it to the intercept the gradient is defined. This idea is rather pointless since any one line is applicable only to a single set of components and flowrates, and therefore only one point on the line is usually needed for the design. The use of an Underwood evaluation to find this point is precisely what we are trying to avoid, and using the Underwood equations in this way requires exactly the same number of evaluations as would a full unoptimised design!

The other approach considered is the derivation of an empirical equation describing the line. This derivation has not been attempted, but several points must be borne in mind. The first of these is that the 'line' that is being approximated is in fact slightly curved which may introduce error since the straight line approximation can only fit exactly at two points, one of these probably being chosen as the intercept on the V axis. Another possible difficulty lies in the likely complexity of the expression for the gradient. Figures 6.21 and 6.22 show that approximate linearity is maintained for either a light or a heavy non-key in the presence of the other, that is for quaternary systems with both light and heavy non-keys. Similarly Figure 6.23 shows that a system with two light non-keys also exhibits linearity. What these graphs and the earlier ones show is that the gradient is a function of several variables, including feed mole fractions of the key components and relative volatility. While these results are not conclusive they do suggest that the gradient we are seeking is a function of all the component flowrates and volatilities, as might also be suggested by the form of the Underwood equations. This in turn also suggests that there might need to be considerable complexity in an empirical expression for the gradient. Although no attempt has been made to derive such a correlation one possibility for simplifying one is considered below.

The method in question is to lump all the light non-keys together into a single pseudo-light non-key and all the heavy non-keys similarly. The parameter describing such a pseudo-component would be a pseudo-relative volatility, α_p. Figures 6.23 and 6.24 show an attempt to check on whether it would be possible to use a simple mole fraction weighted average of the light non-key relative volatilities as α_p in this context. If it were possible then all the results shown in figure 6.23 would lie on the same straight line, that shown in figure 6.24. Figure 6.24 shows

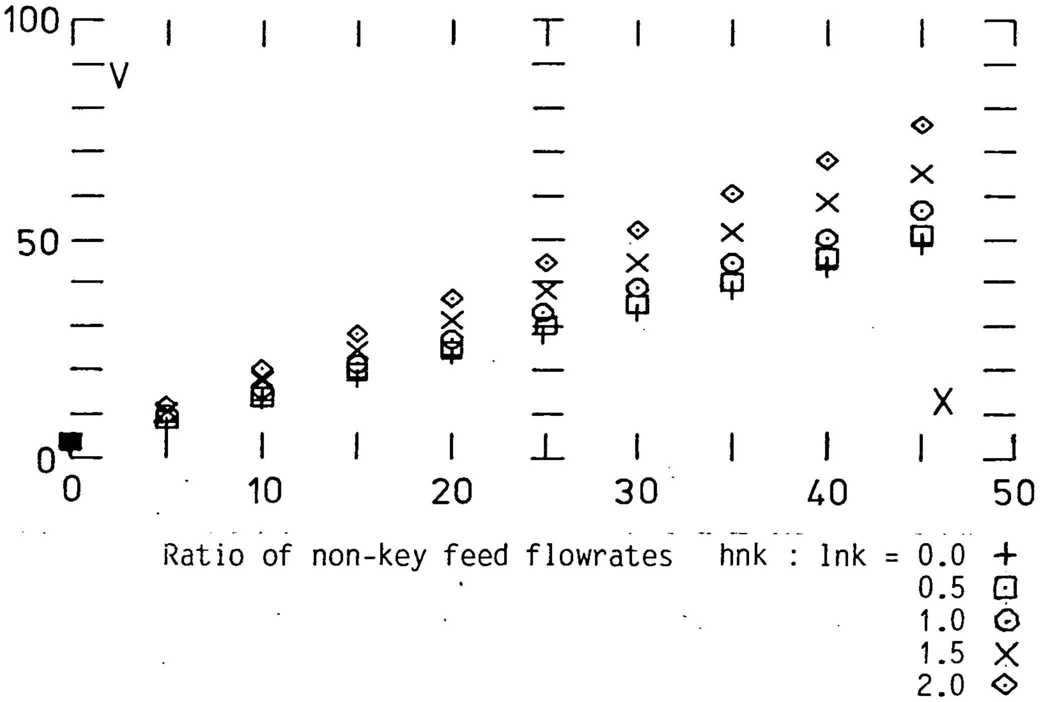


Figure 6.21 Vapour rate in units of key component feed rate vs mole ratio of light non-key in tops, quaternary system, differing concentration of heavy non-key.

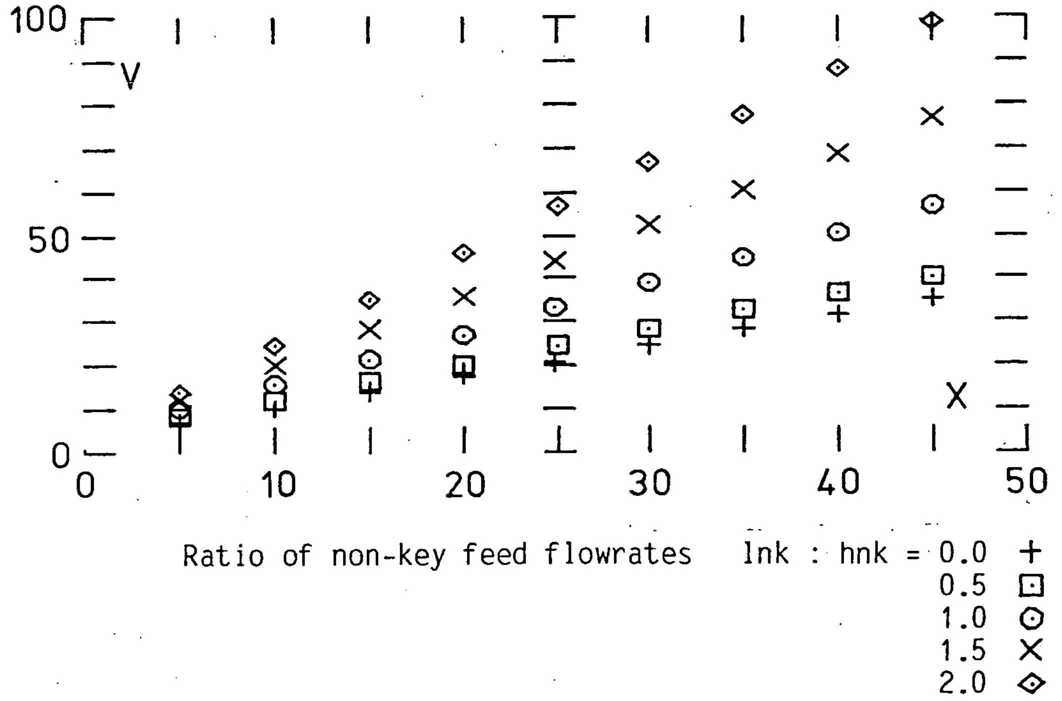


Figure 6.22 Vapour rate in units of key component feed rate vs mole ratio of heavy non-key in bottoms, quaternary system, differing concentration of light non-key.

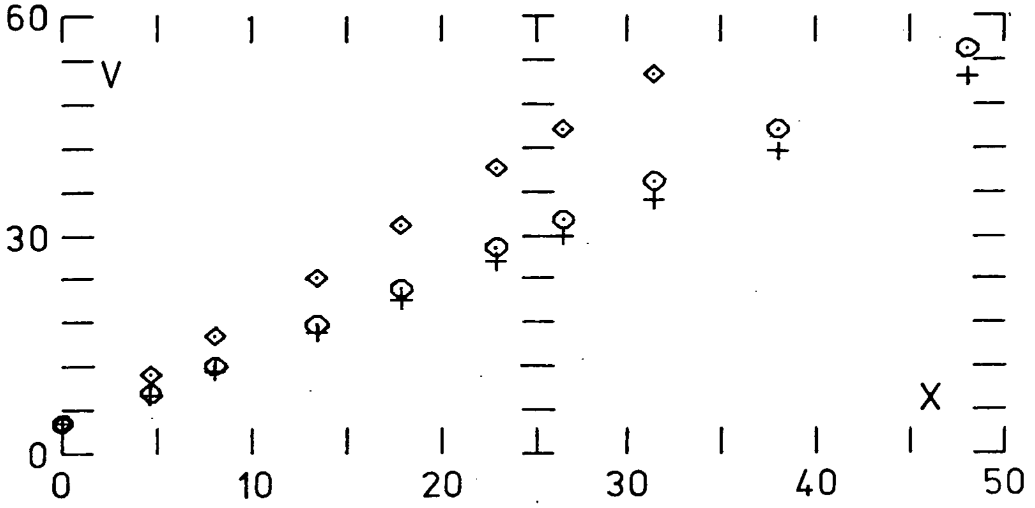


Figure 6.23 Vapour rate in units of key component feed rate vs mole ratio of light non-keys in tops, quaternary system. Mole fraction weighted mean of non-key relative volatilities is constant.

