Evolutionary Synthesis of Separation Processes

This paper deals with the problem of finding, among many alternatives, the sequence of separation units that will isolate desired products from given feed at the minimum cost. Multicomponent products can be specified. An evolutionary procedure is presented. This method consists of two phases. In the first phase, a good initial structure is created by heuristic methods. In the second phase, the initial structure is successively modified by making evolutionary structural changes. This logic has been programmed on a digital computer and has been tested on several problems.

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SCOPE

The optimum sequencing of separation units has been treated in the chemical engineering research literature since the work of Lockhart in 1947 and that of Harbert in 1957. More recently, Hendry and Hughes (1972), Thompson and King (1972), Gomez and Seader (1976), Stephanopoulos and Westerberg (1976) and Seader and Westerberg (1977) have addressed the same problem. This work is an extension of the latest work in the field, combining eight heuristic selection rules to provide an initial structure and five evolutionary rules plus a strategy to refine the separation sequence configuration.

The product set, the key components, the type of separator, the mass separating agent, if required, are chosen in a systematic way with the help of a heuristic evaluation function. The product set and product recoveries are specified by the user, but the program redefines the product set if it is cheaper to isolate separate species in a product and remix. The system handles distillation and extractive distillation separator types (with any of several mass separating agents) and can readily be extended to other separator types. Recovery and recycle of mass separating agent will be delayed, if the cost of processing will be reduced by using the same agent in several serially connected columns. The final criterion of quality is the total annual cost of the complete sequence, fixed charges plus operating cost.

CONCLUSIONS AND SIGNIFICANCE

An evolutionary approach to the synthesis of sharp separations has been proposed. The synthesis problem is decomposed into two phases. In the first phase, an initial feasible structure is created by using heuristic rules. In the second phase, the initial structure obtained in the first phase is successively improved by applying evolutionary rules. The heuristic rules embody knowledge of the general behavior of separation units and separation sequences, and the evolutionary rules question the validity of these heuristic rules for the particular problem under consideration. This approach closely mimics the problem-solving procedures commonly employed by engineers, especially with regard to relatively large systems.

The proposed procedure does not suffer from the shortcomings of the dynamic programming based synthesis procedures such as the one proposed by Hendry and Hughes (1972). The calculations are performed only on the subproblems selected by the heuristic and evolutionary rules thereby reducing the amount of computer time used. The proposed procedure also does not suffer from the problem of "cycling" (Thompson and King, 1972), since separation types are not selected on the basis of estimated costs. The proposed procedure is practical and has been tested on several problems. For each problem, the procedure creates the best separation sequence in a reasonable amount of computational time.

In the course of process design, the problem of isolating species from a given multicomponent feed stream occurs frequently. The task of feed preparation for the reactor and the task of product purification naturally calls for the synthesis of separation sequences. In the present work, an attempt is made to systematize the synthesis of separation sequences by the creation of an arrangement of separation units that will isolate the desired products from a given feed stream at a minimum annual cost.

The problem is difficult to solve, because the number of arrangements which can isolate desired products from a given feed is enormous. As a simple example, consider a stream con-

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taining three components, A, B and C. Isolation of each of the components using distillation (I) and/or extractive distillation with a solvent X (II) is desired. The ranked lists for the components (ranked according to distribution factor, highest first) for both methods are shown below:

RL(I) : A B C
RL(II): A C B X

Nine configurations are possible for this case. For four components and two separator methods, 51 configurations are possible. The number of possible configurations increases exponentially, as the number of desired products increases and as the number of types of separators to be considered increases. Nath (1977) presents a detailed derivation of the number of possible configurations. One of the objectives of process synthesis research is to reduce the combinational space of alternative configurations to be evaluated in the search for the optimum structure.

PREVIOUS WORK ON SYNTHESIS OF SEPARATION SEQUENCES

Lockhart (1947) studied arrangements of distillation sequences for three different feeds for a natural gasoline plant. Each feed was to be separated into three products, thus requiring two distillation columns, which can be arranged either as a direct or an indirect series. For each of the feeds, the optimum arrangement was given as a function of feed composition.

Harbert (1957) pointed out that "heating requirement" is the single most important variable in distillation column arrangements. He proposed two heuristics, "difficult separation last" and "near 50-50 splits," for the optimum arrangements. The examples considered were limited to three product feeds.

Rod and Marek (1959) considered the sequencing problem from the point of view of minimizing total vapor flow in the system. Vapor flows have been calculated by using analytical expressions for the minimum reflux.

Heaven (1970) considered the sequencing problem with a very detailed costing of the distillation units. His studies confirmed the heuristic rules of Harbert for minimum cost sequences. Strong emphasis was placed on three component feeds and one example considered a five-component feed.

Nishimura and Hiraizumi (1971) have considered the distillation system pattern problem by minimizing a simplified cost function for two restricted cases, when either all components are about the same composition, or when one of the components dominates. A three-component system is synthesized as an example.

Powers (1971) has outlined a heuristic method for the creation of a separation scheme. Four heuristics have been proposed. For the process stream under consideration, all possible separation points are identified and the heuristic rules are used to evaluate the desirability of each separation. Numerical values are assigned by each heuristic to the alternative separations. The alternative with the maximum score is selected as the next separation in the process. This procedure is repeated for each process stream. This algorithm has been applied to four industrial separation problems. In two of the cases, the algorithm produced distillation sequences used by the industry. Unfortunately, no comparisons were made with the optimum sequences.

Hendry and Hughes (1972) have used the Dynamic Programming (DP) method to find the optimum arrangement of distillation and extractive distillation units. A simplification was made; any unit which used a Mass Separating Agent (MSA) had the MSA removed in the immediate successor unit. The method is general but has the shortcomings of DP; during suboptimizations all unique subproblems are evaluated. The computational time requirement is almost prohibitive for big problems. The algorithm, however, guarantees optimality.

Thompson and King (1972) have presented a systematic method suitable for computer implementation. A single "cheapest first" heuristic has been used to create several good separation sequences to isolate multicomponent products from a given feed without a large consumption of computer time. In the beginning, a feasible product set is identified based on feed composition and at a typical process temperature. The cheapest separator is picked by comparing predicted costs of alternate separation units. After the sequence is decided, it is simulated and actual costs computed are used to update cost coefficients used for cost prediction. Since computations are performed in the forward direction, the compositions of the streams are known exactly. This procedure faces a problem of "cycling" but has nevertheless been applied successfully to several large-scale examples.

Westerberg and Stephanopoulos (1975) have proposed a branch and bound search technique. Sub-Lagrangians for all possible subsystems are computed first. Based on "choose the potentially cheapest unit first" heuristic, a basic flowsheet is created and dual and primal bounds are computed. Flowsheets whose dual bound exceeds the primal bound of the basic flowsheet are rejected. The remaining flowsheets can be further screened by repetition of the procedure above. The choice of the basic flowsheet and the value of the primal bound for the basic flowsheet are crucial; neither of which is a trivial problem. Two example problems have been solved by the proposed method.

