Synthesis in the Design of Chemical Processes

Until recently, the subject of chemical process synthesis was largely ignored in the chemical engineering literature despite its importance in chemical engineering practice. However since 1968, attention to the subject has greatly increased and important progress has been made. This review article critically surveys the readily accessible literature in chemical process synthesis from two points of view: theory and applications. It is concluded that a number of effective synthesis tools are now available for application in industrial chemical process design. However, much more remains to be done, particularly with regard to the development of synthesis procedures which can lead to novel processing system structures.

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SCOPE

The essential concern, purpose, and culmination of engineering is design (Asimov, 1962). Too often, a final design is achieved without due consideration to all aspects of the design morphology (Alger and Hays, 1964). Proper design procedure includes the three essential stages of synthesis, analysis, and evaluation (Asimov, 1962; Dixon, 1966; and Jones, 1970). The design process is complicated by the interrelationships existing among these stages. Frequently these interrelationships are complex and cause design to be an iterative process (Gibson, 1968). Analysis is a term that is equally familiar to both practitioners and students of engineering. Starting with a postulated system structure, its behavioral and performance characteristics are determined for a given set of design specifications and system operating parameters (Porter, 1969). Generally, the analysis proceeds by decomposing the system into its component parts and investigating each component separately (Edel, 1967). Synthesis is a term that is not so familiar, particularly to students of engineering. In one sense, synthesis is the opposite of analysis in that the goal is to conceive a system structure that will, upon analysis, meet certain requirements or specifications. In the iterative design cycle, synthesis precedes analysis, but analysis may suggest changes to the synthesis. Ideally, proper design procedure leads to the optimal system structure as well as to the optimal operating conditions for that structure. Thus, optimization, during the evaluation stage, is related to synthesis as well as to analysis.

While engineering analysis has been developed deductively and quantitatively to a high degree, engineering synthesis has remained largely an art that involves rather elusive inductive logic. Furthermore, synthesis requires considerable knowledge of and experience with engineering principles of analysis (Dixon, 1966) before it can be applied effectively. For this reason, modern engineering curricula are devoted almost exclusively to analysis, with only a very modest amount of time, if any, allocated to synthesis. By stressing analysis in engineering education, the importance of creativity, innovation, and invention in engineering have been made to appear minimal.

Some progress has been made in recent decades in developing systematic approaches to synthesis in various types of engineering design for fields other than chemical engineering. For example, Middendorf (1969) mentions synthesis procedures for certain electrical networks, including filters and attenuators. Porter (1969) considers synthesis of dynamic mechanical systems.

In chemical engineering, the literature abounds in examples of chemical process design where synthesis is a required consideration, for example, Sherwood (1963), Oliver (1966), Rudd and Watson (1968), Bodman (1968), Russell and Buzzelli (1969), King (1971), Meissner (1971), Smith (1967-1971), Gallup et al., (1970), King et al., (1967), King and Barnes (1971), Forrester and Lynn (1971), Lingafelter and Lynn (1971), Alesandrini and Lynn (1971), and Thompson and King (1971). However, except for isolated treatments of the synthesis of heat-exchange networks and the synthesis of multicomponent distillation separation sequences, the development of formal and systematic approaches to synthesis in the design of chemical processes dates back only to 1968. Since then, though, the body of literature on this subject has grown rapidly and significant progress has been made. The first technical symposium devoted entirely to process synthesis was held at the AIChE 71st National Meeting, Dallas, Texas, February 20-23, 1972 and a textbook on this subject will soon appear (Rudd et al., 1973). In this paper, we present a critical review of the subject and indicate future direction and possibilities for further progress.

CONCLUSIONS AND SIGNIFICANCE

Although points of view of process synthesis are not readily separable, this review considers process synthesis from two points of view, techniques and applications. Papers dealing with techniques of process synthesis are classified and summarized in Table 1, while papers which include applications of process synthesis techniques to specific chemical processes are classified and summarized in Table 2.