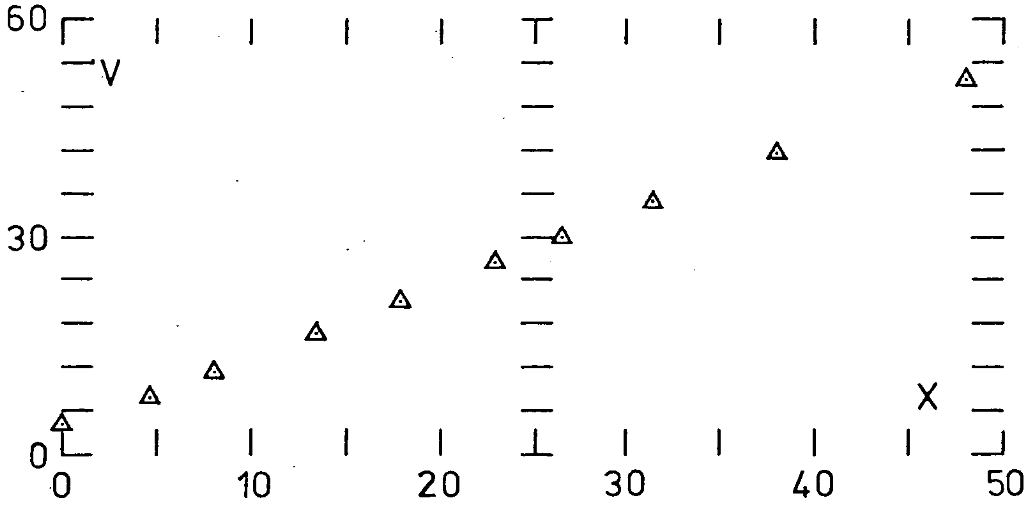


Figure 6.24 Vapour rate in units of key component feed rate vs mole ratio of light non-key in tops, ternary system. Relative volatility of light non-key equal to weighted mean of figure 6.23.

the results for a ternary system where the light non-key has the appropriate mole fraction weighted average relative volatility. The check clearly fails, and an attempt to derive a formula for α_p consistent with the Underwood equations will also fail. Consider. From the form of the Underwood equations what is needed is simply a value of α_p which will satisfy the following

$$\sum_{i \epsilon NK} \frac{x_i \alpha_i}{\alpha_i - \theta} = \sum_{i \epsilon NK} (x_i) \cdot \frac{\alpha_p}{\alpha_p - \theta}$$

where NK is a set of non-key components, light or heavy.

For the simplest non-trivial case, where there are two light non-keys or two heavy non-keys this expression becomes

$$\alpha_p = \frac{\theta\ (\alpha_1 x_1 + \alpha_2 x_2) - \alpha_1 \alpha_2\ (x_1 + x_2)}{\theta\ (x_1 + x_2) - \alpha_1 x_2 - \alpha_2 x_1}$$

The value of α_p is therefore dependent on θ, the Underwood parameter, the determination of which requires precisely the iterative calculation which we are trying to avoid.

6.2.3 Summing Up

In this section we have noted two observations which, taken together at face value, appear to have some use in the synthesis of distillation networks. The first observation is fundamental to distillation and other separation techniques with similar characteristics, namely that every simple configuration consists of the same skeleton of binary separations and that what distinguishes the configurations is the distribution of non-key components around this skeleton. The second observation suggests a simple way of using the first by calculating the marginal cost of adding non-key components to a binary separation. However, although not enough work has been done to make the results conclusive, this particular method for calculating the marginal cost turns out to be rather more complicated than expected and seems unlikely to produce the simplification required.

6.3 Conclusion

The work reported in this chapter, though largely preliminary in nature, gives rise to several interesting points which should be investigated further.

-- The close correlation of minimum vapour rate (V_{min}) and reboiler duty with cost is significant and should be confirmed in systems other than the one reported here. Particular attention should be paid to the effect of using different heating media on the cost of distillation networks.

-- The work on the systems studied by Freshwater and Henry [46] raises two points meriting further work. The first of these is the correlation of cost with tops load, ΣD, for systems other than those whose feed contains a large proportion of heavy product. The second point is the grouping of costs or vapour rate for systems with a non-heavy major component. These observations allow a preliminary screening to be applied in the choice of distillation networks.

-- Finally in the last piece of work consideration is given to a fundamental feature of systems of distillation columns and of other systems with similar characteristics. Unfortunately an evaluation method suitable for utilising the observation in a synthesis method has not been discovered.

The first two points above are dependent on the cost structure of distillation systems, in that for them to be of any use in design then the major cost of distillation must be due to energy use. However, even were this not so, energy use would still govern the material flows in distillation systems and thus their size, a point discussed in chapter 5. The last point is at the moment an observation in search of an application, so even though it is intrinsic to distillation systems it is not yet clear whether it will be of any use in design.

7 Summary of Conclusions and Suggestions for Further Work

7 Summary of Conclusions and Suggestions for Further Work

7.1 Summary of Chapter Conclusions

7.1.1 Literature Review

-- In heat exchange network synthesis, a rather simpler problem than separation scheme synthesis, many different synthesis methods have been tried. Arguably the most successful approach to date has been based on understanding of the basic principles of both the design and costing of heat exchange networks. Methods of analysing the data of particular problems are provided which reveal targets for design, and synthesis methods follow naturally from the nature of the targets.

-- Separation scheme synthesis has been investigated using the same range of methods as employed for heat exchange networks. There has not however been the same sort of success with analysis and target setting as there has been with the earlier work. This may be due to the inherently more complex nature of the separation process, but may also be due to the lack of application of similar approaches by workers in this field. It must be noted that such an approach is reported to be imminent.

7.1.2 S6

-- The S6 program was expected to produce small numbers of separation flowsheets which were either near optimal or contained exciting new features to be investigated. It does neither of these things at present.

-- It does however indirectly point out which multicomponent products may be separated intact and which must be split using a particular set of separation techniques.

-- S6 might be made to work as intended by extensive adjustment of the screening parameters. However the major gain from this is likely to be in the engineer's understanding of his particular problem and of separation scheme synthesis in general. Better training methods could probably be devised...

7.1.3 Minimum Number of Separators

-- The minimum number of separator types required to produce a given product set is not always a particularly useful quantity to know, but more valuable information can be derived. For example it it is possible to determine which separator type or types, other than a given 'base' type, is or are needed to isolate a given set of products. Alternatively which products need be split and which may be isolated intact can be determined explicitly and with rather less effort than required by the S6 program.

-- It is possible to calculate the minimum number of separation units required, using either of two conventions regarding mass separating agent recovery. Examples reveal that the minimum number of units is not necessarily a very good guide to minimum cost.

-- The examples also reveal that separator types utilising a mass separating agent are often used in the optimal flowsheet. However they are not usually used to recover multicomponent products intact and thus minimise the number of separation units required, but are used instead to replace expensive separators using energy separating agents.

7.1.4 Ternary Evaluation Functions

-- This work reveals that the simple external mass flow based functions investigated did not correlate well with cost.

-- The study of reboil related functions showed better correlation with cost, however these functions are unlikely to be useful as alternative evaluation functions due to the relatively large effort required to compute them.

-- The major gain from this study is the understanding that for distillation systems the cost of energy, to which the internal vapour rate is closely linked, comprises a major part of the total system cost.

7.1.5 Further Reboil Investigations

-- Analysis of Thompson and King's results [17] for systems of more than three components suggests that the use of steam supplies at discrete temperature and pressure intervals upsets the correlation of reboiler duty with cost. (The Appendix shows a similar feature for ternary systems.) In such circumstances the correlation of internal vapour rate with cost is better.

-- For distillation systems where one component is in considerable excess, and that major component is not the least or next to least volatile, it is possible to find a group of flowsheets whose costs are likely to be lower than those of other configurations. This group will probably contain the cheapest process.

-- For distillation systems other than those described above then ranking processes by the sum of the top product flowrates of the columns in the flowsheet may provide a preliminary screening method.

-- It is recognised that the differences in cost between different distillation flowsheets for the same duty are caused by the different distributions of non-key components around the common set of key components pairs. The lack of a simple method for calculating the marginal cost of adding non-key components to a binary separation presently prevents the exploitation of this feature in a synthesis method.

7.2 Overall Summary and Suggestions for Further Work

The beginning of the work described here was concerned with a very general synthesis method for separation systems, discarding cost as an evaluation function in the hope of discovering good processes on grounds perhaps more fundamental than cheapness. Although the results proved disappointing they did help spur the second section of the work which covered known sets of separator types. Eventually the focus of the work narrowed to consider only distillation. The facts discovered in the final pieces of work, confirmation that the cost of distillation depends largely on energy use and that that is governed to a large extent by the quantity of material in the top product merit further investigation.

Suggestions for further work must therefore include the following:

-- from S6: It may be worth attempting fine tuning of the method to come nearer to achieving the original aims, but this must be considered a project of relatively low priority.

-- from the investigation of the minimum number of separators and from S6: It would be interesting to extend the idea of looking for unknown separator types by searching for mass separating agents whose use would enable particular products to be isolated or which could be used in separators replacing particularly difficult distillation units. The search would be in two parts. First ranges of physical constants would be calculated for the target specie which would give the desired mixture properties. The second phase would be the searching of a physical properties database for a material with the required constants.

-- from the minimum separators work: It might be possible to discover more useful targets than those already discussed in this work and to develop methods of determining them. Considering the later chapters here some sort of prediction of energy use in distillation would be particularly useful. It is possible that synthesis methods will arise quite naturally when appropriate goals have been defined.

