**Marginal Price: A Unifying Concept for Insights into
Distillation Column Sequence Heuristics**
Ajay Modi, Arthur Westerberg
EDRC 06-113-92

Marginal Price: A Unifying Concept for Insights into Distillation Column Sequence Heuristics

Ajay K. Modi and Arthur W. Westerberg[1]

Department of Chemical Engineering
and
Engineering Design Research Center
Carnegie Mellon University, Pittsburgh, PA 15213

Abstract The marginal price, the change in price of a separations task as a result of performing it in the absence of non-key components, is introduced. The marginal price can be computed using any metric that reflects process economics; vapour flowrate and total annualised cost are typical examples. The utility of the marginal price concept in quantifying and explaining a number of heuristics used for distillation column sequencing is described. As an evaluation function, marginal price is closely related to the cost function employed in the predictor-based ordered search procedure of Gomez and Seader. Its performance in controlling the search in distillation-sequence design problems is compared to that of other commonly employed heuristics.

1. Introduction

Of the many decisions that **need to be made** in designing **a** separations **system,** those affecting the cost of **the artifact are among the most crucial. Since the mass flow pattern plays a** major role in determining the cost of a chemical process, decisions that influence it are important. Of these, the set **of tasks selected to apply to the process streams** is **significant since different** task sets give rise **to sequences** with **different material flows. However, because of the** explosive size of the solution **space, even when the separators** involved **are** simple **and** sharp-splitting, the creation of a **separations sequence is rendered difficult. Consequently, considerable incentive** lies in discovering useful insights and evaluation functions that can provide the basis for methods that yield solutions **with minimal** effort.

We begin by describing the problem and previous work in the domain. Next, a heuristic search method that **employs** a novel evaluation function is introduced for solving the problem. The evaluation function is described in depth and the performance of the method is illustrated with data from test problems ranging in size from four to eight components. Finally, the method presented here is compared to others reported in the literature and their relative strengths and weaknesses contrasted.

---

[1]To whom correspondence concerning this paper should be addressed.

2. Previous Work in the Domain

The problem considered here is the creation of a sequence of simple sharp-splitting distillation columns for the separation of a feed mixture into its constituent components. Several procedures have been proposed by previous workers for solving problems in this class.

The earliest work in the domain consisted mainly of analytical studies performed primarily with the intention of understanding what the significant variables were and how they affected the arrangement of the columns. Lockhart [Lockhart 47] related feed composition to type of sequence for input mixtures consisting of 3 components. The results were obtained by studying the column arrangements for different feeds to a natural gasoline plant. Harbert [Harbert 57] advocated the use of two heuristics in determining the separations sequence: perform the most difficult separation last and favour separations that result in equimolal (50/50) splits. Rod and Marek [Rod & Marek 59] suggested selecting the sequence that minimises the vapour load on the system. They presented analytical expressions for computing the vapour flow. The studies performed by Heaven [Heaven 69] lend support to the heuristics suggested by Harbert in creating separations sequences with minimum cost. On the basis of the investigations of Heaven, King [King 71] proposed four heuristic rules for the sequencing of multicomponent systems. Nishimura and Hiraizumi [Nishimura & Hiraizumi 71] presented three column-arrangement rules for determining the sequence in two situations, either when all the components in the feed mixture are approximately equal in amount, or when when one of the components dominates in quantity.

Powers [Powers 72] presented a heuristic method for solving the problem. For each stream in the process, a heuristic evaluation function is used to select the separation task that should be applied to it. The evaluation function is a composite index of the support provided to a separation task by the four heuristic rules proposed by King. Of all the competing tasks, the one with the maximum function value is selected to apply. This method is used by the AIDES program [Siirola & Rudd 71] for creating a distillation sequence.

Hendry and Hughes [Hendry & Hughes 72] described a dynamic programming approach to determine the arrangement of separation units.

Thompson and King [Thompson & King 72] presented a computer program that uses a "cheapest first" heuristic to select the separation task to apply to a process stream.

Freshwater and Henry [Freshwater & Henry 75] presented the results of a detailed study relating the cost of a system, its configuration, its energy requirements and its recovery to the composition of the feed. The results were obtained by analysing the separation of three, four and five-component hydrocarbon mixtures into relatively pure products.

Westerberg and Stephanopoulos [Westerberg & Stephanopoulos 75] presented a branch and bound method for searching the space of sequences. The procedure first creates a basic flowsheet and computes it primal and dual bounds. The basic flowsheet can be any separation sequence

that produces the desired products. The authors have advocated the use of Thompson and King's "cheapest first[11] heuristic in creating the basic flowsheet. Once the basic flowsheet and its dual and primal bounds are established, further flowheets are then generated and any with a dual bound that exceeds the primal bound of the basic flowsheet are screened out. The result of this procedure is a number of flowsheets, each with a dual bound less than the primal bound of the basic flowsheet. From these, a single candidate may be chosen on considerations such as operability, controllability, etc., or the entire procedure may be repeated by conjecturing the flowsheet with the lowest dual bound as the new basic flowsheet. In a later paper [Stephanopoulos & Westerberg 76], the authors proposed an evolutionary method for the creation of a process flowsheet Given an initial flowsheet created by some other method, a set of evolutionary rules is applied to the flowsheet until no further improvements result

Rodrigo and Seader [Rodrigo & Seader 75] presented a depth-first branch and bound search in which the "cheapest first[11] heuristic is used to select the task to be applied.

Gomez and Seader [Gomez & Seader 76] presented a predictor-based ordered search procedure based on the A^x algorithm ([Hart *et al* 68], [Hart *et al* 72]). This method computes a lower bound on the cost of the remainder of the sequence by relying upon the heuristic that a separation is least expensive when conducted in the absence of non-key components. This paper is discussed extensively later.

Seader and Westerberg [Seader & Westerberg 77] described an evolutionary method for the design of a separations sequence. Six ordered heuristic rules for the creation of the initial sequence and two evolutionary rules to further improve upon it are presented

Tedder and Rudd ([Tedder & Rudd (a) 78], [Tedder & Rudd (b) 78]) studied the separation of three-component mixtures into pure-component products using different column configurations. They identified heuristics based on feed composition and ease of separation for selecting a configuration. Their results lend support to the claim that the design of a separator is not strongly influenced by its location in the sequence.

Nath and Motard [Nath & Motard 81] described an evolutionary method that extends the earlier work of Seader and Westerberg. Eight rules for the creation of the intial sequence and five rules to evolve it further are presented. In addition, the Coefficient of Difficulty of Separation (CDS), an intuitively formulated quantitative measure of the ease of performing a separations task, is also presented.

Tedder [Tedder 81] and Mindennan and Tedder [Minderman & Tedder 82] exhaustively generated all the constituent simple and complex sharp-splitting columns for two example problems, one involving four components and the other involving five. Columns common to both problems were then individually optimised using short-cut models. The optimised columns were then used as the components of a complete sequence for each of the problems. Finally, using the evolutionary method of Umeda [Umeda *et al* 79], the sequences were heat integrated.

From their studies they drew two conclusions: one, that the sizing of the individual columns was more important than the subsequent heat integration; and two, if complex columns were already a part of the distillation sequence, heat integration does not pay.

Nagdir and Liu [Nagdir & Liu 83] presented a heuristic method which employs seven ordered rules for the creation of a separations sequence. The rules are categorised into four groups: method heuristics, design heuristics, species heuristics and composition heuristics.

Muraki and Hayakawa [Muraki & Hayakawa 84] presented an evolutionary method for the creation of distillation sequences to separate a feed mixture into two multicomponent products. The procedure is divided into two stages, which are repeatedly applied until the desired sequence is created. In the first stage, a initial sequence is created that separates the feed mixture into pure components. In the second stage, division and blending tasks are introduced into the sequence to improve it further. The Material Allocation Diagram (MAD) is introduced as a means of representing and facilitating the computations required for the second stage.

Floudas [Floudas 87] and Wehe and Westerberg [Wehe & Westerberg 87] described methods that require the use of optimisation algorithms. In the method by Floudas, a superstructure is first created in which are embedded all possible configurations of separation tasks. The superstructure is then represented as a nonlinear programming problem which is solved to extract the desired sequence. In the method by Wehe and Westerberg, alternative column configurations are first represented as linear programs that are solved to obtain lower bounds on their costs. The sequence with the best lower bound is then formulated as a nonlinear program which is solved to obtain the upper bound on the cost; any configuration whose lower bound is higher than this value is discarded. The bounds of the sequences that are retained are then gradually tightened by solving ever more complex mathematical programming problems. Any sequence whose bounds are within 1% of each other is deemed a solution.

Lien [Lien 88] presented a heuristic method to create distillation sequences. The method employs an evaluation function termed the energy-index to determine if a split should be performed in the presence or absence of non-key components.

Beltramini and Motard [Beltramini & Motard 88] described KNOD, a system that employs a heuristic method to create a distillation sequence. The heuristic knowledge, however, is represented within the system as a belief network. Thus, of all the separation tasks that can be applied to a stream, the one with the highest belief value is selected to apply.

3. Problem Description

In the case of simple sharp-splitting separators, two commonly used schemes for representing the space of sequences are trees and networks. While the former permit more accurate modelling of the separations sequences, the latter allow the sequences to be analysed faster by approximating the quantity of each species in the input to a separation unit to be the same as in the initial process feed or zero. A tree depicting the 5 possible sequences for a 4-component problem is

presented in Figure 3-1. Stream nodes are represented by unfilled boxes while task nodes are represented by filled boxes.

To illustrate how a sequence may be created, consider the problem of separating the stream (ABCDE) into pure components[2]. Since this is the only stream present initially, it will be selected, and the following tasks applicable to it proposed: A/BCDE, AB/CDE, ABC/DE and ABCD/E. If the task ABC/DE is chosen as a consequence of the analysis, its application to the feed will result in the streams (ABC) and (DE). If the former is selected to be separated next, the tasks A/BC and AB/C will be proposed. If task AB/C is now chosen to apply, the streams (AB) and (C) will be generated, and together with the stream (DE), will form the set of competing streams. If the stream (AB) is next selected, the task A/B will be proposed. Being the only option, it will be selected to apply, resulting in the streams (A) and (B). Finally, the task D/E will be proposed to apply to the single remaining stream, resulting in the pure-component streams (D) and (E). Since no more streams remain to be separated, the set of tasks {ABC/DE, AB/C, A/B, D/E} represents an initial solution to the problem. At this point the procedure may or may not terminate depending upon whether the sequence created is deemed satisfactory or not. The traversal of the search tree is depicted in Figure 3-2. The numbers next to the task nodes indicate the order in which the tasks were proposed.

Thompson and King [Thompson & King 72] have shown that the number of possible sequences for separating a N-component feed stream into single-component products using only a single separations technology is given by the following closed-form equation:

$$N_s = \frac{[2(N-1)]!}{N!(N-1)!}$$

(1)

For 10 components, the number of different sequences possible is 4862. For 15 components, it increases to 2,674,440. Thus, it is seen that the number of possible column sequences rises rapidly as the number of components increases, even when the separators involved are simple and sharp-splitting.