The general techniques that have been developed for

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process synthesis have included the heuristic approach based on the use of rules of thumb, algorithmic methods often involving well-known optimization principles, and evolutionary strategies wherein improvements are systematically made to an initially proposed feasible structure. Often these techniques have been used in combination and in conjunction with process decomposition. They have been applied to both homogeneous process structures and to heterogeneous structures, including entire chemical processes. In many cases, the digital computer can be effectively used to implement the methods.

The heuristic approach has been developed by a number of investigators for determining the optimal sequence of distillation columns for a multicomponent separation. Actually at best, the method can only narrow the possible choices. However, the designer then needs to evaluate only a few structures in detail. The useful heuristics are well-stated by King (1971) based on the work of Heaven (1969).

An algorithmic approach to the sequencing of general multicomponent separation operations is available from the work of Hendry and Hughes (1972). The designer must state the types of operations to be considered and supply the necessary separation functions (for example, K-values for distillation). The approach makes use of dynamic programming in conjunction with list processing to determine the optimal separation scheme.

For determining optimal heat-exchange networks, the algorithmic branch-and-bound method of Lee, Masso, and Rudd (1970) and the evolutionary strategy of McGalliard and Westerberg (1972) have been used successfully to solve complex networks involving as many as six process streams. The algorithmic methods of Nishida et al. (1971) and Kobayashi et al. (1971) have been applied to even larger problems but involve some serious restrictions.

The evolutionary strategy can be particularly effective when utilized by skilled process design practitioners. When applied to the development of general energy-transfer networks, King et al. (1972) were able to generate two unique process structures worthy of patent applications.

A beginning towards the difficult problem of synthesizing entire chemical processes has been successfully achieved by Siirola (1970) and Powers (1971) as described in articles by Siirola et al. (1971), Siirola and Rudd (1971), and Powers (1972). Because the techniques are heuristic and not algorithmic in nature, alternative flow sheets are generated rather than the optimum design. Implemented in a digital computer program called AIDES, the method is already being applied in industry. It can be an effective tool in the hands of both novices and experts in process design.

As may be gathered from the above brief summary and the detailed discussion below, significant progress has been made in just a few years (1968-1972) towards providing the process-design engineer with effective and efficient tools for systematic chemical-process synthesis. However, much remains to be done. In particular, several areas may be cited:

1. Improved methods for determining optimal multicomponent separation schemes. The work of Thompson and King (1972) represents an important step.

2. General methods for achieving optimal energy recovery systems. The work of Menzies and Johnson (1972) efficiently combines heuristics with the branch-and-bound technique to reduce the computational effort required.

3. Techniques for establishing optimal reactor networks. This important topic has received almost no attention.

4. Synthesis techniques based on more elementary modules or operations, rather than on certain preconceived and more restricted modules, so that really novel processes could be automatically synthesized. For example, a synthesis program that would be capable of generating a thermally coupled distillation system rather than an ordinary distillation train, when the former was more optimum (for example, see Brugma, 1937; Petlyuk et al., 1965; Stupin and Lockhart, 1971) would be of considerable value. This synthesis area is perhaps of the greatest interest with respect to industrial applications.

PROCESS SYNTHESIS TECHNIQUES

Processing systems are characterized by two distinct features. The first of these is the nature of the process components and component interconnections and the second is the capacities and operating conditions of these process components. Synthesis of optimal processing schemes requires a directed search over the space of configurational alternatives as well as over the design variable space for each particular process configuration. However, while process optimization using established mathematical programming techniques is rapidly becoming a basic tool of the designer, system structure synthesis has been hampered by the lack of sufficient theoretical guidance. Consequently, the aspect of process design which offers perhaps the greatest economic incentive is usually performed quite empirically, new process flowsheets often being developed from previous experience and by analogy with other processes. Some progress has been made in recent years in developing the necessary theoretical guidance. This section presents a review of significant accomplishments in this field. Techniques utilizing process decomposition, heuristics, direct search, and evolutionary strategy have developed singly and/or in combination with each other.