In a later paper, Stephanopoulos and Westerberg (1976) have proposed three evolutionary rules for the synthesis of separation processes. In this work, the MSA is treated unlike other products and is isolated in the immediate successor unit after use. Also, the starting structure is crucial for the success of this search scheme. This approach can be improved by using an n-step look-ahead strategy but at the cost of greatly increased computational time.

Freshwater and Henry (1975) have presented a detailed cost and total energy requirement for three-, four-, and five-component systems as a function of configuration. Detailed analyses were made as a function of feed composition. For an N-component system $N + 2$ different feeds were considered. System feed streams consisted of hydrocarbons in the range $C4$ and $C7$. Only simple distillation units were considered and products were all relatively pure single components. Surprisingly, for most cases considered, the direct sequence was the optimum configuration. The study, however, confirms a direct proportionality between energy requirement and cost.

Rodrigo and Seader (1975) have outlined a modified depth-first method (Nilsson, 1971). In a later paper, Gomez and Seader (1976) have further refined the search procedure by using a modified uniform cost method (Nilsson, 1971).

In a recent note, Seader and Westerberg (1977) have proposed a combined heuristic and evolutionary strategy for synthesis of simple separator sequences. Six heuristics have been suggested for the creation of the initial structure and two evolutionary rules have been suggested. Two example problems have been synthesized manually by the proposed strategy. In the first example, the optimum structure is obtained in two evolutionary steps, but in the second example the proposed strategy fails to produce the optimum flowsheet due to the absence of one evolutionary rule (#5).

In another paper, Bakshi and Gaddy (1977) have analyzed nine separation problems by exhaustive search over the space of configurations. The reflux ratio for each column and the pressure for the first column in the sequence has been optimized by a random search technique. Three commonly used heuristics have been tested, but no strategy for the application of these heuristics to solve a synthesis problem has been proposed.

Another related synthesis problem exists for the cases when complex separation units are considered. Such cases are not considered in the present paper but references are included for completeness. Research performed in this area is limited to the cases isolating single-component products from a ternary feed stream using complex distillation units. Petlyuk (1965) examined four complex configurations and two conventional configurations (direct and indirect arrangement of two distilla-

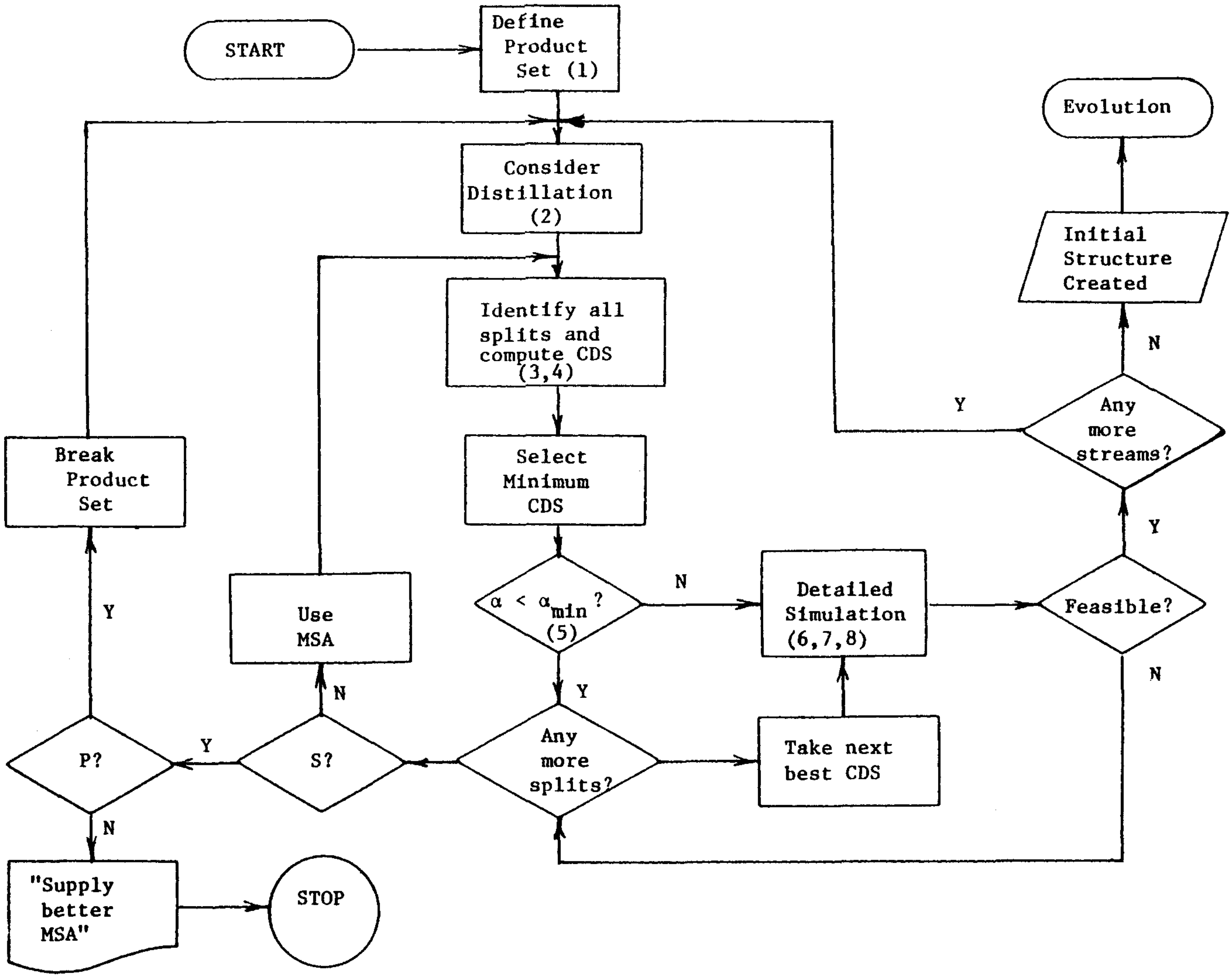

Figure 1. Logic for creation of initial structure. $S? \equiv$ All separation methods considered? $P? \equiv$ All there any multicomponent products?

tion units) for a three-component system. The total specific amount of liquid vaporized was used to compare different configurations. Ten different compositions were considered. For each composition, complex schemes were better than the conventional schemes. Stupin and Lockhart (1972) compared one complex scheme and the two conventional schemes for an equimolar ternary feed stream. The complex scheme had about 20% less vapor boil-up than the conventional schemes. Tedder (1976) has presented a more thorough analysis for ternary feed streams. Six ternary hydrocarbon systems have been considered. For each ternary system, detailed design and costing is performed for seven different compositions and for six complex and two conventional configurations. The results of this study have been summarized on ternary diagrams on which the optimum designs are given as a function of composition.