Because of the large combinatorial problem resulting when even a modest number of components is involved, the use of heuristic rules and evaluation functions is the only viable approach to making the search tractable. Besides being useful in its own right, an effective heuristic method can provide a good initial sequence for other methods that require one in creating a good-quality final solution. The performance of the evolutionary methods by Stephanopoulos and Westerberg [Stephanopoulos & Westerberg 76], Seader and Westerberg [Seader & Westerberg 77], Nath and Motard [Nath & Motard 81] and Muraki and Hay akawa [Muraki & Hay akawa 84], the branch and bound method by Westerberg and

---

[2]A process stream is denoted as a ranked list within parentheses and a separations task as a ranked list with a slash indicating the separation point Thus (PQR) is a process stream with 3 components and XY/Z is a task that separates components X and Y from component Z.
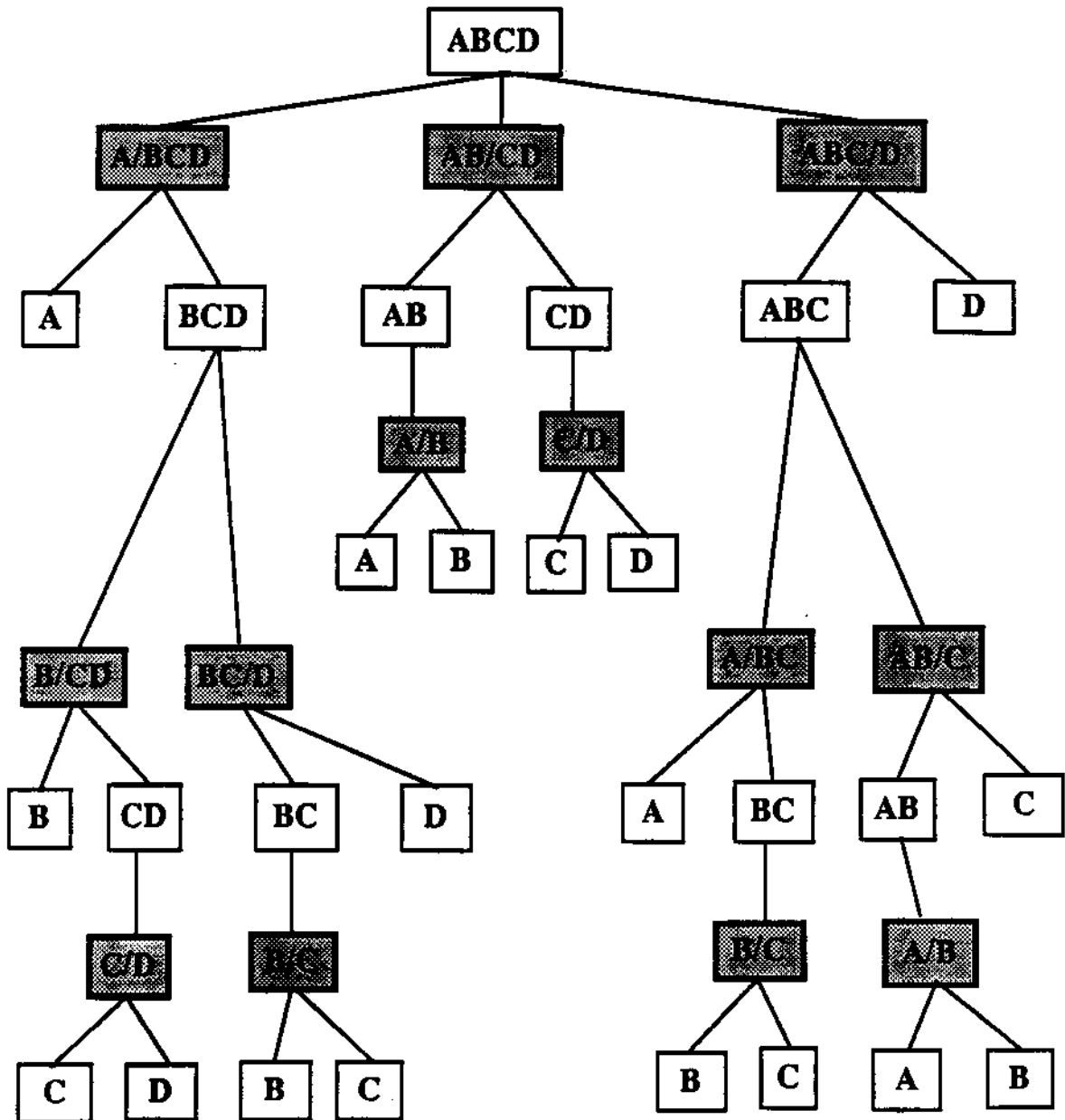
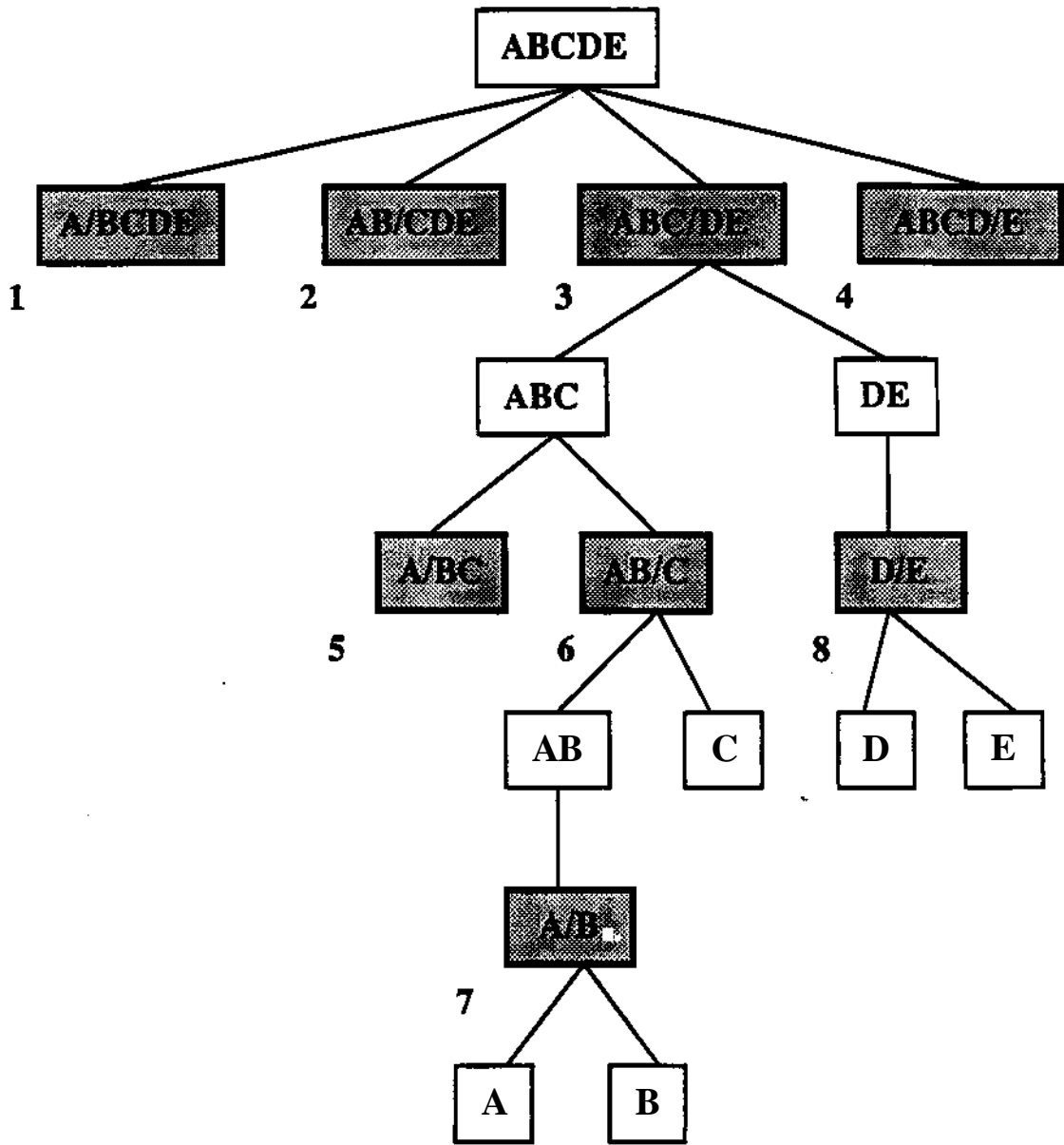Figure 3-1: Search tree for a 4-component problem

Figure 3-2: Example tree traversal for a 5-component problem

Stephanopoulos [Westerberg & Stephanopoulos 75] and the methods by Floudas [Floudas 87] and Wehe and Westerberg [Wehe & Westerberg 87] that employ optimisation algorithms, are all dependent upon good initial solutions or starting points being available.

At the level of representation depicted in Figure 3-1, a key decision encountered repetitively in the creation of a separations sequence is the selection of a task to apply to a process stream. Thus, the evaluation performed in making this choice plays a crucial role in determining both the quality of the solution and the computational overhead required for its creation.

The selection of an evaluation function to decide among competing tasks involves a trade-off between the computational resources used and the solution quality obtained. The greater the effort expended, the higher the solution quality is expected to be. Although true in general, this statement needs to be qualified. Exhaustive search of a space will find the best solution; however, a search that employs information that bounds its options could also result in the same solution, but with lower resource usage. Hence, the claim that greater effort results in better solutions only holds in the absence of search-control knowledge.

When searching the space of distillation sequences, selecting which task to apply to a stream ideally requires evaluating the entire sequence of which the task is a part. Since this demands an exhaustive search, either explicit or implicit, of all the possible sequences, it is not a viable option in most cases. The dynamic-programming based method reported by Hendry and Hughes [Hendry & Hughes 72] carries out an implicit exhaustive search. On the other hand, the decision about which task to select can also be made without conducting a search; in other words, by using local knowledge only. This information is usually available in the form of heuristic rules. The solution methods reported by Powers [Powers 72], Nagdir and Liu [Nagdir & Liu 83] and Beltramini and Motard [Beltramini & Motard 88] employ heuristic rules to select a task as does the evolutionary method presented by Seader and Westerberg [Seader & Westerberg 77] in its creation of an initial sequence.

Although the use of heuristic rules is computationally cheaper than conducting a search, the quality of the decision made and that of the final solution obtained, may not be acceptable. It should be noted however, that the two kinds of evaluation just described represent extreme positions along a computational-overhead/solution-quality trade-off axis. Between these points that involve either exhaustive search or no search, a whole spectrum of alternative evaluation functions exists that require a partial search. The Thompson and King [Thompson & King 72] method conducts a one-step lookahead search and selects the cheapest task to apply, the Nath and Motard [Nath & Motard 81] method selects the task with the lowest CDS and the method presented by Lien [Lien 88] picks the task with the lowest energy index.

Table 3-1 lists the main heuristic rules and evaluation functions that have been proposed to date for the design of distillation sequences.

