A Review of Process Synthesis

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Process synthesis is the step in design where the chemical engineer selects the component parts and how to interconnect them to create his flowsheet. This paper reviews the rapidly growing process synthesis literature of over 190 articles, almost all of which have been produced in the last decade.

The paper first introduces the nature of the synthesis problem and outlines the variety of approaches which have appeared to solve aspects of it. The problems include developing a representation, a means to evaluate alternatives, and a strategy to search the almost infinitely large space of possible alternatives. As the article demonstrates, effective solutions are very dependent on the nature of the synthesis problem being addressed.

The article covers in detail the following five synthesis topics: chemical reaction paths, separation systems, heat exchanger networks, complete flowsheets, and control systems. Readily apparent are the development of industrially significant insights to aid in the design of heat exchanger networks. Reasonable progress exists in the synthesis of separation systems based on nearly ideal distillation technology and in the development of computer aids by chemists for reaction path synthesis leading to desired complex organic molecules. More work is needed for the remaining areas to become industrially significant.

SCOPE

Process Synthesis as a research area is only about a decade old. While several early papers do exist which ask synthesis questions, it was not until the late 1960's, with the work of Rudd and his students, that the question of synthesis was posed as one which was tractable using systematic approaches. Rudd and his students sought to create a computer program called AIDES which could, with limited information, develop almost automatically the structure of a preliminary process flowsheet. They partitioned the problem into, among others, the tasks of reaction path synthesis—the choosing of the correct chemical reactions to use and a step they did not attempt to automate, separation system synthesis, and heat exchanger network synthesis. These subtasks are still among those most commonly considered today.

An example synthesis problem is to select a network of heat exchangers to exchange heat among a given set of process streams, each of which is to be heated or cooled from a source to a target temperature. The goal is to minimize the annual cost of utilities required in the solution plus the annualized cost of the equipment itself. Such a problem allows for an enormous number of alternative solutions. The better solution approaches have recognized that one can predict the minimum utilities required and the probable number of heat exchangers one should have in his/her solution. These approaches thus strive to locate only solutions meeting or approaching these targets.

If one were to extract a general insight from the variety of methods published to date, it is that the only effective approaches to synthesis have taken advantage of every possible special characteristic of the problem being considered. A general purpose algorithm has not been developed. We hope the review will not only help the readers to understand the relevant synthesis literature, but what may be the better approaches to take when developing a synthesis tool for a new problem type.

CONCLUSIONS AND SIGNIFICANCE

Process synthesis is about a decade old and in that decade, over 190 articles and three significant review articles have appeared. The most studied problem by chemical engineers has probably been heat exchanger network synthesis with over 39 articles written on this topic alone. One is already finding testimonial articles by engineers solving industrial problems (for example, Linnhoff and Turner, 1980), indicating that the results in this area do indeed lead to significant savings in *both* operating and capital costs. Thus, one can suggest at least this synthesis topic has matured. Energy conservation system synthesis is not a fully solved problem, however, as most existing efforts have not attempted to consider simultaneous recovery and use of both heat and *mechanical* energy, except for Menzies and Johnson (1971) and Grossmann and Santibanez (1979).

A second area receiving considerable interest is reaction path synthesis, particularly for the discovery of the better alternative reaction sequences to produce a given large organic molecule. Rather impressive interactive computer programs now exist for aiding in this task, but it is far from solved because of the enormous difficulty in "rating" a reaction which has never been experimentally studied.

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Separation system synthesis is a third area which has led to useful insights, but here much work remains to be done. Most of the existing works have dealt with rather well-behaving multicomponent mixtures. The most useful heuristics generated refer to such systems. Work is needed towards a better understanding of nonideal multicomponent mixtures. Computational work should be undertaken to analyze several special cases, so that indicative trends are established. Analytical and thermodynamic work will be needed to develop useful heuristics.

Further research is needed on the synthesis of distillation sequences with heat integration. Due to the size of the problem, effective heuristics are badly needed. The algorithmic branch and bound strategy offers optimistic expectations. The experience gained with a multitude of strategies should be extended to tackling this problem. Most of the reported work has been limited to the traditional utilization of the separators and especially distillation columns. Not much attention has been focused on a very large variety of distillation configurations (columns with side streams, the presence of a prefractionator, etc.).

There is substantial evidence that many of the more "exotic" distillation arrangements offers significant economic potential. More work should be directed towards these possibilities. Furthermore, the nonsharp (sloppy) separations only recently (Motard in Motard and Westerberg, 1978) were brought to the center of active interest. There is almost a complete lack of progress on such separations; intensive work should be undertaken.

Control system synthesis is well along to having its problem defined and to having at least one working model as to how to approach the solution of such problems. More work is clearly needed, however, before industry decides to use these ideas. The synthesis of total flowsheets is still at a very early stage in its development, with existing programs as yet only being able to "play" an elementary but interesting game of design.

INTRODUCTION

Process system synthesis is an act of determining the optimal interconnection of processing units as well as the optimal type and design of the units within a process system. The interconnection of processing units is called the structure of the process system. When the performance of the system is specified, the structure of the system and the performance of the processing units are not determined uniquely. The task is to select a particular system out of the large number of alternatives which meet the specified performance.

The synthesis of systems might be carried out through the following step-by-step decision process.

1. Define objectives of the system.
2. Define criteria for evaluating the system.
3. Select the technology for attaining the objectives.
4. Decompose the problem into a set of interconnected tasks.
5. Realize the tasks within the selected technology.

The first three steps might be termed "process planning" and the last two "process design" (Ichikawa, 1971). This sequence of decision-steps may be referred to as a *hierarchy of decision layers in system synthesis*. A concept of hierarchy appears in the context of a complex decision-making process, which is an extremely common but profoundly important feature of almost any real life decision-making: "Until the higher-level decisions are made, one can never make the lower-level decisions. However, one cannot make the optimal decisions at the higher levels without knowing the optimal lower-level decisions." This fact results in the fundamental dilemma of decision-making in system synthesis. We refer to the higher level decisions as *supremal decisions* and that of the lower levels as *infimal decisions*. In process system design, the supremal decision is viewed as the determination of the structure of the system in terms of subsystem selection; the infimal decision, the realization of the functions required for each subsystem by the supremal decisions.

It is somewhat surprising to find that little attention had been given to developing process synthesis theory before the pioneering work (Rudd and Watson, 1968; Rudd, 1969). This is perhaps due to the combinatorial nature of process synthesis problems, which has been well documented in previous review articles (Hendry et al., 1973; Ichikawa, 1972; Hlavacek (1978); Westerberg, 1979). We can illustrate the problem by considering the synthesis of separation sequences.

For a mixture of N components to be separated into N pure component products, using M separation methods, the number of possible separation sequences is given by (Thompson and King, 1972):

$$R = \frac{[2(N-1)]!}{N!(N-1)!} M^{N-1} \tag{1}$$

Each separator must split its feed mixture into two product streams; each component in the feed must exit in only one of these streams. A consequence is that Eq. 1 is restricted to sequences where any mass-separating agent (like the extractive agent used in the extractive distillation) is recovered for recycle in a separator following the separator into which it was introduced. Any approach based on the complete enumeration and evaluation of all the alternatives is clearly an overwhelming computational task even for reasonable low values of N and M (Table 1). This "combinatorial problem" is very hard to tackle. Thus, major attention must be focused on finding efficient means to avoid the combinatorial nature of the system synthesis problem.

SOLUTION TO PROCESS SYNTHESIS PROBLEM

Three important problems exist in process synthesis (Motard and Westerberg, 1978):

Representation Problem. Can a representation be developed rich enough to allow all alternatives to be included and clever enough to exclude automatically ridiculous options? Can it also be clever enough to aid one directly in solving the synthesis problem?

Evaluation Problem. Can the alternatives be evaluated effectively so they may be compared? Effective evaluation means a balance between speed and accuracy.

Strategy Problem. Can a strategy be developed to locate quickly the better alternatives without totally enumerating all options?

None of these problems is easy to solve.

TABLE 1. NUMBER OF SIMPLE, SHARP SEPARATION SEQUENCES FOR EXAMPLE N AND M VALUES

Number of Components N	Number of Methods M	Number of Sequences
3	1	2
3	2	8
5	1	14
5	3	1134
9	5	558, 593, 750

In the process synthesis literature, two types of approach exist: (1) begin with and seek to improve a feasible flowsheet; (2) no flowsheet exists, thus find a good candidate. Most articles deal with only one of these approaches, but lately some articles have combined them.

There are two basic methods for class 1: evolutionary methods and structural parameter methods.

The idea of an evolutionary design has been central in the modus operandi of process engineers for the improvement of chemical process systems. It consists of making a sequence of design modifications to a previously synthesized process, leading to an improved design. Since processes synthesized by evolutionary methods depend heavily on the initial or starting flowsheets, it is very important for these initial flowsheets to be reasonable which can eventually lead to optimal or satisfactory flowsheets by successive modifications. These initial flowsheets are generated by other more general methods such as heuristic methods which will be discussed later in this section.

The structural parameter approach is to embed all possible alternatives into one integrated "superstructure," in which all possible interconnections among process units are included. The structural parameters, which are essentially split fractions with values between zero and one, indicate the fraction of the flow leaving one unit and going to each of its possible destination units. The superstructure is optimized by means of appropriate nonlinear optimization techniques. A zero-valued structural parameter then indicates that the corresponding interconnection does not exist in the optimal solution.

A typical characteristic of the structural parameter approach is the presence of a large number of constraints associated with structural parameters. As the synthesis problem in chemical engineering involves, in general, a number of nonlinear elements in the subsystem equations and the objective function, it becomes a constrained nonlinear programming problem. The use of structural parameters is relatively simple to set up and apply. Apparent disadvantages in the approach are: it requires a very effective, constrained, nonlinear optimization technique; and the nonlinear complex optimization problem may contain many optima (Westerberg and Shah, 1978).

Methods for class 2 are: breadth- and depth-first methods; bounding methods; heuristic methods; and decomposition methods. Breadth- and depth-first methods are simply tree-search methods. A tree is represented graphically by a collection of nodes interconnected by a set of links. These links establish one and only one path from one node to any other node. Each node represents a particular state for the current problem. If we were to trace the complete tree, we would locate the solution to the problem.

A method frequently used to enumerate completely all the nodes in a tree is a blind-search procedure (Nilsson, 1971). This method offers a systematic path by which all the nodes are explored in an orderly manner. Depending on the order in which the nodes are to be expanded, the blind-search procedures can be further classified into breadth-first and depth-first methods. In the breadth-first method the nodes are expanded in the order in which they are generated, whereas in the depth-first, the most recently generated nodes are expanded first. It is apparent that the blind-search procedure in its pure form is equivalent to a complete enumeration of all problem solutions. As the synthesis problem becomes large, the computation time for the complete enumeration is prohibitive.

To avoid this, tree search methods are usually combined with heuristic or bounding methods. In bounding methods, one determines on each level a bound on the best solution attainable for each node emerging from this level. Many of these bounds will be inferior to the best solution already obtained and the corresponding nodes need not be investigated further. The remaining nodes can be expanded one at a time, until they are found to be inferior to another solution or they prove to be the optimal solution. This method is called the branch and bound method of operations research. The branch and bound method is a solution strategy that has emerged over the last decade as one of the major practical tools for the solution of real life optimization problems. The attractiveness of this method stems from its ability to eliminate implicitly large groups of potential solutions to a problem without explicitly evaluating them.

The success of a branch and bound algorithm hinges to a great extent on the manner in which the bounds are calculated. Since the bound is to be calculated many times, the bound has to be calculated easily. However, one also wants a bound that is reasonably close to the actual value. If it is possible to obtain bounds for the problem that are mathematically rigorous, the branch and bound method offers proof of optimality. However, such mathematical bounds are usually difficult to obtain in the process synthesis problem because of its complex nature.

In order to overcome the need to examine all possible alternative flowsheets before we can identify the optimum or a nearly-optimum one, various plausible but fallible screening rules have been suggested. These rules, usually known as rules-of-thumb, allow the designer to locate quickly one or several acceptable solutions. The heuristic methods are in fact the older methods, and they are based on the experience of the process engineer in designing similar systems and on his insight into the physical and chemical phenomena relating to the various unit operations. The heuristic method, of course, provides no guarantee of optimality.

The decomposition method is the sequential decomposition of the design problem into subproblems which are simpler to solve and which eventually reach the level of available technology. A survey of this method is given by an earlier review paper of Hendry et al. (1973).

Each of these methods has its advantages and disadvantages. To develop effective methods for process synthesis, it is desirable to find a method which combines the best of each of these approaches. Several combinations of methods can be considered and have been developed by previous investigators, for example, heuristic evolutionary, branch and bound heuristic, evolutionary bounding search, depth-first heuristic, etc. The advantages and disadvantages of each method explain why these combinations are taken. Many of the techniques developed during these six years are very sophisticated because of their hybrid use of various methods.

Several classifications of the synthesis literature are possible. The one used here is by the nature of the synthesis problem—reaction path synthesis, heat exchanger network synthesis, separation system synthesis, reactor network synthesis, entire flowsheet synthesis, and control system synthesis. Within each, we shall often subclassify methods as to whether they are for locating a good first candidate solution and/or are seeking to improve an existing solution. We shall also frequently subclassify methods as to whether they are basically heuristic or algorithmic in nature.

Various process synthesis methods are surveyed which have been applied to a number of structured chemical processes. Articles by Hendry et al. (1973) are not discussed here except for comparison purposes.

SYNTHESIS OF REACTION PATHS

For the more efficient utilization of raw materials, intermediate by-products, wastes, etc., the chemical process industries feel more and more the need for discovering and developing novel reaction paths. Significant effort is made by the research and development departments for the synthesis, analysis and evaluation of alternative reaction paths, which are considered as potential processing systems for the production of desired products.

Considerable advances have been made mainly by organic chemists towards the systematic and automated generation of feasible chemical routes for given molecules. Several computer-aided systems are already in operation. For chemical engineers very little has been accomplished during the last 5-6

years in reaction path synthesis even though it has gained a prominent role in the problem of process synthesis. The reason is simple: the high dimensionality of the resulting problem and the extreme difficulty in developing a means to compare the value of the alternatives. It should be emphasized that for the development of an efficient process, the generation of the most efficient reaction path is not enough. Problems of separations, energy requirements, capital investment, cost of raw materials, etc., weigh very heavily in the final selection.

This section deals with the methods that organic chemists and recently chemical engineers have developed for the automated generation of organic chemical reaction paths. This discussion has drawn substantial material from the Ph.D. work of Agnihotri (1978) and a personal communication with Motard (1979). Possible future growth of interest for a process designer is discussed also.

The literature on the synthesis of reaction paths is very extensive. The reader is referred to the following works which include several articles and an extensive analysis of the subject:

- *Computer-Assisted Organic Synthesis*, W. T. Wipke and W. J. Howe, eds., ACS Symposium Series, Vol. 61, 1977.
- *Computer Representation and Manipulation of Chemical Information*, W. T. Wipke, S. R. Heller, R. J. Feldmann, and E. Hyde, eds.; John Wiley and Sons, 1974.
- "Computer Assisted Design of Organic Synthesis," Report of Table Ronde Roussel UCLAF, Maison de la Chimie, Paris, 1977.
- *Topics in Current Chemistry*, J. B. Hendrickson, "Approach to Reaction Path Synthesis," Vol. 62, 1976.

In attempting to review the previous work on the subject, one is immediately confronted with the magnitude of the task and the diversity of features under which to classify them. Two main ingredients characterize the various methods, especially the integrated ones: representation of the chemical molecules and the strategy of the synthesis of the alternative paths. As several works have dealt with only one of the two ingredients, we have decided to concentrate primarily on the methods which constitute complete methodologies, leaving the rest for a future undertaking.

The existing approaches for the synthesis of reaction paths can be divided into two categories (Agnihotri and Motard, 1979):

Logic Centered
—Ugi's MATCHEM-EROS
—Hendrickson's Method
Direct Associative
—Corey's LHASA
—Govind, Powers' REACT
—Wipke's SECS
—Gelerntner's SYNCHEM

The logic-centered methods generate sets of intermediates which can be converted to the target molecule. Iteration of this procedure for all generated intermediates develops a synthetic tree. The direct associative methods recognize the fact that a number of structural subunits are available and, if they are brought together using some standard reactions, the target molecule can be synthesized.

Ugi and his coworkers (Ugi et al., 1971; Ugi et al., 1972; Dugundji and Ugi, 1973; Blair et al., 1974; Gasteiger et al., 1974; Brandt et al., 1977) have developed a very simple representation for the chemical molecules. It is based on the concept of isomerism for simple molecules and for sets of molecules. It allows the generation of a rigorous mathematical model for the underlying chemical foundation of the various reactions.

The chemical molecules are described by three parameters: (a) the identity of every atom in the molecule; (b) the covalent bonds between atoms; and (c) the free electrons for bonding at the outermost orbit of each atom. A set of atoms forming a molecule is represented by the BE-matrix (Bond-Electron connection matrix), Figure 1. A set of molecules are represented by

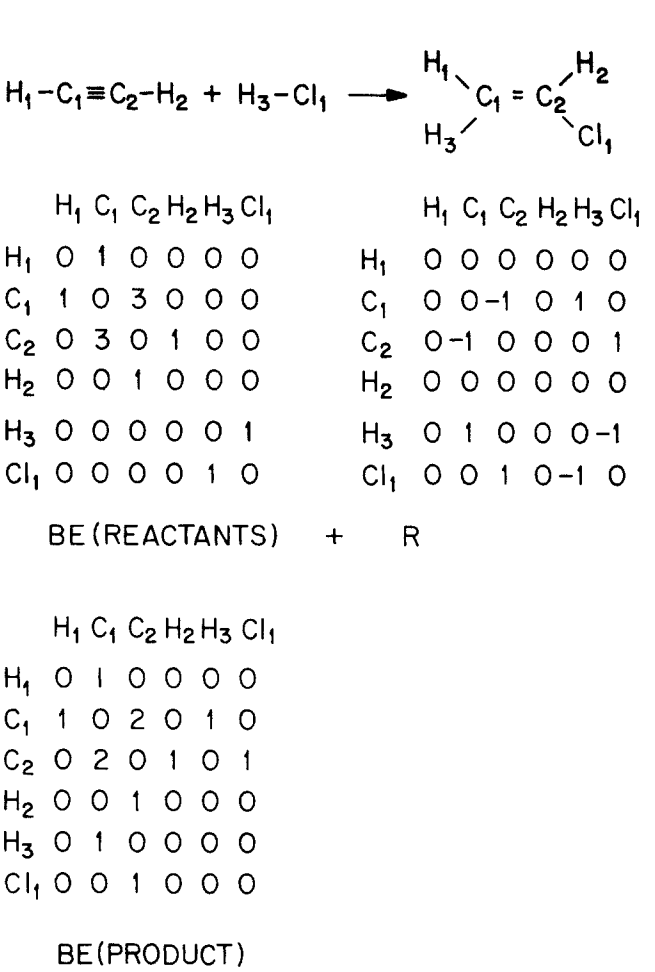


Figure 1. BE and R matrices for HC ≡ CH + HCl → $H_2CCl = CH_2$.

a block diagonal matrix, where each submatrix along the diagonal constitutes the representation of a set of atoms for the particular molecule. Such a matrix is called an EM (ensemble of molecules) matrix. If the molecules of an EM-matrix do not react, they retain their BE-matrices. If on the other hand they undergo reaction, the BE-submatrices change to denote their new electron-bonding constitution (making and breaking bonds). Consequently, when a reaction takes place, the EM-matrix is transformed by a linear operator R (reaction matrix) which denotes how the molecules react with each other. By changing the form of the operator R for the same EM-matrix, we can generate all alternative reaction schemes involving the same initial molecules. This rigorous mathematical formulation of the molecule constitution and of the reaction pattern allows significant analysis, and many useful results have been derived.