Problem Definition

Given a feed stream of known conditions (i.e., composition, flow rate, temperature, pressure), it systematically synthesizes a process that can isolate the specified products from the feed at minimum cost.

$$Min\ [\Sigma C_i]$$

$i \epsilon I$ = a feasible separation unit
C_i = the total annual cost of separation unit i
I = a subset of S
S = the set of all possible separator configurations that can produce the desired products

The desired products are such that a specie is required to be present in only one product. The separation types are restricted to conventional distillation and extractive distillation methods. For the extractive distillations, the solvents used are to be recycled.

A second objective which is most important, is to arrive at a final configuration with a minimum computational effort.

Proposed Method

An evolutionary method is presented. The method consists of two phases. In the first phase, a very good starting structure is created on the computer by eight heuristic rules. In the second phase, the starting structure is modified by making evolutionary changes. The evolutionary changes are made by following five evolutionary rules. These rules are applied in a hierarchical manner until no further modification can be detected. The structure thus obtained is termed the "best structure."

Definitions

Some of the terms used in this paper have special meaning as defined here. A *separation factor* between two components is the ratio of the distribution coefficients of those two components. A *distribution coefficient* or *distribution factor* is the ratio of component mole fraction in the lighter phase to that in the heavier phase, at equilibrium, or in solvent phase to feed phase, if it applies. A *mass separating agent* (*MSA*) is a component added to a separation unit to affect the desired separation. A *ranked list* is the ranking of components in the feed in order of decreasing distribution coefficients. *Sequence, arrangement, structure, process, flowsheet* and *configuration* have been used interchangeably and refer to an arrangement of separation units. *Process stream* refers to a stream in the process which needs further processing. *Column, separation unit* and *separator* are used interchangeably to refer to distillation or extractive distillation equipment. *Product set* is a mapping of components (by number) onto product numbers. A *direct sequence* removes the products successively in the distillate or overhead stream of each column in the sequence.

CREATION OF INITIAL STRUCTURE

The initial structure in evolutionary synthesis is crucial to the success of the evolutionary synthesis procedure. The initial structure is the starting structure on which structural changes are made successively by evolution. If the starting structure is too unlike the optimum structure it would require many more iterations to converge. Since the evolutionary rules do not guarantee optimality in a rigorous mathematical sense, a structure too unlike the optimum would perhaps lead to a local optimum rather than the global optimum. A better initial structure may, on the other hand, arrive at the optimum in less iterations and with a higher probability of reaching the global optimum.

We propose a set of heuristic rules that in a systematic way would create a good starting structure. Most of the heuristic rules presented here have been available in the literature, but in a qualitative manner. We have attempted to make the rules quantitative and to order their application in a manner suitable for computer implementation.

Sharp Separators as List Splitters

For sharp separations, a separator separates the input stream into two output streams, the top stream and the bottom stream, each of which contains a predominant fraction of a species in the feed or almost none at all. Each stream consists of a list of components and the separator can be viewed as a list splitter which splits the input list into two smaller output lists. The components in the feed stream can be ranked in the order of decreasing distribution factors for a particular separation method. The arranged list is called the *ranked list*. In the ranked list, the component with the largest value of the separation factor is called the *lightest component* and the one with the smallest value of the separation factor is called the *heaviest component*. A *split* is defined by specifying two adjacent components, the lighter of the two is called the *light key* and the heavier one is called the *heavy key*. A sharp separator splits the ranked list corresponding to its input stream into two lists. The list corresponding to the top stream contains the light key and all the lighter components and the list corresponding to the bottom stream contains the heavy key and all the heavier components.

This concept of visualizing a separator as a list splitter has been described in detail by Hendry and Hughes (1972).

Heuristic Rules

Heuristic rules for the creation of the initial structure are given below:

1) Favor the smallest product set
2) Favor distillation
3) Easiest separation should be done first
4) A separation method using a mass-separating agent (*MSA*) cannot be used to isolate another *MSA*
5) A separation with $\alpha_{LK-HK} < \alpha_{min}$ is not acceptable
6) Operating pressure should be close to ambient
7) Set split fractions of the keys to those specified by the user
8) Set operating reflux ratio equal to 1.3 times the minimum reflux ratio for each column

Heuristic rule 1 concerns the product set definition. This rule is simple and is the first step in the creation of the initial structure. The product set is defined from the designer's requirements. Heuristics 2, 3, 4 and 5 form a group of rules (group II) which provide guidelines for selection of the separation method and split point for each stream starting from the feed stream. Heuristics 6, 7 and 8 form another group of rules (group III) which provide specifications for the detailed simulation of the design objective obtained by the previous group of rules and as such they are not strictly heuristics.

The proposed procedure for the creation of initial structure is shown in Figure 1 where the numbers within parentheses identify the heuristic rules.

Heuristic Rule 1. For a process in which the desired products are all single components, the product set is unique and trivial. If, however, the desired products include one or more multicomponent products, there is more than one product set. In such cases, during the creation of the initial structure strong preference is given to the smallest product set, i.e., the user-supplied product definition. Intuitively, a smaller product set suggests a structure with fewer separation units and potentially less total cost.

In the cases when the initial structure cannot be completed either because a multicomponent product cannot be isolated directly or because the separation unit isolating the multicomponent product violates heuristic rule 5, the multicomponent product is split to produce a new product set. With this product set, the creation of the initial structure is attempted again.

This heuristic may not lead to the best structure; therefore, heuristic rule 1 is challenged during the evolution of structures.

Heuristic Rule 2. Distillation or methods using energy-separation agents are favored because they minimize the number of separation units in the structure. For each separation unit using an *MSA*, an additional separation unit is required to isolate the *MSA*. In addition, the internal flow rates are usually much higher for the separator using an *MSA*. These disadvantages are offset, if the separation using the *MSA* provides a much larger separation factor or if it makes the isolation of a multicomponent product feasible when it is otherwise infeasible.

During the creation of the initial structure, for any stream under consideration, distillation is tried first. Only when distillation does not give any feasible design alternative, separation methods using an *MSA* are considered.

This heuristic may not lead to the best process and the application of this heuristic in the creation of the initial structure may be negated by evolutionary changes later on.

Heuristic Rule 3. During the creation of the initial structure, for each stream having more than one possible split, we are faced with the problem of picking the one that is likely to be in a near-optimum structure. We propose here to select the split which is the easiest. The easiest separation is qualitatively defined as the one which in general is in accordance with the following four heuristics.

a) Favor large α_{LK-HK}
b) Favor a balanced column
c) Favor sloppy splits of the keys
d) Favor less distillate product

Each of the above heuristics has merit. We will now consider them one by one. Later on, a quantitative formula that combines the above four heuristics is presented.