Heuristic	Reference
Perform the easiest separation first (Perform the most difficult separation last)	[Harbert 57] [Rudder *al* 73]
Remove the most plentiful component first	[Nishimura & Hiraizumi 71] [King 71] [Rudder *al* 73]
Favour the 50/50 split	[Harbert 57] [Heaven 69] [King 71]
Perform the cheapest separation first	[Harbert 57] [Rudder *al* 73]
Perform the separation with smallest CDS first	[Nath&Motard81]
Perform the separation with smallest CES (Coefficient of Ease of Separation) first	[Liu 87]
Perform the separation with smallest energy-index first	[Lien 88]
Favour the direct sequence	[King 71]
Favour a separation in the absence of non-key components	[Heaven 69] [King 71] [Gomez & Seader 76]

Table 3-1: Main heuristic rules and evaluation functions proposed for the design of distillation sequences

4. Marginal Price Method

From the discussion in the previous section, it is seen that a key facet in devising a search method for creating a separations sequence involves discovering a task-selection evaluation function that trades off computational cost and solution quality in an effective way. A good function will consume few computational resources, yet compromise little on solution quality. One evaluation function that meets these criteria is the marginal price of a task. This is defined as the price of performing the task less the price of performing the same separation without any

non-key components present, i.e., its binary analogue or split[3]. It thus follows that the marginal price of a binary task is zero.

The marginal price of a task measures the effects of the presence of non-key components on the resources required to perform it Consequently, the smaller the marginal price of a task, the smaller the effect additional components have on resource usage. Thus, the marginal price of the task A/BC is the effect on the price of the separation A/B of the component C in the mixture (ABC). Similarly, the marginal price of the task DEF/GH is the effect on the price of the separation F/G of the components D, E and H in the mixture (DEFGH).

As noted earlier, all the distillation sequences in the solution space apply the same set of splits to the feed mixture. The difference among the alternatives, however, lies in the order in which they apply the splits. Choosing the task to apply to a stream thus implies deciding whether to apply a split at the current moment or to delay its application to later in the processing. In other words, it involves deciding whether to perform a separation in the presence or absence of non-key components. In this regard, the concept of marginal price is a powerful device since it provides a quantitative basis for deciding whether to postpone a task or not. For a task with a large marginal price, delaying the application of its corresponding split will be beneficial since the absence of additional components will result in a big decrease in resource consumption. On the other hand, for a task with a small marginal price, delaying the application of its corresponding split will not result in large gains since the presence of additional components does not significantly increase resource use. The use of marginal price as an evaluation function for choosing which task to apply next thus involves computing the marginal price of each of the competing tasks and selecting the one with the smallest

The concept of marginal price can be employed as an evaluation function irrespective of whether the effects of the non-key components are positively monotonic, i.e., as their number increases, resource consumption also increases; or negatively monotonic, as their number increases, resource consumption decreases. The marginal price evaluation function is closely related to the commonly employed heuristic that a separation is least expensive when conducted in the absence of non-key components ([King 71], [Gomez & Seader 76]).

The marginal price of a task can be computed using any property that reflects resource consumption. A commonly used metric is the total annualised cost *(TAC)* of a task and the marginal price based on it, the marginal total annualised cost (AfTAC), is thus the effect of the non-key components on the *TAC* of performing the separation without any non-keys present.

In many instances, however, the vapour flow through the system is also a good indicator of

---

[3]It should be noted that the same convention has been used to represent both splits and tasks. A split, defines a separation in terms of the key components only, whereas a task defines it in terms of all the components. Consequently, multiple tasks can implement the same split. For example, the tasks ABC/D, BC/D, C/D and C/DE all implement the split C/D.

process economics since it directly impacts both the capital and operating costs of the equipment. Lower vapour flows result in smaller columns and possibly lower utility usage. Hence, vapour rate has also **been** proposed as an alternative evaluation metric [Douglas 88]. Analogous to *MTACy* the marginal vapour rate (AfV) of a task is the change in the vapour rate of the separation as a consequence of the non-key components. Hence, like *MTAC, MV* can also be used as an evaluation function for selecting among competing tasks.

Similar to deciding what evaluation function to employ, selecting the metric used to rate alternative choices also strongly influences both the quality and character of the solution and the computational overhead required for its generation. This can be illustrated using the two evaluation metrics discussed in this section: total annualised cost and vapour flowrate. Design alternatives evaluated using total annualised cost will reflect actual process economics more closely than those evaluated using vapour flowrates. However, total annualised costs are more expensive to compute than vapour flowrates since computing the former first necessitates computing the latter. Hence, selecting an evaluation metric, like choosing an evaluation function, also involves a trade-off between solution quality and computational overhead.

Table 4-1 outlines the steps in the marginal price method. In presenting the method, the following nomenclature is employed: {P} is the set of desired product streams, {T} is the set of tasks to be applied to create the set of desired products, {U} is the set of unprocessed streams, *v* is the stream currently selected to be separated and x is the task chosen to apply to 0).

1. **Identify the stream to be separated and rank the components** in order of **decreasing volatility.**

2. **Identify the desired product streams and place them in the** set {P}. Initialise {U} **and** {T} as **empty** sets.

3. Place **the feed stream in** {U}.

4. Is {U} **identical to** {P}? **If** yes, go **to** step (7). **If no,** select **a** non-product stream from {U} **and label it D.**

5. **Generate each task applicable** to *x>* **and** estimate its marginal price. Label the task **with the smallest marginal price x. Remove** *v>* **from** {U}.

6. **Apply task** x **to** stream *x>* **and** place the streams generated in {U}. Place x in {T} **and go to** step (4).

7. The set {T} contains the tasks to be applied **to** the feed stream and hence defines **the** separations sequence.

Table 4-1: Marginal price method

Since the marginal price of a task is an integral part of the search method introduced, the scheme

used to compute it has a large effect on both the quality of the solution and the effort expended in creating it The following section describes two methods for computing the marginal price of a task. The first **can** be used to estimate the *MV* of a task while the second can be employed to estimate its *MTAC.*

Many models have been presented in the literature for computing the vapour rate in a distillation column ([Perry & Chilton 73], [Douglas 88]). These include analytical expressions, short-cut models such as Underwood's and rigorous stage-by-stage models. Analytical expressions are faster to solve than short-cut models, which in turn are quicker than stage-by-stage models. On the other hand, the accuracy of the models in predicting column behaviour decreases from stage-by-stage models to short-cut models to analytical expressions. Similarly, many models and empirical correlations, varying in accuracy and solution speed, have been reported in the literature for calculating the cost of a column ([Peters & Timmerhaus 68], [Guthrie 69], [Perry & Chilton 73], [Douglas 88]).

In the schemes described for computing the marginal price of a task, the selection of the particular vapour rate and cost models used has been made for illustrative purposes only. The proper choice of an appropriate model, like the selection of an evaluation function, involves a trade-off between computational overhead and solution quality. Unlike many methods reported in the literature for designing distillation sequences, the marginal price method does not impose restrictions on the models used. This is important since model-selection decisions are strongly dependent upon the context in which the design is to be performed.

5. Marginal Price as a Unifying Concept

This section describes how marginal price can be used to unify many earlier concepts in the selection of distillation column sequences. However, the section^ begins by showing that marginal price is an alternative formulation of the evaluation function employed by the search method proposed by Gomez and Seader [Gomez & Seader 76].

5.1. Gomez and Seader Evaluation Function

In their predictor-based ordered search procedure for the sequencing of distillation columns, Gomez and Seader proposed that a competing separations task, $\dot{y}$, be evaluated by the following function:

$$F(J) = TAC(j) + TAC^{X}(J) \tag{2}$$

where. *TAC(J)* is the· cost of performing the task and *TAC^x(f)* is the estimated cost of the partial sequence from task *j* to the end. Gomez and Seader have. proposed that the latter quantity be approximated as the sum of the costs of the remaining separations performed in the absence of their non-key components.

To illustrate the Gomez and Seader function, consider the mixture (ABCD) to which the tasks

A/BCD, **AB/CD and ABC/D** are applicable. Their evaluations are respectively given by:

$$G(A/BCD) = TAC(A/BCD) + TAC(B/C) + TAC(C/D)$$

$$G(AB/CD) = TAC(AB/CD) + TAC(A/B) + TAC(C/D)$$

$$G(ABC/D) = TAC(ABC/D) + TAC(A/B) + TAC(B/C)$$

If the quantity $TAC(A/B) + TAC(B/C) + TAC(C/D)$ is subtracted from the RHS of each of the above equations, the following equations are respectively obtained:

$$T(A/BCD) = TAC(A/BCD) - TAC(A/B)$$

$$T(AB/CD) = TAC(AB/CD) - TAC(B/C)$$

$$U(ABC/D) = TAC(ABC/D) - TAC(C/D)$$

It should immediately be recognised that $t(j)$ is the marginal price of task j.

Hence, it is seen that if the search is controlled using the Gomez and Seader evaluation function, the same sequence will be generated as the method presented in Table 4-1.

5.2. Estimating and Using the Marginal Vapour Rate

The *MV* of a task may be approximated by a simple analytical expression that quantifies two commonly employed heuristics, "perform easiest task next" and "remove most plentiful species next."[1] The expression, which relates the marginal vapour rate to the flowrates and relative volatilities of the components in the feed mixture, will also allow the heuristic that suggests using the direct sequence when all else is equal to be justified.

The flowrates of the distillate and feed streams, *D* and *F* respectively, for a distillation column whose performance is characterised by constant relative volatilities, are given by Underwood's equations([Underwood 46], [Underwood 48]):

$$\sum_{i=l}^{h} \frac{\alpha_i d_i}{\alpha_i - \phi} = D(R_{min} + 1) \tag{3}$$

$$\sum_{i=l}^{h} \frac{\alpha_i f_i}{\alpha_i - \phi} = F(1 - q) \tag{4}$$

where l is the lightest component in the feed, h is the heaviest component, α_i is the relative volatility of component i, d_i is its flowrate in the distillate, f_i is its flowrate in the feed, R_{min} is the minimum reflux ratio of the column, ϕ is the root of the equations and q is the thermal condition of the feed, which is equal to one for a saturated liquid.

The vapour rate of a column, V, is related to its reflux ratio, R, by:

$$V = D(R + 1) \tag{5}$$

Similarly, the minimum vapour rate of a column is defined by:

$$V \ll * - * > < * \bullet * + D \tag{6}$$

The reflux ratio of a column is given by:

$$* - K^{TM} \qquad \bullet \tag{7}$$

where P is a parameter that usually lies between 1.03 and 1.30.

Substituting equations 7 and 6 into equation 5 and rearranging gives:

$$V = \beta V_{min} - (\beta - 1)D \tag{8}$$

A column operating at high recovery may be modelled satisfactorily as a perfect splitter. In this case, the compositions of the distillate and bottoms are respectively given by:

$$\mathcal{A}_i = \begin{cases} r \, f_i & \text{if } l \leq l \leq lk \\ 0 & \text{otherwise} \end{cases} \tag{9}$$

$$b_i = \begin{cases} I \, f_i & \text{if } hk \leq i \leq h \\ o & \text{otherwise} \end{cases} \tag{10}$$

where lk is the light key, hk is the heavy key and b_i is the composition of component i in the bottoms.

Substituting equations 9 6 into equation 3 gives:

$$V^{TM} = \sum_{i=l} \frac{\wedge}{\alpha_i - \phi} 4 \tag{11}$$

Substituting equation 11 into equation 8 gives:

$$V = \beta \sum_{i=l}^{lk} \frac{\alpha_i f_i}{\alpha_i - \phi} - (\beta - 1) \sum_{i=l}^{lk} f_i \tag{12}$$

since $D = S \, l_i // i$ - This equation relates the vapour rate of a task to volatilities of the species in the distillate stream.