It should be noted that the methodology proposed by Ugi and his coworkers can be very easily extended to systems which involve dissociated species, ions and other molecular fragments.

Hendrickson's work (1971, 1975a,b, 1976) presents a constructive representation of the chemical molecules and offers a systematic development of new reactions. For the molecule representation, Hendrickson uses as parameters the type of atoms that the molecule contains and the topological structure of the bonds among the atoms of the molecule. Particular emphasis has been placed on the types of carbon atoms in a molecule according to the types of bonds that the carbon forms with other elements and the nature of these elements. Four types of carbon sites have been identified by Hendrickson.

During a reaction, a molecule undergoes transition from one state (initial, reactant) to another state (final, product). Hendrickson has identified sixteen operators which operate on a carbon site and six operating on two carbon sites. These operators are rationally necessitated by the transformation of one organic molecule to another. Such operators, introduced

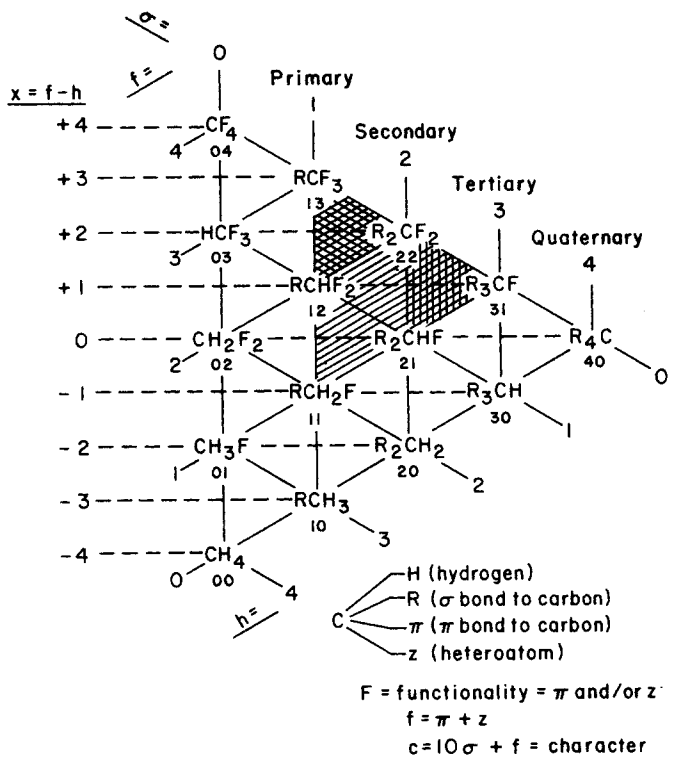


Figure 2. Character triangle for carbon sites and interconversions. The character C, is shown beneath each carbon site. The shaded areas indicate possible double bond sites (C=C). ▨; possible triple bond sites (C≡C), ▩; and possible aromatic sites ▥.

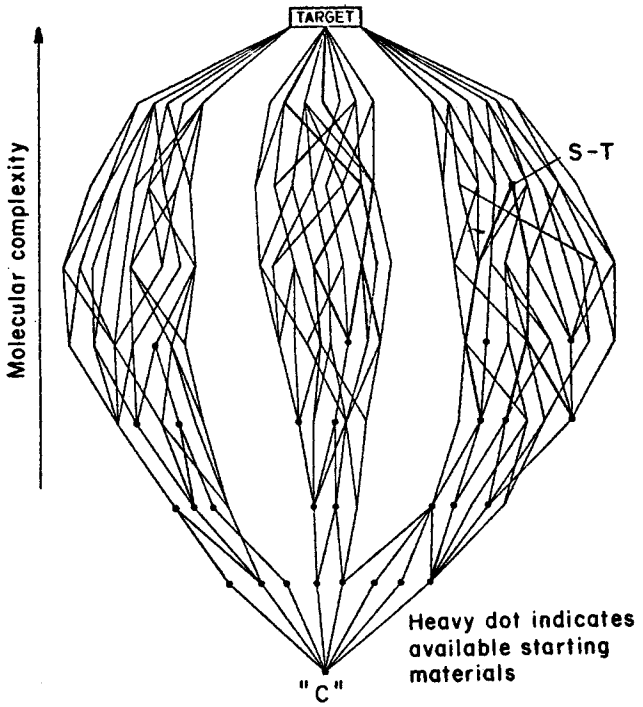


Figure 3. The synthesis tree.

initially in the area of artificial intelligence, are used in many similar problems, where the objective is to reconcile differences between various states.

A single carbon site can be transformed in many different ways (e.g., in terms of the type of bond with another carbon, in terms of the atom that it is bonded to, etc.). Each transformation necessitates a special operator. Hendrickson has developed the "character triangle" (Figure 2) that identifies all possible states for a single carbon site and consequently allows the enumeration of all possible transformations. Using this triangle, it is a straightforward task to develop all possible reaction paths leading to any given carbon site, starting from any other site. Hendrickson's character triangle reveals very easily the enormous combinatorial problem that someone is faced with during the synthesis of reaction paths. In order to develop a single functionality at a single carbon site, Hendrickson found 16 single step transformations and 56 two-step transformations.

In Figure 3, we have presented the logical evolution of various molecules undergoing reaction towards the target molecule. One could start from the most elemental species C (carbon) and proceed forward in a "synthetic" manner to produce alternative routes of chemical transformation leading to the target molecule. On the other hand, one could start from the target molecule and through a series of chemical transformations decompose it by functionality or by skeleton until available starting material (molecules) are encountered. This strategy being opposite to the synthetic is known as "retrosynthetic." Both procedures have advantages and disadvantages that Hendrickson has discussed (1976). He has favored the retrosynthetic approach and the decomposition of the molecular pattern by skeleton. For the selection of the specific bonds which will be tackled in the decomposition by skeleton, he has proposed seven heuristic rules. These rules allow a reduction of the search space and develop meaningful reaction paths.

The LHASA (Logic and Heuristics Applied to Synthetic Analysis) program was developed by Corey and his coworkers (Corey et al., 1969; Corey, 1971; Corey et al., 1972a,b,c; Corey and Peterson, 1972; Corey et al., 1974; Corey et al., 1975; Corey et al., 1976; Pensak et al., 1977). It is an interactive program allowing continuous communication between man and computer. It follows the retrosynthetic approach, using available chemical transformations, generates all the precursor states for the target molecule or any other intermediate state. Each precursor generated by a chemical reaction is used as the current target molecule to generate the next level of precursor molecules. The chemical transformations, used to develop the precursor molecules of the current target molecule, have been defined *a priori* and constitute a set of approximately 600 chemical reactions.

For the representation of the molecules, the LHASA program includes a logic that determines first what atoms or bonds possess a particular property. Such simple information can be combined to recognize various functional groups which characterize the nature of the carbon atoms.

The LHASA program is heuristic in its strategy while the representation of the molecules is rather complex and esoteric. Its most attractive feature is the man-computer interaction and the fact that it can systematize the synthesis of quite complex molecules in an automatic manner. On the other hand, the usage of specific chemical reactions does not allow for the generation of innovative alternative reaction paths.

The SYNCHEM program was developed by Gelernter and his coworkers (Gelernter et al., 1971; 1973), and it bears close similarity to the LHASA program. It is basically heuristic in its nature, using techniques developed within the area of artificial intelligence for the efficient screening of the alternative reaction paths.

The SECS (Simulation and Evaluation of Chemical Synthesis) program developed by Wipke and his coworkers (Wipke et al., 1974; 1977) has borrowed its basic structure and philosophy from the LHASA program. It is also a man-computer interactive system with graphic capabilities. It uses an *a priori* prescribed set of chemical transformations. The synthesis strategies employed follow the retrosynthetic approach, and they select the transform either to fit the target molecule or to generate a desired precursor molecule. The second strategy is directed towards available starting raw materials.

The REACT program developed by Powers and Govind (1977b) represents the first serious effort to automate the gener-

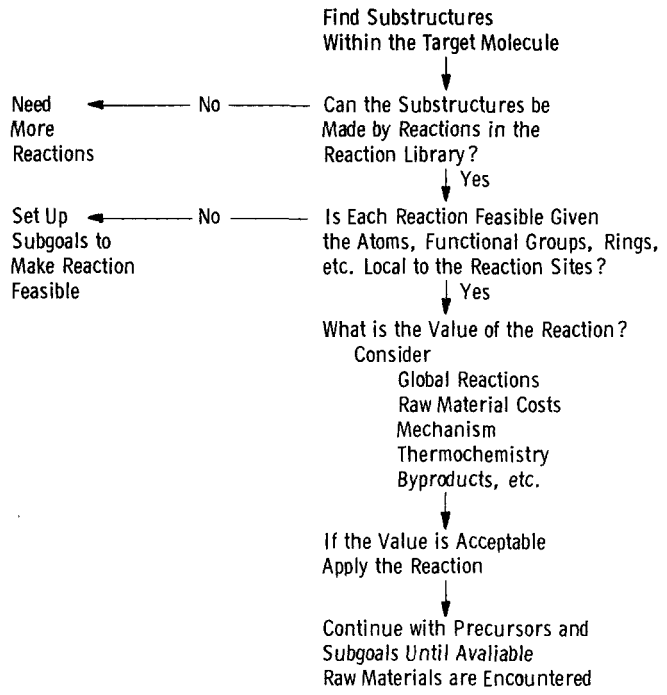


Figure 4. Steps in applying a transform based on the substructures which change during reaction.

ation of chemical reaction paths for the petrochemical industry. To represent a molecule, it uses a connection table where the type atoms (or atom group type) and the bond type have been coded.

The strategy employed by the REACT program is basically retrosynthetic. Starting with the target molecule the program identifies in it various functional groups. Attempting to match the functional substructures identified in the target molecule with the substructures available from the encoded chemical transformations, various alternative routes are identified. Redundancies are checked and eliminated. Two subsequent levels of checking identify and reject infeasible alternatives. The generated sequences of precursors are then assigned a heuristic score of desirability according to the size of the resulting precursor skeleton and the number of functionalities present in the precursor. This heuristic weighting reduces significantly the search space. The procedure ends when available starting materials are encountered as precursors. The logic of the strategy is shown in Figure 4.

The REACT program has scaled-down objectives when compared with more advanced synthesis systems but it serves also different types of reaction synthesis problems, i.e., it directs its search towards the synthesis of rather smaller and simpler molecules from similar precursor molecules as against the large and very complex molecules that the organic chemists attempt to synthesize.

The latest development on the synthesis of reaction paths is credited to Agnihotri (1978), resulting in the generation of the CHIRP program. This program uses the Ugi representation discussed earlier. The question which is addressed here is: "What products are formed if one mixes certain molecules at a certain temperature and pressure and what reaction paths produced them?"

Initially, the user specifies the starting chemicals and the allowable reactions to be performed, selecting among a group of 10 general reaction schemes. The reactions selected determine the bonds to be broken and the bonds to be made, as well as the atoms to be affected. The strategy is synthetic in nature, and for every permutation in the R operator a new reaction is generated. The reaction proceeds so long as new product sets are generated. A thermodynamic screening though limits the alternatives to the "n most thermodynamically feasible steps," where n is variable and selected by the designer according to the size of the search space he intends to generate. The n most thermodynamically feasible steps correspond to the n steps with minimum Gibbs free energy. The Gibbs free energy for a particular molecule is computed after the molecule has been identified from a table of 400 entries currently available in the program.

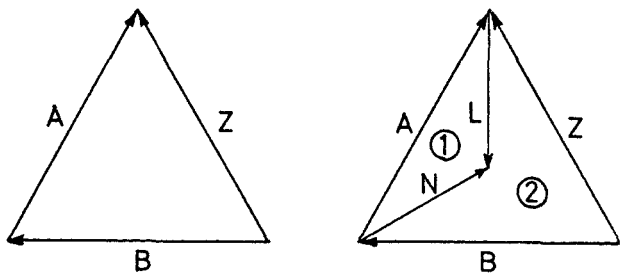
The CHIRP program is basically heuristic in its nature and the first one that attempts a preliminary screening of the alternative reaction steps in terms of their effects on the anticipated chemical process (through the effect of the Gibbs free energy). The program is under continuing expansion and development.

A final comment is in order. As Agnihotri and Motard (1979) have observed: "The field of reaction path synthesis still holds a great deal of promise. In particular, familiarization by chemical engineers is an opportunity to interact with synthetic chemists and to close the gap between the chemical laboratory and the process engineering division. The result of this dialogue should be the orientation of chemical research along paths that hold the promise of cost-effective process development."

All the previous methods attempt to develop systematic strategies for the synthesis of open-reaction paths. A number of reactions (among which the Sovay soda ash process occupies historically a central position) are of a closed-cycle type and they use "clusters of intermediate chemicals and chemical reactions to bypass an important but unwilling chemical reaction" (May and Rudd, 1976).

The synthesis of such clusters of chemical reactions was undertaken by Rudd and May (May and Rudd, 1976; Rudd, 1976) and represents a typical and characteristic example of the problems that a chemical engineer faces during the synthesis of reaction paths. The clusters of reactions are represented by polygons and nested polygons, with the edges (A, B, Z, N, L in Figure 5) denoting chemicals (reactants or products) and the closed loop of edges representing a reaction. Figure 5(b) shows two reactions that constitute a cluster. This cluster was formed by nesting polygons inside the polygon of the overall reaction, Figure 5(b). This procedure can be repeated several times producing a large class of feasible clusters of reactions.

Once simple geometric constructions for the synthesis of the alternative clusters have been generated, a thermodynamic analysis follows to screen the most feasible and optimistic among them. A cluster of reactions is acceptable if no reaction in the cluster has a change in the Gibbs free energy larger than 10 kcal/g-mole. This requirement along with the concept of the common difference allows the construction of free-energy vs. reaction conditions (temperature) diagrams that aid the screen-



(1) A + B = Z

(a)

(1) A + L = N

(2) N + B = Z + L

(b)

Figure 5. Polygon representation of reactions.

ing of alternatives. Consider the cluster shown in Figure 5(b). From reaction (1) $N - L = A$ and from reaction (2) $N - L = Z - B + A$ (common difference. Then if $\Delta G_1 = G_N - (G_A + G_L) \leq 10$ kcal/g-mole and $\Delta G_2 = (G_L + G_N) \leq 10$ kcal/g-mole, the cluster is feasible. Figure 6 depicts the feasible operating regions that satisfy the above conditions, between T_L and T_1 and between T_2 and T_H. The region of T_1 to T_2 is infeasible.

The authors have expanded the work to include cases that: (1) include unrelated compounds through the introduction of an artificial common difference; and (2) allow combination of two reaction sets, each with distinct common difference, by allowing multiple common differences. The approach is very fast and efficient. It synthesizes rapidly a large number of feasible Solvay clusters of reactions for industrially important problems.

PROGNOSTICATIONS FOR REACTION PATH SYNTHESIS

The future belongs to the Logic Centered methods. The Direct Associative (Information Centered) were early developments in the evolution of the subject and offer limited opportunities within the scope of "known" chemistry. In contrast, the Logic Centered methods are rigorous, mathematically based search strategies, which can develop exhaustively all alternative reaction paths. Despite the obvious obsoleteness of many of them (something that is not encountered in the economizing nature of the Information Centered methodologies), it provides the opportunity for the creation of really novel reaction paths. The presently available computer facilities can handle satisfactorily the most demanding of the Logic Centered methods.

For chemical engineers, the efficient incorporation of the process aspects in selecting the most promising reaction routes, is the most pressing challenge. Although, simple thermodynamic criteria, like the heat of reaction, the change of free energy, the required temperature and pressure, can provide some initial screening of the least promising alternatives, still, aspects related to the economics of processing systems (fixed capital, operating cost) or their operability are not quantifiable in a simple manner, neither can they be described by the above thermodynamic measures. More work in this direction should be focused on the development of "working" heuristics, which will incorporate the processing aspects into the problem. It is the opinion of these authors that any general development, that accounts efficiently for all the processing aspects during the screening of reaction paths, will not come in the immediate future.

Another direction that may prove worthwhile to pursue is, a more in-depth analysis of the topological characteristics of the reaction paths. A recent work by Rotstein, Stephanopoulos and Resasco (1980) has revealed several properties possessed by alternative reaction paths in the ΔG (free energy change) vs. T space. For example, let us consider the four chemicals A, B, C and D which participate as raw materials or by-products in a reaction path that produces one mole of the desired product E. The following properties have been proved:

- The lines ΔG vs. T which correspond to various stoichiometries for A, B, C and D, have a common point (ΔG^+, T^+) characterized only by the nature of the chemicals.
- Substitution of the product chemical, E, by a different one, E', results in a vertical shift of the point (ΔG^+, T^+), at constant T^+, for any E'.
- Substitution of the chemical, A, by any other chemical results in a new (ΔG^+, T^+) point which is on the same straight line, characterized by the other chemicals. Similar geometric loci can be developed if the chemicals B, C or D are substituted by any other chemical.
- The above properties are invariant to any permutations in the order of the chemicals.

Such properties allow us to "move" in a prespecified, desired region of the (ΔG, T) space by making specific changes in the pattern of the reaction path. Thus, the development of desired reaction paths can follow a systematic procedure and become free of exhaustive, large-scale computations.

The above procedure has been extended recently to cover the cluster reaction schemes (May and Rudd, 1976) leading to an algebraic search and screening procedure.

It is believed that more properties can be developed by expanding the space of concern to include the heats of reaction. Particularly helpful for future work is expected to be the methodology of Ugi and his coworkers (1972) due to the vigorous mathematical framework they provide for such analysis.

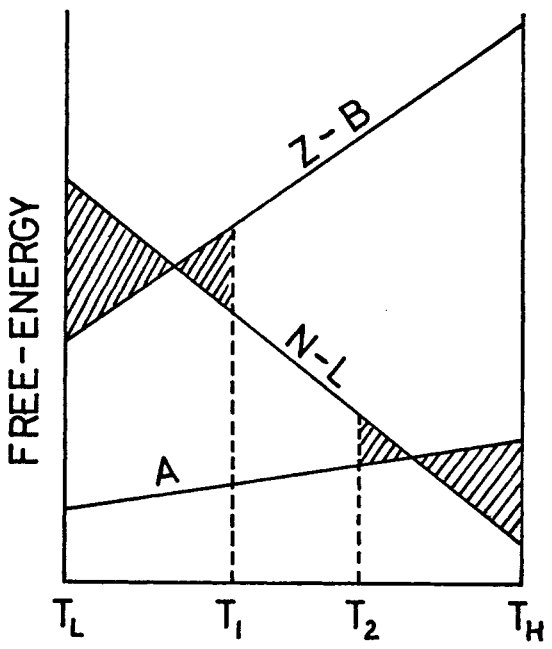


Figure 6. The ladder pattern to identity. Solvay clusters that satisfy the maximum free energy criterion (May and Rudd, 1976).

SYNTHESIS OF SEPARATION SEQUENCES

Chemical processes frequently include multicomponent separation sequences which produce more than two product streams. As Hlavacek (1978) has pointed out, such separation systems arise because there is a need for: feed preparation, product separation, product finishing, and waste treatment. Because separation processes are a significant portion of the total capital investment and annual operating cost for a plant, a great deal of interest has been generated for the development of systematic and rational methods which will produce "optimum" separation schemes. In terms of its potential for industrial implementation, the synthesis of separation schemes ranks second only to the synthesis of networks of heat exchangers.