The heuristics 3a) and 3c) are widely accepted. Favoring large α_{LK-HK} and sloppy splits in the beginning of the synthesis defers the difficult separations, i.e., those with low α_{LK-HK} and high recovery of the keys, towards the end of the synthesis. Difficult separations are best done when the stream flow rate is low (requiring a smaller diameter for the separator) and when most of the nonkey components are absent. The cost of large columns at high reflux rates is most sensitive to the presence of nonkey components.

Heuristic 3b) is favored when the amounts of overhead and bottoms products can be made about the same. The reflux ratios in the sections above and below the feed will be better balanced, which leads Harbert (1957) to call this heuristic "the advantage of 50-50 split" and to justify it on the basis of minimum heat requirement.

Heuristic 3d) favors, other things being equal, a separator with less distillate product. The operating costs for a column vary directly with the amount of distillate. Consequently, a split with a smaller amount of distillate implies lower operating expense and is preferred if other factors are equal. A sequence generated by this heuristic alone would result in a direct sequence of separators.

For any proposed split where all four heuristics are favored, we can be reasonably sure that the separation would lead to a good structure. However, in most cases the heuristics will be in conflict. For example, if, for a given stream, the split gives the largest α_{LK-HK}, the two heuristics 3a) and 3b) point toward different decisions. Such conflicts can be resolved by giving each split a numerical value proportional to the difficulty of separation. This value should be such that it represents the four heuristics in a quantitative fashion. We propose here such a function, called the Coefficient of Difficulty of Separation (*CDS*):

$$CDS = \frac{\log \dfrac{sp_{LK}}{1-sp_{LK}} \cdot \dfrac{sp_{HK}}{1-sp_{HK}}}{\log \alpha_{LK-HK}} \cdot \frac{V}{V+L} \cdot \left(1 + \left|\frac{V-L}{V+L}\right|\right) \quad (1)$$

The first term is the minimum number of theoretical stages and takes into account heuristics 3a) and 3c) realistically. The second term is the fraction distillate and accounts for heuristic 3d). The last term is a penalty term which penalizes unbalanced columns. When $V = L$, the column is balanced and this term is unity. For any other values of V and L, the term is always greater than 1. The last term is a mathematical analog of heuristic 3b). This function itself becomes a heuristic rule in lieu of 3a) through 3d). The choice of a triple product and of unity weighting factors among the terms is arbitrary. However, our experience with the *CDS* function does not suggest that any higher complexity is needed. An alternative form of a *CDS* function might be developed using minimum reflux as the first term instead of minimum theoretical stages. However, either choice of first term is equally sensitive to the degree of difficulty and minimum theoretical stages are easier to compute.

With this *CDS* function in hand we are in a position to evaluate each possible split for a stream in a quantitative way. The split with the smallest value of *CDS* is tried for detailed simulation; if it is infeasible, the split with the next larger value is tried. This procedure is followed for each stream that needs to be processed in the structure, resulting in the creation of the initial structure. A split becomes infeasible during detailed simulation if $\alpha_{LK-HK} < \alpha_{min}$ at the actual conditions in the column, or if a multicomponent product is split, or if an extractive distillation results in an immiscibility between solvent and residue in the bottom of the column.

Heuristic Rules 4 and 5. Heuristic rule 4 prohibits the use of a separation using an *MSA* to isolate another *MSA*. This agrees with heuristic rule 2, favoring distillation. Moreover, by this rule, we have eliminated the possibility of absurd sequences having an indefinite number of columns in which an *MSA* is isolated using another *MSA* which is isolated using another *MSA* and so forth.

Heuristic rule 5 is more of an assumption, separators with α_{LK-HK} less than α_{min} are rejected. This assumption is quite sensible since very low values of α result in extremely expensive columns. For the present study, α_{min} is arbitrarily set to a value of 1.1. This heuristic is applied to eliminate very difficult separations, both during the separator selection and during separator simulation.

Heuristic Rules 6, 7 and 8. These rules provide specifications for selecting the operating conditions for a separation unit. Operating pressure is set close to the ambient, the reflux ratio is set to 1.3 times the minimum and the design of the column is performed for prespecified values of recovery for each key. After the best structure has been obtained by evolution, some of the operating conditions are optimized to meet product recovery and purity specifications by the user and to choose the most economic reflux ratio. The optimization step is not discussed in this paper but the details are available in Nath (1977).

EVOLUTION OF STRUCTURES

Evolution of structures is the second step in the synthesis procedure by evolutionary methods. The initial structure created by heuristic rules is successively modified by the application of the evolutionary rules. The evolutionary rules suggest structural modifications which usually question the validity of heuristics or other assumptions used in the creation of the initial structure. If the assumptions or heuristics were not appropriate for specific applications, corrective measures are taken by the evolutionary rules.

The evolutionary rules are applied in an hierarchical order. If a particular rule does not suggest any structural modification, the next evolutionary rule is applied. The evolution stops when no more modifications are possible. If, however, an evolutionary rule suggests a modification at some point in the structure, the downstream structure is destroyed and a new structure is created by repeating the first step. The upstream structure remains unchanged. If the new structure is superior to the starting structure, the new structure replaces the starting structure and is evolved further. If the new structure is inferior to the starting one, the starting structure is restored in the computer memory and is evolved further.

Evolutionary Rules

The evolutionary rules used in the proposed method are as follows:

Rule 1: Challenge heuristic 1.

Rule 2: Examine the neighboring structures, if: a) the *CDS* is within 10%; and b) refrigeration is required to condense the reflux.

Rule 3: Challenge heuristic 2.

Rule 4: Examine neighbors to decide if the *MSA* removal should be delayed.

Rule 5: Challenge heuristic 3, if: a) R_{min} of the immediate successor $>>$ R_{min} of the unit under consideration; and b) the cost of the immediate successor $>>$ the cost of the unit under consideration.

Evolutionary Rule 1. For the creation of the initial structure, the smallest feasible product set was chosen in accordance with heuristic rule 1. To retain the smallest product set, sometimes a separation unit using an *MSA* is used in the process. A separation unit using an *MSA* needs an additional separator to isolate the *MSA* for recycle. For such cases, there is a possibility which could lead to a superior flowsheet. This possibility is to break the multicomponent product which makes the use of an *MSA* necessary in the first place. The two products defined by breaking the multicomponent product may both be isolated using distillation units, in which case the new process will have exactly the same number of separation units and may be superior. Evolutionary rule 1 checks for such possibilities in the structure undergoing evolution.

A simple example illustrating the application of evolutionary rule 1 is considered next. For a ternary feed stream containing species *A*, *B* and *C*, isolation of species *A* and *B* as product 1 and specie *C* as product 2 using two separation methods is desired. The separation methods available are distillation (I) and extractive distillation using solvent *X* (II). The ranked list corresponding to these two separation methods is given below:

RL (I) : $A\ C\ B$
RL (II) : $A\ B\ C\ X$

The initial structure for this problem using the smallest product set requires extractive distillation followed by a distillation step to recover the solvent. Evolutionary rule 1 is applicable; the product set is altered by breaking the multicomponent product. The new structure would require two distillation steps with remixing of components A and B to make product 1. The new structure would be economically superior to the starting structure if the separation factors for extractive distillation are not sufficiently higher than those for simple distillation to overcome the additional cost of processing the added solvent.