-- from the work on ternary distillation: Since the functions which approximated best to cost required considerable effort to calculate other simpler functions might be investigated as alternatives to cost. The possible danger here is that such an approach may not lead to a greater understanding of why distillation costs are distributed in the way that they are, but will give rise instead to purely empirical design methods.

-- from the further investigation of reboil functions: Work must be continued on understanding the correlation between cost or vapour rate and tops load. The important point here is that it is more expensive to take a component in the overhead rather than in the bottom product.

-- finally, also from the further investigation of reboil functions: Work should continue based on the observation that in networks of distillation columns it is the non-key rather than the key components which determine the cost of the system.

Appendix: Costs of Ternary Distillation Schemes

Appendix: Costs of Ternary Distillation Schemes

Several workers have studied the cost of ternary distillation systems. Most have looked only at the two simple sequences. Here the cost results from chapter 5 are compared with those of other workers presented in a standard format.

A.1 Results of This Study

The results reported in this appendix were obtained using the same mixtures of species and the same methods as those reported in chapter 5. In particular the feed mixtures are those of Tedder and Rudd [39,40] and the design and costing method that of Rathore et al [52,53] -- see sections 5.2.1.1 and 5.2.2. The format of the results as ternary diagrams is the same as used in that chapter also, see section 5.2.1.3. There are three variable factors covered in this study, namely the way the composition of the top product is related to that of the feed, the pressure of the system and the temperature of steam used in the reboilers of the system. These are examined below.

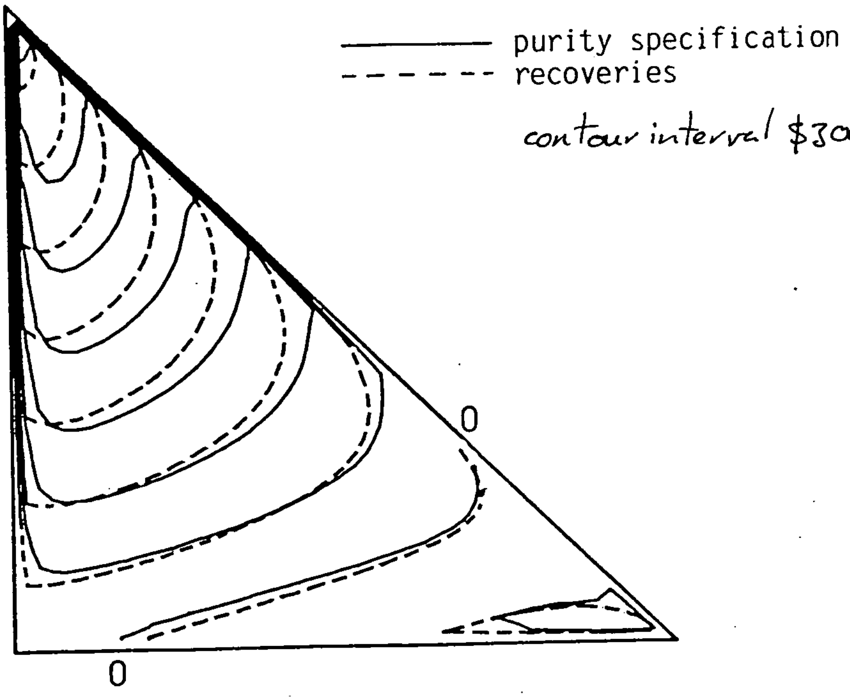
A.1.1 Top Product Specification

It is possible to specify the top product in at least two ways: one is to specify the recovery fraction of components in the two products, for instance that 95% of the light key component appears in the top product; the other is to specify the concentration of components in the product, for instance that the top product will be no more than 1% of the heavy key component. These two methods might typically be used in different situations, for instance recoveries might be used when designing a recycle stream, and the purity method in a product finishing train.

As to results, figure A.1 shows the contours of cost difference for feed number 1 (see table 5.1 for details) for two different situations, one where the top and bottom products were specified to have only 1% concentration of the 'wrong' key component, and the other where 95% of each key in the feed appeared in the appropriate product. It is easily seen that there is little effect on the shape or position of the contours.

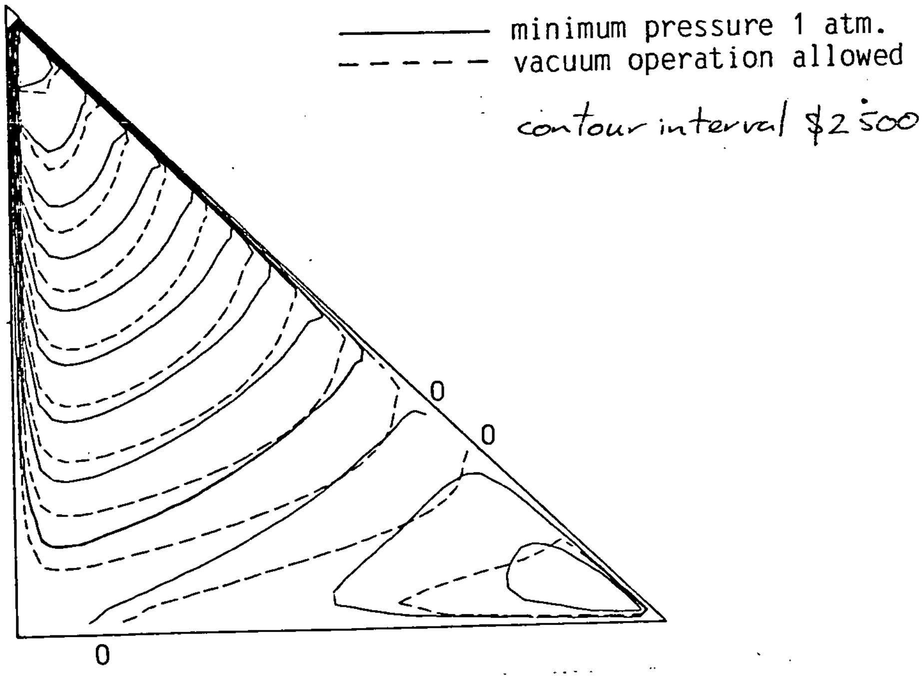
A.1.2 Effect of Pressure

In this study two options were available for setting the pressure in the two columns of the system. In every case the pressure was initially chosen to be the lowest at which cooling water could be used for condensing the reflux (see chapter 5 for an explanation of this). In some cases this would result in the pressure in one or both of the columns of the system being set to below atmospheric pressure. The option allowed this probably unrealistic situation to be modified so that the pressure was set at 1 atmosphere. The effect of not allowing vacuum operation is a little more marked than that of the method of product specification,



contour interval $3000

Figure A.1 Effect of product specification, feed number 1



contour interval $2500

Figure A.2 Effect of allowing vacuum operation though more restricted since it affects only systems with higher boiling point components. The effect is to raise the cost of the inverted relative to the direct sequence, thus making the direct sequence cheaper over more of the diagram, and thus moving the zero contour toward the BC edge. Figure A.2 shows this effect for feed number 1.

A.1.3 Effect of Steam Specifications

There are two possibilities for specifying how steam is supplied to the reboilers of the distillation system. One of these is the method used by Tedder and Rudd [39,40] in a similar study to this one, where steam is considered to be available at a set of predefined temperatures with associated costs. This is the 'discrete steam levels' case. The other case, with 'smooth steam levels', is where steam is considered available at any temperature at which it is required, the cost varying accordingly. In both cases the specification of reboiler temperature difference is a further variable.

A.1.3.1 Smooth or Discrete Steam Levels

It is obvious that the use of discrete steam levels will introduce discontinuities into the cost function as the steam temperature employed changes from one level to another. Often, however, the same steam levels will be used over the whole extent of the triangular diagram. This situation will be more likely when the boiling points of the components are more widely separated. Figure A.3 shows the cost difference contours for the six feed mixtures studied by Tedder and Rudd when the columns were designed using discrete steam levels. In fact, of the six feeds studied only numbers 1 and 3 (figure A.3(a) and A.3(c)) showed any discontinuities in the cost contours. These mixtures have the highest range of boiling points of those studied, and the discontinuities occur towards the AC edge where the feed mixture will have the highest boiling range. As can be seen from figure A.4 these discontinuities disappear when smooth steam levels are used instead of discrete.

In an attempt to discover a relationship between the properties of the feed mixture and the position of the zero contour figure A.5 has the zero contours from figures A.3(a)-(f) plotted on one diagram. There is no readily discernible pattern of variation of the zero contour with the ESI or MESI of the feed mixture. (For discussion of the terms ESI and MESI see chapter 5). On the other hand, for systems with smooth steam levels, as shown in figure A.6, there is a visible correlation of cost with ESI or MESI, and figure A.7 reveals a roughly hyperbolic variation in the cases with smooth steam levels. It is obvious that the use of discrete steam levels disturbs any ordering of zero contour position with ESI or MESI.