Process Design Decomposition

The chief difficulty associated with process structure synthesis is that an enormous number of equipment arrangements exist for most processing tasks. Examination of each of these is not possible, and conventional optimization techniques designed to handle discrete variables (representing equipment connections) are generally limited to fairly small problems. Thus, process synthesis beginning at the detailed equipment level is generally not possible. Recognizing this problem, Rudd (1968) proposed an approach to process system synthesis based on decomposition whereby a design problem for which no previous technology existed is broken down into a sequence of subdesign problems until the level of available technology is reached.

Desired system performance is specified by constraints on the set of variables $\mathbf{X}$, which represent, for example, availability of raw materials, required product purity, etc. The region of available technology is represented by $\mathbf{R}$, so that for values of $\mathbf{X}$ within $\mathbf{R}$, system performance can be handled by available equipment and, thus, no synthesis problem exists. For this case, the economics of technology are denoted by $E(\mathbf{X})$. However, if $\mathbf{X}$ does not lie within $\mathbf{R}$, system performance cannot be accomplished using one

piece of existing technology and a composite process must be synthesized. This synthesis must be carried out so as to optimize system economics, that is, subtasks $\mathbf{X}_j$ are sought such that

$$O^*(\mathbf{X}) = \underset{(\mathbf{X}_j)}{\text{Opt}} \ [\Sigma E_j(\mathbf{X}_j)] \tag{1}$$

$$\mathbf{X} = \cup_j \mathbf{X}_j \tag{2}$$

$$\mathbf{X}_i \cap \mathbf{X}_j = 0, \quad \text{for all } i \neq j \tag{3}$$

$$\mathbf{X}_j \subset \mathbf{R}, \quad \text{for all } j \tag{4}$$

Rudd indicated a method by which the solution to the above design problem might be synthesized. The method consists of a decomposition of the original problem into two smaller problems $\mathbf{S}_I$ and $\mathbf{S}_{II}$ such that

$$\begin{aligned} \mathbf{S}_I &= \mathbf{X}_I \cup \mathbf{T} \\ \mathbf{S}_{II} &= \mathbf{X}_{II} \cup \mathbf{T} \\ \mathbf{X} &= \mathbf{X}_I \cup \mathbf{X}_{II} \\ \mathbf{X}_I &\cap \mathbf{X}_{II} = 0 \end{aligned} \tag{5}$$

where $\mathbf{T}$ is a set of artificially imposed tear constraints which unite the two subtasks $\mathbf{S}_I$ and $\mathbf{S}_{II}$ to accomplish the original task defined by $\mathbf{X}$. Rudd pointed out that if a selection could be made between the alternate structures arising from the decomposition of a task into subtasks, then the entire system could be synthesized merely by sequentially decomposing the subtasks to such an extent that existing technology is reached, that is,

$$\mathbf{S}_j \subset \mathbf{R} \tag{6}$$

The basis for such a selection is given by

$$O^*(\mathbf{X}) = \underset{\substack{\text{tear location} \\ \text{tear value}}}{\text{Opt}} \ [O(\mathbf{S}_I) + O(\mathbf{S}_{II})] \tag{7}$$

$$= \underset{\substack{\mathbf{S}_I \text{ and } \mathbf{S}_{II} \\ \text{selection}}}{\text{Opt}} \ [\text{Opt}_{\mathbf{T}}(O^*(\mathbf{S}_I) + O^*(\mathbf{S}_{II}))] \tag{8}$$

The original task $\mathbf{X}$ is divided into tasks $\mathbf{X}_I \cup \mathbf{T}$ and $\mathbf{X}_{II} \cup \mathbf{T}$, and the terms $O^*(\mathbf{X}_I \cup \mathbf{T})$ and $O^*(\mathbf{X}_{II} \cup \mathbf{T})$ are the optimal objective functions that can be obtained by the solution to the subtasks, given the values of $\mathbf{X}_I \cup \mathbf{T}$ and $\mathbf{X}_{II} \cup \mathbf{T}$. The tear constraints $\mathbf{T}$ are free to be adjusted, and the interior optimization merely adjusts $\mathbf{T}$ to optimize the sum of the optimum objective functions of the two parts *I* and *II*. The exterior optimization is over the distribution of $\mathbf{X}$ between $\mathbf{X}_I$ and $\mathbf{X}_{II}$ and constitutes an optimization over system structure.