Evolutionary Rule 2. This evolutionary rule takes into consideration some of the shortcomings of the *CDS* function. The *CDS* function defined above is an evaluation function which assigns a numerical value proportional to the difficulty of separation for each possible split of a process stream. This evaluation is approximate since it does not consider the operating reflux ratio and the operating pressure of the separation unit in the evaluation. This approximation to some extent is compensated for in the following rules:

- *Rule 2a)*: Because of the approximate nature of the *CDS* function, it cannot be used to distinguish between very competitive design alternatives. To compensate for this, designs with *CDS* values within 10% of each other are treated alike. Therefore, during the creation of a structure, the splits which are within 10% of the one selected for the creation of the structure are saved. During evolution, these alternative splits are considered to create additional structures.
- *Rule 2b)*: The cost of refrigeration to condense the reflux is extremely high. The *CDS* function cannot predict the presence of refrigeration for a separation and consequently is not a good representation of the difficulty of separation in such cases. To rectify this shortcoming, during the creation of an initial structure, a separation using refrigeration to condense the reflux is tagged. During evolution, for these tagged separations other design alternatives are considered.

Evolutionary Rule 3. During the creation of the initial structure, strong preference was given to the use of distillation units in the flowsheet, in accordance with heuristic rule 2. Evolutionary rule 3 questions this heuristic. For each distillation unit in the scheme, alternative separation units using an *MSA* are considered. Separation methods using an *MSA* require an additional separation unit to isolate the *MSA* for recycle. A separation unit using an *MSA* will become economically superior to a distillation unit, only if the separation unit using the *MSA* would have a separation factor (α) between the *LK* and the *HK* sufficiently larger than the one for distillation. How much larger should the value of α be for the separation using an *MSA* compared to the value of α for distillation? No rigorous answer can be given to answer this question; however, a semiquantative analysis will give an approximate.

As a first approximation, we can assume that the actual number of stages, N, for a specified split is:

$$N \propto \frac{1}{ln\ \alpha}$$

If α for the separation using an *MSA* has a magnitude equal to the square of α for distillation, the number of stages for the separation unit using an *MSA* would be half the number of stages for the distillation unit. To a first approximation, the cost of the separation using an *MSA* would be about half of the cost of the distillation unit. In this case, the combined cost of the separation unit using an *MSA* and the separation unit isolating the *MSA* may be less than the cost of the distillation unit.

Based on the above semiquantitative reasoning, the following criteria for considering a separation using an *MSA* instead of distillation for a split is recommended:

$$\alpha_{MSA} \geq \alpha^{1.95}$$

where, α_{MSA} refers to the separation factor between the *LK* and *HK* for the separation method using the *MSA* and α refers to the separation factor between the *LK* and *HK* for distillation.

For a process stream, in general, several splits are possible. The criterion above is generalized for such streams:

$$\underset{\text{All Splits}}{\text{Max}} (\alpha_{MSA}) \geq \underset{\text{All Splits}}{\text{Max}} (\alpha)^{1.95}$$

i.e., a separation using an *MSA* may be a superior alternative to distillation, if the value of the largest α for the separation using an *MSA* is at least equal to 1.95 power of the value of the largest α for distillation for the stream under consideration.

Evolutionary Rule 4. The *MSA* is generally a fairly heavy polar solvent with a low value of distribution coefficient. During the creation of structures, strong preference is given to easy separations in accordance with heuristic rule 3, and there is a strong tendency to isolate the *MSA* immediately after its use in a separation unit. At times, this leads to structures in which there are two or more separation units using the same *MSA* which are arranged such that the top stream from the separator isolating the *MSA* for the first separator using an *MSA* is fed to the second separator using the same *MSA* and so on. The same result can be obtained using fewer separation units by delaying the isolation of the *MSA*. Evolutionary rule 4 searches for patterns in which the isolation of the *MSA* can be delayed and creates better structures by delaying the isolation of the *MSA*. Our Example 1, shown later, illustrates the application of this rule.

Evolutionary Rule 5. For multistage structures which are generated by the present synthesis program, the easiest separation at any stage will not always lead to the optimum structure. A very easy separation at a particular stage may in fact make the next separation very difficult and may lead to a flowsheet more expensive than the one obtained when both the separations are moderately difficult. In particular, unanticipated composition effects on relative volatility would explain such a situation. Evolutionary rule 5 is based on the above logic. In a flowsheet, the occurrence of an easy separation followed by a very difficult one is detected; for each such occurrence, new flowsheets are created by increasing the difficulty of the first separation.

For the flowsheet undergoing evolution, all pairs of adjacent separation units are checked to find if one of the following conditions is met:

a) The minimum reflux ratio of the immediate successor is much greater than (for example, nine times) the minimum reflux ratio of the unit under consideration.

b) The cost of the immediate successor units is much greater than (for example, five times) the cost of the unit under consideration.

If either of the above conditions is true, the split for the first separation is changed. This also alters the split for the second separation and the new structure may be superior to the starting structure.

Evolutionary Strategy

Each of the evolutionary rules described in the previous section suggests a structural modification to improve the starting structure. The evolutionary rules can be applied in a variety of ways. A strategy to apply these rules is given next.

The definition of the product set is the single most important decision in the synthesis of separation sequences. For the creation of the initial structure, the product set was defined on the basis of heuristic rule 1. Evolutionary rule 1 questions the validity of this heuristic rule and is applied before any other evolutionary rule to resolve the question of the product set definition. Evolutionary rules 2, 3, 4 and 5 are treated equally, but of course cannot be applied at the same time. Therefore, starting from the feed stream forward, evolutionary rule 2 is applied next. If any modification is suggested by this rule, it is adapted in the starting structure and a new structure is produced. The new structure or the starting one, whichever is superior, is evolved further by applying rule 2 to the portion of the structure not checked by rule 2 in the earlier application. Evolutionary rule 3 is applied starting from the feed stream forward, after no further structural modifications are suggested