The problem can be defined as (Nath and Motard, 1978): given a feed stream of known conditions (i.e., composition, flow rate, temperature, pressure), synthesize systematically a process that can isolate the desired (specified) products from the feed at minimum venture cost. Thus,

$$\underset{I,X}{\text{Minimize}}\ \Phi = \sum_{i} C_i(x_i) \qquad (2)$$

where $i \epsilon I$ denotes a feasible separation unit; C_i, the total annual venture cost of separator i; I, a subset of S; S, the set of all possible separator configurations that can produce the desired products; x_i, the design variables of separator i, and X = union $(x_i)^n$.

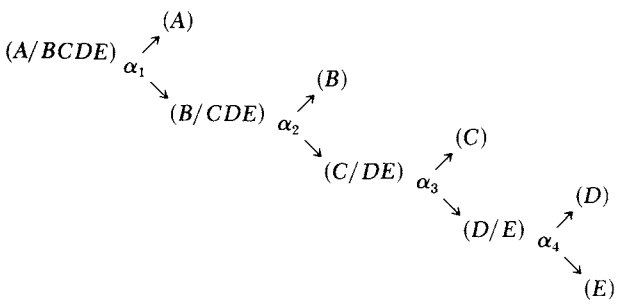
The above problem is a mixed-integer, nonlinear mathematical programming problem, where the discrete decision is made

over all subsets I that can be generated from S and the continuous decision is made over the continuous range of values of the design variables x_i. Thus, the designer is confronted with two questions: 1) find the optimum sequence of separations and the nature of each separator, and 2) find the optimum values for the design variables (sizes, operating conditions) of each separator.

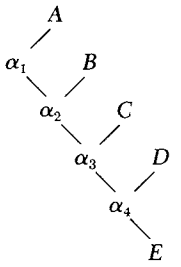
Most of the existing publications deal with synthesis of separation sequences involving what we shall label "simple sharp" separator units. A simple separator unit is one which splits a single feed stream into two product streams. A sharp separator unit is one where each entering component exits in only one product stream. An ordinary two product distillation column which operates with very high recovery of adjacent light and heavy key components is an example of a simple sharp separation unit. Most of this section will deal with such separators only.

Let us discuss now the general representation of separation sequences based on simple, sharp separation units and define various terms that we will be using thereafter. Consider a mixture of components A, B, C, D, and E. The "ranked list" (Hendry and Hughes, 1972) of these components with respect to a given separation method is a list of the components in decreasing order with respect to the value of the physical property exploited by the separation method (e.g., boiling point for distillation, solubility for extraction, size of solid particles for screening). The ranked lists provide an easy way to represent a mixture and help to identify all the possible sharp separations between *adjacent* key components.

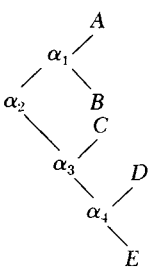
The basic structural skeleton of a separation sequence can be represented by a binary tree, which is composed of "operators" (separators) and "operands" (intermediate or final product streams). For example the following separation sequence:



can be represented by the binary tree:



The operator α_1 determines the split between the first two key components A/B using separation method α. Similarly, α_2 determines the split B/C, α_3 the split C/D, etc. Consequently, if we want to perform the split B/C before the A/B, we can reverse the relative order of the operators α_1, and α_2 on the binary tree and write:



Over the last 30 years, various methods and approaches have been proposed, with the majority of them developed during the last 10 years. All these methods can be divided into three categories: (a) heuristic methods which use rules-of-thumb resulting from the long engineering experience and the insights in the physics and chemistry of the separation methods; (b) evolutionary strategies which attempt to identify the best separation system through a sequence of evolutionary improvements; (c) algorithmic techniques which employ various algorithms developed in the area of nonlinear mathematical programming. Each class of methods has its relative advantages and disadvantages that we will discuss in detail in the following paragraphs.

Heuristic Methods. Within this category of synthesis methods we can distinguish two subclasses: those which deal only with the synthesis of distillation sequences; and those which allow, in addition to distillation, for extractive distillation, extraction, crystallization, absorption and other separation methods.

Heuristic Synthesis of Distillation Sequences. The earliest attempt to synthesize optimum distillation sequences is credited to Lockhart (1947). He examined the three-product distillation sequences encountered in the processing of natural gasoline.

The total heating requirement was considered by Harbert (1957) as the single most important factor in deciding the optimum arrangement of distillation columns, because it dominated the economic evaluation. In a variation to Harbert's approach, Rod and Marek (1959) identified the total vapor flow in the distillation sequence as the dominant economic factor.

Heaven (1969), in an extensive computational investigation with detailed sizing and costing of the distillation columns, identified several heuristic rules that he used in his synthesis strategy. A similar heuristic approach with relative weighting of the various heuristics was proposed also by Powers (1971).

Freshwater and Henry (1975) undertook an extensive computational work to analyze the validity and the regions of applicability of the four heuristics suggested by Heaven. Thus, they studied the effect of: (a) composition of the feed, (b) volatility of components, and (c) degree of recovery of the feed components on the total annual cost of every possible distillation sequence for the separation of three, four and five component feeds. From their computational experience they made the following observations:

(1) Considerable variation occurs in the cost differences among all possible distillation sequences for a given separation problem. This remark justifies *a posteriori* all the work undertaken and described in this section.

(2) Smaller variations (in the cost differences of alternative sequences) are in most cases restricted to feeds in which the least volatile components are predominant, while large cost differences occurred in the reverse situation.

(3) The absolute value of the cost of all distillation sequences for a given separation exhibits considerable variation with the composition of the feed.

A very striking observation was made when they noticed that in many instances the magnitude of the variations in total cost over all possible configurations was not greater than 10%. This feature was particularly evident where the highest recovery levels were required and in many feeds in which the least volatile components were predominant. According to Hlavacek (1978), similar trends have been observed by Kafarov et al. (1975). Additional observations from the numerical results include the following:

(4) If there are no difficult separations, the components should be removed in decreasing order of volatility one by one as overhead products (direct sequence), except in the cases where one component is predominant; in that case, this component should be removed first.

(5) Leaving the most difficult separation last was found in several cases to be the dominant factor.

Doukas and Luyben (1978) undertook a computational study of the alternative distillation configurations, similar to that of Freshwater and Henry. They studied four different configurations for the separation of a ternary mixture into three products. In addition to the classical direct and indirect sequences, they also included the following two configurations: a single column with side stream product and a prefractionator column followed by a sidestream column (i.e., column with three product streams). Several values of the relative volatilities and the feed compositions were explored. Their studies did not cover the wide spectrum of all possibilities but they pointed to the following aspects: (a) The single side stream column is the most economical when the concentration of the most volatile component is very low (less than 10%). As its concentration increases above 10%, it becomes rapidly uneconomical. (b) The effect of the changes in relative volatilities were very pronounced, while (c) the heating and cooling costs seem to dominate the decision making.

All the above heuristic methods have provided a very simple basis for an easy and quick screening of all the distillation sequences. The main drawback is the fact that we either have one predominant heuristic rule which can lead to serious miscalculations, or a set of conflicting heuristics which must be weighed against each other in a very subjective manner. Consequently, a trend has developed for the last 5-7 years that tends not to develop the search framework based on heuristics but to imbed them in an evolutionary or algorithmic strategy as we will discuss in subsequent paragraphs. In such strategies only heuristics of very wide applicability are used to reduce the set of alternative distillation sequences, thus reducing significantly the risk of serious failure, and in a manner that these heuristics are challenged continuously.

Heuristic Synthesis of General Separation Sequences. In this section we will examine the synthesis methods which allow the usage of more than one separation technique, i.e. distillation, extractive distillation, absorption, extraction, crystallization, etc. This is the general problem that one encounters during the design of a chemical plant, and at its discrete level requires the answer to two questions: (i) what separation method will be used (selection problem); and (ii) what is the sequence of the splits to be performed (sequencing problem).

The earlier methods developed to answer the above two questions of selection and sequencing were heuristic in nature. The first attempt should be credited to Rudd and his coworkers (Siirola and Rudd, 1971; Powers, 1971, 1972; Siirola, Powers and Rudd, 1971; Rudd, Powers and Siirola, 1973). They developed an heuristic approach for the synthesis of multicomponent separation sequences, as a part of a general strategy that synthesized alternative process flowsheets in an adaptive manner. This program, known as the AIDES system, employed heuristics at two distinct levels; in the first to select the separation method and in the second to decide on the best sequencing.

Thompson and King (1972a,b), about the same time, developed an alternative heuristic strategy for the synthesis of general separation sequences. The major drawback of the method by Thompson and King is the fact that the values of the heuristic parameters are independent of the pressure or absence of nonkey components in the separator. The approach though is easily implementable and has been demonstrated to lead to very good separation sequences. Being heuristic, it is "liable to complete failure when confronted with a sufficiently complex and unusual situation" (Hendry et al., 1973).

Mahalec and Motard (1977a,b) have proposed the so-called BALTAZAR procedure for the synthesis of entire chemical processes, whose detailed discussion will be presented in a later section. In the BALTAZAR procedure, alternative separation sequences are developed in an effort to bridge the discrepancy between the multicomponent streams encountered at certain parts of the flowsheet being developed and the products streams required by the design specifications. Mahalec (1976) suggested the following function for the heuristic evaluation of the separation sequences involved in the design of total initial process flowsheets:

$$GS = N_S \sum_{J\{S\}} N_J \sum_{I^J} (S_I - 1)M_I \qquad (3)$$

where:

$\{S\}$ = set of separation sequences
N_S = number of separation sequences
N_J = number of separations in sequence J
S_I = number of outlet streams in separation I
M_I = mass load on separation I

Minimizing the function GS, the separation sequences with the following characteristics are preferred: (i) separation sequences with the smallest product set; (ii) separation sequences where the components that dominate the feed are separated first. The heuristic evaluation function (3) constitutes a very crude tool for the selection of the optimum separation sequence, but its purpose was to provide an easy and quick way to use measure for the evaluation of alternative total flowsheets (see also Motard and Westerberg, 1978).

Hartmann and his coworker (Hartmann and Hacker, 1979; Hartmann, 1979) have employed an heuristic strategy for the synthesis of optimum separation sequences as a part of the synthesis of total process flowsheets. They have divided the heuristics into two groups. The first group includes rules that determine the structure, i.e., the selection and the sequencing of the splits, while the second group involves heuristics which determine the operating condition of the separators. The group of the structural rules is based on a set of heuristics. The group of parametric rules includes: (i) choose operating conditions which are close to the ambient; (ii) use as reflux 1.2 times the minimum reflux; (iii) choose the reflux in such a way that the maximum realizable number of trays is required. None of the employed heuristics is new but all have been suggested and used before by other workers. Hartmann and his coworkers have assigned subjective weights to the above heuristics and their goal is to synthesize the separation sequence with the maximum sum of weights. There are no guidelines given on how to weigh the various heuristics, and it is left to the designer to decide on an ad hoc basis for each problem. Their approach has been used effectively on an 18-component mixture with 10 desired product streams.

The generation of appropriate simple and effective heuristics for the synthesis of optimal, or nearly optimal separation sequences, has provided the first breakthrough towards the solution of the problem, but it is a limited and restrictive approach for future growth and development. Heuristics have provided the incentive for a closer examination of the separations' problems and motivated significant analytic and computational work, and, in several instances, this work has led to more effective approaches for the synthesis of separation sequences like those of Seader and Westerberg (1977) and Nath and Motard (1978). These two works depend heavily on the use of heuristics but they will be discussed in the next section since the central strategy employed is an evolutionary strategy.

EVOLUTIONARY STRATEGIES

The evolutionary synthesis of separation sequences includes the following three subtasks (Stephanopoulos, 1974); generate an initial separation sequence; identify the evolutionary rules; determine the evolutionary strategy. In the following paragraphs we will examine how the various evolutionary methods for the synthesis of separation sequences have treated the above three subtasks.

Development of an Initial Separation Sequence for Design Goals

The generation of the initial sequence has a very profound effect; the better it is, the closer we are to the optimum sequence and therefore the faster we attain it. Many of the heuristic methods discussed in the previous section could be viewed as techniques to generate the initial separation sequence in an

evolutionary scheme. Other methods which are algorithmic in nature could be used for the synthesis of the initial sequence, as discussed later. In case that a similar separation problem has been solved in the past, the available literature and industrial practice could provide a suitable separation sequence to start the evolutionary strategy.

Stephanopoulos and Westerberg (1976) have used as the initial separation sequence the one generated by selecting the next separator as the one with the minimum "sublagrangian." The "sublagrangian" was employed in the design of a basic separation sequence that was used in their branch and bound strategy, as mentioned later. It is a variation of the heuristic "choose the cheapest as the next separator" suggested by Thompson and King (1972a,b).

Seader and Westerberg (1977) have suggested six heuristics to guide the generation of the initial sequence and the evolution to better ones in later stages as follows.

(1) Identify the forbidden splits. As such they have forbidden splits with the relative volatility between the key components less than 1.05, etc.

(2) Easiest separations as characterized by their relative volatilities should be done first.

(3) When the mole percentage of the feed components varies widely but the relative volatilities do not, sequence the splits to remove components in the order of decreasing molar percentages in the feed.

(4) When neither relative volatility nor molar percentage in the feed varies widely, favor the direct sequence, i.e. remove the components one by one as overhead products.

(5) When a msss separating agent is used (e.g. extractive distillation), remove it in the separator immediately following the one into which it is introduced.

(6) When multicomponent products are specified favor the sequence that produces the smallest product set.

These six heuristics are used in the order presented and only if one heuristic is not applicable the following is considered. As the authors caution the reader, the ordering of the heuristics is itself an heuristic. Clearly the above heuristics are not new and they have been tested earlier. Heuristics (1) and (6) have been used by Thompson and King (1972a,b); (2) and (3) have been used by Heaven (1969) for distillation sequences; heuristic (4) has been verified in a way by the computational work of Freshwater and Henry (1975), while heuristic (5) has been used by Hendry and Hughes (1972).

It should be noted, of course, that for all these works the heuristics constitute the only decision-making mechanism, while, for Seader and Westerberg, they constitute the vehicle for developing an initial separation sequence from which one can evolve to better systems.

A similar heuristic approach has been adopted by Nath and Motard (1978) for the creation of the initial separation sequence, considering only distillation and extractive distillation. The heuristic rules employed by the above two authors for the creation of the initial structure are: (a) favor the smallest product set; (b) favor distillation; (c) easiest separations should be done first; (d) a separation method using a mass-separating agent (MSA) cannot be used to isolate another MSA; (e) a separation with a relative volatility between the two key components less than a minimum is not acceptable; (f) operating pressure should be close to ambient; (g) set splits fractions of the key components to prespecified values; (h) set the operating reflux equal to 1.3 times the minimum reflux ratio for each column.

We notice that there is a common set of heuristics used by Seader and Westerberg, Nath and Motard for the generation of initial sequences. Heuristics (a), (b), (c), (d) and (e) provide the guidelines for selecting the separation method and the split sequencing in the initial structure, while heuristics (f), (g) and (h) provide guidelines for the design of the separators of the initial sequence.

Identify Evolutionary Rules

Once an initial separation sequence has been generated, the evolution can begin. Evolutionary rules are needed to generate all the permissible structural changes in a separation sequence. They should possess the following properties.

- Efficiency, i.e., inventing separation sequences which are feasible.
- Completeness, to guarantee by repeated application the generation of all possible sequences.
- Intuitive reasonableness, to generate separation sequences which do not differ significantly from the current sequence undergoing evolution.

The evolutionary rules specified by Stephanopoulos and Westerberg (1976) and modified by Westerberg (Motard and Westerberg (1978)) have all the above properties. First, we characterize the separation task accomplished by a simple sharp separation unit by identifying the separation method it uses (e.g., ordinary distillation, extractive distillation with extractive agent E_2) and by its (adjacent) key components.

Rule 1: Move a separation task one position earlier in the separation sequence.
Rule 2: Change the separation method used by a task.

In each case, the moved or new task will create altered products, and downstream structures for separating these products are constructed to use the same task sequencing as the original structure has. Seader and Westerberg (1977) adopted the unmodified evolutionary rules of Stephanopoulos and Westerberg (1976).

Nath and Motard (1978) have employed a different idea. The evolutionary rules they are using challenge directly the first heuristics that were used for the synthesis of the initial structure. The idea is very interesting since it cuts through the main weakness of any heuristic approach, namely the fallibility of the heuristic rules. The challenge is limited to the first three heuristics since these are the most critical in determining the selection of the separators and the sequencing of the splits.

Challenging the first heuristic is not new. Thompson and King (1972a,b) have incorporated a similar step in their synthesis approach. The rational behind this rule lies on the fact that the smallest product set might force upon the design the performing of a very difficult separation which could be avoided all together if the heuristic was relaxed. Challenging the second heuristic, the logic of using distillation for very close boiling points is challenged, and more freedom is introduced in the decision-making. To challenge the third heuristic is a very reasonable objective in view of Freshwater and Henry's work. Challenging and relaxing the above three heuristics develop a family of alternative separation sequences which are examined and evaluated. In the next paragraph, we will see how these alternatives are rejected or retained.

Determine Evolutionary Strategy

This will determine how the system evolves to better and better designs. Several alternatives lend themselves. Stephanopoulos (1974) has suggested the following four.

- Using the evolutionary rules generate all feasible separation sequences resulting from the current sequence through one modification. Size and cost all the alternatives rigorously and select the cheapest one as the next current sequence. This approach belongs to the general class of the "breadth-first" search techniques.
- A simple variation on the above strategy allows one to employ heuristics for the selection of the next flowsheet among all the alternatives generated from the evolutionary rules.
- Apply the evolutionary rules selectively and repeatedly (i.e., only Rule 1) until an apparent optimum has been encountered; then, apply the other Rule(s) to break the impasse and

generate improved sequences. This is known as "depth-first" strategy.

- The breadth-first strategy could be employed in two successive steps, i.e., by selecting the next separation sequence among all the alternatives that are generated from the current separation sequence through two successive modifications.

Stephanopoulos and Westerberg (1976) have employed the selective breadth-first strategy using the equivalent of Rule 1 until an apparent optimum separation sequence has been encountered. Then they employed a rule similar to Rule 2.

Seader and Westerberg (1977) have adopted a heuristic strategy of the breadth-first type. According to their approach, an alternative separation sequence generated from the current structure using the three evolutionary rules is retained if: (i) it could have been generated by a heuristic if the heuristics ranked above it had been relaxed; (ii) it is almost an equivalent choice using the same heuristic. All the retained separation sequences are ranked according to the heuristic responsible for its retention. Thus, sequences that were retained using the first heuristic are higher than the sequences through the use of the second heuristic. Finally, all the retained flowsheets are evaluated until a better flowsheet is found. In that case, this better sequence becomes the current flowsheet and the evolution starts all over again. The method of Seader and Westerberg uses heuristics without including the high risk of their fallibility. This safety feature is the result of the evolutionary strategy.

The evolutionary strategy of Nath and Motard (1978) is also heuristic in nature. After each evolutionary step where their first three heuristics are successively challenged, a number of alternative separation sequences are generated. To assess the results of challenging the first heuristic, two factors are taken into account. The first one is called Coefficient of Difficulty of Separation (CDS) as it is used as a measure of the relative difficulty for each separation.

Thus, examine and evaluate a generated sequence if the CDS is within 10% of the CDS for the current sequence or if refrigeration is required to condense the reflux. Refrigeration was excluded before due to its high cost. In this evolutionary step this restriction is relaxed.

Challenging the second heuristic allows the introduction of extractive distillation columns in the current separation sequence. The alternatives generated from this and the challenge of the third heuristic are evaluated in a breadth-first screening manner.

The evolutionary strategies constitute very serious tools in the hands of experienced engineers (Hlavacek, 1978) since they allow the repetitive use of general-purpose simulation programs, in a man-computer interactive environment, to improve entire processing systems by sequential modifications. Furthermore, the evolutionary strategies allow a very flexible usage of algorithmic and heuristic techniques.

Table 2 lists a number of references together with a summary of the heuristics used to develop separation sequences.