A component material balance over the whole column gives:

$$f_i \wedge d_i + b_i \qquad V / = /,.., h \tag{13}$$

Substituting equations 13, 10 and 9 into equation 3 gives:

$$y \quad \wedge \; + \; T \quad \underline{\quad} = F(1\text{-}q) \tag{14}$$

Substituting equation 11 into equation 14 and rearranging the result gives:

$$V_{min} = F(1-q) - \sum_{i=hk}^{h} \frac{\alpha_i f_i}{\alpha_i - \phi} \tag{15}$$

Substituting equation 15 into equation 8 gives:

$$\pounds \underset{i=1}{\quad} \underset{t=}{\quad} \quad - (\beta - 1)(F - B) \tag{16}$$

since $D = F - 5 . F = Jj.J.$ andfi $= I?_{sAJk} f_i.$

Equation 16 may be rewritten as:

$$V = (1 - \beta q)\sum_{i=1}^{h} f_i + (\beta - 1)\sum_{i=hk}^{h} f_i - \beta \sum_{i=hk}^{h} \frac{\alpha_i f_i}{\alpha_i - \phi} \tag{17}$$

As opposed to equation 12, this equation **relates the vapour rate** of a task to the condition of the bottoms stream.

The marginal vapour rate of the task l..lk/hk..h is given by:

$$MV\{L.lk/hk..h) \; - \; V(l..lk/hk..h)\text{-}V(lk/hk) \tag{18}$$

where $V(l..lk/hk..h)$ is the vapour rate of the task Llk/hk..h and $V(lk/hk)$ is the vapour rate of the separation Ik/hk.

If the vapour rates are computed using equation 12, the marginal vapour rate of a task is given by:

$$MV(l..lk/hk) = \beta \sum_{i=1}^{lk} \frac{\alpha_i f_i}{\alpha_i - \phi_{l..lk/hk..h}} - (\beta - 1)\sum_{i=1}^{lk} f_i \tag{19}$$
$$- \beta \frac{\alpha_{lk} f_{lk}}{\alpha_{lk} - \phi_{lk/hk}} + (\beta - 1) f_{lk}$$

However, if the vapour rates are computed using equation 17, the MV of a task is given by:

$$MV(lk/hk..h) = <1\text{-}p^*)\pounds/\} \ + \ <p\text{-}1)\pounds/, \qquad (20)$$
$${}_{i=l} \qquad\qquad {}_{i=hk}$$

$$\_py \frac{h}{iftk \ \ ^{a},\text{-}\!\!\!\!\not\!\!\!>/..tt/A^*..A} \ 1 \pm \frac{af\text{-}}{}$$

$$- (1-\beta q)\sum_{i=lk} f_i \ - \ (\beta-1)f_{hk} \ + \ \beta \ \frac{\alpha_{hk}f_{hk}}{\alpha_{hk}-\text{\^c}_{Ylktkk}}$$

Since both of the roots $\S_{lk/hk}$ and $<!>/../^*/\ll,.^*$ lie between ct_{lk} and $cc^\wedge$ it can be assumed they are approximately equal. Hence, equations 19 and 20 may be written as:

$$MV\{L.lklhk) = \underset{i=l}{p\pounds} \ \underset{\alpha_i-}{-2\pounds}. \ - \ (p\text{-}1)\overset{lk}{\underset{i=l}{Y/;}}\text{-} \qquad (21)$$

$$-\beta \ \frac{\alpha_{lk}f_{lk}}{\alpha_{lk}-\overline\phi} \ + \ (\beta-1)f_{lk}$$

$$MV(lklhk..h) = (1\text{-}p<7)\overset{h}{\%/;} \ + \ (P\text{-}1)\overset{h}{\underset{i=hk}{X/,}} \ - \ \overset{h}{\underset{i=hk}{PX}} \ \text{-}i\overset{\alpha \ f.}{1}i\text{-} \qquad (22)$$

$$-(1-\beta q)\sum_{i=lk}^{hk} f_i \ - \ (\beta-1)f_{hk} \ + \ P \ \frac{\alpha_{hk}f_{hk}}{\alpha_{hk}.-\overline\phi}$$

where $\underset{lk}{a} \ \leq \ \overline\phi \ \leq \ \alpha_{hk}$.

Finally, equations 21 and 22 can respectively be simplified to:

$$MV\{l..lklhk) = \underset{i=l}{p^{\prime*}2} \ \underset{\alpha_i-\overline\phi}{\overset{-1}{-\ \wedge}} \ - \ - \ (P\text{-}D\overset{l}{\underset{i=l}{\wedge/}}i \qquad (23)$$

$$MV\{lklhk..h) = (1\text{-}P\underset{\nu}{d}Y./J \ + \ p(1\text{-}\wedge)\overset{h}{2}/, \qquad (24)$$

$$\textbf{-PI}^{h} \ \frac{\text{af-}}{}$$
$$\phantom{\textbf{-PI}^{h} \ }{}_{i}$$

It should be **noted** that the former equation relates the *MV* of a task to the condition of the distillate stream while the latter relates it to the condition of the bottoms stream.

The *MV* of a task with non-key components that are both lighter than the light key and heavier than the heavy key is obtained by combining equations 23 and 24:

$$MV(i..ik/hk..h) = \beta \sum_{i=l}^{lk-1} \frac{\alpha_i f_i}{\alpha_i - \bar{\phi}} - (\beta - 1) \sum_{i=l}^{lk-1} f_i + (1 - \beta q) \sum_{i=l}^{lk-1} f_i$$

$$+ pa-?) \, i \, /; - P \sum_{i=hk+1}^{h} \frac{\alpha_i f_i}{\alpha_i - \bar{\phi}} \tag{25}$$

The marginal minimum vapour rate MV_{min} of a task is another variation of marginal vapour rate. Similar to the other marginal prices, it is defined as the V_{min} of performing the task less the V_{min} of performing the same separation without any non-key components present. The MV_{min} of the task l..lk/hk..h can be calculated using the following equation, which is obtained by setting p to one in equation 25:

$$MV_{min}(l..lk/hk..h) = \sum_{i=l}^{lk-1} \frac{\alpha_i f_i}{\alpha_i - \bar{\phi}} + (1 - q) \sum_{i=l}^{lk-1} f_i + \tag{26}$$

$$d<7) \, \overset{h}{X} \, /,- \, \overset{h}{I} \, _{l} \, \frac{\alpha_i f_i}{\alpha_i - \bar{\phi}}$$

For a saturated liquid feed, equation 26 reduces to:

$$MV_{min}(l..lk/hk..h) = \overset{lk-}{J}{}^{1} \, -2i^{\wedge}L \, - \sum_{i=hk+1}^{h} \frac{\alpha_i f_i}{\alpha_i - \bar{\phi}} \tag{27}$$

since $q = 1$.

Figure 5-1 pictorially illustrates the relationship among the a_t and $\bar{\phi}$ for all the sharp-splitting tasks applicable to a mixture ABCD. $\bar{\phi}_{i/j}$ is the value of $\bar{\phi}$ for the split i/j. For example, the marginal effect of component A on the split B/C is given by:

$$MV_{min}(AB/Q = \frac{\alpha_A f_A}{\alpha_A - \bar{\phi}_{B/C}} \tag{28}$$

Letting $\bar{Q}_{B/C} *\bullet 0.5 (a^{\wedge} + a_c)$ gives a very simple evaluation function which incorporates only the ease of separation (relative volatilities) and the quantities of the species. Like the CDS function of Nath and Motard [Nath & Motard 81], it quantifies the decision making for heuristics such as "perform the easiest task next" and "remove the most plentiful species next."

5.2.1. Explaining **the "Direct Sequence" Heuristic**
A commonly applied heuristic rule in the design of distillation sequences states, "All things being equal, choose the direct sequence, i.e., at each decision point, remove the lightest key first." This heuristic can easily be justified using the concept of marginal price. To illustrate this,
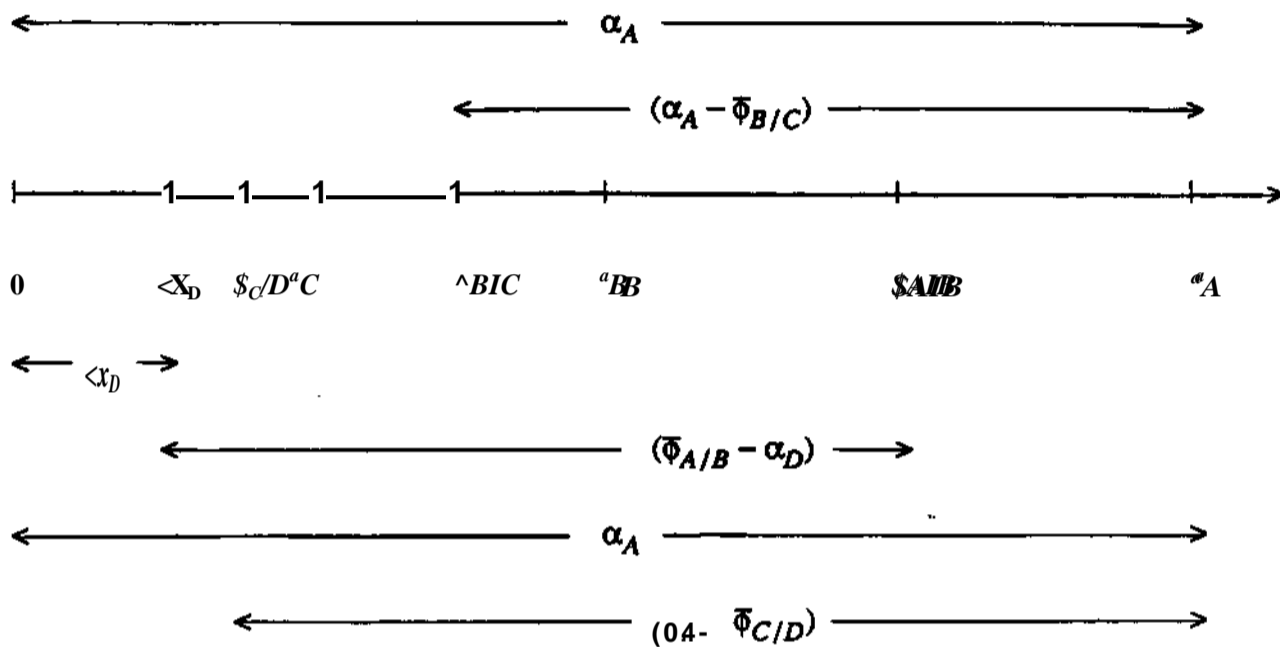
Figure 5-1: Relationship among a,- and $ for a 4-component problem

consider the mixture ABC. The MV_{min} of the task A/BC, whose selection results in the direct sequence, is given by:

$$MV^\wedge A/BQ = \frac{acfc}{\$A/B \,{}^{\cdot\cdot}\,{}^{a}C} \tag{29}$$

Correspondingly, the $MV_{m}j_n$ of the task AB/C, whose selection results in the indirect sequence, is given by equation 28.