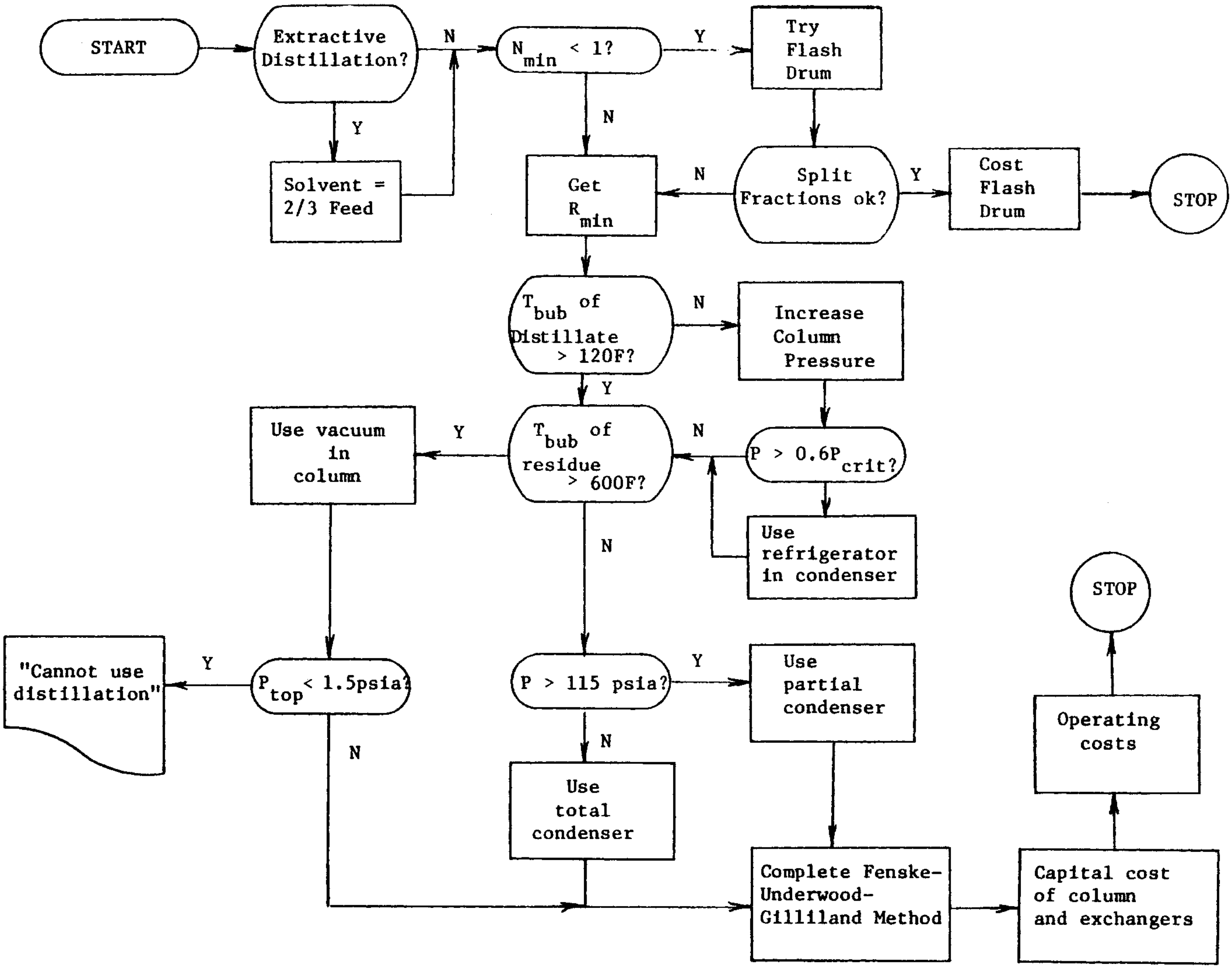

Figure 2. Logic of detailed simulation.

by rule 2. If rule 3 suggests a modification, it is adapted to the structure undergoing evolution, whichever is superior, is evolved further. Rule 2 is applied again only to the modified portion of the structure. Rule 3 is applied again to the portion of the structure not checked by rule 3 in the earlier application. This process is repeated by applying all the evolutionary rules.

DETAILED SIMULATION

Distillation and extractive distillation units are designed by the Fenske-Underwood-Gilliland short-cut method. For extractive distillation, solvent is added to the extent of 67 mol % of the total liquid feed. The column pressure selection rules are obtained from Thompson and King (1972), Figure 2. Costing of columns and heat exchangers are based on the work of Heaven (1970) as coded by Thompson and King (1972). A summary of the Heaven procedure may also be found in Rathore, van Wormer and Powers (1974). An equation for the Gilliland relationship may be found in Molokanov et al. (1972). Vapor-liquid equilibrium and liquid-liquid immiscibility detection are based on the work of Thompson and King (1972). Total annual cost (TAC) is one-fourth of the installed capital cost plus the annual operating expense.

SAMPLE SEPARATION SEQUENCES

Example 1: C6 Separation

Consider the C6 separation synthesis problem described by Rodrigo and Seader (1975). The feed stream consists of three C6 components, each of which is to be isolated in a fairly pure form. Ordinary distillation and extractive distillation using phenol as the *MSA* are suitable. The details of the problem are given in Table 1.

Creation of Initial Structure. The feed stream is considered first. Two tentative designs are selected:

TABLE 1. EXAMPLE 1: PROBLEM DEFINITION

FEED (Stream 1):

Component	Component Name	Mole Fraction
1	*n*-Hexane	0.3333
2	Benzene	0.3333
3	Cyclohexane	0.3334

Total Flow Rate = 170.1 kg mol/h
Temperature = 37.8°C
Pressure = 1.033 kg/cm² abs.

Desired Products:

Product	Component
1	1
2	2
3	3

Separation Methods Available Are:
I) Distillation
II) Extractive distillation using phenol (component 4)
Initial Ordering of Components at 54.4°C:
I) 1, 2, 3
II) 1, 3, 2, 4

Design #1: $LK = 1$ $HK = 2$ $ST = \mathrm{I}$ $\alpha = 1.36$
$CDS = 1.222$
Design #2: $LK = 2$ $HK = 3$ $ST = \mathrm{I}$ $\alpha = 1.18$
$CDS = 4.461$

The first design has a lower CDS value and is tried for detailed simulation, producing two new streams, 2 and 3. Stream 2 contains mostly product 1, and stream 3 contains the rest. Stream 3 needs further processing:

Design #1: $LK = 2$ $HK = 3$ $ST = \mathrm{I}$ $\alpha = 1.18$
$CDS = 2.508$

During detailed simulation the properties stream 3 are recomputed at actual operating conditions. The α_{LK-HK} is 1.04 which is below the acceptable level. Consequently, this design is abandoned and an additional design is proposed:

Design #1: $LK = 3$ $HK = 2$ $ST = \mathrm{II}$ $\alpha = 1.76$
$CDS = 0.750$

Detailed simulation returns a feasible design resulting in two new streams, 4 and 5. Stream 4 contains product 3 and stream 5 contains product 2 and the MSA used in separator 2.