TABLE 2. HEURISTICS USED IN HEURISTIC AND EVOLUTIONARY STRATEGIES FOR SYNTHESIS OF SEPARATION SEQUENCES

Reference	Type Separators Applied	Heuristics Used
Lockhart (1947)	Distillation	1,6
Harbert (1957)	Distillation	2, 3
Rod & Marek (1959)	Distillation	4
Heaven (1969)	Distillation	1, 2, 3, 5
Rudd and his co-workers (1971-73)	General	1, 2, 3, 6, 8, 12, 13
King (1971) and Thompson and King (1972a, b)	General	1, 2, 3, 7, 11
Stephanopoulos (1974), and Stephanopoulos and Westerberg (1976)	General	7 plus evol. rules
Freshwater & Henry (1975)	Distillation	1, 2, 3, 5, 6
Mahalec (1976) and Mahalec & Motard (1977a, b)	General	6, 11 plus evol. rules
Seader & Westerberg (1977)	General	1, 2, 3, 9, 11, 12, 13, 18 plus evol. rules
Nath & Motard (1978)	General	9, 10, 11, 14, 15, 17, 19 plus evol. rules
Doukas & Luyben (1978)	Distillation	1
Hartmann (1979) and Hartmann and Hacker (1979)	General	1, 2, 3, 6, 8, 16, 19

Heuristic rules as numbered above.
1. Remove components one-by-one as overhead products.
2. Save the most difficult spearation for last.
3. Favor 50-50 splits.
4. Sequence with the minimum total vapor flow.
5. Make high recovery fractions last.
6. Separate the more plentiful components first.
7. Choose the cheapest as the next separator.
8. Remove the thermally unstable and corrosive material early.
10. Perform least-tight separation first.
11. Favor sequences with the smallest product set.
12. Avoid separations using a mass-separating agent (MSA).
13. Remove a MSA from one of the products in another, subsequent separation process.
14. A separation method using a MSA cannot be used to isolate another MSA.
15. Favor distillation.
16. Separate first the components which might undergo undesirable reactions.
17. Set splits fractions of the key components to prespecified values.
18. Avoid extreme processing conditions.
19. Favor ambient operating pressure.

ALGORITHMIC METHODS

In this class, we will consider methods which attempt to synthesize the best separation sequence using well-known optimization methods. All the algorithmic methods are in principle rigorous and infallible, but all at the same time cumbersome and overwhelming in terms of the computational time and effort required. Thus, it is not surprising to find algorithmic methods which make use of general and well-accepted heuristics to reduce the size of the search space. We will also consider various strategies which have been proposed for the sequencing of distillation columns. These methods have been classified as heuristic but they differ significantly from the heuristic methods described. They possess a certain analytic and algorithmic content and they will be presented here under the heading, parametric methods for the structuring of distillation sequences.

Parametric Methods for Sequencing of Distillation Columns

The earlier work of this kind is credited to Petlyuk and his coworkers (Petlyuk et al., 1965; Petlyuk and Platonov, 1964). They undertook an extensive study of the thermodynamic optimality of multicomponent distillation sequences using as the objective the minimization of thermodynamic irreversibilities through a proper selection of the distillation sequence.

Nishimura and Hiraizumi (1971) proposed an evaluation function which takes into account the volume of the distillation column and the heat consumption rate at the reboiler. They formulated the problem of the synthesis of distillation configurations as an optimization problem with optimization variables the structure (configuration) of the system and the system parameters (sizes and operating conditions). Their conclusions supported two earlier heuristic rules: when everything else is the same, select the direct sequence and remove first the dominant component.

Recently Tedder and Rudd (1978a,b) undertook an extensive study of eight distillation configurations separating ideally behaving ternary feeds of light hydrocarbons. The eight distillation sequences they examined include the simple direct and indirect sequences, single columns with side streams (three product streams), sequences with nonsharp (sloppy) separations with and without prefractionator and thermally-coupled columns as

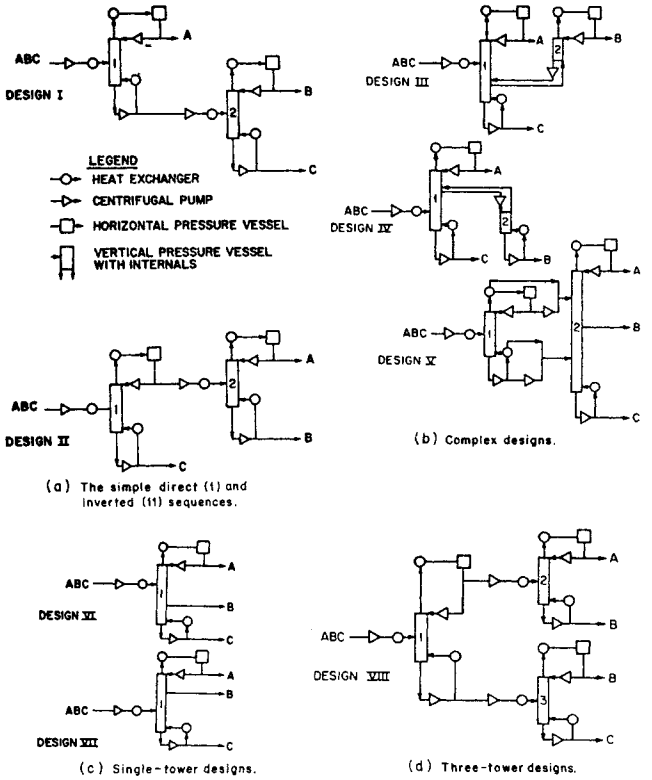


Figure 7. Eight different configurations (Tedder and Rudd, 1978a).

shown in Figure 7. They identified two parameters as the most critical for deciding on the optimal distillation sequence; the feed composition and the Ease of Separation Index (ESI). Two general triangular diagrams were constructed with the expected regions of optimality for ESI < 1.6 and ESI ≥ 1.6. The three sides of each diagram represent the concentrations of the three components of a ternary feed.

Each diagram (Figure 8) is separated into regions where a particular distillation configuration is expected to be optimal. These regions of optimality have been defined by the intersection lines for the minimum venture cost surfaces of two different configurations. For a given ternary feed with known composition, it becomes a trivial task to identify the optimal distillation sequence. As the authors state very clearly, these expected regions of optimality and the resulting conclusions are somehow uncertain, because the cost of the various configurations depends upon many variables in a complex way, and not only on the ESI and the feed composition. The value of the work by Tedder and Rudd lies primarily on the fact that it provides the designer with a simple tool to help him define the most likely optimal candidates for a distillation configuration. Furthermore, it can be extended to N component separation problems to sequence pseudoternary separations by performing the most difficult ternary separations last. The authors have developed a

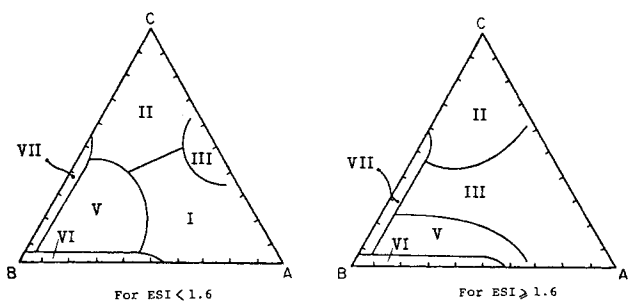


Figure 8. Expected regions of optimality (Tedder and Rudd, 1978a).

list of specific cases for which optimal distillation configurations are proposed.

All the above three methods share a common feature: they are approximate quantitative approaches to develop guiding rules in the synthesis of distillation sequences. As such they attempt to identify the critical parameters and analyze their impact on the optimal configuration. Due to the multitude of factors that affect the cost of the distillation columns, such parametric approaches have limited applicability. Their principal value consists of the heuristic guidelines they provide for a quick and simple estimate. The fact that they have a quantitative basis helps to determine numerical ranges of accuracy and bounds on their applicability. The above parametric methods are usually at their best when they are used to analyze and design systems where various parameters take on extreme values: e.g., one component is dominant in the feed; one separation is much harder than the rest; one product is required with extreme purity; the mixture is ideal, all the components are at equimolal compositions and all splits are of about the same difficulty (relative volatility). Nevertheless, such efforts should continue since they provide invaluable insight of the synthesis problem and the separations themselves.

Algorithmic Methods for Synthesis of General Separation Sequences

As mentioned earlier, the problem of the synthesis of separation sequences is that of a minimization problem (Eq. 2). The algorithmic methods attempt to solve this optimization problem by employing various algorithms developed in the area of the mathematical programming.

The earliest purely algorithmic synthesis method is credited to Hendry and Hughes (1972). Their approach makes use of the dynamic programming in attempting to identify rigorously the optimal sequence. The rationale behind this method lies in the fact that the number of separators increases much slower than the number of process flowsheets. Thus, if N is the number of components in a mixture and $R(N)$ and $Q(N)$ are the number of resulting distinct separation sequences and separators, respectively, it can be shown (Stephanopoulos, 1974) that:

$$\frac{R(N+1)}{R(N)} \to 4 \quad \text{and} \quad \frac{Q(N+1)}{Q(N)} \to 1$$

Although the method of Hendry and Hughes requires the examination and evaluation of all possible separators, the branch and bound strategy proposed by Westerberg and Stephanopoulos (1975) is capable of finding the optimal separation sequences, without having to search over the entire space of all possible separators.

Consider a three-component mixture A, B, and C, to be separated using two separation methods α and β. Let the ranked lists for these two methods be:

$$RL(\alpha) : ABC$$
$$RL(\beta) : ACB$$

In Figure 9, the tree of all possible separators is shown and consequently all the possible sequences can be found by following the branches of the tree. Consider the sequence of separators 1 and 5 (Figure 9). Let the cost of this sequence (not the optimal cost) be C_u. This value is an upper bound to the optimal cost of the sequence that we can generate for this problem. Let C_l be a lower bound of the cost of the unknown optimal sequence. (We will discuss later how to compute this lower bound.) Then,

$$C_l \leq C \text{ (optimal sequence)} \leq C_u$$

If any sequence has $C_l' > C_u$, it is clear that this sequence cannot be optimal, since we attempt to minimize the cost. The above remarks summarize the branch and bound strategy of Westerberg and Stephanopoulos. Thus,

(i) Using the ranked lists develop the tree of all possible

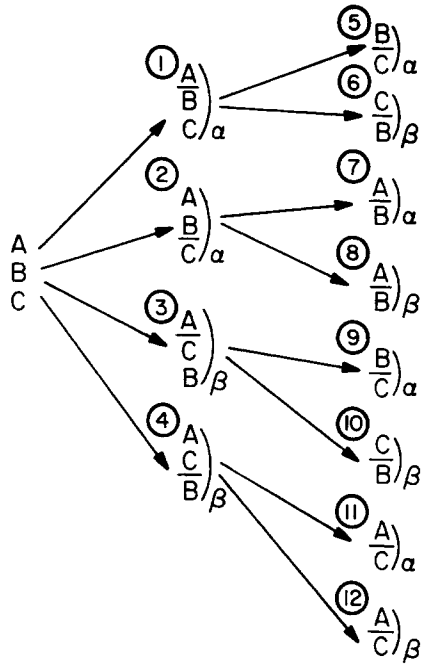


Figure 9. A tree of all possible separators with a 3-Component mixture, A, B, and C to be separated using two separation methods α and β.

separators and separation sequences.

(ii) For a promising separation sequence compute an upper and a lower bound for the optimal cost (this is called the basic sequence).

(iii) Any other complete or partially developed separation sequence which has a lower bound larger than the upper bound of the basic sequence cannot be optimal and it is disregarded.

The major advantage of this approach is that it can eliminate separation sequences before they have been completed, thus not requiring the evaluation of all possible separators as required for the dynamic programming strategy of Hendry and Hughes.

The lower bound on the cost of the optimal separation sequence is found to be the dual function of the corresponding Lagrangian problem. Consider the Optimization problem:

$$\text{Minimize } \phi = \sum_{i=1}^{N-1} \phi_i(x_i, u_i)$$

$$\text{Subject to } x_1 = x_1^o = \text{given conditions of feed}$$

$$x_{i+1} = f_i(x_i, u_i)$$

where, ϕ_i is the cost of the i-th separator in a sequence; x_i is the vector of all states for separator i; u_i is the vector of design variables for separator i. The Lagrangian of the above problem is:

$$\mathscr{L} = \sum_{i=1}^{N-1} l_i(x_i, u_i, \lambda_i; \lambda_{i+1}) + \lambda_1^T x_1^o \tag{4}$$

where,

$$l_i = \phi_i - \lambda_i^T x_i + \lambda_{i+1}^T f_i \tag{5}$$

and λ_i is the vector of Lagrange multipliers associated with the equality (connection) constraint $x_i = f_{i-1}(x_{i-1}, u_{i-1})$. Let,

$$h(\lambda_1, \lambda_2, \ldots, \lambda_{N-1}) = \underset{x_i, u_i; i=1,N-1}{\text{Minimize}} \mathscr{L} = \sum_{i=1}^{N-1} \underset{x_i, u_i}{\text{Min}}\, l_i \tag{6}$$

for given Lagrange multipliers. The function h is called dual function and has the property

$$h \le \phi^* = \text{minimum cost of sequence}$$

i.e., constitutes a lower bound of the cost of the optimal sequence. If $\underset{x_i, u_i}{\text{Min}}\, l_i > 0$ for all $i = 1, 2, \ldots, N-1$, it is clear that $\underset{x_i, u_i}{\text{Min}}\, l_i \le h \le \phi^*$. This last inequality implies that we can eliminate partially developed sequences, if $\underset{x_i, u_i}{\text{Min}}\, l_i > C_u =$ upper bound on the cost of the optimal sequence. Unfortunately, the inequality $\text{Min}\, l_i > 0$ does not hold for arbitrary values of the Lagrange multipliers and care should be exercised in the screening of partially developed sequences.

Rodrigo and Seader (1975) have developed an ordered branch search which, in conjunction with two rules, reduces the search space for the location of the optimum separation sequence. The search space is conveniently represented by a tree, and the reduction of the search space is accomplished with the aid of two rules. (1) Identify certain forbidden separations, thus eliminating other separators which stem from these forbidden splits. (2) Identify multiplicate separators and analyze them no more than one. After the reduced space of the acceptable separation sequences has been defined, a branch and search procedure is initiated. Three heuristics are employed to aid the search: (a) generate or select the subproblem where the first species in the ranked list is separated from the remaining species; (b) generate or select the subproblem where the split of species between the two product streams is nearly equimolar; (c) generate or select the subproblem with the lowest cost (cheapest first). These heuristics are not new, but as we have seen earlier in this section they were used by Heaven (1969), Thompson and King (1972a,b), and others.

The ordered branch search proceeds in two stages. During the first stage, the search proceeds throughout the search space using the above heuristics until a complete feasible sequence has been generated. As the authors point out, although the initial sequence is not optimal, quite often it is nearly optimal (due to the heuristics). The cost of this initial sequence is an upper bound to the cost of the optimal sequence. In the second stage, a backtracking starts in an effort to lower the cost of the initial sequence by identifying promising structural variations. The major advantage of this method is an organized and systematized usage of the heuristics.

In an apparent improvement over the last method, Gomez and Seader (1976) have proposed an ordered search which makes use of predicted lower bounds on the cost of the optimal sequence. Their method achieves considerable reduction in the search space and nearly-optimal separation sequences are readily developed. The procedure of Gomez and Seader uses a state space representation of all the alternative separation sequences which is elegantly described by a sequence graph (Figure 10). Each node in the graph represents a step towards the development of complete separation sequences. The solid arcs represent the separators. The beginning node (B) is the initial multicomponent feed mixture and the terminal node (T) is the set of the desired products. Associated with each node are two measures:

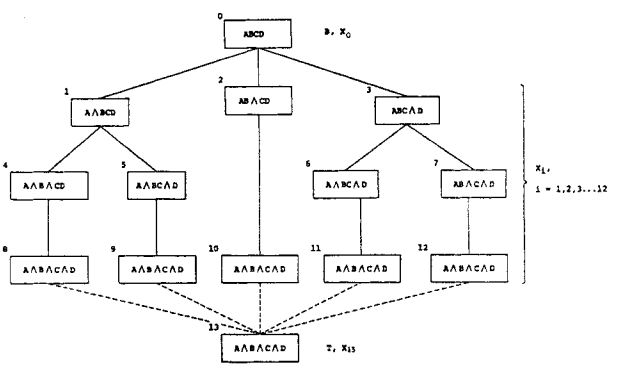


Figure 10. A state space representation of all the alternative separation sequences (Gomez and Seader, 1976).

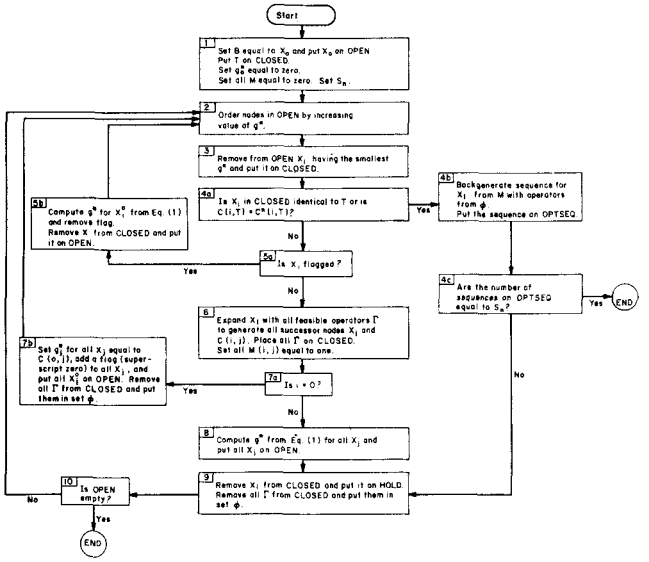


Figure 11. The predictor-based ordered search algorithm of Gomez and Seader (1976).

COLD STREAMS \ HOT STREAMS	A	B	C	D	A B	B C	C D	A C	B C	A B C	A B C D
B	VR	4	F	F	F	4	F	F	4	F	
C	F	VR	4	F	VR	F	4	F	F	F	
D	F	F	VR	4	F	VR	F	VR	F	F	
E	F	F	F	VR	F	F	VR	F	VR	VR	
B C	VR	F	4	F	3	4	3	F	4	F	
C D	F	VR	F	4	VR	3	4	4	F	F	
D E	F	F	VR	F	F	VR	3	VR	4	4	
B C D	VR	F	F	4	4	F	4	4	4	F	
C D E	F	VR	F	F	VR	4	F	4	4	4	
B C D E	VR	F	F	F	4	F	F	4	F	3	

Figure 12. The energy match matrix for a 5-Component mixture. F means the match is feasible. VR indicates that vapor recompression is required for the match to be feasible. A number denotes the rule which indicated the match was infeasible. (Rathore et al, 1974b).

(1) The actual cost of the partially completed sequence beginning with (B) and ending in that node.

(2) The evaluation function which estimates the cost of the complete sequence passing through that node.

The evaluation function is an heuristic function and is defined by:

$$g_j^*(B, T) = g(B, i) + C(i, j) + C^*(j, T)$$

where $g_j^*(B, T)$ is the estimated cost of the completed sequence going from B to T and passing through node i; $g(B, i)$ is the actual cost of the partially completed sequence from node B to node i; $C(j, i)$ is the minimum actual cost from node i to node j by an appropriate separation type, evaluated for every successor j; $C^*(j, T)$ is the estimated cost of the separators involved from node j to node T, i.e., the cost of all remaining binary subproblems evaluated in the absence of nonkeys. It is clear that $g_j^*(B, T)$ is a lower bound on the total sequence cost if $C^*(j, k) \leq C(k, j)$ for all k's for which the algorithm converges to an optimal solution. The difficulty is that different separator classes require that $C^*(j, T)$ be evaluated for every feasible separation, and a failure of the heuristic cost function is not detectable (Motard and Westerberg, 1978). Gomez and Seader also point out that close to 50% of all subproblems must be evaluated. The predictor-based ordered search algorithm of Gomez and Seader is shown in Figure 11. The admissibility of this algorithm is proven in their work. This strategy has been applied to several separation problems and the results were very promising. The search space was reduced significantly and nearly-optimal sequences were readily developed.