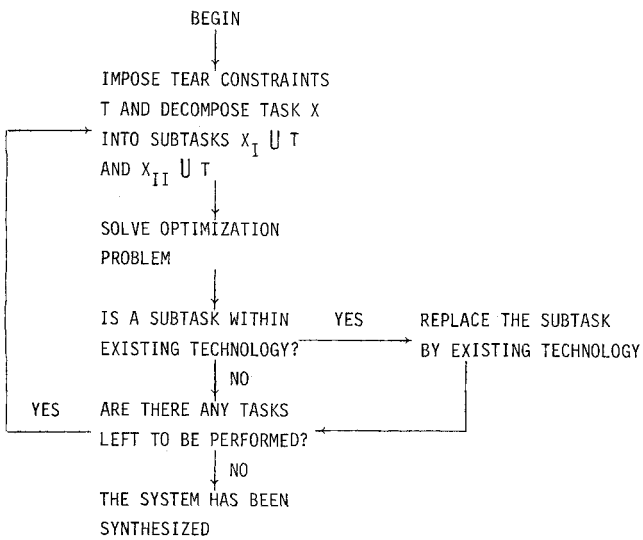

Fig. 1. The synthesis algorithm.

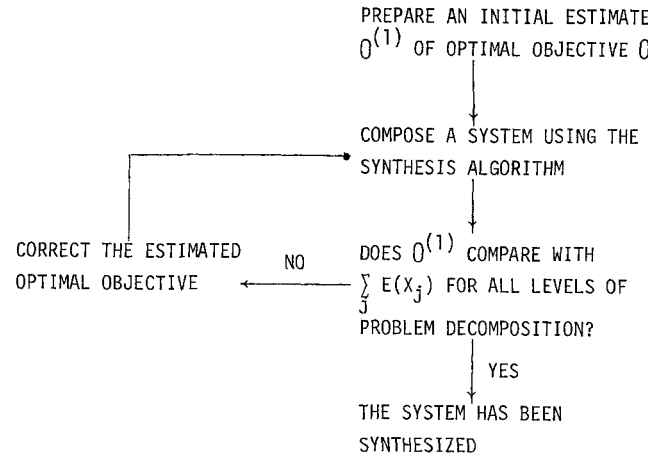

Fig. 2. The accumulation of experience.

TABLE 1. PROCESS SYNTHESIS TECHNIQUES

Technique	References
A. Decomposition	Rudd (1968), Nishida et al. (1971), Kobayashi et al. (1971), Umeda et al. (1972), Menzies and Johnson (1972).
B. Heuristics	Lockhart (1947), Herbert (1957), Masso and Rudd (1969), Nishida et al. (1971), Nishimura and Hiraizumi (1971), King (1971), Siirola et al. (1971), Siirola and Rudd (1971), Menzies and Johnson (1972), Powers (1972), Thompson and King (1972).
C. Algorithmic (optimization)	Rod and Marek (1959), Hwa (1965), Kesler and Parker (1969), Lee et al (1970), Kobayashi et al. (1971), Menzies and Johnson (1972), Umeda et al. (1972), Goto and Matsubara (1972), Hendry and Hughes (1972), Umeda and Ichikawa (1972), Thompson and King (1972).
D. Evolutionary	King et al. (1972), McGalliard and Westerberg (1972), Ichikawa and Fan (1972).