Suppose the relative volatility of C, the heaviest component, is equal to some value tt In a situation in which "all things are equal," the ratios of the relative volatilities of adjacent components will be equal. Thus, the relative volatilities of components B and A will respectively be given by $k\overline{a}$ and $k^2\overline{a}$ where k is a constant greater than one. Furthermore, the amounts of each of the components will also be equal, say to some value f. Substituting these values into equations 29 and 28 results in the following equations respectively:

$$MV_{min}(A/BQ = \frac{-\wedge-}{\overline{\phi}_{A/B} - \overline{\alpha}} \tag{30}$$

$$MV_{min}(AB/Q = \frac{*W}{*^2\overline{\phi} - {}_{B/C}} \tag{31}$$

If $\$_{Aj_B} \approx 0.5(a^\wedge + a_B)$ and $\$5/c^a 0,5(a_5 + a_c)$, equations 30 and 31 may respectively be simplified to:

$$MV_{min}(A/BC) = \frac{2}{k^2 + k - 2} \tag{32}$$

$$MV_{min}(AB/Q = {}^\wedge j {}^{\wedge} \tag{33}$$

Using L'Hospital's theorem, it can easily be shown that:

$$\lim_{k \to 1} \frac{\dfrac{2k^2 f}{lk^2 - k - 1}}{\dfrac{2f}{k^2 + k-2}} = 1 \tag{34}$$

Hence, for values of k greater than 1, $MV_{min}(A/BQ$ will always be less than $MV_{min}(AB/C)$, thus leading to the direct sequence. Figure 5-2 depicts how the ratio of $MV_{min}(AB/Q$ to $MV_{min}(A IB O$, represented by r, varies with k.

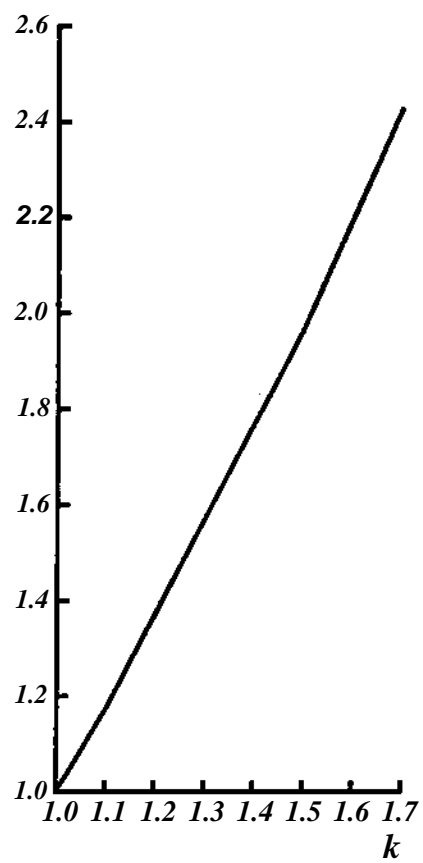The observations used to verify the "all things being equal, choose the direct sequence" heuristic

Figure 5-2: Variation of *r* with *k*

can easily be extended to mixtures involving more than three components. Under the conditions governing the heuristic, the additional price incurred as a consequence of performing a split in the presence of a component heavier than the heavy key is usually much less than the extra price incurred as a result of performing a split in the presence of a component lighter than the light key. Consider the 4-component mixture illustrated in Figure 5-1 once again. The additional price of performing the split A/B (which leads to the direct sequence) in the presence of component D is approximately $\frac{*D/D}{*}$. Correspondingly, the additional price of performing the split C/D

(which leads to the indirect sequence) in the presence of component A is approximately$\frac{o_{A/A}}{}$

Since $./^ = f_D$, it can be seen from Figure 5-1, which is drawn to scale, that:

$$\frac{-22*}{\$A/B\sim^aD} < \frac{2**}{^aA\sim\$C/D} \tag{35}$$

Hence, the split leading to the direct sequence will be preferred over the one leading to the indirect sequence.

5.3. Computing **the Marginal Total Annualised Cost**

The marginal total annualised cost of a task is defined as the *TAC* of performing the task less the *TAC* of performing the same separation in the absence of non-key components. Appendix A lists a set of equations that can be used to compute the *TAC* of a column. The minimum reflux ratio of the column is obtained by solving Underwood's equations. Fenske's expression [Douglas 88] is used to compute the number of stages at total reflux and Gilliland's correlation ([Robinson & Gilliland 50]], [Eduljee 75]) is used to compute the actual number of stages. The heating and cooling requirements are estimated using simple energy balances with constant overall heat-transfer coefficients. Guthrie's correlations ([Guthrie 69], [Guthrie 74]) are used to calculate the cost of the equipment.

Computing the *MTAC* of a task requires that the composition of the input streams to both the task as well as its corresponding binary separation be known. However, the composition of the feed stream to a binary column will only be known if the performance of the columns which precede it in the sequence has already been determined. Doing so, however, requires these columns to first be designed. This poses a dilemma since the purpose of computing the *MTAC* is to allow a decision to be made without having to create the entire sequence. This problem can be solved by assuming that the quantity of each species in the feed to a task is the same as in the initial process feed or zero. As Hendry and Hughes [Hendry & Hughes 72] have argued, this approximation is justified for columns operating at high recoveries since the presence of relatively small quantities.of non-key components has only a .slight effect on the performance of a column. Although this approximation is only required for determining the costs of the binary tasks, its application to the remaining tasks will further reduce the computational effort required in designing a sequence. Nath and MotardfNath & Motard 81] have recommended that a reevaluation of any sequences selected be performed since the build-up of trace species, which

affect product purities, differs from one sequence to the next

6. Efficiency of the Search

To quantify the efficiency of a search method, Rodrigo and Seader [Rodrigo & Seader 75] have introduced the unique subproblem search factor:

$$F_{U*P} - 77 = \frac{N_u}{}$$ (36)

where $N_u{}^\wedge{}_a$ is the number of unique subproblems or tasks analysed and $N^\wedge$ is the number of unique tasks in the problem. $F^\wedge$ is a measure of the computational savings the search method allows over a full graph search. The lower its value, the more efficient the search.

Rathore *et al* [Rathore *et al* 74] have shown that the number of unique tasks in a problem is given by:

$$N_{usp} = \frac{N^3 - N}{6}$$ (37)

where N is the number of components in the feed mixture.

If the marginal price (total annualised cost or vapour rate) of a task is computed by subtracting the price of its corresponding binary separation from its price, the maximum number of unique tasks that will be analysed for a given problem is given by:

$$\wedge = \frac{}{2}$$ (38)

This limit is derived in Appendix B.

However, if the *MV* of a task is computed by linearly combining the individual effects of the additional species, the value $of N_{uspa}$ is given by:

$$N_{uspa} = (N - 1)(N - 2)$$ (39)

This equation[4] is derived in Appendix B.

Table 6-1 displays how N_{iapa9} $N^\wedge$ and F_{usp} vary with N. It can be seen that as the size of the problem increases, the search efficiency also increases. UN_{wva} is computed using equation 38, it can be shown that:

---

[4]It should be noted that $N^\wedge{}_{pa}$ as computed by equation 39 does not correspond directly to the number of tasks analysed. In this case, $N^\wedge{}_{pa}$ is the number of times equation 25 is solved during the search. The actual number of tasks analysed is given by equation 38. If the *MV* of a task is computed by linearly combining the individual effects of the non-key components, $N^{\wedge\wedge}$ computed using equation 39 will more accurately reflect the number of distinct models solved and hence will be a better measure of the search efficiency.

$$\lim \frac{\dfrac{N^2 + iV\text{-}4}{2}}{\dfrac{N^3 - N}{6}} = N \tag{40}$$

Similarly, if N_{mpa} is computed using equation 39, the following limit is derived for $F^\wedge$:

$$\lim_{N \to \infty} \frac{v - iw - ni}{\dfrac{N^3 - N}{6}} \quad \overline{N} \qquad\qquad m$$

It should be noted, however, that since the complexity of the models used in analysing a task is not accounted for, the computational effort expended in solving a model is not factored into the efficiency metric.

N	Nusp	Nuspa eqn. 38	F_{usp} eqn. 38	Nuspa eqn. 39	F_{uso} eqn. 39
3	**4**	**4**	1	3	0.75
4	10	8	0.8	6	0.6
5	20	13	0.65	12	0.6
6	35	19	0.543	20	0.571
7	56	26	0.464	30	0.536
8	84	34	0.405	42	0.5

Table 6-1: Number of tasks analysed, number of unique tasks and the unique subproblem search factor for problems of different sizes

7. Performance of the Marginal Price Method

To evaluate the performance of the marginal price method, a number of test problems ranging in size from four to eight components were solved. The mixtures consisted of normal, branched-chain and cyclic hydrocarbons. Their relevant chemical and physical properties are summarised in Table 7-1. T_b is the normal boiling point of the component, H_{vap} is its heat of vapourisation at the normal boiling point, MW is its molecular weight and $anta_9$ antb and $ante$ are its Antoine vapour-pressure-equation coefficients. Since well-behaved problems were used as a base case for testing, it is expected that the heuristics will perform no better than they do for these problems.

The models and equations used to size and cost the columns are presented in Appendix A. For each problem the split fractions of the light and heavy keys in the distillate and bottoms products respectively were assumed to be 99.9%, the feed, distillate and bottoms streams were assumed to

Species	T_b(K)	Hvap (cal/mol)	MW	anta	antb	ante
dimethyl butene	314.4	6130.0	84.162	15.3755	2326.80	-48.24
hexane	314.9	6896.0	86.178	15.8366	2697.55	-48.78
benzene	353.3	7352.0	78.114	15.9008	2788.51	-52.36
heptane	371.6	7576.0	100.205	15.8737	2911.32	-56.51
octane	398.8	8225.0	114.232	15.9426	3120.29	-63.63
nonane	424.0	8823.0	128.259	15.9671	3291.45	-71.33
decane	447.3	9388.0	142.286	16.0114	3456.80	-78.67
dodecane	489.5	10430.0	170.340	16.1134	3774.56	-91.31

Table 7-1: Relevant properties for the test problem components

be saturated liquids, the column pressures were set to 1 atm, and their reflux ratios to 1.2 times their minimum. The sizing and costing data used is summarized in Appendix A,

Three different marginal prices, the marginal minimum vapour rate (MV_{min}), the marginal vapour rate (MV) and the marginal total annualised cost $(MTAC)$ were evaluated in their capacities as evaluation functions- MV_{min} was computed using equation 27, MV was computed using the vapour rates obtained by solving Underwood's equations and $MTAC$ was computed using the equations presented in Appendix A. The performance of each evaluation function was determined by comparing the solution obtained by using it to control the search to the solution obtained by doing an exhaustive search. This comparison was carried out for all the test problems and the results are summarized in Tables 7-3, 7-4 and 7-5. The first and second columns of each table respectively indicate the problem size (in terms of the number of components in the feed mixture) and the percentage of problems in which the marginal price method obtained the same solution as that obtained by conducting an exhaustive search. The third column indicates the difference in the TAC of the distillation sequence obtained by the marginal price method from the TAC of the sequence obtained via an exhaustive search averaged over all the test problems.