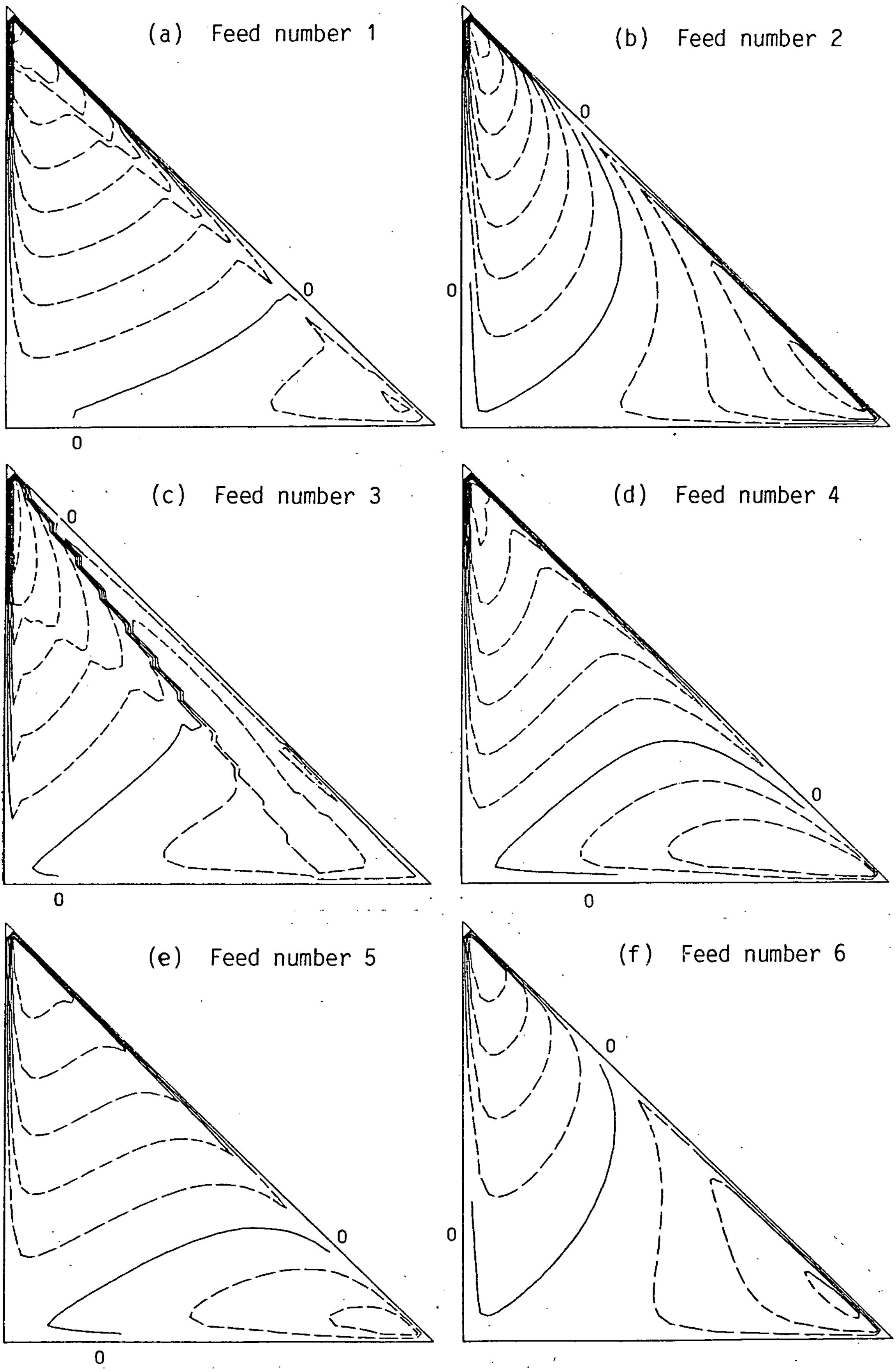
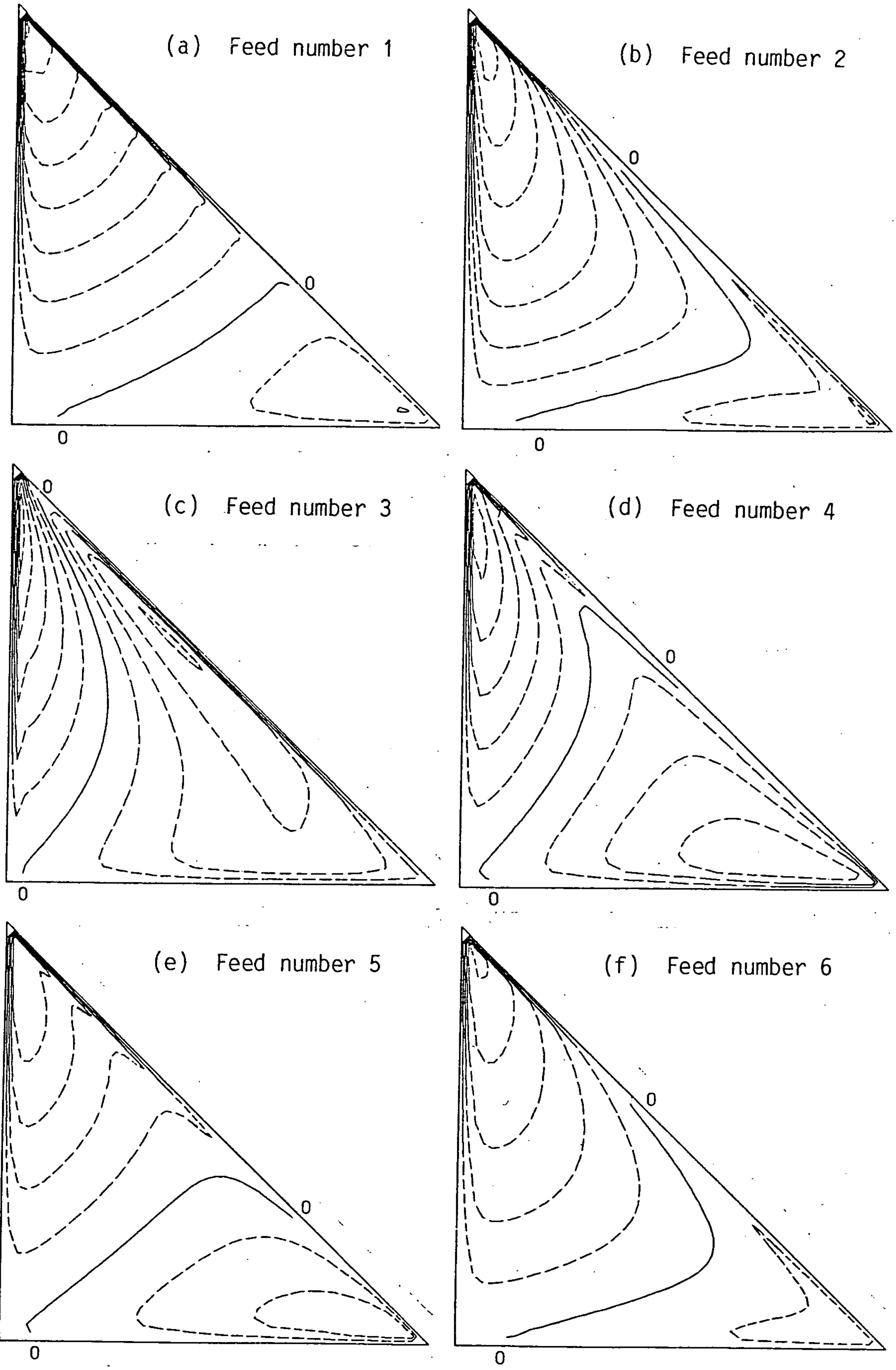


Figure A.3 Cost contours for designs using discrete steam levels

contour intérval $2500



(a) Feed number 1

(b) Feed number 2

(c) Feed number 3

(d) Feed number 4

(e) Feed number 5

(f) Feed number 6

Figure A.4 Cost contours for designs using smooth steam levels

contour interval $2500

Figure A.5 Comparison of zero contours from figure A.3, discrete steam levels

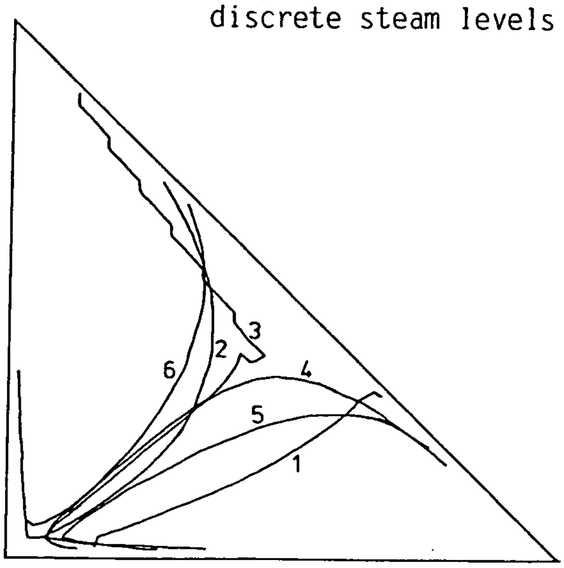
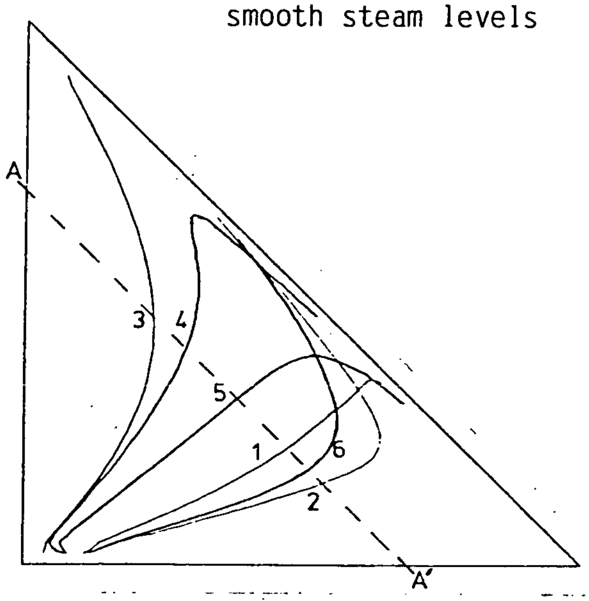