While Equation (8) is a theoretically valid basis for system synthesis, certain difficulties exist which may preclude its implementation. These difficulties arise from the requirement that the optimal objective function be obtainable for any task, including those which do not lie within the region of available technology. For tasks within this region

$$O^*(\mathbf{S}_j) = E(\mathbf{S}_j) \tag{9}$$

where $\mathbf{S}_j \subset \mathbf{R}$. Subject to Equation (9), Rudd's synthesis procedure is illustrated in Figure 1.

Furthermore, Rudd has suggested that synthesis by sequential design decomposition may also be implemented for $\mathbf{S}_j \not\subset \mathbf{R}$ by replacing $O^*(\mathbf{S}_j)$ by some estimated optimal objective function $O^{(1)}(\mathbf{S}_j)$, perhaps formed on the basis of previously solved designs in the particular processing area. This estimate would be used in the synthesis algorithm along with a suitable iteration scheme designed to promote convergence of successive estimates $O^{(1)}$, $O^{(2)}$, $O^{(3)}$, to O^*. The suggested iteration scheme is shown in Figure 2.

Rudd goes on to point out the two kinds of error which may result from the use of an estimated optimal objective function. These are

1. The tear constraint sets $\mathbf{T}$ will not be adjusted to

their optimal numerical values.

2. The tear constraint sets $\mathbf{X}_I \cup \mathbf{T}$ and $\mathbf{X}_{II} \cup \mathbf{T}$ will not decompose the assigned problem into the optimal arrangements of existing technology.

The first kind of error involves erroneous system optimization and may be removed by application of standard optimization techniques to the particular system structure. The second kind of error involves erroneous system configuration, which Rudd maintains may be detected by an observed difference between the estimated optimal objective function, $O^{(1)}$ and the actual objective function of the composed system, $O_{act.}$.

$$O_{act.}(\mathbf{X}) = \sum_j E(\mathbf{X}_j) \tag{10}$$

where $\mathbf{X}_j \subset \mathbf{R}$. Any such difference indicates a need for refinement of the estimated optimal objective function and further iteration. However, it may well be that although the estimated optimal objective function is in agreement with the actual objective function of the composed system, this system is not the optimal structure. For instance, if the estimated optimal objective function accurately represented the objective function of one particular feasible structure and unfairly penalized all other feasible structures, then it would be quite possible that the structure with the accurately represented objective function would be selected as being optimal. Thus, agreement between estimated and observed objective functions cannot guarantee that some other structure with superior system economics does not exist, and so the proposed convergence criterion of Rudd's synthesis algorithm may not always lead to an optimal system structure.

The chief advantage of a process design decomposition approach to system synthesis is that it provides a framework for the systematic synthesis of solutions for quite general design problems. The technique of breaking large, unmanageable tasks into subtasks which may be handled with existing technology or broken down further is a realistic means of coping with the synthesis of complex processing systems. Although certain difficulties arise in attempting to ensure solution optimality using this approach, it nevertheless provides a means by which reasonable solutions may be obtained for problems which would otherwise appear quite intractable.

Attempts to overcome the difficulties associated with the selection of optimal tear locations and tear values have led to the use of heuristic techniques for process synthesis.

Heuristic Synthesis Techniques

Masso and Rudd (1969) indicated a means by which the design decomposition approach to process synthesis could be modified to make use of heuristic problem-solving techniques. A heuristic method is one which seeks the discovery of the solution to a problem by means of plausible but fallible guesses. Heuristic rules of thumb are quite common in chemical engineering practice, examples being the six-tenths-power-law cost approximation, minimum approach temperature in heat exchange, estimates of optimal reflux ratios, and economic fluid velocities. Masso and Rudd utilized heuristic synthesis techniques as a means of overcoming problems associated with evaluation and selection of subtask decompositions according to Equations (7) and (8) and also proposed a modification of Rudd's original decomposition procedure designed to aid in the application of synthesis heuristics.