Table 7-2 presents some information about the test problems. For each problem size, the number of problems solved is indicated in the second column. The third column indicates the difference in the TAC of the worst or most expensive sequence in the search space from the TAC of the best or least expensive sequence averaged over all the test problems. The average maximum TAC difference is important in placing the average TAC difference in perspective. It is likely that if the total annualised costs of the sequences embedded in the search space differ by only small

Problem Size(N)	No. Problems Solved	Av.Max.TAC Difference (%)
4	48	49.49
5	48	20.97
6	32	28.99
7	10	37.22
8	5	34.47

Table 7-2: Test problem data

amounts from the *TAC* of the best sequence, the resulting small average *TAC* difference could be a misleading indicator of the performance of the solution method unless the average maximum *TAC* difference is also presented to qualify it.

From Tables 7-3, 7-4 and 7-5, it can be seen that *MV* performs better than MV_{min} as an evaluation function and *MTAC* performs the best of all. This was expected since *MTAC* better accounts for process economics than *MV* which in turn is better than $MV^$. The improvement in performance from MV_{min} to *MV* to *MTAC* is reflected both in an increase in the number of best solutions obtained as well as an increase in the quality of the solutions obtained in those cases where the best solutions were not found. The performance of all three marginal prices decreases as the problem size increases. Again, this was as expected.

8. Comparison of the Marginal Price Method to other Heuristic Methods

The same set of test problems used to evaluate the marginal price method was solved using three other commonly employed heuristic methods. In the first, the cheapest task is always selected to apply next. This is equivalent to performing a one-step lookahead search. In the second, the task with the lowest Coefficient of Difficulty of Separation (CDS) is selected to apply, and in the third, the easiest task, i.e., the one with the lowest relative-volatility ratio between the key components, is selected to apply. In each case the format used in presenting the results is similar to that employed for the marginal price method. The column costs were determined using the equations presented in Appendix A.

Tables 8-1 and 8-2 contain the results for the "cheapest first[11] and "easiest first" heuristics respectively.

Table 8-3 contains the results using the CDS evaluation function, which was defined by Nath and Motard [Nath & Motard 81] as:

Problem Size (N)	% Best Solution	Average TAC Difference (%)
4	31.3	3.52
5	8.3	6.22
6	0	6.19
7	10	3.72
8	20	1.42

Table 7-3: Performance *of MV$_{min}$* as an evaluation function

Problem Size (N)	% Best Solution	Average TAC Difference (%)
4	62.5	1.17
5	72.9	0.25
6	50	.O84
7	10	*Ti*38
8	20	"094

Table 7-4: Performance of *MV as* an evaluation function

Problem Size(N)	%Best Solution	Average TAC Difference (%)
4	95.8	0.04
5	77.1	0.06
6	50	0.53
7	20	2.82
8	40	0.68

Table 7-5: Performance of *MTAC* as an evaluation function

$$\cos - \hat{}\hat{}u + i\hat{}n \qquad (42)$$

where D is the distillate flowrate, B is the bottoms flowrate and N_{min} is the minimum number of stages in the column (See Appendix A.)

From Tables 8-1, 8-2 and 8-3, it can be seen that as evaluation functions, all three marginal prices presented in the previous section outperform the heuristics presented in this section. The improved performance is reflected both in the number solutions found as well as the quality of the solutions found, as measured by their total annualised costs.

Table 8-4 summarizes the performance of all the methods over all 143 test problems. The average maximum TAC difference over all the example problems was 33.95%.

9. Discussion

The concept of marginal price can be employed as an evaluation device in a number of design situations other than those described thus far. Since the evaluation function places no restrictions on the models used to characterise the behaviour of a distillation column, it can also be used for non-ideal mixtures, i.e., mixtures whose component volatilities are temperature, pressure and composition dependent, provided the ordering of the components in the stream does not change in the range of operating conditions considered.

The concept of marginal price can also be used as an evaluation function for design problems in which separations technologies other than distillation are considered. The idea here is exactly the same as for distillation-based systems. For each alternative, the price of performing the separation with and without the non-key components is estimated and the one with the smallest marginal price is selected to apply. Multiple competing technologies for a separation can also be assessed on the basis of marginal price. Instead of creating a single component ordering for a process stream, multiple orderings, each corresponding to a different separations technology, are generated to determine the competing separation tasks, which are then evaluated on the basis of their marginal prices.

Andrecovich and Westerberg [Andrecovich & Westerberg 85] have described how a distillation column may be represented, as illustrated in Figure 9-1, using a temperature-energy (T-Q) diagram. This representation is based on the view that a distillation column is a device that degrades energy (2nd law) rather than just on the conventional view of a distillation column as a device that consumes energy (1st law). The energy injected into a distillation column, Q_R, vapourises the liquid entering its reboiler at the temperature T_R. This energy is then degraded to the temperature T_c and subsequently expelled from the column through its condenser. If the feed, distillate and bottoms streams are all saturated liquids and the separations task is moderately difficult, it can be shown that the energy removed via the column condenser, Q_c, is approximately equal to Q_R. Thus, a distillation column can roughly be characterised as a device

Problem Size (N)	% Best Solution	Average TAC difference (%)
4	14.6	6.18
5	0	10.85
6	0	13.20
7	0	8.98
8	0	4.96

Table 8-1: Performance of the "cheapest first" heuristic

Problem Size (N)	% Best Solution	Average TAC Difference (%)
4	18.8	6.15
5	2.1	8.86
6	0	13.22
7	0	8.52
8	0	11.63

Table 8-2: Performance of the "easiest first" heuristic

Problem Size (N)	% Best Solution	Average TAC Difference (%)
4	16.7	4.52
5	4.2	9.04
6	6.3	7.64
7	10	3.92
8	0	2.33

Table 8-3: Performance of *CDS* as an evaluation function

| Heuristic	% Problems Best Solution	Average TAC Difference (%)
Cheapest first	4.9	9.47
Easiest first	7.0	9.00
CDS	9.1	6.62
MVmin	14.7	4.96
MV	58.0	0.97
MTAC	72.0	0.37

Table 8-4: Summary of the performance of the heuristic methods

that degrades an amount of heat Q$(= Q_R \ast > Q_c)$ over the interval $AT = T_R\text{-}T_C$.

Since the metric QAT characterises the resource usage of a separations task, a corresponding marginal QAT can be used to decide what task to apply to a mixture. Similar to the other marginal prices, Mg_{Ar}, the marginal QAT of a task, is defined as its QAT value less the QAT value obtained by performing the same separation in the absence of the non-key components. A task with a smaller Afg_{Ar} value would be preferred over a task with a larger M_{Q6T}. A marginal price based on QAT is likely to be a more accurate evaluation function than a marginal price based solely on Q since it implicitly takes into account the fact that heating costs are not only a function of mass load, but also of temperature.

A large amount of the research effort in process design is devoted to discovering and reporting new design methods, and it was in following this tradition that the work leading to the marginal price method was carried out. However, like all other methods that have been reported for solving process design tasks, the marginal price method can only be used in particular contexts. Its primary purpose is to aid in making a specific decision, the order in which to perform the splits on a given feed mixture. Hence, if the design context only calls for this decision to be made, the marginal price method can be used on a stand-alone basis. If the design context also requires additional decisions to be made, it goes without saying that the marginal price method will have to be used in tandem with others. Also, in contexts in which additional resources are available for solving the design problem, the solution created using the marginal price method can be used either for bounding any subsequent search that is performed or as a starting point for an improved solution.

It should also be recognised that the marginal price method is primarily an addition to an existing suite of methods for deciding the order in which to apply the splits to a feed stream. However, the method does have a number of advantages over the others: it is not afflicted by the problem of conflicting heuristics, it does not place restrictions on the models used to compute column
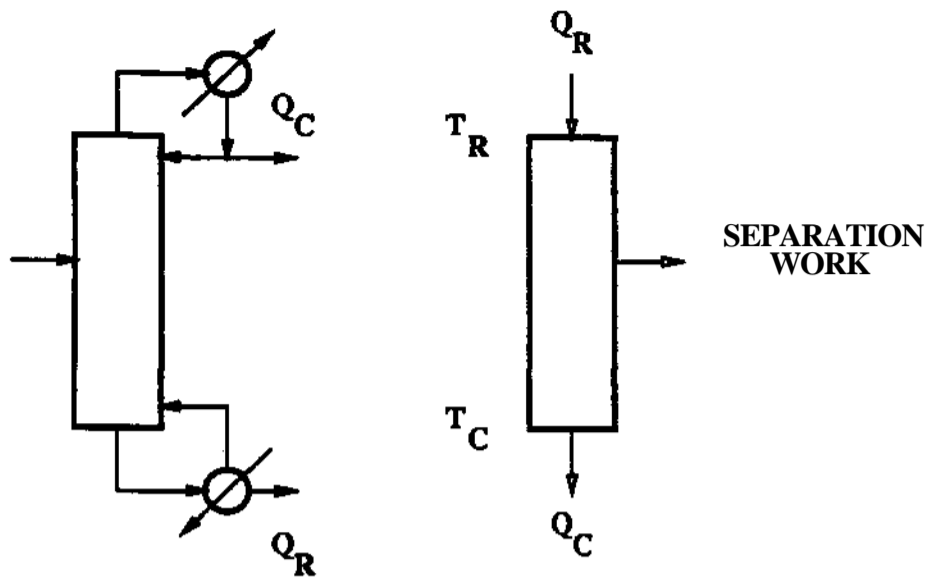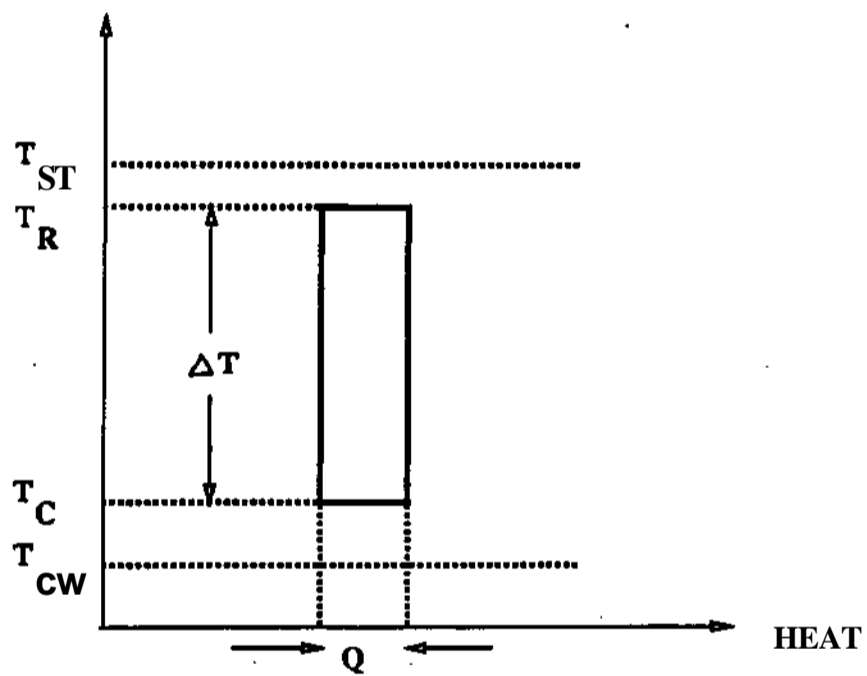
Figure 9-1: T-Q representation of a distillation column

performance, it can be employed to solve large-sized problems within reasonable lengths of time, and it results in high-quality solutions for mixtures whose vapour-liquid equilibrium behaviour is nearly ideal. However, even with these intrinsic strengths, it will be emphasised that the marginal price method is not being advocated as the method to be used under all design circumstances. The choice of a solution method is highly dependent upon the context in which the design problem is to be solved, and until a theory of process design is available, the selection of an appropriate method will require the competing solution methods to be reasoned about.