Figure A.6 Comparison of zero contours from figure A.4, smooth steam levels



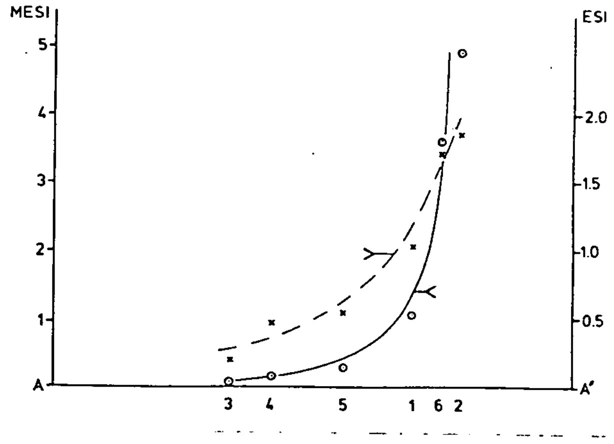


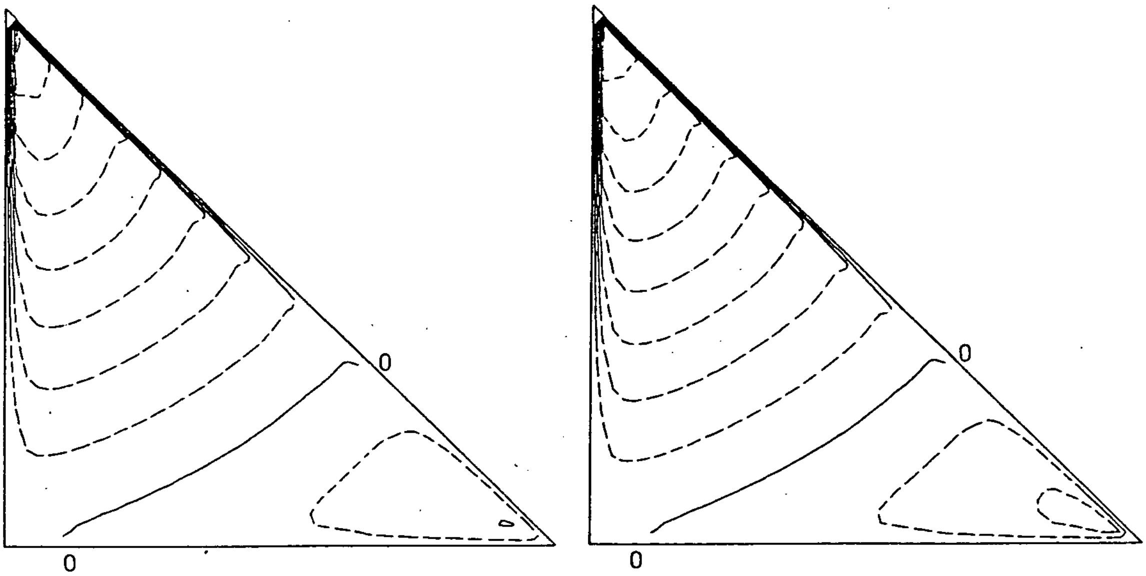
Figure A.7 Section A-A' from figure A.6 showing position of zero contours and correlation with ESI and MESI

A.1.3.2 Reboiler Temperature Difference

For the smooth case we will now turn to the effect of reboiler temperature difference. Figures A.8, A.9 and A.10 have the contours for feeds number 1, number 5 and number 6 at reboiler temperature differences of 20, 29 and 50 degrees C. The effect on the general shape of the contours is negligible, although the zero contour moves slightly, though not systematically with ESI or MESI. The effect on the position of the contours is to deepen them, the costs of both configurations increasing.

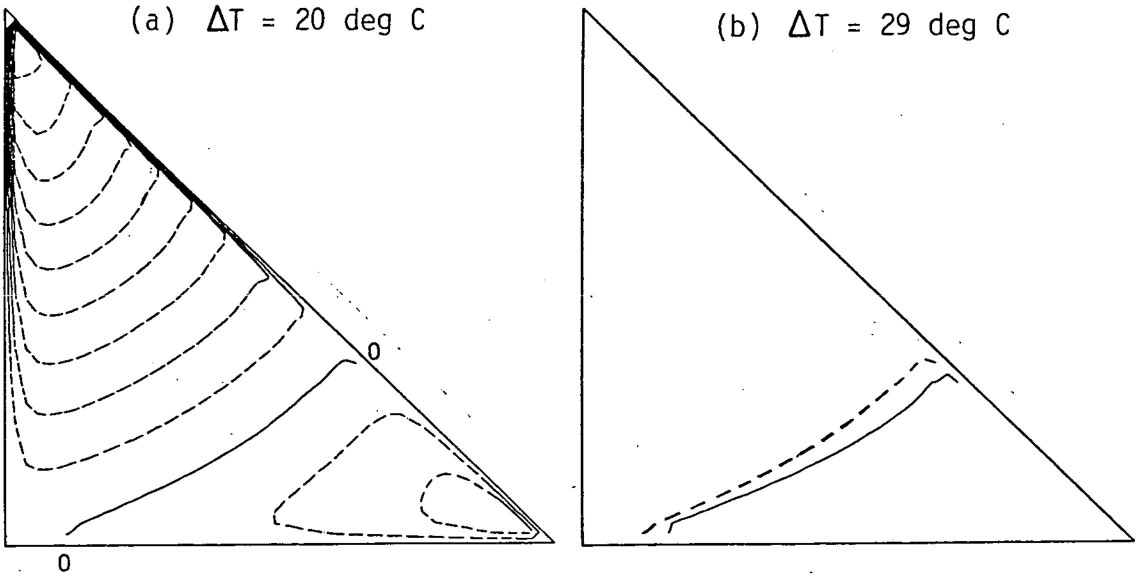
A.1.4 Conclusion

The major conclusion from this work is that there is a recognisable trend of zero contour position on the ternary diagram with feed ESI or MESI for the case where steam is considered available at any required temperature: as ESI or MESI increases the direct sequence is favoured over more of the feed domain, the zero contour moving from the AB edge toward the BC edge. However where steam is available at only predetermined levels the discontinuities introduced into the cost surface are enough to disturb this trend. Vacuum operation of distillation columns also has an effect, though not so large a one as discrete steam levels. On the other hand reboiler temperature difference and the method of specifying the top product composition have relatively small effects.