For stream 5, only one design is proposed:

Design #1: $LK = 2$ $HK = 4$ $ST = \mathrm{I}$ $\alpha = 14.4$
$CDS = 0.117$

After detailed simulation two new streams, 6 and 7, are generated. Stream 6 contains product 2 and stream 7 has the MSA which is recycled back to separator 2. There are no more streams left which need further processing. The initial structure has been created, Figure 3. The total annual cost (TAC) for this structure is \$274,803/yr. This structure is saved for future reference.

Evolution of Structures. Evolutionary rules are applied for possible modifications to the initial structure. Evolutionary rule 1 does not apply. Rule 2 does not suggest any changes. Rule 3 suggests that a change of separation method for separator 1 may result in a better structure. Consequently, the structure downstream of stream 1 is destroyed. Synthesis is restarted by implementing the proposed modification to the initial structure. A new structure E-1 is created. This structure has a TAC = \$214,675/yr, Figure 4. Since the structure E-1 is better than the initial structure, structure E-1 is saved for future reference. Now structure E-1 is evolved. Evolutionary rules 2 and 3 do not suggest any change. Rule 4, however, suggests that a change in the split for separator 2 to delay the MSA removal may improve the structure. Consequently, the structure downstream of separator 2 is destroyed. Synthesis is restarted by implementing the proposed modification to structure E-1. A new structure E-3

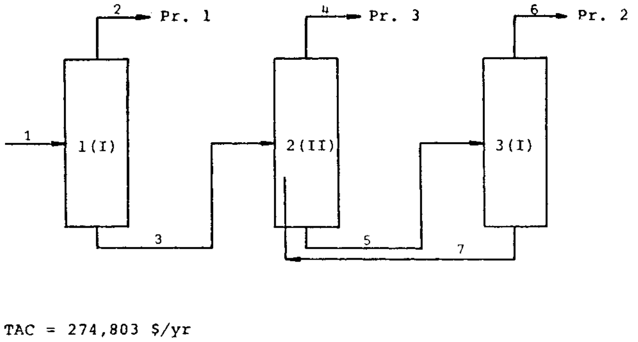

Figure 3. Example 1: initial structure.

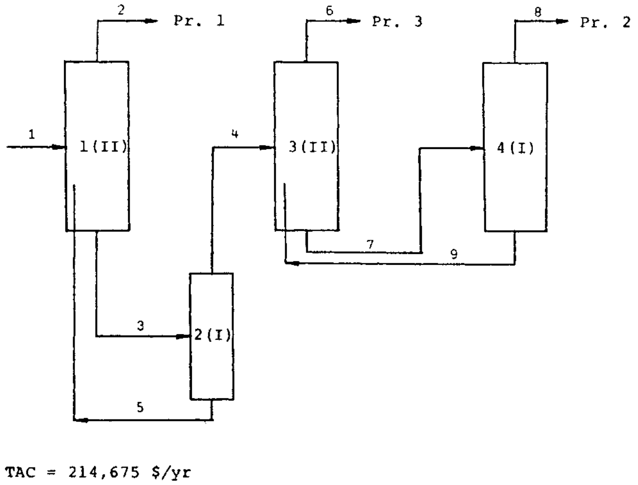

Figure 4. Example 1: structure E-1 (TAC = \$214,675/yr).

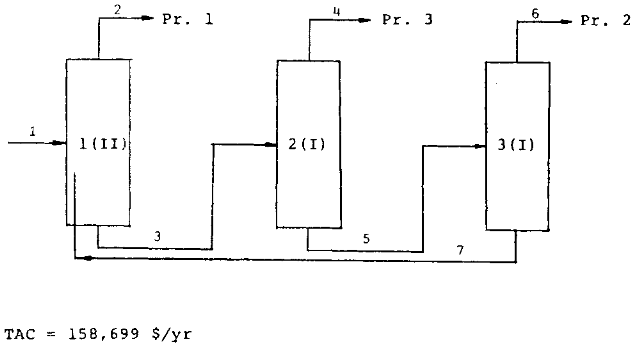

Figure 5. Example 1: structure E-2, optimal structure (TAC = \$158,699/yr).

TABLE 2. COMPARISON OF VARIOUS METHODS FOR EXAMPLE 1.

$N_s = 9$
$N_{usp} = 16$

Method	N_{sa}	N_{uspa}
Rodrigo and Seader (1975) OBS*	4	10
Gomez and Seader (1976) PBOS†		8
Proposed Method	3	6

* Ordered branch search.
† Predictor based ordered search.

TABLE 3. EXAMPLE 2: PROBLEM DEFINITION

Feed (Stream 1):

Component 1	Component Name	Mole Fraction
1	Propane	0.0147
2	n-Butane	0.5029
3	Butene-1	0.1475
4	Trans-Butene-2	0.1563
5	Cis-Butene-2	0.1196
6	Pentane	0.0590

Total Flow Rate = 303.04 kg-mole/hr.
Temperature = 53.89°C
Pressure = 5.62 kg/cm^2 abs.

Desired Products:

Product	Component(s)
1	1
2	2
3	3, 4, 5
4	6

Separation Methods Available Are:
I) Distillation
II) Extractive Distillation Using Furfural (Component 7)
Initial Ordering of Components at 54.4°C:
I) 1, 3, 2, 4, 5, 6
II) 1, 2, 3, 4, 5, 6, 7

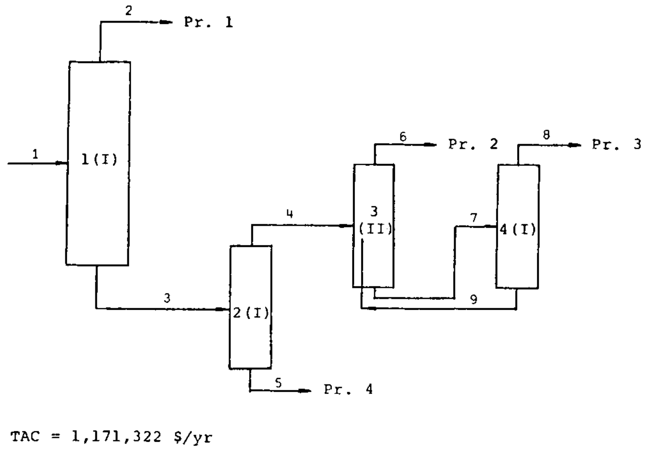

Figure 6. Example 2: initial structure using initial product set (TAC = $1,171,322/yr).