Almost all software systems developed to date for supporting process design can only reason about a few of the many decisions that must be made in solving a design task. Those decisions that cannot be handled by the support systems are usually hardwired into the systems when they are first created. Furthermore, almost all the decisions that can be reasoned about pertain to the artifact itself and not to the processes undertaken in creating the artifact's description. However, both kinds of decisions are equally important in solving a design problem. Any system that is to be truly useful for aiding in the solution of a separations system design task should be capable of making all decisions pertaining to the task, including those pertaining to the design processes applied, e.g., the decision-making methods to be used. Given the appropriate knowledge, the system should be capable of either executing a design method itself or invoking another tool thai directly implements the selected method. Modi [Modi 91] describes how such a system can be constructed using the Soar architecture.

10. Summary

The utility of the marginal price method was demonstrated for the problem of determining the mass flow pattern of a distillation sequence. Its performance was illustrated with data from test problems ranging in size from four to eight components. A discussion of how the use of marginal price as an evaluation function could be extended to other problems was also presented. These include problems involving mixtures whose components are not characterised by constant relative volatilities, problems involving separations technologies other than distillation and problems in which the columns are to be internally integrated.

The performance of the marginal price method was also demonstrated to be superior over that of a number of other commonly employed heuristic methods. When *MTAC* was used as the evaluation function, the solution obtained was the same as that obtained via an exhaustive search in 72% of the cases.

Besides being useful in its own right, the marginal price method can also be used to generate good initial sequences or starting points for other more exhaustive strategies such as the branch and bound, evolutionary and mathematical-programming based methods that have also been proposed as solution methods. The solutions generated by the marginal price method can also be used as upper bounds that constrain the search in richer solution spaces containing non-distillation based separations units, complex and thermally-coupled columns and heat-integrated sequences, for example.

11. Acknowledgements

Appendix A. Sizing and Costing a Distillation Column

A.I. Models and Correlations

The flowrates of the feed, distillate and bottoms streams, F, D and B respectively, are given by Underwood's equations ([Underwood 46], [Underwood 48]):

$$\sum_{i=i}^{h} \wedge \quad + = D_1(R_{min} \tag{43}$$

$$\underset{.}{X} \; \frac{\alpha_i J_i}{- \; -\!\tilde{\text{m}}} = F(\backslash\text{-}Q) \tag{44}$$

$$\sum_{i=i}^{h} \frac{\alpha_i \, b_i}{\alpha_i - \phi} = -S_{min}B \tag{45}$$

where / is the lightest component in the feed, h is the heaviest component, a_i is the relative volatility of component i, d_i is its flowrate in the distillate, f_i is its flowrate in the feed, b_i is its flowrate in the bottoms, $R^\wedge$ is the minimum reflux ratio of the column, S_{mil} is the minimum reboil ratio of the column, ϕ is the root of the equations and q is the thermal condition of the feed.

The vapour rate of a column, V, is given by:

$$V = D(R+l) \tag{46}$$

where R, the reflux ratio of the column, is given by:

$$* = Wmin \hspace{4cm} '' \tag{47}$$

where (3 is a parameter that usually lies between 1.03 and 1.30.

The reboil ratio of the column, 5, is given by:

$$s = \frac{V - F(1-q)}{B} \tag{48}$$

The minimum number of trays in the column, iV_{min}, is given by Fenske's expression [Douglas 88]:

$$N_{''mm} \bullet = \frac{\ln SF}{\ln \frac{a_{lk}}{a_{hk}}} \tag{49} \quad {}_{v^{*y)}}$$

where SF, the separation factor, is defined as:

$$SF = \frac{d_{lk}}{d_{hk}} \frac{b_{hk}}{b_{lk}} \tag{50}$$

Here *lk* and *hk* **refer to the light** and heavy keys **respectively.**

The actual number of trays in the column, N_m, is given by Gilliland's correlation ([Robinson & Gilliland 50], [Eduljee 75]):

$$v = 0.75(1 - X^{0.5688}) \tag{51}$$

where v and X are respectively given by:

$$v = \frac{N_{TR} - N_{min}}{N_{TR} + 1} \tag{52}$$

$$\lambda = \frac{R - R_{min}}{R + 1} \tag{53}$$

The diameter of the tower, d_T, is given by:

$$d_T = \delta^{0.5} \tag{54}$$

where δ is defined as:

$$\delta = \frac{4 D_{mass}(R + 1)}{\rho_G \pi v_F f_f} \tag{55}$$

where D_{mass} is the mass flowrate of the distillate, ρ_G is the density of the column vapour, v_F is the flooding velocity of the column and f_f is the flooding factor.

The column-vapour density is approximated by:

$$\rho_G = \frac{P W_F}{R_U T_F} \tag{56}$$

where P is the column pressure, W_F is the mole-averaged molecular weight of the feed, R_U is the universal gas constant and T_F is the bubble point of the feed.

The flooding velocity of the column is given by:

$$v_F = C T^{0.5} \tag{57}$$

where *c* is a constant and *y* is defined as:

$$Y = \frac{}{\rho G} \tag{58}$$

ρ_L is the density of the liquid in the column.

The height of the tower, Hj, is given by:

$$H_T = h_{space}(N_{TR} - 1) + h_{extra} \tag{59}$$

where h_{space} is the spacing between the trays and h_{extra} is the extra spacing at the ends of the tower.

The condenser and reboiler duties, Q_c and Q_R respectively, are given by:

$$Q_c = H_D D(R + 1) \tag{60}$$

$$Q_R = H_B SB \tag{61}$$

where H_D and H_B are the mole-averaged heats of vapourisation of the distillate and bottoms streams at their bubble points.

The condenser and reboiler heat-transfer areas, A_c and A_R respectively, are given by:

$$A_C = \frac{Qc}{U_C(T_C - T_{CW})} \tag{62}$$

$$A_R = \frac{Q_R}{U_R(T_{ST} - T_R)} \tag{63}$$

where U_c and U_R are the overall heat-transfer coefficients in the condenser and rebolier respectively, T_c and T_R are bubble-point temperatures of the distillate and bottoms respectively, T_{cw} is the temperature of the cooling water and T_{ST} is the temperature of the steam.

The utility costs of the column, C_{U9} can be estimated from:

$$Q/ = Qc^{\wedge}CW + QR^{\wedge}ST \tag{64}$$

where C_{cw} and C_{ST} are the unit costs of cooling water and steam respectively.

The total cost of the equipment,' $C_{E}Q_9$ is given by:

$$EQ — v^{\wedge}y \ i \smile TR \cdot \smile C \cdot \smile R \tag{W^}$$

where C_r, C_{TR}, C_c and C_{R9} the cost of the tower, trays, condenser and reboiler respectively, are

given by Guthrie's correlations [Guthrie 69].

Finally, the total annualised cost of the column, *TAC*, is given by:

$$TAC = C_v(1\text{-}tax) + \frac{C_{EQ}}{I} \tag{66}$$

where *I* is the investment-payback period and *tax* is the current tax rate.

A.2. Test Problem Data

For each column, key-component recoveries were assumed to be 99.9% and non-key component recoveries to be 100%. The feed, distillate and bottoms streams were assumed to be saturated liquids. The bubble point temperature of a liquid stream was calculated using Raoult's law and the Antoine equation. The former relates the partial vapour pressure of a component to its mole fraction while the latter relates the bubble point of a liquid mixture to its pressure.

Table A-1 summarizes the sizing and costing data used in the test problems.

Parameter	Value
P	1.2
$\mathcal{Q}$	1
p	1 atm
f_f	0.8
d_{space}	24 in
d_{extra}	20 ft
UR	250 btu/hr-ft^2- °F
U_C	250 btu/hr-ft^2- °F
c_{cw}	20 \$/kw-yr
c_{ST}	80 \$/kw-yr
T_{cw}	300 K
TST	500 K
tax	0.48
I	2yr
ρ_L	850 kg/m^3

Table A-1: Sizing and cost data for the test problems

Appendix B. Computing the Number of Unique Subproblem Analysed for the Marginal Price Method

B.I. Deriving Equation 38

If, during the search of a distillation-sequence space, the decision about which task to apply to a process stream is made by analysing each competing task in turn, the maximum number of tasks that will be examined, N_x, is given by:

$$N_x = \sum_{i=1}^{N-1} i \tag{67}$$

where N is the number of components in the feed stream.

If the tasks are evaluated by computing their marginal prices, $N - 1$ binary tasks will also have to be examined. Hence, the maximum number of unique tasks that will be analysed, $N_{USD(P}$ is given by:

$$N_{uspa} = N_x + (N-1) \tag{68}$$

Substituting equation 67 into equation 68 and simplifying gives:

$$N_{uspa} \simeq \frac{N^2 + N - 4}{2} \tag{69}$$

which is equation 38,

B.2. Deriving Equation 39

Table B-1 depicts all the different instances of equation 39 that have to be evaluated in order to determine the marginal effects of all the components (listed as rows) on all the possible separations (listed as columns) that can be applied to a 5-component mixture. Hence, to determine the effect of component B on the split C/D, the expression in the second row and the third column will have to be evaluated. As can be seen, for a 5-component mixture, the space of expressions is a 5 by 4 matrix with the two leading diagonals empty. In this case, the number of elements in each filled triangle is $3 + 2 + 1 = 6$. Since there are two triangles, the total number of expressions is 12.

This example result can easily be generalised. For N components, the number of elements in each triangle, N_{y9} is given by:

$$N_y = \sum_{i=1}^{N-2} i \tag{70}$$

Doubling equation 70 (since there are two filled triangles) and simplifying gives:

	A/B	B/C	C/D	D/E
A		AB/C - B/C	ABC/D - BC/D	ABCD/E - BCD/E
B			B/CD - C/D	BCD/E - CD/E
C	A/BC - A/B			CD/E - D/E
D	A/BCD - A/BC	B/CD - B/C		
E	A/BCDE - A/BCD	B/CDE - B/CD	C/DE - C/D	

Table B-1: Determining the marginal effects of all components on all separations for a 5-component mixture

$$JV_{sep} = (N-2)(N-1)$$

which is equation 39.