(a) ΔT = 20 deg C

(b) ΔT = 29 deg C



(c) ΔT = 50 deg C

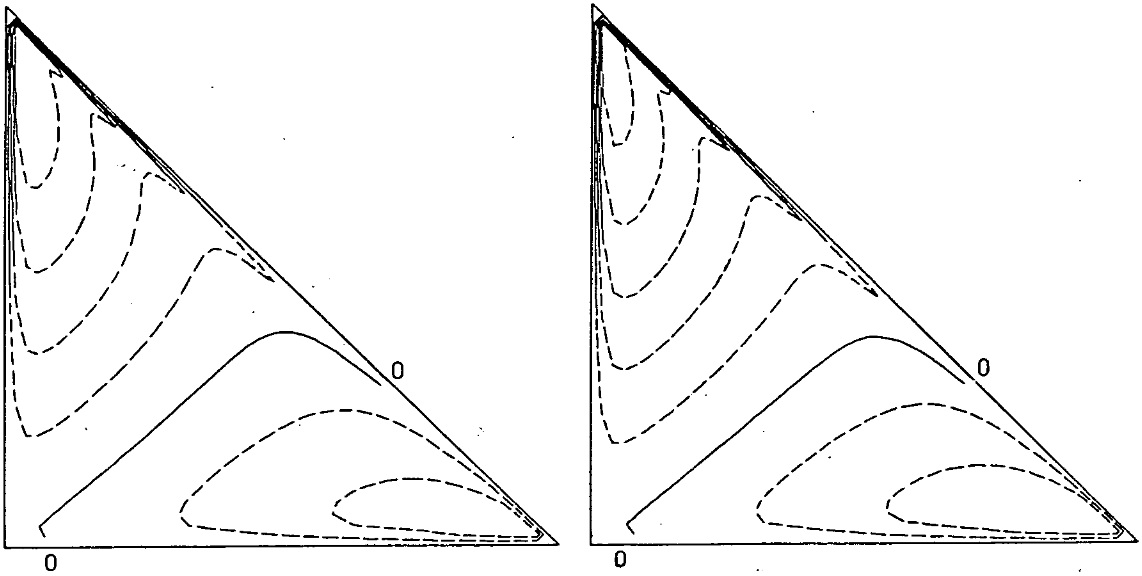
(d) Comparison of zero contours

———— discrete

- - - - - { smooth, ΔT = 20
smooth, ΔT = 29
smooth, ΔT = 50 }

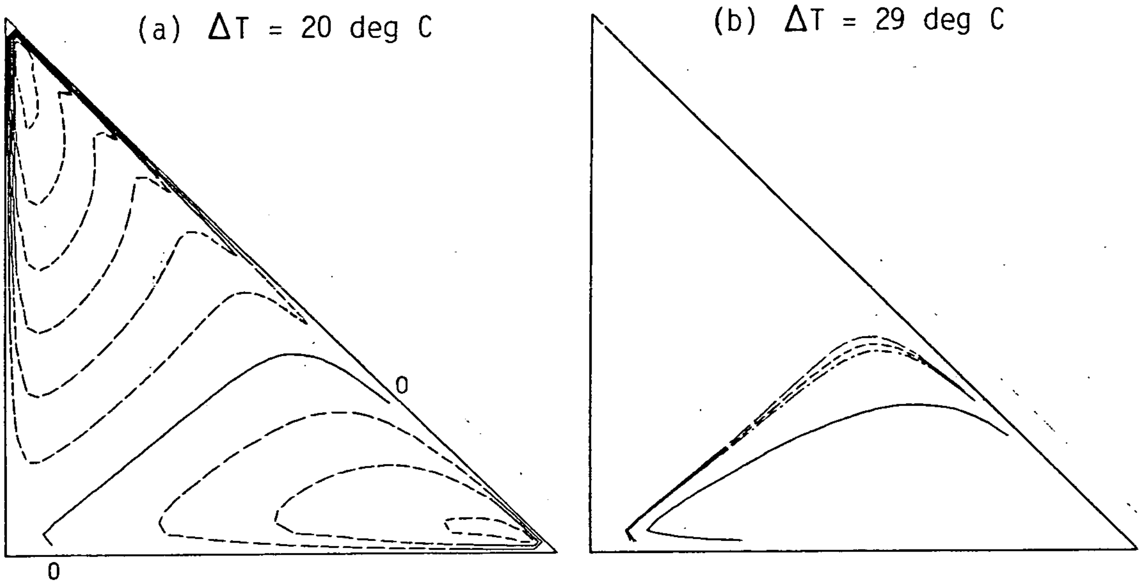
Figure A.8 Effect of reboiler temperature difference, feed number 1

contour interval $2500



(a) ΔT = 20 deg C

(b) ΔT = 29 deg C



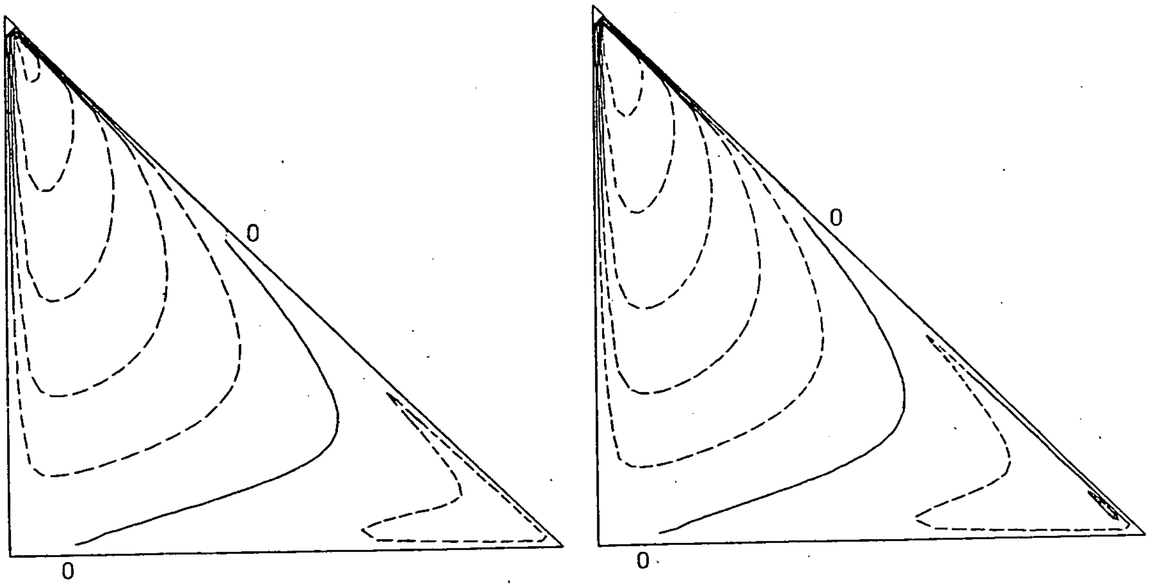
(c) ΔT = 50 deg C

(d) Comparison of zero contours

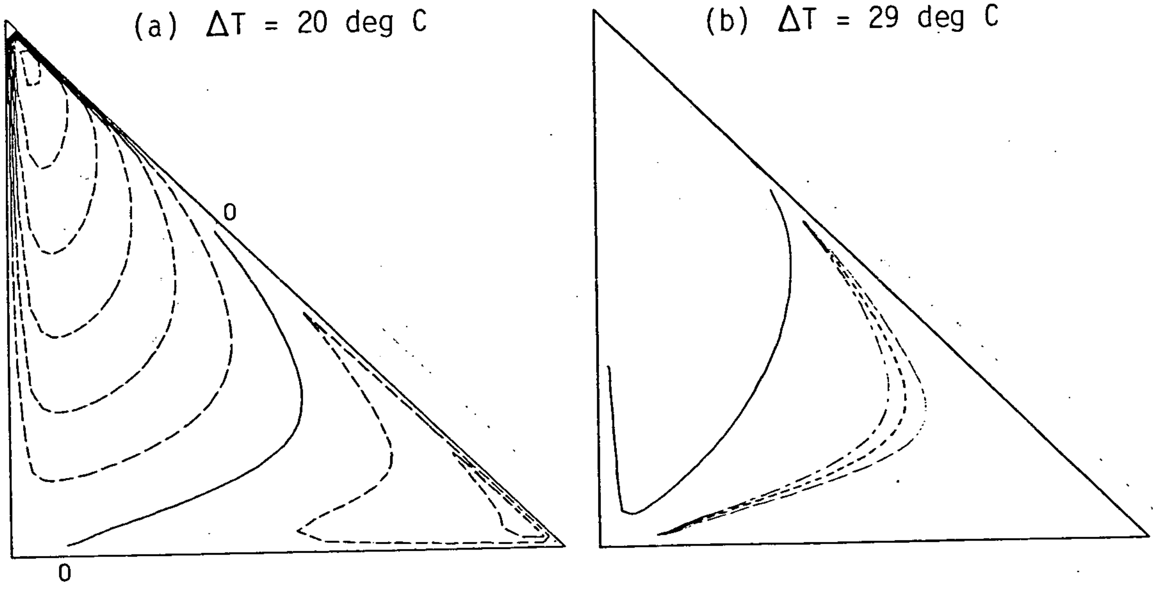
———— discrete
— — — smooth, ΔT = 20
- - - - - - - smooth, ΔT = 29
—·—·— smooth, ΔT = 50

Figure A.9 Effect of reboiler temperature difference, feed number 5

contour interval $2500



(a) ΔT = 20 deg C (b) ΔT = 29 deg C



(c) ΔT = 50 deg C (d) Comparison of zero contours

——— discrete
— — — smooth, ΔT = 20
- - - - smooth, ΔT = 29
—·—·— smooth, ΔT = 50

Figure A.10 Effect of reboiler temperature difference, feed number 6

contour interval $2500

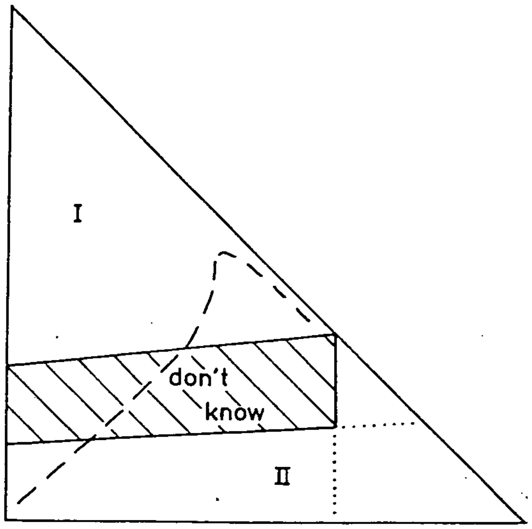
A.2 Comparison with Other Workers' Results

A.2.1 Lockhart, 1947

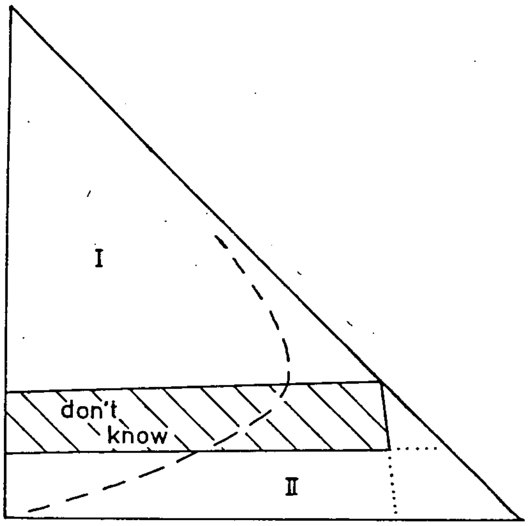
Lockhart [47] studied two systems, iso-butane, n-butane and 12lb Reid Vapour Pressure gasoline, and propane, iso-butane and 30lb RVP gasoline. He presents his results (apparently based on designs at 4 feed composition points for the first system and two for the second, hardly surprising in 1947) in terms of volume %. He divides the triangular diagram into three regions, one where the direct sequence is favoured, one the indirect, and one region where no discrimination is possible. His results are shown converted into terms of mole fractions in figure A.11. The conversion assumes that the properties of 12lb RVP gasoline lie somewhere between those of n-pentane and n-hexane, and those of 30lb RVP gasoline a little nearer to i-pentane than to n-butane. These assumptions suggest ESIs for the feeds of about 0.3 and 1.5, and MESIs of 0.15 and 3.0. Shown superimposed on figure A.11 are ficticious zero contours such as would be produced in the present work for feeds with those ESIs or MESIs. The trend of change with feed property is similar and general agreement as good as might be expected considering the difference between the two studies.