SYNTHESIS OF DISTILLATION SEQUENCES WITH HEAT INTEGRATION

With two exceptions, all the works discussed previously did not consider any thermal coupling among the separators for energy conservation. Due to the significant portion that utilities contribute to the venture cost of a distillation column, there is a great economic incentive to synthesize distillation sequences with heat integration among the columns of the sequence. The size of the resulting problem is overwhelming. The alternatives increase rapidly, since the sequencing of the columns and the heat integration cannot be solved separately, but they have to be tackled simultaneously. The heat integration of two distillation columns is possible because there are heat sources (overhead vapor and the bottoms product) and heat sinks (feed and recycle stream through the reboiler).

Petlyuk et al. (1965) considered thermally coupled columns but their analysis did not go very far. Stupin and Lockhart (1972) presented a case study without suggesting any general rules for the synthesis.

The first systematic effort was undertaken by Rathore et al. (1974a,b) who employed list processing techniques and a strategy of dynamic programming as suggested by Hendry and Hughes (1972). The first of the two works dealing with sequences operating at the same column pressure is of little value since the great opportunities for heat integration appear when the columns of a sequence are allowed to operate at different pressures. The second work lifts this restriction, and it is of more interest.

In the first step, Rathore, Vanwormer and Powers use list processing techniques to identify all the hot (overhead product streams) and all the cold (bottoms product stream flowing through the reboiler) streams. Figure 12 presents the energy matrix for a five-component mixture with all the feasible heat matches among all the hot and cold streams. Vapor recompression has been excluded. Four additional rules eliminate other matches since they lead to infeasible separation sequences. All the feasible heat matches shown in the energy matrix define all the thermal integration possibilities for a given system. In the next step, all 2-, 3-, ... , $(N-1)$-, N-component separators are identified and optimized. Also all the two-column heat integrated systems are identified and optimized. Finally, in a manner identical to the one used by Hendry and Hughes (1972), the optimal integrated sequence is synthesized. The formulation of an exclusion principle helps to identify potential three-column integrations. It should be noted that the exclusion principle, as applied to the screening of three-column integration, is incorrect (Sophos, Stephanopoulos, Morari, 1978). The procedure suffers from the curse of dimensionality and it still needs to be affirmed through its application to various systems.

Branch and bound strategies similar to the one discussed earlier by Westerberg and Stephanopoulos (1975) have been developed by Sophos, Stephanopoulos, and Morari (1978) and Faith and Morari (1979). The first work has developed a sequence of rules to aid in the reduction of the search space. The second work using the concept of the free-market economic interaction among the integrated columns has developed an interesting and effective estimation scheme for the Lagrange multipliers that enhances the screening capabilities of the strategy. Both these works are still under development.

Freshwater and Ziogou (1976) have performed extensive computations on four systems of four components and one of five components. All were nearly ideal mixtures and a wide range of feed compositions were considered. They found that when no heat integration was considered the direct sequencing was the optimal configuration in all but four instances. When heat integration was allowed nearly half of the four-component cases and one third of the five-component cases required optimal sequences distinctly different. This work being computational in its nature and dealing with very specific cases does not provide any rules for synthesis but only indications of possible optimal patterns.

Recent work by Sophos, Stephanopoulos and Linnhoff (1981) has showed that a two-level screening procedure is feasible for the synthesis of distillation sequences with heat integration. At the first level, a small number of very good unintegrated distillation sequences is identified. It is shown that these sequences offer the largest potential for heat integration and that the optimum sequence with heat integration is a member of this small group. The central argument of the above conclusion is that the nearly optimum unintegrated sequences offer the smallest heat loads and the smallest temperature differences between the hot and cold streams, thus leading to feasible heat matches without excessive deviations of the operating pressures from their optimum values.

At the second level, the development of achievable targets and bounds allows the designer to identify very quickly the most promising integrated sequences. Further screening can be performed after that but with diminishing returns.

PROGNOSTICATION FOR SEPARATION SYSTEM SYNTHESIS

Separation system synthesis results have occurred principally for systems of simple, sharp separators. A more general separation synthesis problem was suggested by Rudd, Powers and Siirola (1973), and it has yet to be solved adequately. The more general separations problem is to transform several mixtures available as sources into a different set of several mixtures desired as products. One should use flash units, columns having components which distribute among two or more product streams, mixers, simple stream splitters, absorbers not followed immediately by solvent recovery units, etc., to solve this more general problem. Significant economies can result from not using sharp separations when the product mixtures do not require them (Motard in Motard and Westerberg, 1978).

To date, the only attempts to solve this problem are in the approaches developed for the synthesis of total flowsheets (e.g., Siirola et al., 1971; Mahalec and Motard, 1976, 1977a,b), and they do not appear to yield the quality of solution which must be possible. Add to this more general problem the desire for heat integration and one has an even more interesting synthesis problem, one yet to be solved.

With the continued attack apparently being made on these problems, one has to assume that industrially useful approaches will be forthcoming, likely within two to five years. The approaches developed will likely involve heuristics, in a significant way, yielding excellent but not probably optimal solutions.

HEAT EXCHANGER NETWORK SYNTHESIS

Solution of this particular synthesis problem has progressed substantially since it first appeared. Thirty-nine publications are listed in this review specifically on this topic. While not yet a fully solved problem, available results are directly usable in the design of energy-efficient processes. It is time to present an overview of this work.

Three major results are available with two of them tracing back to an unpublished Ph.D. thesis of Hohmann (1971) who did his work with Lockhart; the third was hinted at in this thesis but not explored. The first two of these results are that one can predict a minimum utility usage target and the probable fewest number of heat exchangers required prior to developing an actual network design. These targets can usually be met with actual designs which turn out to be the more economic ones. Linnhoff and coworkers (Linnhoff, 1979; Linnhoff and Flower, 1978a; Boland and Linnhoff, 1978) systematized and proved these first two results. Also the conjecture by Hohmann that both targets could be always met, if stream splitting were permitted, was shown by Linnhoff not to be true by counterexample.

The third major result is that one can locate "bottlenecks" in a process design which preclude further heat integration, again prior to developing a network solution, and discovery of these bottlenecks can be used to alter the process design (Linnhoff, 1979; Umeda, Niida and Shiroko, 1979; Umeda, Harada and Shiroko, 1979). These results are industrially significant and would by themselves justify the research expended to date on the entire area of process synthesis.

Problem Specification

The form of the heat exchanger network problem is as follows. A set of process streams are given; for each is specified an inlet temperature, a desired outlet or target temperature, a flow rate and a heat capacity. The temperature effect on heat capacity is usually ignored, but most algorithms can be easily modified to overcome this assumption. Also given are available utility streams such as steam and cooling water, but here flow rates are not specified and typically not limited. Also specified are heat transfer coefficients which usually can be stream/stream match dependent, that each exchanger will be countercurrent, a cost correlation for the investment cost of a heat exchanger vs. its area and finally the annual cost per unit flow for each of the utility streams.

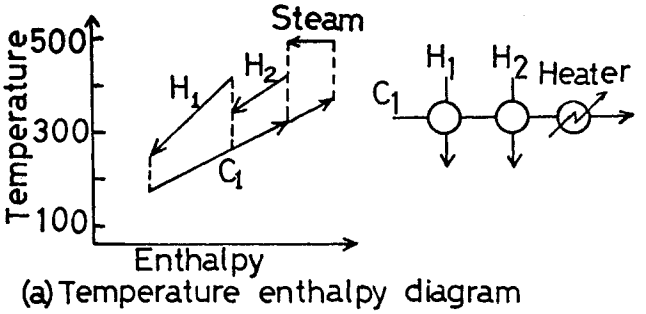
The above specification for a heat exchanger network synthesis problem corresponds to the information typically available from a process flowsheet which has not as yet been heat-integrated but for which the heat and material balances are completed. Thus, it represents a reasonable and useful problem.

A very large number of alternative heat exchanger network configurations can exist even for very small problems (for example, Motard and Westerberg, 1978). The synthesis problem has been to find the network of heat exchangers which has the least annualized cost and which brings each process stream from its inlet to its target temperature. Cost includes the investment cost for the heat exchangers, converted to a cost per year figure, and the annual cost of the necessary utilities. The problem is typically one of trading investment costs against utility costs.

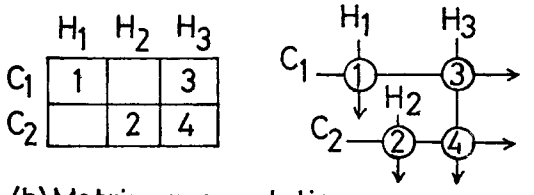
Problem Representation

A variety of different representations have been used in developing heat exchanger networks. Perhaps, the oldest is the "temperature/enthalpy diagram" (Whistler, 1948). Figure 13a illustrates this representation. Temperature (ordinate) for each stream is plotted against its enthalpy (abscissa). The enthalpy scale is only relative; thus, streams may be moved to the right or left on this diagram. A match between two streams is represented by placing a cold stream (one which is to be heated in the match) directly below a hot stream (one which is to be cooled). Where the streams overlap, the match takes place. By construction, the overlapped portions are in heat balance. The match is also thermodynamically feasible as the hot stream is hotter than the cold stream in all places along the match. The vertical distance between the streams is the temperature difference experienced along the match. Figure 13a illustrates a set of matches and the corresponding heat exchanger network using this representation.

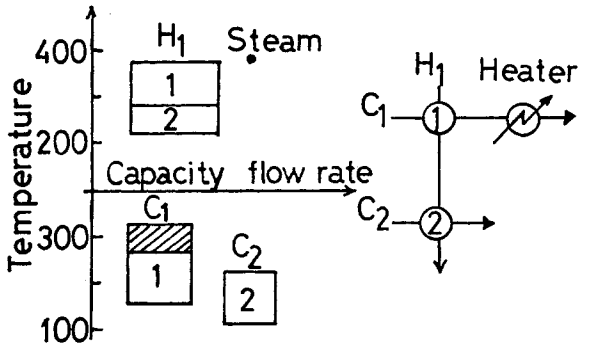
A second representation introduced by Pho and Lapidus (1973) is the "simple match matrix," Figure 13b. Here, the networks are limited to those which can be represented as an ordered *sequence* of matches among the streams, with only one match allowed between any stream pair.



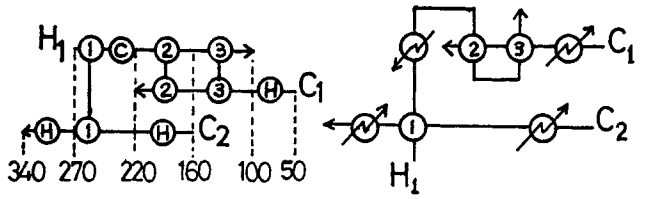
(a) Temperature enthalpy diagram



(b) Matrix reprentation



(c) Heat content diagram



(d) Temperature interval diagram

Figure 13. Representation of heat exchanger networks (a)—(d).

A third representation is the "heat content diagram" of Nishida, Kobayashi, Ichikawa (1971), Figure 13c. Each stream is represented by an "area" with the vertical scale being the temperature and the area width the flow rate × heat capacity of the stream at that temperature. The area is then:

$$Q = \int_{T_1}^{T_2} FC_p \, dT$$

which is the heat to be added to or removed from the stream by the network to be designed. A match between two streams is represented by assigning equal areas, one from a hot stream and one from a cold stream, to the match. Figure 13c illustrates a heat content diagram with matches assigned and the corresponding network. The temperature/enthalpy diagram is the integral of the heat content diagram.

Another very convenient representation by Linnhoff and Flower (1978a) directly represents the network structure, Figure 13d. A match between streams is indicated by placing a pair of circles on each of the streams and connecting them by a vertical line.

TABLE 3. MINIMUM UTILITY BOUNDS USED IN LITERATURE

UB-1: Net difference between heating needed for cold streams and cooling needed for hot streams.
UB-2: Same as UB-1 but modified to account for portions of hot (cold) streams colder (hotter) than any existing cold (hot) process stream.
UB-3: Exact bound accounting for uniform minimum allowed approach temperature.
UB-4: Exact bound accounting for uniform minimum allowed approach temperature and user stated disallowed stream/stream matches.
UB-5: Same as UB-4 but with match dependent minimum allowed approach temperatures.

Synthesis Algorithms

The heat exchanger network synthesis problem can be conveniently partitioned into the following three major steps.

I. Preanalysis to set targets, limitations.
II. Network invention.
III. Evolution.

The first two of these steps can be further subdivided, both into the same set of substeps, but with each emphasizing the substeps differently. The substeps are as follows.

A. Partition the synthesis problem.
B. Merge *equivalent* heat sources, sinks.
C. Select stream/stream matches.
D. Select network capable of producing desired matches.

We will show how the different synthesis algorithms, published for heat exchanger networks, fit into and contribute to the above problem "generalization."

Step I—Preanalysis. Preanalysis involves establishing targets for the network to be designed. These targets are: (1) the least amount of utilities which are needed; (2) assuming all heat transfer coefficients are equal, the minimum area required; and (3) the probable, but not guaranteed, fewest number of heat exchangers needed. Experience has shown that networks satisfying targets (1) and (3) are economically very attractive solutions. Target (2) is usually only approached, but not reached by the optimal solutions.

Least utility targets have been set five ways, some of which only establish lower bounds. Table 3 lists the alternatives. An obvious bound (UB-1) is to establish the difference in heat needed for heating the cold streams and the heat available when cooling the hot streams, a bound used by one of the early papers. The bound has sometimes been refined slightly (UB-2) where one accounts for cold streams which require heating above any temperatures available by the hot process streams or any hot streams requiring cooling below any temperature available by cold process streams (Rathore and Powers, 1975; Nishida, Liu and Lapidus, 1977; Grossmann and Sargent, 1978). However, exact targets (UB-3) are possible which can *always* be met, and these account for both the difference in heating and cooling required and the temperature levels of the streams (Hohmann, 1971; Linnhoff and Flower, 1978a; Greenkorn, Koppel and Raphaven, 1980).

The essence of the method to establish the minimum utility bound (UB-3) is to merge all hot streams into a single hot "superstream" and all cold into a single cold "superstream." Plotting these two superstreams on a temperature/enthalpy diagram, one moves the cold superstream under the hot until the minimum vertical distance exactly equals the minimum ΔT allowed anywhere in the exchanger network, which in the limit may be zero, or until one of the superstreams is matched in its entirety by the other (Umeda, Itoh and Shiroko, 1978). The unmatched portions of each of the streams represent the minimum heating and cooling required.

Cerda (1980) in cooperation with B. Linnhoff, D. Mason and A. Westerberg calculates minimum utility requirements. The extension permits one to exclude matches between designated

pairs of streams, either in total or over certain ranges of temperature (UB-4). He also extended the ideas to allow the minimum approach temperature to differ with each stream/stream pair (UB-5). He observed that the problem is directly formulated as a "network flow" problem in linear programming and that earlier methods for problems without matches being excluded correspond directly to the calculations involved in solving a simple network flow problem.

The target of "minimum area" (Nishida, Kobayashi and Ichikawa, 1971; Hohmann, 1971) can be discovered rather quickly using the superstreams. We slightly refine these ideas here. If exactly matched by the introduction of utilities as needed and if all heat transfer coefficients are assumed equal, the so-called "minimum area" solution corresponds to a countercurrent exchanger designed for these superstreams. The area calculation involves integrating

$$A = \frac{1}{U}\int_1^2 \frac{dQ}{\Delta T}$$

where A is the area, U is the heat transfer coefficient, Q the heat transferred and ΔT the vertical distance between the two superstream curves at the point that the incremental heat dQ is transferring.

The fewest exchangers needed in a network is generally one less than the total number of streams involved in the two superstreams, including the utility streams (Hohmann, 1971). Figure 14 illustrates why. If one wishes to accomplish the heat exchange among all the streams, each match proposed *in an ordered sequence* must totally eliminate one of the two streams in the match. In Figure 14, the first match starting at the left is 50 heat units, eliminating the cold stream. This match leaves the hot stream with 55 units of heat still to be exchanged. These 55 units are totally eliminated in the second match, etc. Note four matches can occur if, for each match, one stream is eliminated except for the last when both are simultaneously eliminated because of overall heat balance. Linnhoff, Mason and Wardle (1979) discuss in detail and refine the minimum unit ideas.

The preanalysis steps for the existing methods are compatible with the four substeps (A through D) stated earlier. To establish utility bounds and/or area bounds, the problem is usually partitioned according to key temperature intervals (Linnhoff and Flower, 1978a). First, if ΔT_{min}, a user prescribed "minimum approach temperature," is added to all cold stream temperatures (Linnhoff and Flower, 1978a), the prescribed inlet and outlet temperatures for all streams, *including utilities*, are ordered into decreasing order. Each pair of temperatures on the list represents an interval for the hot streams. Cold stream intervals are the same but with the temperatures decreased by ΔT_{min}. If no stream/stream matches are excluded by the designer, within each interval the contributions for each substream can be merged to create hot and cold superstreams.

The last step taken to calculate minimum utilities is to select the stream/stream matches among these merged substreams. With only one hot stream and one cold stream, matching is a trivial step. No actual network need be proposed, so step *D* is not required for preanalysis.

Step II—Network Invention. As stated earlier, the network invention step can also be subdivided into four substeps: (A) partitioning the synthesis problem; (B) merging equivalent heat sources, sinks; (C) selecting stream/stream matches; and (D) selecting the heat exchanger network capable of producing the desired matches. Not all of these substeps are necessarily a part of each of the published algorithms.

Partitioning, if it occurs, is of two types. The problem may be partitioned by temperature intervals, as explained already for the preanalysis step. The TI method of Linnhoff and Flower (1978a) uses this partitioning. If one has done a preanalysis and has located where in the problem a temperature pinch point exists, one may also partition the overall problem into two problems, one above the point of the pinch and one below. Heat

TABLE 4. SEQUENTIAL MATCH OPTIONS

Search Strategy Employed
- SS-1: Total Enumeration
- SS-2: Branch and Bound
- SS-3: Heuristic
- SS-4: Other

Heuristics Used (If SS-3 Employed)
- HR-1: Select hot stream with highest inlet temperature and cold stream with highest target temperature.
- HR-2: Select hot stream with coldest target temperature and cold stream with coldest inlet temperature
- HR-3: Select match giving least value to ΔT_{ave}
- HR-4: Select match giving least value to estimated upper bound on overall network cost.

Match Restrictions
- MR-1: Disallow stream splitting
- MR-2: Disallow stream/stream rematching (cycling)
- MR-3: Disallow if match precludes predicted minimum utility usage
- MR-4: Disallow if match precludes network having predicted fewest number units

Stream Heat Selection Decisions (h = for Hot Stream, c = for Cold Stream)
- HS-1*h*, *c* Take heat from or supply heat to hottest end of stream.
- HS-2*h*, *c* Take heat from or supply heat to coldest end of stream.
- HS-3*h*, *c* Take heat from or supply heat to intermediate portion of stream.

cannot be exchanged between these two partitions, if one wishes to develop networks requiring minimum utility usage. This partitioning was implicitly a part of the TI method (Linnhoff and Flower, 1978a) and is used explicitly by Grimes, Rychener and Westerberg (1980).