(71)

List of Symbols

anta   Antoine equation coefficient

antb   Antoine equation coefficient

ante   Antoine equation coefficient

A_c Condenser area [m^2]

A_R Reboiler area [m^2]

B Bottoms flowrate [kmol/hr]

c Constant in equation 57

C_c Condenser cost [$]

C_{cw} Cooling water cost [$/kw-yr]

$C_E Q$ Equipment cost [$]

C_R Reboiler cost [$]

C_{ST} Steam cost [$/kw-yr]

C_{TR} Cost of trays [$]

CDS   Coefficient of difficulty of separation

d_t Flowrate of component i in distillate [kmol/hr]

d_T Tower diameter [m]

D Distillate flowrate [kmol/hr]

D_{mass} Mass flowrate of the distillate [kg/hr]

f_t Flowrate of component i in feed [kmol/hr]

f_j-   Column flooding factor

F Feed flowrate [kmol/hr]

G Gomez and Seader cost function [$/yr]

U Function identical to *MTAC* [$/yr]

F_{usp} Unique subproblem search factor

h_{space} Column tray spacing [in]

h_{extra}	Extra spa^g at column ends [ft]
H_B	Bottoms stream heat of vapourisation [kcal/kmol]
H_D	Distillate stream heat of vapourisation [kcal/kmol]
H_T	Tower height [m]
H_{vnn}^{vap}	Heat of vapourisation of species [kcal/kmol]
I	Investment-payback period [yr]
$MQ\&I$	Marginal QAT [kw-K]
$MTAC$	Marginal total annualised cost [$/yr]
MV	Marginal vapour rate [kmol/hr]
MV_{min}	Minimum marginal vapour rate [kmol/hr]
MW	Molecular weight of species
TV	Number of components
N_{min}	Minimum number of trays
N_s	Number of sequences
N_{TR}	Number of trays
$^{\wedge}usp$	Number of unique subproblems
N_Uspa	Number of unique subproblems analysed
N_x	Variable defined by equation 67
N_y	Variable defined by equation 70
P	Column pressure [atm]
q	Thermal condition of the feed stream
Q_c	Condenser duty [kw]
Q_R	Rebolier.duty [kw]
QAT	Product of column duty and temperature drop [kw-K]
R	Reflux ratio
R_{min}	Minimum reflux ratio

Rfj	Universal gas constant [m^3 atm/kmol-K]
S_{min}	Minimum reboil ratio
SF	Separation factor
tax	Tax rate
T_b	Normal boiling point of species [K]
T_c	Condenser temperature [K]
T_{cw}	Cooling water temperature [K]
T_F	Feed temperature [K]
T_R	Reboiler temperature [K]
T_{ST}	Steam temperature [K]
TAC	Total annualised cost [\$/yr]
$TAC^x(j)$	Estimated TAC of partial sequence from tasky· to end [\$/yr]
U_c	Overall condenser heat transfer coefficient [Btu/hr-ft^2- °FJ
U_R	Overall reboiler heat transfer coefficient [Btu/hr-ft^2- °FJ
V	Vapour flowrate [kmol/hr]
V_{min}	Minimum vapour flowrate [kmol/hr]
W_F	Molecular weight of feed

Arguments   and   Subscripts

h	Refers to the heaviest component
hk	Refers to the heavy key
l	Refers to a component
j	Refers to a task
l	Refers to the lightest component
Ik	Refers to the light key

GreekLetters

α_i	Relative volatility of component/
β	$* '' *' '' * * * * *_, *$
γ	Variable defined by equation 58
δ	Variable defined by equation 55
ΔT	Difference between reboiler and condenser temperatures [K]
$\lambda,$	Variable defined by equation 53
ν	Variable defined by equation 52
ν_F	Flooding velocity [m/s]
ρ_G	Vapour stream density [kg/m^3]
ρ_L	Liquid stream density [kg/m^3]
ϕ	Root of Underwood's equations

References

[Andrecovich & Westerberg 85]
Andrecovich, M. & A, W. Westerberg.
A simple synthesis method based on utility-bounding for heat-integrated
distillation sequences.
AIChE Journal **31:363,1985.**

[Beltramini & Motard 88]
Beltramini, L. & R. L. Motard.
Knod - A knowledge based approach for process design.
Computers & Chemical Engineering **12(9):939-958,1988.**

[Douglas 88] Douglas, J. M.
Conceptual Design of Chemical Processes.
McGraw-Hill, San Francisco, CA, 1988.

[Eduljee 75] Eduljee, H. E.
Equations replace Gilliland's plot.
Hydrocarbon Processing **54(9): 120,1975.**

[Floudas 87] Floudas, C. A.
Separation synthesis of multicomponent feed streams into multicomponent
product streams.
AIChE Journal 33(4):540-550,1987.

[Freshwater & Henry 75]
Freshwater, D. C. & B. D. Henry.
Optimal configuration of multicomponent distillation systems.
The Chemical Engineer :533, Sept., 1975.

[Gomez & Seader 76]
Gomez, M. A. & J. D. Seader.
Separator sequence synthesis by a predictor based ordered search.
AIChE Journal **22:970,1976.**

[Guthrie 69] Guthrie, K. M.
Capital cost estimating.
Chemical Engineering **76(6): 114,1969.**

[Guthrie 74] Guthrie, K. M.
Process Plant Estimating Evaluation and Control.
Craftsman Book Company, Solana Beach, CA, 1974.

[Harbert 57] Harbert, V. D.
Which tower goes where?
Petrol. Refiner. **36:169-174, 1957.**

[Hart et al 68] Hart, P. E,y N. J. Nilsson & B. Raphael.
A formal basis for the heuristic determination of minimum cost paths.
IEEE Trans. SSC **4, 1968.**

[Hart *etal* 72]   Hart, P. E., N. J. Nilsson & B. Raphael.
Correction to "A formal basis for the heuristic determination of minimum cost paths".
SIGARTNewsletter 37,1972.

[Heaven 69]   Heaven, D. L.
Optimum Sequencing of Distillation Columns in Multicomponent Fractionation.
Master's thesis, University of California, Berkeley, 1969.

[Hendry& Hughes 72]
Hendry, J. E. & R. R. Hughes.
Generating separation process flowsheets.
Chemical Engineering Progress 68:69,1972.

[King 71]   King, C. Judson.
Separation Processes.
McGraw-Hill, New York, NY, 1971.

[Lien 88]   Lien, K.
Expert Systems Technology in Synthesis of Distillation Sequences.
PhD thesis, Norges Tekniske Hogskole, Trondheim, Norway, 1988.

[Liu 87]   Liu, Y. A.
Process synthesis: Some simple and practical developments.
Recent Developments in Chemical Process and Plant Design.
In Liu, Y. A., H. A. McGee, Jr. & W. R. Epperly,
John Wiley & Sons, New York, NY, 1987.

[Lockhart 47]   Lockhart, F. J.
Multi-column distillation of natural gasoline.
Petrol. Refiner. 26:104,1947.

[Minderman & Tedder 82]
Minderman, P. A., Jr. & D. W. Tedder.
Comparisons of distillation networks: Extensively state optimized versus extensively energy integrated.
In *AIChE Symposium Series.* AIChE, New York, NY, 1982.

[Modi 91]   Modi, A. K.
On the Development of an Engineering Design System within an Integrated Problem-Solving and Learning Architecture.
PhD thesis, Carnegie Mellon University, Pittsburgh, PA, 1991.

[Muraki & Hayakawi 84]
Muraki, M. & T. Hayakawa.
Separation process synthesis for multicomponent products.
Journal of Chemical Engineering of Japan 17(5):533-538,1984.

[Nagdir&Liu83]Nagdir,V.M.vfcY.A.Liu.
Studies in chemical process design and synthesis:. Part V. A simple heuristic method for systematic synthesis of initial sequences for multicomponent separations.
AIChE Journal 29:926,1983.

[Nath&Motard81]
>Nath, R. & R. L. Motard.
Evolutionary synthesis of separation processes.
AIChE Journal 27:321,1981.

[Nishimura & Hiraizumi 71]
Nishimura, H. & Y. Hiraizumi.
Optimal system pattern for multicomponent distillation systems.
International Chemical Engineering 11:188,1971.

[Perry & Chilton 73]
Perry, R. H. & C. H. Chilton (editors).
Chemical Engineer's Handbook.
McGraw-Hill, New York, NY, 1973.

[Peters & Timmerhaus 68]
Peters, M. S. & K. D. Timmerhaus.
Plant Design and Economics for Chemical Engineers.
McGraw-Hill, New York, NY, 1968.

[Powers 72]   Powers, G. J.
Heuristic synthesis in process development.
Chemical Engineering Progress 68:88,1972.

[Rathore *et al* 74] Rathore, R. N. S., K. A. Van Wormer & G. J. Powers.
Synthesis of distillation systems with energy integration.
AIChE Journal 20(5):940,1974.

[Robinson & Gilliland 50]
Robinson, C. S. & E. R. Gilliland.
Elements of Fractional Distillation.
McGraw-Hill, New York, NY, 1950.

[Rod & Marek 59] Rod, V. & J. Marek.
Separation sequences in multicomponent rectification.
Collect. Czech. Chem. Commun. 24:3240,1959.

[Rodrigo & Seader 75]
Rodrigo, B. F. R. & J. D. Seader.
Synthesis of separation sequences by ordered branch search.
AIChE Journal 21:885, 1975.

[Rudder *al* 73]   Rudd, D. F., G. J. Powers & J. J. Siirola.
Process Synthesis.
Prentice-Hall, Englewood Cliffs, NJ, 1973.

[Seader & Westerberg 77]
Seader, J. D. & A. W. Westerberg.
A combined heuristic and evolutionary strategy for synthesis of simple
separation sequences.
AIChE Journal 17:951,1977.

[Siirola&Rudd71]
>   Siirola,J.F.&D.F.Rudd.
>   Computer-aided synthesis of chemical process designs.
>   *Industrial & Engineering Chemistry Fundamentals* 10:353,1971.

[Stephanopoulos & Westerberg 76]
>   Stephanopoulos, G. & A. W. Westerberg.
>   Studies in process synthesis - II. Evolutionary synthesis of optimal process flowsheets.
>   *Chemical Engineering Science* 31:195,1976.

[Tedder 81]   Tedder, D. W.
>   The computer-aided design of an optimal deethanizer sequence.
>   In *Proceedings of Summer Computer Simulation Conference.* Washington, D.C., 1981.

[Tedder&Rudd(a)78]
>   Tedder, D. W. & D. F. Rudd.
>   Parametric studies in industrial distillation, Part I. Design comparisons.
>   *AIChE Journal* 24:303,1978.

[Tedder & Rudd (b) 78]
>   Tedder, D. W. & D. F. Rudd.
>   Parametric studies in industrial distillation, Part II. Heuristic optimization.
>   *AIChE Journal* 24:316,1978.

[Thompson & King 72]
>   Thompson, R. W. & C. J. King.
>   Systematic synthesis of separation schemes.
>   *AIChE Journal* 18:941,1972.

[Umeda *etal* 79]   Umeda, T., K. Niida & K. Shiroko.
>   A thermodynamic approach to heat integration in distillation systems.
>   *AIChE Journal* 25:423,1979.

[Underwood 46]   Underwood, A. J. V.
>   Fractional distillation of multicomponent mixtures - Calculation of minimum reflux ratio.
>   */. Inst. Petrol.* 32:614,1946.

[Underwood 48]   Underwood, A. J. V.
>   Fractional distillation of multicomponent mixtures.
>   *Chemical Engineering Progress* 44(8):603,1948.

[Wehe& Westerberg 87]
>   Wehe, R. R. & A. W. Westerberg.
>   An algorithmic procedure for the synthesis of distillation sequences with bypass.
>   *Computers & Chemical Engineering* 11(6):619-627,1987.

[Westerberg & Stephanopoulos 75]
                    Westerbcrg, A. W. & G. Stephanopoulos.
                    Studies in process synthesis - 1. Branch and bound strategy with list
                        techniques for the synthesis of separation schemes.
                    Chemical Engineering Science 30:963,1975.