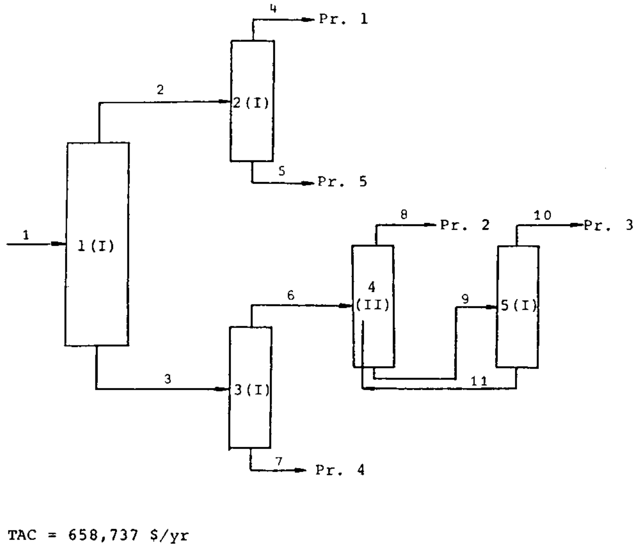

Figure 8. Example 2: Structure E-2 using product set 2, optimal structure (TAC = $658,737/yr).

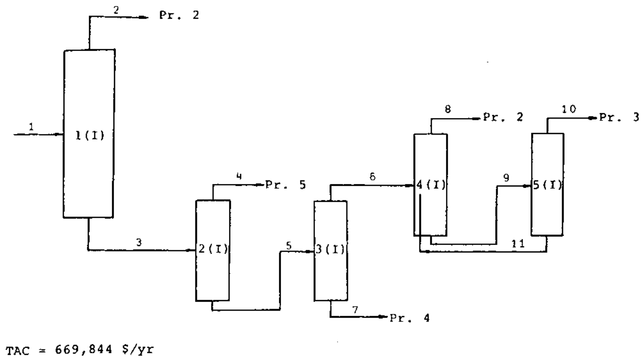

Figure 7. Example 2: structure E-1 using product set 2 (TAC = $669,844/yr).

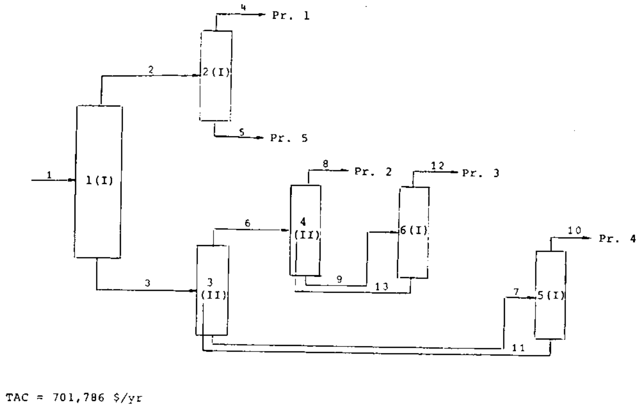

Figure 9. Example 2: structure E-3 using product set 2 (TAC = $701,786/yr).

is created. This structure has a TAC = $158,699/yr, Figure 5.

Now the evolution of structure E-2 is attempted but evolutionary rules 2, 3, 4 and 5 do not suggest any further modification. Rodrigo and Seader (1975) have obtained the same structure as the optimal structure. The costs reported by Rodrigo and Seader (1975) are different since a slightly different procedure for computation of costs has been used. Table 2 gives a comparison of the various methods used to solve this problem.

TABLE 4. COMPARISON OF VARIOUS METHODS FOR EXAMPLE 2.

N_s = 376
N_{usp} = 92

Method	N_{sa}	N_{uspa}	
Hendry and Hughes (1972), *DP*	4	64	Certain Splits Were Prohibited, Reducing N_s = 227, N_{usp} = 64
Westerberg and Stephanopoulos (1975), *B&B**	4	43	
Stephanopoulos and Westerberg (1976) Evolutionary	10 19		Initial Structure Obtained by *B&B* Initial Structure Obtained As Direct Sequence
Rodrigo and Seader (1975), *OBS*	14	23	
Gomez and Seader (1976), *PBOS*		21	Some More Splits Were Prohibited, reducing N_s = 33, N_{usp} = 30
Seader and Westerberg (1977), *H&E*†	3	11	
Proposed Method	4	12	

* Branch and bound method.
† Heuristic and evolutionary method.

Example 2: *n*-Butylene Purification System

Consider the *n*-butylene purification problem studied in detail by Hendry and Hughes (1972). The feed stream is the stabilized output from a butane dehydrogenation unit in a butadiene processing plant and consists of four components. Four products are to be isolated in relatively pure form. Ordinary distillation and extractive distillation using furfural as the *MSA* are suitable. The details of the problem are given in Table 3.

Creation of Initial Structure. The synthesis program creates an initial structure by following the heuristic rules. The initial structure has a total annual cost of $1,171,322/yr and is shown in Figure 6.

Evolution of Structures. Evolution of the initial structure is attempted and rule 1 suggests the breaking up of product 3 since an *MSA* was used to isolate product 3 in the initial structure. The new product set is given below:

Product Set 2

Product	Components
1	1
2	2
3	4,5
4	6
5	3

Using the product set 2, the procedure for the creation of the initial structure is repeated and a new structure, structure *E*-1 is created, Figure 7. This structure has a total annual cost of $669,844/yr and is superior to the initial structure.

Structure *E*-1 is evolved next. Rules 2, 3 and 4 do not suggest any modifications. Rule 5, however, suggests changing the split in separator 1. The structure downstream of separator 1 is destroyed. The proposed modification is implemented and a new structure, *E*-2, is created, Figure 8. This structure has a total annual cost of $658,737/yr and is superior to structure *E*-1.

Structure *E*-2 is evolved next. Rules 2, 3 and 4 do not suggest any modifications. Rule 5, however, suggests a change in the split for separator 3. The structure downstream of separator 3 is destroyed and the proposed modification is implemented. A new structure, *E*-3 is created. This structure has a TAC = $701,786/yr, Figure 9. Since structure *E*-3 is inferior to structure *E*-2, structure *E*-2 is retained.

Structure *E*-2 cannot be improved any further. Hendry and Hughes (1972) have obtained the same structure as the optimal structure. The costs reported by Hendry and Hughes (1972) are different, since different procedures for design and costing have been used. Table 4 gives a comparison of the various methods used to solve this problem. Additional examples are given by Nath (1977).

NOTATION

$A, B, C,$ = products $A, B, C, \ldots$ or refers to components $A, B, C, \ldots$
C_i = cost of column ($/yr)
CDS = coefficient of difficulty of separation
HK = heavy key
L = flow rate of bottom product
LK = light key
max = maximum
MS = number of separators using an MSA
MSA = mass separating agent
N = number of components or number of stages
N_s = number of possible configurations
N_{usp} = number of possible unique subproblems
N_{sa} = number of configurations actually developed
N_{uspa} = number of unique subproblems actually developed
R = reflux ratio
R_{min} = minimum reflux ratio
RL = ranked list
sp = split fraction
S = number of separators available
ST = separation type
TAC = total annual cost ($/yr)
V = flow rate of distillate product
α = separation factor
1, 2, 3 = separators 1, 2, 3, . . .
I, II, = separation types (I, II, . . .)
ϵ = belongs to
Σ = summation
$\propto$ = proportional to
{ } = set

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