A.2.2 Rod and Marek, 1959

Rod and Marek [49] present an evaluation method which, although it is not a costing method, will bear comparison with the



(a) i-butane, n-butane, 12lb RVP gasoline



(b) propane, i-butane, 30lb RVP gasoline

Figure A.11 Lockhart's results

other results reviewed here. They assume cost to be a linear function of the vapour flow, and proportionality constant to be the same for every feed mixture and equipment configuration. They then make various assumptions and calculations and derive a comparison criterion based only on the feed flowrates and relative volatilities of the lightest and heaviest components. This criterion is called Δ and for systems where the reflux ratio is 1.25 times the minimum it is defined as

$$\Delta = \frac{(\alpha_{AC} + 0.25)\ x_A - 1.25\ x_C}{\alpha_{AC} - 1}$$

where x_A and x_C are the feed mole fractions of light component A and heavy component C and α_{AC} is their relative volatility.

The direct sequence is favoured if Δ is positive and the indirect if it is negative. This results in a zero contour for Δ which bisects the ternary diagram when $\alpha_{AC} = 1$, moving towards the BC edge as α_{AC} increases. Figure A.12 is an adaptation of figure 2 from [49] and shows the effect of changing the value of α_{AC}. Figure A.13 compares Rod and Marek's criterion with results from this work.

A.2.3 Freshwater and Henry, 1975

These workers [46] considered only one three component feed in their study of simple distillation systems, which happens to be the same mixture as feed number 2 in this work, namely n-butane, i-pentane and n-pentane, ESI of 1.72, MESI of 4.93. They find the

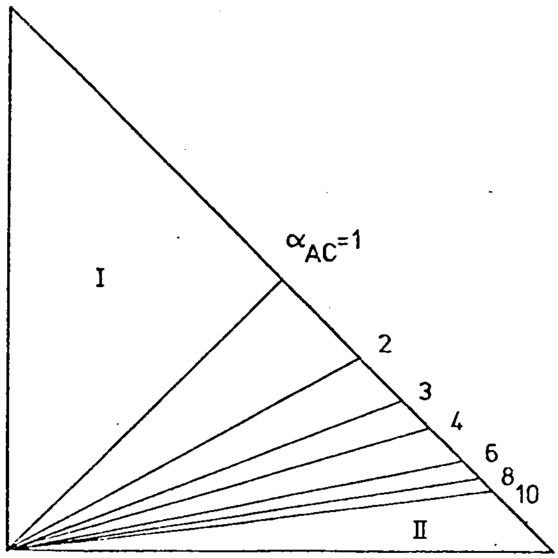
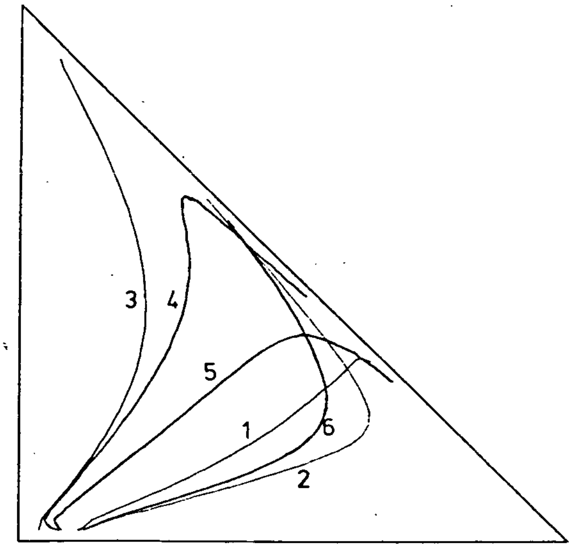


Figure A.12 Figure 2 from Rod and Marek [49]

(a) zero contours of cost, smooth steam levels, figure A.6



(b) Δ lines for same six feeds

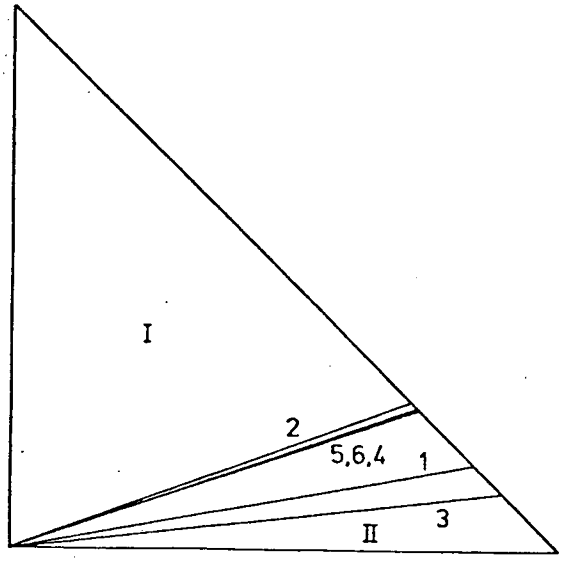


Figure A.13 Comparison of Rod and Marek's results with this work direct sequence the cheaper at all the points shown in figure A.14, and therefore by inference over the whole range of feed composition. Their designs use a reflux ratio of 1.25 times the minimum, similar to this study, but specify a constant pressure in every column. This may account for the different result, or it may be due to the very low component recoveries specified (90%).

A.2.4 Bakhshi and Gaddy, 1977

This work [44] is similar to that of Freshwater and Henry in that it considers the cost of ternary distillation systems at a few discrete points. They also optimised pressure, but this was a single pressure for the whole system, not for individual columns. They considered two feed compositions for a mixture not covered by other workers, namely n-pentane, n-hexane and n-octane, and three points for Tedder and Rudd's feed number 6, propane, i-butane and n-butane. Their results are shown in figure A.15, together with those from this work for feed number 6 and estimated results for the first feed. Their results are obviously in agreement with the present ones but with so few points no firm conclusions can be drawn about more extensive agreement.

A.2.5 Tedder and Rudd, 1976

Tedder and Rudd [39,40] present a more extensive study than any other worker, having looked at six different feed mixtures at at

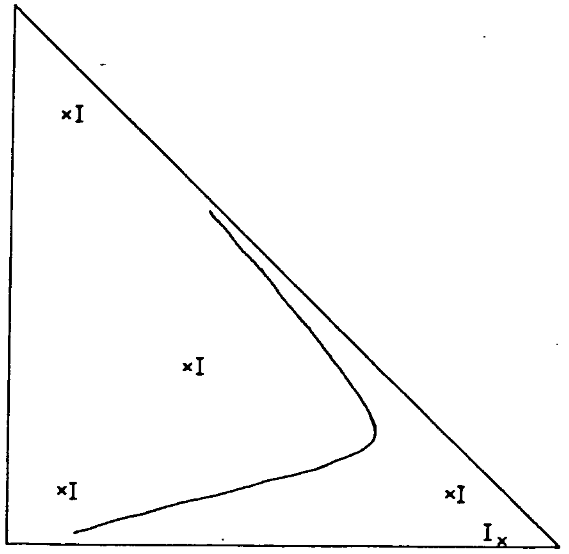
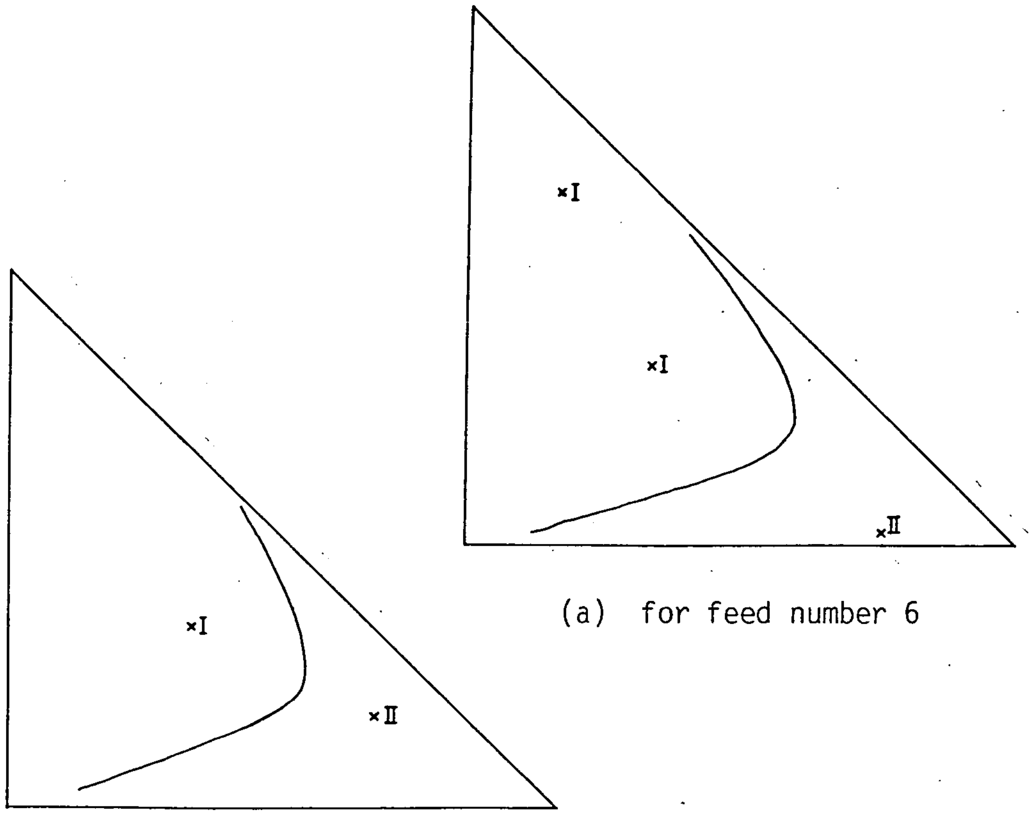