A class of algorithms (e.g., Nishida, Kobayashi and Ichikawa, 1971) next proposes that streams with equivalent heat be merged. This step is exactly that of producing superstreams mentioned earlier and thus moves these algorithms into the class performing a preanalysis to find the minimum area target for a problem.

Next, the different algorithms select which stream/stream (or superstream/superstream) matches to make. Many accomplish this step as a sequence of match decisions, while others make their stream/stream match decisions in parallel. We shall discuss this step at length because of the different ways the various algorithms accomplish this step.

Sequential Match Decision Algorithms

For algorithms in this class, the principal questions are the search strategy employed, how to select the next match, and what is meant by a match.

Table 4 lists the variety of search strategies (SS), of heuristics for selecting the next match (HR), of match restrictions (MR), and of heat selection rules (HS) used by the various algorithms.

The essence of algorithms which develop the network as a *sequence* of match decisions is to develop a tree of networks, where the initial node is the network with no process stream/process stream matches. All heating and cooling is done explicitly or implicitly by utilities for this node. The children of this node are all networks containing exactly one process stream/process stream match. Their children contain exactly two process stream/process stream matches, etc.

One of the earlier algorithms (Pho and Lapidus, 1973) suggests developing the entire tree of networks which their rules allow to be generated, but the tree becomes excessively large for a 10-stream problem. They suggest a fallible "look ahead" procedure to reduce to a feasible size, the number of branches of the tree to be examined. Others propose using a branch and bound scheme (McGalliard, 1971; Rathore and Powers, 1975; Grossmann and Sargent, 1978; Greenkorn, Kop-

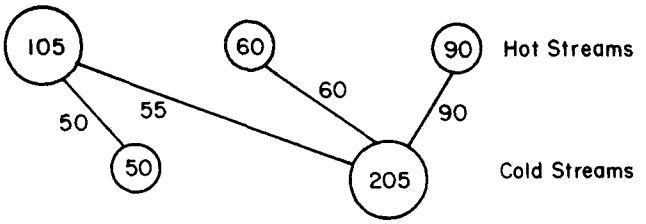


Figure 14. Minimum number of matches for a five-stream problem. Numbers in circles are heats available or needed. Numbers along edges are heat transferred in match.

pel and Raghavan, 1978) to search the tree. To use this scheme a lower bound on cost has to be estimated for all nodes emanating from a given node. This lower bound is usually the annualized capital cost for the heat exchangers already involved in the network plus a lower bound on the cost of utilities still required (calculating minimum utilities using one of the utility bounds UB-1 to UB-3). The fastest, but obviously most fallible, algorithms simply use one of the heuristics HR-1 to HR-4 to select a single next match.

A number of possible matches are disallowed by match restrictions (MR). For example, no stream splitting may be allowed (MR-1) or no stream rematching (MR-2). Rematching is when two streams exchange heat in two or more noncontiguous exchangers. If the algorithm was developed with an awareness that one can calculate the minimum utility usage bound (UB-3), it may eliminate any match which if made, would preclude reaching this bound. Detection of such a match is done by making the match and then running a "preanalysis" on the unmatched streams to detect the minimum utility usage needed for them. If this prediction indicates an increase in the use of utilities, the match can be excluded.

The algorithm may also disallow matches not leading to the predicted fewest number of heat exchanger units. To reach such a solution each match made in the sequence must eliminate one of the two streams entirely and leave a single residual from the other (Greenkorn, Koppel and Raghaven, 1978). If the match cannot accomplish such an elimination because of temperature limitations, it need not be considered.

The heat selection options (HS-1h,c to HS-3h,c) can have a profound effect on the network developed. Earlier algorithms matched the hottest portion of the hot stream against the coldest portion of the cold (HS-1h/HS-2c). This match decision is the most thermodynamically irreversible, leaving the hot end of the cold stream and/or the cold end of the hot stream for subsequent heating or cooling. Ponton and Donaldson (1974) appear to be first to advocate matching the hot end of the hot stream against the *hottest* portion of the cold stream (HS-1h/HS-1c). This heat selection rule is quite reasonable for above ambient networks as it is more thermodynamically reversible and it tends to allow the lower-temperature hot utilities to be used. It does this using lower-temperature cold utilities. This trade is sensible since above-ambient hot utilities are more expensive than above-ambient cold ones.

Some of the later papers describe sequential algorithms simultaneously requiring minimum utility usage (MR-3) and accomplishing all this in the predicted minimum number of heat exchanger units (MR-4) (Greenkorn, Koppel and Raphaven, 1978; Grimes, Rychener and Westerberg, 1980).

Simultaneous Match Decision Algorithms

Two approaches published so far establish the stream/stream match decisions "simultaneously." The first is one of the earliest published, using a method based on the "assignment algorithm" in linear programming. Three publications have appeared based on this approach (Kesler and Parker, 1969; Kobayashi, Umeda and Ichikawa, 1971; Cena, Mustacchi and Natali, 1977).

The second is the thermodynamic-combinatorial (TC) algorithm of Linnhoff (1979) and Flower and Linnhoff (1980).

In the assignment algorithm approach, every stream is first partitioned into a set of usually quite small substreams, each substream having a heat content of "Q" units. Kesler and Parker (1969) and Cena, Mustacchi and Natali (1977) require this partitioning to be sequential (equivalent to no stream splitting), whereas Kobayashi, Umeda and Ichikawa (1971) permit parallel partitioning (equivalent to stream splitting). "Q" is chosen so every stream is partitioned, to an adequate approximation, into an integer number of substreams. Utilities are also added as a generous number of substreams, each again with a heat content of Q units. The assignment problem is to assign hot substreams to cold ones in a manner which minimizes the sum of the costs associated with each assignment. Constraints precluding certain assignments are readily added, and they reflect thermodynamic considerations (the hot substream must be hotter than the cold one). They could also represent user restrictions. Very quickly a large number of substreams and constraints are generated, even for small problems. Fortunately the "assignment algorithm" of linear programming is effective at handling large problems.

The cost of an assignment is the weakest aspect of this approach. It can easily include the cost of utilities by assigning, as part of the cost for a match involving a utility substream, the cost of Q units of that utility. The cost must also include the annualized cost of the heat exchanger area, but area costs should reflect overall heat exchanger size, since large exchangers cost much less per unit area than small ones. Unfortunately overall size is unknown *a priori*. No mathematically correct solution to this dilemma has been proposed.

The solutions resulting from this approach require further work as they simply provide one with assignments of the small substreams to substreams. It is up to the user to translate these into a practical set of heat exchangers corresponding only approximately to the substream assignments. This method apparently results in quite good answers.

As mentioned above, the second simultaneous match decision algorithm is the TC algorithm of Linnhoff (1979) and Flower and Linnhoff (1980). This algorithm will generate all heat exchanger networks satisfying match restrictions MR-1 (no stream splitting), MR-3 (minimum utility usage) and MR-4 (fewest number of heat exchanger units). It is readily shown that such networks are acyclic; that is, if one were to disregard the direction of stream flows and then trace the pipework for such a network, one cannot find a path which starts with a given heat exchanger and which subsequently returns to it. Figure 14 may aid in believing this property. The best description of this algorithm is in Linnhoff (1979). First the minimum amount of utilities is predicted, along with required utility streams. Then like an odometer ticking off the miles, the algorithm generates all stream/stream match structures (one of which is illustrated in Figure 14) which do not contain cycles. Note this is a "parallel" match decision step, not a "sequential" one. Then calculating the heat loads for each match, entire structures are eliminated if the heat loads suggested by these matches are not all positive (they require that heat must always transfer from hot streams to cold streams). Finally the algorithm systematically creates heat exchanger networks for each match structure. No stream splitting is allowed. The temperatures are calculated and the structure is kept only if the temperatures are satisfactory throughout; that is, temperature driving forces larger than some prescribed minimum ΔT occur in all matches. The algorithm may terminate with no satisfactory structure. It is apparently very fast, with the 10 stream problem (10SP1) creating some 800 alternative solutions and being solved in about 50 seconds on a DEC system 10 computer (personal communication by Linnhoff (1980)).

Select Network. The final step of each of the algorithms is to select the actual heat exchanger network. For the sequential match decision algorithms this step is accomplished at the same time that the stream/stream matching decisions are made; for the parallel match algorithm, it is a separate step. The algorithm of Nishida, Kobayashi and Ichikawa (1971), where equivalent heats are merged, has only selected which merged heat sources

are to supply heat to which merged heat sinks. Subject to guidelines given, it is up to the user to choose among the alternatives possible to develop the actual network.

Evolution. Many publications present steps to aid in improving a heat exchanger network, once one is developed. An early publication by McGalliard and Westerberg (1972) presented a method to determine if a modification to a process flowsheet leads to an improved flowsheet without requiring either the original or the modified flowsheet to be fully optimized. The method was based on primal/dual bounding, extending ideas from the two-level algorithm of Brosilow and Lasdon (1965). They applied the ideas to several example heat exchanger networks. The modifications themselves were developed by the person inventing the network.

Shah and Westerberg (1975) presented a paradigm for evolutionary development of a heat exchanger network plus some evolutionary rules to modify a network in a "small" way. The algorithm was to make a small change, if it led to an improvement, move to the improved network and repeat. The crucial issue was what type of modifications constitute small changes. Three modifications were suggested.

Nishida, Liu and Lapidus (1977) present a set of evolutionary rules to improve networks generally developed with the aid of the earlier minimum area algorithm of Nishida, Kobayashi and Ichikawa (1971). The minimum area algorithm requires one to develop a network containing far too many actual heat exchangers, and the evolutionary rules were to aid one to reduce the number of exchangers.

The ED (evolutionary development) method of Linnhoff and Flower (1978b) is a set of rules to aid one when making small changes to a heat exchanger network, such as those developed by their TI method. The rules tell one how to detect easily if the suggested small step will lead to a thermodynamically feasible network. While suggesting that one's goal should be to reduce the number of heat exchangers to a minimum, the paper was not intended to give strategies to apply the evolutionary steps.

The thesis by Grimes (1980) uses heuristics to develop a network subject to the targets implied by match restrictions MR-3 (network must use minimum utilities) and MR-4 (network must use predicted fewest exchangers possible). Two evolutionary rules then allow one to find neighboring structures also satisfying these same two match restrictions. He gives theorems which show *all* such structures can be reached from any starting structure through evolution using only his two rules. Applying these ideas by hand, and thus with the user's insights and prejudices included, Grimes improved many of the networks thought to be optimal by others. He, for example, improved a network which could not reach both the minimum utility and fewest exchanger targets. He relaxed the minimum utility target rather than the fewest exchanger target as others had done earlier.

Tables 5, 6 and 7 list the references which deal with heat exchanger network synthesis. Indicated for each reference are the various features which they contain. Also indicated with an asterisk is our belief that the feature or insight originated with this work.

PROGNOSTICATION FOR HEAT EXCHANGER NETWORK SYNTHESIS

The minimum utility usage problem is solved adequately, and it can already be very useful as an industrial design tool. Synthe-

TABLE 5. HEAT EXCHANGER NETWORK SYNTHESIS ARTICLES

Topics Covered	New Representation	Preanalysis	Network Invention	Evolution	Review	Case Study
Whistler (1947)	*					
Hwa (1965)			*			
Kesler & Parker (1969)			*			
Masso & Rudd (1969)			*			
Lee et al. (1970)			*			
Kobayashi et al. (1971)			*			
Nishida et al. (1971)	*	*				
Hohmann (1971)	x	*	x			
Menzies & Johnson (1972)			x			
McGalliard & Westerberg (1972)			x	*		
Pho & Lapidus (1973)	*		*			
Siirola (1974)					x	
Ponton & Donaldson (1974)			*			
Rathore & Powers (1975)			*			
Shah & Westerberg (1975)			x	*		
Hohmann & Lockhart (1976)						x
Donaldson et al. (1976)			*			
Hwang & Elshout (1976)						x
Nishida et al. (1977)		x	x	*		
Wells and Hodgkinson (1977)			x			
Stephanopoulos (1977)					x	
Cena et al. (1977)			*	*		
Linnhoff & Flower (1978a)	*	*(proved ideas)	*			
Linnhoff & Flower (1978b)				*		
Flower & Linnhoff (1978)		x	*			
Greenkorn et al. (1978)		x	*			
Grossmann & Sargent (1978)		x	*			
Boland & Linnhoff (1979)					x	
Hohmann & Nash (1978)						x
Motard & Westerberg (1978)					x	
Nash et al. (1978)						
Elshout & Homann (1979)						x
Linnhoff et al. (1979)					x	
Umeda et al. (1979)					x	
Shah & Westerberg (1980)			x			
Cerda & Westerberg (1980)		*				
Grimes et al. (1980)		x	*	*		

*New Contribution; x = Topic Covered

TABLE 6. HEAT EXCHANGER NETWORK INVENTION OPTIONS SELECTED BY VARIOUS ALGORITHMS

	Partitioning	Temp. Interval at Pinch	Small Equal Substreams	Stream Matching	Sequential	—Search Strategy (SS)	Min Utility Bound Used (UB)	—Heuristics Used (HR)	—Match Restrictions (MR)	—Heat Section (HS)	Parallel	—LP Assignment	—Other	Comment
Hwa (1965)														
Kesler & Parker (1969)			*									*		Sequential Stream Partitioning
Masso & Rudd (1969)						3			1, 2	1h, 2c				Only paper describing "learning"
Lee et al. (1970)						2			1, 2	1h, 2c				Developed all stream paths
Kobayashi et al. (1971)			*									*		Sequential/Parallel stream splitting
Pho & Lapidus (1973)						I			1, 2	1h, 2c				Too restrictive, large tree to search
Ponton & Donaldson (1974)						3		1	1	1h, 1c				Very fast but fallible
Rathore & Powers (1975)						2	2		1, 2	1hc 2hc				Generates enormous tree
Donaldson et al. (1976)						3		1	1	1h 1c				
Cena et al. (1977)			*									*		Sequential stream splitting, Followed by evolution
Linnhoff & Flower (1978a)		*	*			4	3							Followed by evolution
Flower & Linnhoff (1978)									1, 3, 4	1hc 2hc			*	Detailed in text
Greenkorn et al. (1978)		x				2		4	3, 4	1hc 2hc				
Grossmann & Sargent (1978)						2		1	1	1h 1c				Corrected oversight in Ponton & Donaldson (1974)
Grimes et al. (1980)		x	x			3		3	3, 4	1hc 2hc				Followed by evolution

* = New Contribution; x = Used; i = Option i

sizing heat exchanger networks which use minimum utilities and which require the fewest number of exchangers, or nearly the fewest, is very well understood and close to being a solved problem as stated. Useful design strategy insights will still be forthcoming which will allow larger problems to be more readily solved, perhaps even by hand.

The solutions resulting may result in more difficult control and operability problems for the operator. First looks at these problems have already been made by Douglas (1980) and Morari et al. (1980). The operability and control problem will certainly receive more attention in research under way at the present time.

Future work needs to consider other forms of energy integration simultaneously with heat integration for many process designs. Other forms include production and use of mechanical shaft work and/or electricity. Merging this problem with thermally integrating separations systems adds even further dimension to the energy integration synthesis problem. These problems are likely to be the ones worked on in the near future and, in the authors' opinions, will likely have significant results developed for them.

REACTOR NETWORKS

A large number of optimization studies on chemical reactors have been conducted to investigate the effect of temperature distribution, residence time distribution, catalyst dilution profile, etc., on reactor performance. These studies usually considered a single reactor with specified mixing patterns within a reactor. Studies on the optimal configuration of chemical reactors are very scarce. Levenspiel (1962) dealt with the typical two extreme mixing patterns, corresponding to the ideal plug-flow tubular reactor (PFR) and the perfectly mixed or continuous-stirred-tank reactor (CSTR). He proposed several rules on yield and selectivity in these two reactors. The work by Aris (1964, 1969), who applied dynamic programming to determine the optimal amount of by-pass and cold shot streams in a multi-stage reaction system, may be considered as one of the earliest attempts to find the optimal structure among a limited number of possible structures even though it appears to be concerned only with the optimization of a process with a fixed structure.

Horn and Tsai (1967), Jackson (1968) and Ravimohan (1971) treated the optimization of chemical reactor networks with respect to flow configuration. These authors investigated what can be achieved by deliberate manipulation of global and local mixing patterns. Global mixing may be caused by by-pass, recycle, or general feed by-pass. Local mixing is studied by replacing a small part of the reactor with a perfectly mixed cell of equal volume. Using the adjoint variables of optimization theory, these authors proposed an evolutionary method for optimizing the performance of reactor networks by successive adjustments in the mixing patterns. Although their approaches are very promising, the application of their results may be restricted to simple reaction systems such as isothermal, parallel and/or consecutive reactions with fewer than two or three independent reaction steps. For more complex reaction systems, prohibitive computations on the adjoint variables appear to be needed to find the optimal configuration.

TABLE 7. REFERENCES ON EVOLUTION IN HEAT EXCHANGER NETWORK SYNTHESIS

	Rules	Explicit	Implicit	Strategy	Explicit	Implicit	Min Utilities	Min Units	
McGilliard & Westerberg (1972)			x						Evolution Method Based on Primal/Dual Programming
Shah & Westerberg (1975)		*							Present 3 Evolutionary Rules
Nishida et al. (1977)		*				x		x	Purpose to Reduce Number of Units
Cena et al. (1977)			x			x		x	Purpose to Reduce Number of Units
Linnhoff & Flower (1978b)		*				x		x	Purpose to Reduce Number of Units
Grimes et al. (1980)		*				x			Produce Alternative Mine. Unit Solutions

Hartmann (1979) employed the structural parameter method for the synthesis of optimally structured reactor systems as a part of the synthesis of total process flowsheets. He developed a generalized system structure with a PFR and a CSTR using structural parameters. This integrated reactor system structure is essentially the same as the structure used by Umeda and Ichikawa (1972). Several optimal structures were obtained depending on the reactions, the conversion and the ratio of the specific reactor volume cost of the PFR and the CSTR. This work proposed no new systematic method for determining the optimal reactor network structure.

PROGNOSTICATION FOR REACTOR NETWORK SYNTHESIS

Over the last seven years, it appears no work has focused on this synthesis problem. Most of the reported work has been limited to the selection of isothermal stirred tank and tubular reactors. Further, the reaction schemes treated in these works have been very simple first order reactions. There are many reactor configurations (multi-bed adiabatic reactors, reactor with recycles, reactors with heat integration, etc.) and reaction schemes (higher-order reactions, parallel and consecutive reactions, etc.). For the synthesis task, reactors with recycle and with heat integration are very important. The theory and heuristic rules for the synthesis of these types of reactor with various reaction schemes should be developed. They will likely be established using various advanced optimization techniques.

ENTIRE CHEMICAL PROCESSES

The techniques for the systematic synthesis of entire chemical processes, including reactors, separators, energy-transfer equipment, etc., will be classified into: (1) approaches without an initial structure, and (2) structural parameter or integrated approaches.

Approaches without an Initial Structure. Siirola, Powers and Rudd (1971), Siirola and Rudd (1971), and Powers (1972) developed a computer program called AIDES (Adaptive Initial Design Synthesizer), which utilizes systematic heuristic procedures for process synthesis. Since several good reviews (Hendry et al., 1973; Hlavacek, 1978; Westerberg, 1979) on AIDES have been presented, only a comparison of this procedure and the BALTAZAR procedure by Mahalec and Motard (1977a,b) will be made later.

Mahalec and Motard (1977a) proposed a procedure for the sunthesis of promising initial designs of chemical processing systems using the techniques employed for mechanical theorem proving. Underlying this method is the resolution principle (Rovinson, 1965) where the designer attempts to derive conflicts among a set of facts (premises and axioms of chemical processing systems) and the desired goals (desired feasible flowsheet).