Figure A.14 Freshwater and Henry's results



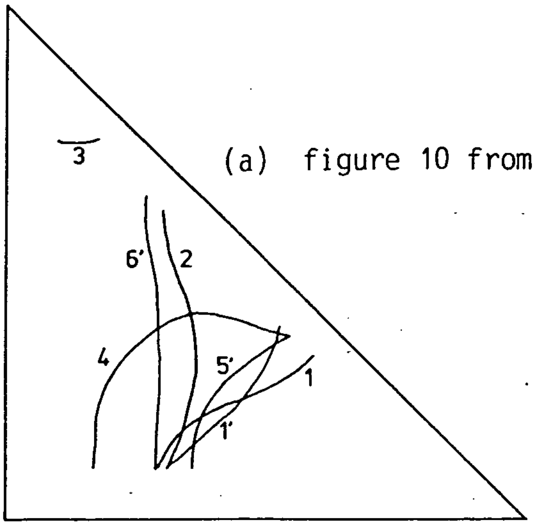
(a) for feed number 6

(b) for n-pentane, n-hexane, n-octane

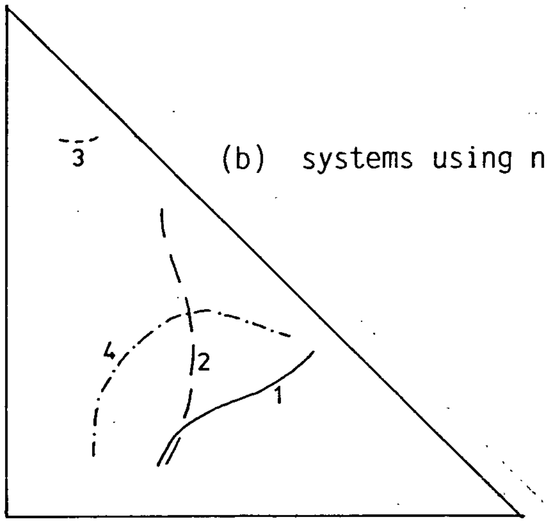
Figure A.15 Bakhshi and Gaddy's results

least seven composition points for each one and comparing eight different equipment configurations, in some cases using two different steam costs. In this case, however, we are only interested in the two simple distillation configurations. Figure A.16 reproduces figure 10 from [39, Part I] and shows the zero contours of cost difference for the two simple configurations for all six of the feeds studied. The results labelled with a prime ('), namely numbers 1', 5' and 6' are all calculated with utility costs at 10 times the values used for the other cases. They suggest that in fact this increase in utility cost has little effect on the position of the zero contour, a conclusion indicative of the large part energy costs play in the total cost of distillation systems, and borne out by the work reported in chapter 5: compare this observation with the similarly small effect produced by increasing the reboiler temperature differences in this study. However in one instance Tedder and Rudd's conclusions are completely at odds with those of this work.

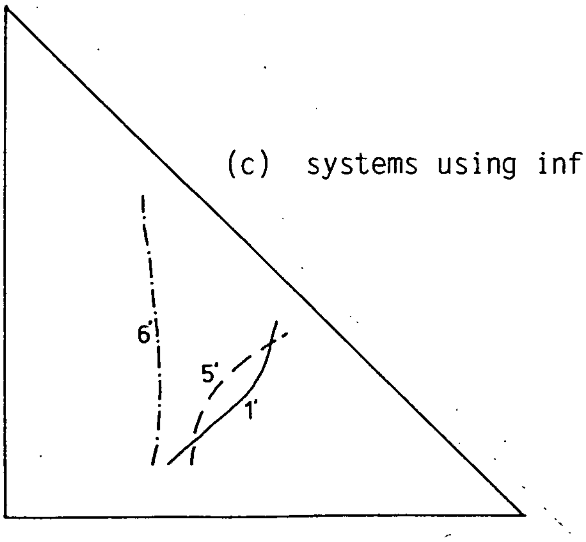
Tedder and Rudd suggest a general trend of zero contour with ESI as shown in figure A.17. This is directly contrary to the trend discovered in this study and shown in figures A.6 and A.7. A possible explanation for this phenomenon is that Tedder and Rudd use discrete steam levels in their study and the discontinuities introduced by this procedure have already been shown to disturb the positions of the zero contours: comparison of figure A.16 with figure A.5 shows a similar lack of order. They suggest that a high ESI will tend to move the zero contour towards the AB edge, while a low ESI will move it towards the BC edge. This study suggests



(a) figure 10 from [39]



(b) systems using normal utilities costs



(c) systems using inflated utilities costs

Figure A.16 Tedder and Rudd's cost results

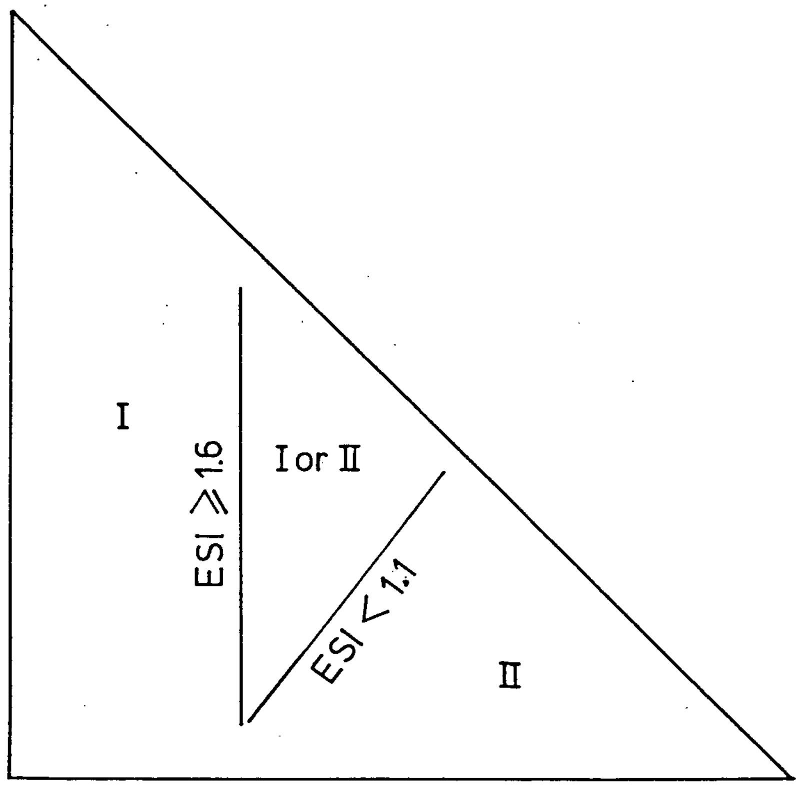


Figure A.17 Tedder and Rudd's suggested trends precisely the opposite trend, which is supported by the following simple reasoning: A high ESI means that the BC split is relatively more difficult than the AB split. This means that the direct sequence will tend to be favoured over the inverted sequence since the column with the more difficult split will be processing less material in the direct sequence. This will tend to make the direct sequence cheaper over more of the triangular diagram. It would seem then that Tedder and Rudd were misled by their assumption that discontinuities in the cost surface caused by discrete steam temperature levels would have little or no effect on the position of the zero contours of cost difference.

A.3 Conclusion

Perhaps due to the increasing availability of computing power the work reported here investigates the cost of ternary distillation systems in greater detail than that of other workers. Their results have not been so conclusive, and in one case appear to have been in error. However for the most part conclusions reported here have been in agreement with those of previous workers, and a reason is suggested for one case of obvious disagreement. The main conclusion is that the zero contour of cost difference moves from the AB edge of the triangular diagram toward the BC edge with increasing feed ESI or MESI. Secondary conclusions are that allowing the use of vacuum operation will tend to favour the direct sequence over more of the ternary diagram; that the method of top product specification and the choice of reboiler temperature difference have little effect on the zero contour; but that the use of discrete steam temperature levels has an unpredictable effect on the zero contour position.

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