The procedure begins with the consideration of production goals (desired product streams) one at a time, and ends with a process flowsheet which is feasible in terms of mass and energy balances. Using a sequential depth-first procedure, the following structural rules are contained: (a) use the compositionally most similar source process streams to generate product streams, (b) give first preference to by-product streams already generated, and (c) reduce the mass load on separation sequences. The same authors (1977b) extended this procedure to generate good (optimal) limiting process flowsheets. The procedure is implemented as a computer program called BALTAZAR.

Redundant portions of the initial feasible flowsheet, which are obtained by the depth-first heuristic search, are eliminated by an analysis of already generated portions of a process structure using a look-back strategy. This look-back strategy is supported by an evaluation function which leads to the optimal integration of separation tasks (GS), as described in the previous section on synthesis of separation sequences.

The structure is further improved by evolutionary search. They described three evolutionary rules: (i) replace a substructure producing a mixture in a process flowsheet by another substructure satisfying the same requirements; (ii) use a by-product stream to satisfy intermediate product stream requirements; and (iii) replace the structure producing stream "S" by another substructure, where stream "S" is produced by the mixing of a recycle stream with another stream.

The BALTAZAR program was successfully employed to produce alternate process flowsheets for the design of monochlorodecane by the direct chlorination of decane. Three different flowsheets were obtained. Two of these flowsheets were obtained earlier by Powers (1971) using the AIDES program.

As pointed out by Mahalec and Motard (1977b), there are fundamental differences between the AIDES procedure and the BALTAZAR procedure as follows:

(1) AIDES performs the stream source/destination matching for the entire flowsheet in one step. It separately considers the flow of each *species* within the flowsheet, developing for each a scoring function which rates each possible source stream/destination stream match. The scoring attempts to account for potential separation costs which might result if the match is made, whether the source has a sufficient amount to supply the destination in terms of that species, etc. After scoring matches for all species, the entire stream/stream matching is done in a single "parallel" step by solving a linear program to optimize the sum of match scores. In BALTAZAR, each destination stream is considered one at a time and is matched to the goal stream which is most similar in composition. This matching procedure is thus done sequentially rather than in parallel.

(2) The AIDES procedure requires the user to determine a species allocation scheme before any process structure is developed. This requirement imposes tight constraints on the possibilities for structure generation by the program in that a conceptual structure is already predicted. In BALTAZAR the procedure allocates resources and matches goals sequentially and stream by stream, creating structure as each match is selected. With no preliminary species allocation, the tight structural constraints do not arise.

The works of Johns (1977), and Johns and Romero (1979) were aimed at the early stages of process development when it is not feasible to optimize equipment dimensions, temperatures, pressures, etc. They qualitatively represent streams within their process by using a number of binary 0/1 flags. A flag may represent high (1) or low (0) temperature, high (1) or low (0) pressure, the existence (1) or nonexistence (0) of a species, etc. Their approach is to select the optimal equipment configuration to transform given raw material streams into desired product streams using a mixture of dynamic programming and branch and bound arguments. The paper does not describe the details of this procedure. The qualitative representation is justified because the program is meant to suggest alternatives at the very early stages of process development. The effort required by the program grows rapidly with the number of binary flags used per stream. It can handle streams characterized by up to about 12 binary flags.

Simple sharp separation problems are well characterized using this representation and are the type of problem first considered by Johns (1977). It is doubtful the approach is suitable for heat exchanger network synthesis.

Integrated Approaches. Since first proposed by Ichikawa, Nishida and Umeda (1969), a number of papers using "structural parameters" have appeared. These methods can be divided into three categories: (a) the analytic and algorithmic methods which employ the necessary condition for the optimal system and then develop a specific algorithm on the basis of necessary conditions; (b) the decomposition and/or transformation methods which decompose or transform the synthesis problem into smaller problems so that the smaller problems are solved separately and their solution coordinated in some way to assure the final solution of the individual problems coincides with that of the overall problem; and (c) the direct application of optimization techniques of nonlinear programming.

For the analytical treatment of the structural parameter approach, Linsley and Davis (1971) introduced a set of structural parameters so that a general structure for discrete, multistage chemical processes was formulated and then derived a necessary condition for a stationary extreme with respect to these structural parameters. They have not proposed any specific algorithm for attaining the optimal structure.

Ichikawa and Fan (1973) derived necessary conditions for the optimal system using the structural parameter approach. An evolutionary search for the optimal structure (ESOS) was developed, starting from a simple feasible structure. Although their method has been shown to be adequate to solve a simple reactor-separator synthesis problem, no attempt has been made to handle a large-size problem.

Decomposition techniques may be one possible way to solve the structural parameter synthesis problem. To ease the difficulty of computations for structure optimization problems, several authors have proposed decomposition techniques. Osakada and Fan (1973) used an infeasible two-level technique in conjunction with the structural parameter approach. Their method was applied to the synthesis problem of a simple reactor-separator synthesis problem. Stephanopoulos and Westerberg (1975) developed an infeasible two-level method, into which Hestene's method of multipliers was incorporated. A penalty term is used to guarantee the success of the method in the presence of functional non-convexities often encountered in chemical process design (Stephanopoulos and Westerberg, 1973). Nishida and Powers (1978) proposed a feasible two-level method, which consists of the first-level and the second-level problem. In the first level, the objective function of the system is minimized with respect to design variables with a given set of structural parameter values. In the second-level, the structural parameters are updated to minimize the same objective function. These previous authors (Osakada and Fan; Stephanopoulos and Westerberg, and Nishida and Powers) tested their two-level methods on a simple reactor separator problem which had been originally solved by ESOS of Ichikawa and Fan (1973). The methods of Osakada and Fan, and Stephanopoulos and Westerberg (1975) required a great number of iterations to obtain the optimal structure, while the two-level method of Nishida and Powers (1978) required only three iterations in the second level calculation using the conjugate gradient method.

Several authors have used nonlinear optimization techniques to solve various synthesis problems of chemical engineering interest. Umeda et al. (1972) used a direct search technique, namely Box's Complex method to synthesize a chemical process system consisting of two reactors, two distillation columns and several heat exchangers. Mishra et al. (1973a,b) used the simplex pattern search technique to synthesize a biological wastewater treatment system composed of a trickling filter, an activated sludge aeration vessel, and a secondary clarifier. Mehrotra and Kpur (1974) applied the integrated approach to determine the optimal configuration of a flotation circuit for separation of mineral species. Himmelblau (1975) tested several currently available nonlinear programming (NLP) codes in order to test their applicability to the solution of the structural nonlinear optimal synthesis problem. These NLP codes were applied to a synthesis problem of a biological wastewater treatment system of Mishra et al. (1973a,b). Himmelblau reported that three penalty function type NLP codes performed poorly, and the only successful NLP code was the Generalized Reduced Gradient (GRG) method of Abadie and Guisou (1969).

Sargent and Gaminibandara (1976) suggested the use of integrated approach to determine the optimal configuration of multiple distillation column systems. The general system for four components which they considered are shown in Figure 15. It can be shown readily that all possible functionally distinct configurations are contained in the system shown in Figure 15. Any particular configuration can in principle be obtained from this general system by deletion of column heat-transfer devices, or interconnecting streams. Clearly, to attempt an optimization over this system results in a nonlinear program with an enormous number of variables and probably a prohibitive computational effort. As they described, their studies of multiple-column systems are at an exploratory stage and powerful techniques to deal with large-scale nonlinear programming problems are badly needed.

Nishida and Powers (1978) used a simultaneous method in conjunction with the Box's Complex method to synthesize a biological wastewater treatment system. They have shown that by the use of a transformation in constrained structural parameters, all the constraints on the structural parameters are eliminated, and this transformation makes it possible to ease significantly the computational difficulty of handling constrained structural parameters. Furthermore, an important observation was reported by Shah and Westerberg (1977), when applying the structural parameter approach to a problem with inequality constraints. If the structural parameters are to be used for a synthesis problem, and if inequality constraints are involved, the problem has to be formulated very carefully to make it a continuous one (Shah and Westerberg, 1977).

As a conclusion of this section, we refer to state of the art reviews on the methods using structural parameter approach by Nishida and Powers (1978): (i) for a small-size problem, the Box's Complex method with a transformation in constrained structural parameters as proposed by Nishida and Powers, and the Generalized Reduced Gradient method of Abadie et al. (1969) work favorably, but the decomposition may not be effective; (ii)

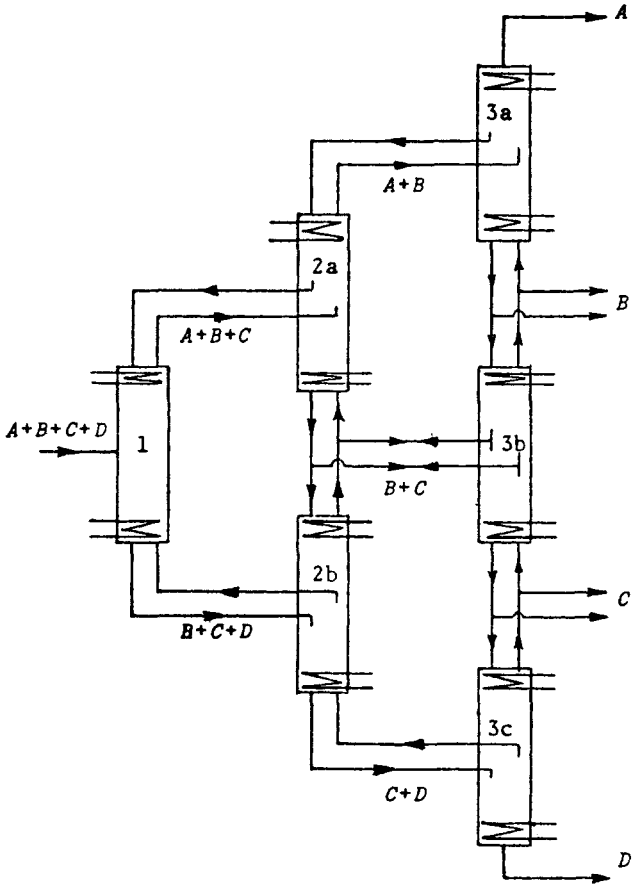


Figure 15. General system for four components.

for a large-size problem, no conclusion about the method to be adopted can be drawn, since no computational experience with a large-size example has been reported; and (iii) the exploitation of powerful methods using structural parameters to large-size problems must be left to future work.

PROGNOSTICATIONS FOR ENTIRE PROCESS SYNTHESIS

Recent research on the use of the second law of thermodynamics for the analysis of process systems is very promising. Second law analysis will be a powerful tool to compare the energy efficiencies of suggested schemes. Since the second law of thermodynamics itself cannot be directly used as a synthesis tool, the initial and/or alternate flow schemes have to be generated or modified by other strategies such as the use of heuristics or evolutionary rules. Therefore the second law analysis will be effectively used in the framework of the evolutionary strategy of process synthesis. The efficient combined use of computer-assisted heuristic-evolutionary approach (like AIDES and BALTAZAR), and the second law analysis of thermodynamics holds promise.

The success of the integrated approach depends highly on the successful use of nonlinear programming methods. Therefore the size of problems, which can be solved using the integrated approach, will become larger with the development of efficient nonlinear programming methods and their computer codes.

SYNTHESIS OF CONTROL STRUCTURES FOR CHEMICAL PROCESSES

The recent critique papers and workshops on process control (Foss, 1973; Douglas, 1973; Foss and Denn, 1976; Kestenbaum et al. 1976; Pallai, 1975; Lee and Weekman, 1976) have all pointed out the lack of a systematic approach towards the synthesis of control structures for a complete chemical plant.

Existing control theories assume that measured and manipulated variables have been selected, and the control structure is thus specified. Most of the emphasis is then placed on the design performance aspects of the controllers. Due to the lack of a theoretical systematic approach, the design engineer is usually guided by his experience to decide which variables to measure and manipulate.

Only very recently attempts have been made to elevate process control from the unit operations control level to the higher level of plant control (Govind and Powers, 1976, 1977a; Morari, 1977; Umeda et al. 1978). Before presenting the most recent results in process control structure synthesis, let us now formulate the control synthesis problem.

Chemical processes are large scale structured systems composed of unit operations interconnected through process streams flowing between them. The independent input variables to a chemical process can be classified as disturbances and manipulated variables. Disturbances are either random noise-like variations or persistent changes in the variables external to the process boundary.

The other inputs over which we have complete control and can freely adjust are the manipulated variables. It is through the manipulation of such variables that certain control objectives are met in the presence of measured or unmeasured disturbances entering the process.

The other important process variables are the outputs which are further classified as measured and unmeasured. Quite often, the output variable of primary interest for control cannot be measured and there exist other process outputs that are easier to measure and used to infer the values of unmeasured output variables.

Once the process variables are classified as above, a *control structure* will be composed of:

i) a set of control objectives;
ii) a set of variables to be controlled to achieve the control objectives;
iii) a set of variables which can be measured for monitoring the behavior of the plant;
iv) a set of manipulated variables;
v) a structure interconnecting measured and manipulated variables (control loops).

The main task of chemical process control synthesis is to structure a dynamic system of measurements and manipulated variables so that certain control objectives are satisfied in the presence of disturbances. The control structure synthesis will then explore the question of which variables should be measured, which variables should be manipulated and how these two sets of variables should be interconnected to form the control loops.

We next proceed through a sequence of steps by defining each element of control structure.

Selection of Control Objectives. The nature of control objectives will depend on both the process characteristics and the plant management policy. The first class of objectives requires certain process variables to be kept at given set-points or within specified bounds, in the face of external disturbances. These objectives will usually originate from the following requirements:

- Product quality specifications
- Safety aspects
- Operational requirements
- Environmental regulations, etc.

The second class of objectives stems from:

- Economic considerations.

The first class objectives defines usually the regulatory control tasks while the second class of objectives defines the optimizing control attitudes.

Selection of Controlled Variables. The regulation objectives directly dictate the controlled variables such as reactor temperatures, product concentration, etc., which will lead to the satisfaction of the given objectives. On the other hand, translation of

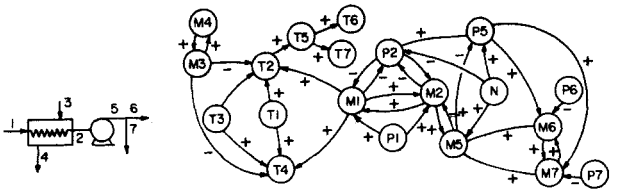


Figure 16. Interconnection of the cause-and-effect graphs for individual units in a process to obtain the graph for the complete system (Govind and Powers, 1976).

economic optimization objectives into process controlled variables is not a trivial task, and under what conditions this can be accomplished will be a subject to be explored in the present section.

Selection of Measurements. The control objectives identify the controlled variables as the primary set of measurements which should be made. These theoretically desirable measurements are not always available to monitor our control objectives, and they have to be replaced by secondary measurements. Through secondary measurements, primary variables can be estimated using a process model.

During the synthesis of control structures, alternative sets of measured variables are to be developed. Which one of these alternative sets should be finally selected becomes an important question for the selection of the best control structure.

Selection of the Manipulated Variables. Proper selection of the manipulated variables will strongly affect the capability of the controller to maintain the control objectives at desired levels in the face of disturbances.

Selection of Structure Interconnecting Measured and Manipulated Variables. The existing structures range from single-input, single-output, noninteracting loops, to multivariable control schemes such as decoupling, modal, optimal or robust controllers. The structure selection will be guided by engineering, cost and theoretical considerations, and the freedom to choose a structure will influence the selection of variables.

A Steady-State Design Procedure Using Cause-and-Effect Digraphs

Govind and Powers (1976, 1977a), in their control system synthesis, try to develop a piping and instrumentation diagram from a steady-state process flowsheet.

Control Synthesis Procedure. The first step is to obtain the steady state flowsheet for the process together with equipment design specifications and steady state operating conditions. For the purpose of control system synthesis, the following modeling aspects were found to be most suitable. (i) Input-output models representing steady state mass, energy, momentum balances through cause-and-effect graphs. (ii) First order lags plus dead time models to represent dynamic relationships between the process variables.

Cause-and-effect relationships between different variables of a system can be represented by a digraph called the cause-and-effect graph of the process flowsheet. Nodes in the graph are the process variables and the edges show the relationships between variables. The edges can carry information about cause-and-effect relationships such as the steady state gains between the variables. The gain between two variables of the cause-and-effect graph is defined as:

$$\text{Gain} = G(A, B) = \left.\frac{\partial B}{\partial A}\right|_C$$

where A and B are the cause-and-effect variables, respectively, and C is the independent variable.

The cause-and-effect digraph for a heat exchanger-pump system is shown in Figure 16 where directed edges indicate how process variables affect each other as indicated by steady state energy and momentum balance relationships.

The dynamic relationship between two variables is modeled as a first order response with dead time, with a transfer function:

$$G_i(s) = \frac{e^{-T_{Di}s}}{1 + T_i s}$$

Parameters T_{Di}, T_i can be either experimentally measured or calculated from models using model reduction techniques. Cause-and-effect relationships between variables can then be solved by following the edges between process variables to calculate the important parameters such as gains, time constants and dead time.

After the cause-and-effect graph for a complete process is generated, the next step is to determine the constrained variables.

Constrained Variables. The control objectives define the process variables to be maintained at desired set-points, and these variables are called the constrained variables. Furthermore, those variables which violate production, safety or operational limits due to disturbances are also identified as constrained variables. Disturbance effects are calculated using linearized process models. After all the constraints have been identified, the next step is to establish a constraint logic.

Constraint Propagation Logic. The goal of propagating the control constraints through the cause-and-effect graph is to locate alternative sets of measured and manipulated variables in the graph to satisfy objective constraints. Constraint variables can be propagated in the forward or reverse directions of process information flow. The direction of process information flow is defined by the directed edges on the cause-and-effect graph. As the constraints are propagated, process variables are encountered along the edges which are classified as ANDed or ORed by the following rules:

1) Input variables are ANDed if all the input variables are required to be controlled in order to control the constrained variable. Two variables (A, B) are ANDed with respect to a given constraint C if both A, B directly affect C and the process information flow is in the same direction as the control information flow. The direction of control information flow is from the measured to the manipulated variables.

2) Input variables are ORed if control of either of the input variables is sufficient to control the constrained variable. OR condition arises only if the variables are not ANDed by the above rule.

After all the candidate control structures are formed from alternative sets of measurements and manipulated variables, they are evaluated based on selection heuristics to reduce the alternatives to a smaller subset. Any detailed analysis and dynamic simulation is performed on this subset to obtain the most satisfactory structures.

The procedure of Govind and Powers suffers from several weaknesses: (i) it cannot identify integrators which need special attention to guarantee stability; (ii) it does not allow for flexible operation at various set-point levels during optimizing control; (iii) it has no efficient method for identifying the best (stability or dynamically) set of secondary measurements to estimate the unknown (unmeasured) control objectives.

Multilevel Design Approach to Synthesize Regulatory Control Structures

Umeda and Kuriyama (1978) developed a different approach to synthesize regulatory control structures for a large scale chemical plant. The synthesis activity is decomposed into a series of process design activities. The first level of design activities starts by generating alternative control structures for each single unit in the plant. The second level of design activities is concerned with coordinating the unit operations' control structures to arrive at a plant control strategy by minimizing the conflicts among the control attitudes in the various unit operations.

Mathematical Formulation. Let C_{nj} define the jth alternative control structure for the nth unit. C_{nj} ($j = 1, \ldots J$) is an

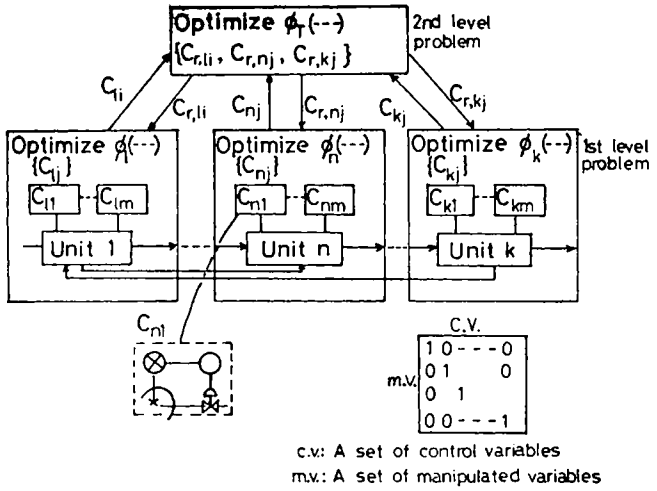


Figure 17. Hierarchical structure for process control system synthesis.

alternative control structure composed of several control loops c_{ni} ($i = 1, \ldots m$) such as pressure, temperature, flow rate control:

$$C_{nj}: \{c_{n1}, c_{n2}, \ldots, c_{nm}\}$$

The problem of the synthesis of a control structure for the nth unit in the first level is then formulated as follows:

$$\underset{C_{nj}}{\text{Optimize}}\ \Phi_n(c_{n1}, c_{n2}, \ldots c_{nm})$$

subject to the operational constraints for the nth unit.

The control synthesis problem for each unit in the plant is to select its best control structure so that its control performance Φ_n is optimal. The alternative control structures are generated by finding the degrees of freedom after analyzing material, energy, momentum balances in each unit and introducing control objectives to eliminate these degrees of freedom. Heuristic rules, dynamic simulation, economic evaluation guide the designer to select the best control structure for each unit.

Since the first level considers the controller design for each unit independently, conflicts among these controllers are expected to arise when they are put together to form the final plant control structure. The combination of unit controllers to form the overall control system will reduce the degrees of freedom and overspecification has to be avoided.

Hence, the design activity at the second level attempts to revise the best set of unit control structures so that overall control objectives are satisfied or the overall performance is optimal.

The second level problem is mathematically formulated as:

$$\text{Optimize } \Phi_T(C_{1i}, C_{2j}, \ldots, C_{Nm})$$

$$\{C_{r,1i}, C_{r,2j}, \ldots, C_{r,Nm}\}$$

Subject to overall operational constraints

where $C_{r,nj}$ is the revised set of control structures as given by the coordinator to the nth unit.

The iterative and multilevel synthesis ends when the revised control structures $C_{r,1i}, \ldots, C_{r,Nm}$ are the same as the best control structures designed at the first unit operations control level, i.e., $C_{1i}, \ldots, C_{Nm}$. The overall synthesis strategy is shown in Figure 17. The information about the unit control structures is passed to the second level through a control structure matrix with diagonal elements defining the control loops. The second level activities then involve logical processing of this given information.

Other Procedures for Synthesis of Control Structures

Before any systematic approach to the control structure synthesis problem was attempted, the plant control versus unit operations control had been first mentioned by Buckley (1964). Casting important views from industry, control structure synthesis was studied in the context of two main process control requirements for plant control: i) product quality control, and ii) material balance control.

Product Quality Control. From the plant manager's standpoint, the plant must produce the product with a quality meeting sales specification. The product quality control system must act as a regulator for random disturbances and as a servo system for product quality set-point changes (dictated by the management).

Material Balance Control. This control objective includes the following aspects:

1) Inventories must be maintained between minimum and maximum limits.

2) Production rate must be adjusted to exactly equal, on a long-term basis, the sales rate (or rate of shipments).

3) The process flow adjustments must be smooth and gradual not to upset equipment.

The measured variables for material balance control are accumulations (i.e., liquid levels) and the manipulated variables are flow rates. Material balance is satisfied when the accumulation is not changing. Buckley points out two types of material balance control: control in the flow direction and opposite to the flow direction. The material balance control opposite to flow has the advantages of faster response to production rate changes with less product inventories.

Having established the main control requirements, the synthesis of the process control structure is composed of the following two activities:

1) With the process flowsheet in hand, the process engineer starts from the product end of the process following the stream flow back to the raw material storage and constructs the material balance control by locating measurements and manipulations for the intermediate equipment. In doing so, either a) control in the flow direction or b) control opposite to flow direction is chosen.

2) After the material balance control system is developed, one starts looking at the product quality control. Starting from the product end and tracing backwards along the process streams, product quality controls are located. Measurements and manipulations are chosen using heuristics similar to those we have discussed in other approaches. Steady-state and dynamic evaluation of control loops are done using frequency response methods.

A more recent approach towards the synthesis of control systems as suggested by J. M. Douglas is based on the quick estimates of control economics. The objective is to obtain quick estimates of the economic impact of disturbances on the process and find the control structure to compensate for their effects.

Since the procedure depends on order of magnitude arguments, it is formulated in terms of specific case studies, lacking a general systematic approach. The specific case study is the acetone recovery process (Douglas, 1977a). The procedure starts with determining the effects of disturbances on the total plant operating cost at steady-state conditions. After several approximations one obtains the sensitivity of the total operating cost to different input variables (Douglas, 1977b):

$$\text{Operating Cost TAOC} \cong 91{,}300 + 11{,}700\ \Delta Y_{in} + 10{,}800\ \Delta L + 80{,}500\ \Delta G + 433{,}900\ \Delta T_L$$

where for the absorber:

Y_{in} = feed mole fraction
L = liquid rate mol/h
G = gas rate mol/h
T_L = inlet liquid temperature

Operating costs are most sensitive to liquid temperature disturbances whose effects have to be compensated by the control structure to be synthesized. From minimization of the operating

costs, stationarity conditions yield the following relation:

$$\frac{L}{mG} \cong 1.38[1 + 0.066\ \Delta Y_{in} - 0.421\ \Delta T_L]$$

The conclusion is that for reasonable changes in disturbance Y_{in} and especially in T_L, L/mG remains constant which suggests the following control policy: control L/mG at fixed design set-point in the face of disturbances.

The economic incentive can then be calculated to determine whether or not to install a controller to maintain L/mG constant. After establishing the incentive for steady-state control, more detailed dynamic analysis considering transient behavior can be carried out to estimate the incentive for installing more sophisticated feedback controllers such as optimal or other types of multivariable control.

An Integrated Approach to Synthesis of Process Control

In the previously outlined control synthesis methods (Govind and Powers, 1976, 1977a; Umeda and Kuriyama, 1978; Douglas, 1977a,b), such important features as follows have not been incorporated into the synthesis approach: i) The differentiation of regulatory and economic control objectives and their implications have not been analyzed. Although regulatory control objectives are most of the time well-defined by product quality, safety, etc., formulation of economic objectives in terms of the controlled variables is not trivial. ii) The above classification of control objectives implies decomposition of the control tasks into: (1) regulatory and (2) optimizing ones. The synthesis procedure must then involve constructing regulatory and optimizing control structures. iii) Classification of disturbances in terms of their impact on process performance has to be established to assess the need for regulatory and optimizing control structures. iv) Decomposition methods are necessary because the dimensionality of the problem is overwhelming. How to decompose the plant becomes a central question. Whereas the finest decomposition is to the unit operations, this requires excessive coordination either during the multilevel design method (Umeda and Kuriyama, 1978) or more importantly, during the on-line optimization of the process. Some of the unit operations can be combined to form a larger subsystem with a common functional goal in terms of control and economics. After the process is decomposed to its subsystems, the synthesis is simplified since regulatory controllers will now have to be structured within each subsystem.

The integrated approach developed by Morari (1977) and Morari, Arkum and Stephanopoulos (1978) incorporates all those aspects into the synthesis. Within the hierarchical control concepts, useful criteria are developed to classify the disturbances, to translate economic objectives into controlled variables, and to decompose the control tasks and the process. Multilevel optimization theory together with engineering motivations are used to develop the decomposition criteria. Decomposition aspects and structuring of optimizing controllers will be the subject of the next section where the motivation and rigorous mathematical developments will be given.

Modeling. The structural characteristics in the form of functional dependencies between process variables were used to describe the processing units and the interactions between units.

The structural presentation not only simplifies modeling but also constitutes the starting point for structural controllability and observability concepts which lead to the generation of alternative control structures as we will see later.

Decomposition for Synthesis of Regulatory Control Structures. After the process is decomposed into its subsystems (Morari, 1977), the regulators will be designed for each subsystem. For the synthesis of regulators, the subsystems will be further decomposed to facilitate the synthesis. The following operations will be applied to a subsystem of units obtained from the process decomposition: (1) precedence ordering and grouping of units; (2) decomposing further the chain of groups to sections based on the following conditions:

a) The number of manipulated variables is at least equal to the number of control objectives in each section.

b) The effect of manipulating a variable in one section is too slow to control the objective in another one.

(3) The irreducible groups along the chain will be decomposed according to the principles in (2).

The streams leaving a "slow" unit will be conceptually torn; in other words, the intensive properties of those streams will be regarded as disturbances on the interconnected units and not as manipulated variables.

The decomposition approach eliminates the synthesis of undesirable control structures from the beginning, thus reducing the potential combinatorial problems encountered in other approaches (Govind and Powers, 1976).

Generation of Alternative Control Structures via an Algorithmic Procedure. The main concepts used here are those of structural controllability and observability. For the following developments consider the following structural dynamic model,

$$\dot{x} = Ax + Bm,\ x \epsilon R^n,\ m \epsilon R^m$$

with outputs

$$y = Cx \qquad y \epsilon R^n$$

and A, B, C structural matrices (with elements 1 or 0).

Theorem 1: The structural pair (A, B) is structurally controllable if and only if the following two conditions are satisfied:

1) Each state node is accessible from at least one control node.

2) $\rho_G[AB] = n$. Where $[AB]$ is the composite structural matrix formed from the n columns of system matrix A and the m columns of controller matrix B.

Theorem 2: The structural pair (C, A) is structurally observable if and only if the structural pair (A^T, C^T) is structurally controllable. If all state nodes are accessible, pole/zero cancellation can occur only at the origin. The generic rank condition $\rho_G(AB) = n$ is equivalent to testing for the existence of a pole/zero cancellation at the origin. It detects pure integrators which are not controllable with a given set of manipulated variables. For stable systems, accessibility only to the states which are pure integrators is important.

Extended Controllability and Observability Concepts for Feedback Structures. For the regulatory tasks let us consider that we will control our objectives at set-points using PI control. Inclusion of integral action will result in the following dynamic system:

$$\begin{bmatrix}\dot{x}\\ \dot{z}\end{bmatrix} = \begin{bmatrix}A & 0\\ C & 0\end{bmatrix}\begin{bmatrix}x\\ z\end{bmatrix} + \begin{bmatrix}B\\ 0\end{bmatrix}u$$

where $z(t_1) = \int_{t_0}^{t_1} Cx dt = \int_{t_0}^{t_1} y dt$ and $u = K\begin{pmatrix}x\\ z\end{pmatrix}$.

For the feasibility of such a control structure,

$$\left[\begin{pmatrix}A & 0\\ C & 0\end{pmatrix}, \begin{pmatrix}B\\ 0\end{pmatrix}\right]$$

has to be controllable.

Theorem 3: The pair

$$\left[\begin{pmatrix}A & 0\\ C & 0\end{pmatrix}, \begin{pmatrix}B\\ 0\end{pmatrix}\right]$$

is structurally controllable if and only if the following conditions are satisfied:

1) (A, B) is structurally controllable

2) $\rho_G\begin{pmatrix}A & B\\ C & 0\end{pmatrix} = n + r,\ r = dim(y)$.

During the synthesis procedure, alternative sets of manipulated variables can be generated satisfying the above feasibility conditions for a given set of observations. A synthesis based on the cause-and-effect checks the accessibility only. Consequently, the method of Govind and Powers (1976) may generate infeasible control structures where the generic rank test will fail, while accessibility is satisfied.

The Algorithm. The algorithm to generate feasible alternative regulatory control structures will now be presented:

1) Choose the controlled variables resulting from control objectives as your observations. Replace any unmeasured desired variable with secondary measurements.
2) Treat each subsystem i obtained through the process decomposition separately in the following steps.
3) Test for accessibility using the selected observations (C_i). Augment C_i if necessary to achieve accessibility (C_i^*).
4) Form the structural matrix:

$$S_i = \begin{bmatrix} A_i & B_i \\ C_i^* & D_i \end{bmatrix}$$

where all the feasible "convenient" manipulated variables make up the columns of $[B_i]$.
5) Delete columns from

$$\begin{bmatrix} B_i \\ D_i \end{bmatrix}$$

such that the number of remaining variables

$$\begin{bmatrix} B_i^* \\ D_i^* \end{bmatrix}$$

is equal to the number of observations.
6) Test for accessibility for the system (A_i, B_i^*). Choose a different B_i^* if necessary to achieve accessibility.
7) Find an output set assignment for S_i by the method described above. All manipulated variables which do not appear in the output set can be eliminated.
8) A manipulated variable in the output set can only be eliminated if after deleting the corresponding column, an alternative output set is found using "Steward path" (Steward, 1962; Westerberg et al., 1979).
9) The manipulated variables corresponding to the columns of B_i^* and observations corresponding to the rows C_i^* form a feasible control structure.

The above algorithm is easy to implement on a computer. It allows a very quick development of all possible control structures. The screening of these alternatives that follows the synthesis part proceeds at different levels of sophistication:

a. Use engineering heuristics to reject unacceptable solutions.
b. Use steady-state considerations (static interactions among input-outputs, gains, etc.)
c. Use simple dynamic measures (time constants, time lags, etc.)
d. Introduce screening based on dynamic interactions between inputs-outputs.
e. Do a dynamic simulation

As we proceed from step a. to step e. fewer and fewer alternatives remain valid.

PROGNOSTICATIONS FOR CONTROL STRUCTURE SYNTHESIS

To a large extent, the synthesis of control systems for complete chemical plants is a problem within a steady state environment. As such, future work should be oriented in two main directions:

- Establish the critical variables which will affect in a profound manner the basic structure of a control system, and develop the central feedforward and feedback configurations for material balance and quality controls.
- Identify the points of a process where there is significant but exploitable interaction between the design characteristics of the process and its steady state control.

Douglas (1973) has tackled recently these two problems using order-of-magnitude arguments with very useful results.

An additional important problem is the construction of simple single or multiple screening criteria which can incorporate the steady state or dynamic considerations for a process at various levels of complexity. Presently, the selection is:

- Either analytic and mathematical but limited to a single unit operation, or
- Heuristic and ad hoc for larger processing systems.

Eventually, the synthesis of control systems for complete plants should include the following important features:

- Allow fast and smooth transitions of the operation of the plant.
- Permit flexibility for automatic start-up, shut-down and change-over in its operation.
- Incorporate a flexible safety system.
- Permit the on-line change of the structural interconnections between measured and manipulated variables.

Although such capabilities will find their place in a few years, work should start now and be directed towards the satisfaction of the above goals.

Finally, any work towards the synthesis of control systems for complete plants should try to exploit the available technology of digital systems. The current applications of digital systems for process control are limiting such systems to trivial number crunching and leave untouched the tremendous abilities for developing "thinking" systems. In this direction, the theory of hierarchical systems (multilevel, multiechelon decompositions); the construction of rational monitoring systems which exploit temporal or/and spatial redundancies (for data reconciliation, identification of gross errors, determination of external gross disturbances); the concept of the variable structure control configurations, can be found very useful in promoting and complementing the basic premises for the design of control systems for the plants of the future.

In all the above areas, it is the process engineer who should take the initiative, formulate the problems and provide the solutions. Standardized, already available methodologies are (very often) inefficient.

OTHER APPLICATIONS

It is often necessary to consider the synthesis of the chemical processes with auxiliary performance indexes other than the total cost such as the dynamic behavior, control considerations, safety features, parameter uncertainty, etc.

The optimal synthesis of dynamic process systems was studied by Nishida and Ichikawa (1975). A wide variety of important chemical process operations, such as the start-up or shutdown of equipment, the batch or semibatch operation of processing units, etc., fall into the category of dynamic process systems. They extended the structural parameter approach for the synthesis of steady state process systems to the synthesis of dynamic systems where the behavior of a processing unit is described by a set of ordinary differential equations. By means of ordinary differential calculus, necessary conditions were derived for an optimal dynamic system. Based on the necessary conditions, they proposed and applied a gradient method to the synthesis of a simple semibatch reaction system. Since such a gradient-based method is time consuming, the application of their method is limited to small problems.

Several authors considered the synthesis of chemical processes under uncertainty in the system parameters or in the system evaluation. Nishida et al. (1974) presented an approach for the optimal synthesis of steady state process systems with

uncertain parameters using min-max concepts. Using a structural parameter approach, they derived the necessary conditions for the optimal performance system structure and presented an algorithm for implementing the synthesis method. The results were applied to the optimal synthesis of a reactor-separator system. Nishida et al. (1976) extended these results to dynamic process systems with uncertain parameters. They have demonstrated that whenever the uncertain parameters have a significant influence on the dynamic behavior of the process to be synthesized, the minimax structure will be different from the optimal structure found using the nominal values of the uncertain parameters. The time needed for the minimaximization of the performance function limits the size of the system to which their method can be successfully applied. Liu et al. (1976) employed a structural parameter approach to solve for the optimal system with multiple performance indices. A scalarized technique was used to find a number of noninferior structures for the problem, and it was applied to the synthesis of a simple reactor-separator system.

Nishitani and Kunugita (1979) considered the design problem of selecting among the various flow patterns possible for an arbitrary number of effects in a multiple effect evaporator system. They posed it as a multiple objective problem. The methods for nonlinear multiple objective problems usually require much computation time, since a number of single synthesis problems with a scalarized objective function have to be solved to generate all noninferior solutions. The application of the methods will thus be restricted to fairly small or special structured problems.

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Forces on Immersed Tubes in Fluidized Beds

The external forces imparted by the bed material on tubes in a fluidized bed contribute to failure of the tubes and their support systems. The objective of this investigation was to provide data on tube forces to be used in structural design. Forces on tubes of various lengths were measured in fluidized beds operating at room temperature. The parameters varied in the experiments were superficial gas velocity and tube array height above the gas distributor. The force-time histories consisted of a series of pulses, whose magnitudes were approximately linearly proportional to tube length. Spectral analyses of the time series indicated that the primary frequency composition of the load was below 25 Hz.

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SCOPE

In a variety of technological circumstances, it is necessary to immerse objects in a fluidized bed. In the case of a fluidized bed combustor, the immersed heat exchange tubes are subjected to random loading. The external forces imparted by the bed material on the heat exchange tubes contribute to failure of the tubes and supports. To have a basis for structural design, a definition of the load environment is required. Information on the magnitude and frequency composition of the applied forces is necessary in order to predict the fatigue life of a structure. Data pertinent to the structural design of tubes in a fluidized bed is very limited (Nguyen and Grace, 1978).

The objective of this investigation was to provide data on tube forces to be used in the design of tubes and their support systems in a fluidized bed with primary application toward heat exchange tubes in a combustor facility. Forces on tubes of various lengths were measured over a range of fluidization conditions. Three cold (310°K), fluidized bed facilities were used in the tests. The three beds had the following cross-sectional dimensions: 0.30 m by 0.30 m (1 ft by 1 ft), 0.91 m by 0.91 m (3 ft by 3 ft), and 2.4 m by 0.30 m (8 ft by 1 ft).

Two different types of load cell mechanisms, specifically designed for this application, were used to measure the forces on tubes in these beds. Forces on 5-cm (2-in.) diameter tubes horizontally aligned with lengths of 25, 71, or 244 cm (10, 28 or 96 in.) were measured. The parameters varied in the experiments were superficial gas velocity [1.5, 2.1, 2.7, and 3.3 m/s (5, 7, 9 and 11 ft/s)] and tube array height [25 and 51 cm (10 and 20 in.)] above the gas distributor.

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