Problem decomposition, according to Equation (5), is based on a generation of new problems that are smaller than the original problem but which may require further decomposition for solution. Masso and Rudd introduced the requirement that decomposition of the original problem (or any unsolved part of it) must produce subproblems at least one of which is immediately solvable with the available technology, that is, some subtask $\mathbf{X}_j$ is sought which satisfies the relation

$$(\mathbf{X}_j \cup \mathbf{T}_j) \subset \mathbf{R} \tag{11}$$

and, thus, for which there is available an exact economic evaluation $E(\mathbf{X}_j)$. Structuring then proceeds according to

$$O^*(\mathbf{X}) = \underset{(\mathbf{X}_j \cup \mathbf{T}_j) \subset \mathbf{R}}{\text{Opt}} \left[\underset{\mathbf{T}_j}{\text{Opt}} \{O^*((\mathbf{X}_j \cup \mathbf{T}_j) \subset \mathbf{R}) + O^*(\overline{\mathbf{X}}_j \cup \mathbf{T}_j)\} \right] \tag{12}$$

where $\overline{\mathbf{X}}_j$ is the complement of $\mathbf{X}_j$, that is, $\mathbf{X} = \mathbf{X}_j \cup \overline{\mathbf{X}}_j$, $\mathbf{X}_j \cap \overline{\mathbf{X}}_j = 0$. The result of this modification is that optimum structuring must now proceed according to an optimal sequence of soluble subtask selection decisions. Obviously, Equation (12) requires knowledge of $O^*(\overline{\mathbf{X}}_j \cup \mathbf{T}_j)$, the optimal objective function for the unsolved subproblem. However, rather than estimating this with some sequence $O^{(1)}, O^{(2)}, O^{(3)} \ldots$, Masso and Rudd proposed that structuring decisions should be made using heuristic selection of $\mathbf{X}_j$ and $\mathbf{T}_j$ at each stage, that is, a heuristic selection of the subtask to be immediately solved with available technology and heuristic estimation of the variables linking this subtask to the remaining unsolved problem.

Masso and Rudd implemented this heuristic decomposition synthesis procedure as an iterative learning scheme designed to promote convergence of a sequence of trial structures to the optimum processing system. Each of the rules available for subtask selection was weighted, and the set of rules was sampled at every level of decomposition to specify a single base on which the next soluble subproblem would be isolated. Based on evaluation of the exact objective function

$$O^{(i)*}_{act.}(\mathbf{X}) = \sum_j E(\mathbf{X}_j) \tag{13}$$

defined by a sequence of structuring decisions for the ith trial, weights associated with the heuristics used in that trial were adjusted. Thus the selection weights were distorted to reflect experience gained in improving estimates of the optimal objective function.

The procedure was illustrated by application to the synthesis of heat-exchange networks. Here decomposition decisions were concerned with the selection of pairs of streams for heat exchange. The tear variables associated with such a problem decomposition are the intermediate stream temperatures, but Masso and Rudd avoided any necessary optimal selection of values for these variables by heuristics, fixing them so that exchanger duty was maximized subject to a specified minimum approach temperature. The procedure was considered to have converged to $O^*(\mathbf{X})$ when no further improvement could be made after a reasonable number of iterations. Further discussion of their examples is given below. Results of their study showed that while heuristic synthesis techniques appear to have great potential as a means of solving complex design problems, a great deal of further investigation is required before such techniques will be of any practical use. Specifically, Masso and Rudd found that it was necessary to include a random selection rule in the set of decomposition heuristics to guard against possible systematic exclusion of certain types of structures and also to indicate insensitivity of the optimal structure to the particular heuristics used.

In addition, they found that weighted heuristics from the solution of similar problems did not necessarily correspond, that is, there was not necessarily any transfer of experience from one problem to the next. Masso (1968) argued that this was probably due to the fairly specific nature of the heuristics tested and that use of more general heuristics would permit such a transfer. Masso and Rudd concluded that a need existed for systematic compilation of the logic employed in particular design areas and for further development of learning and/or synthesis search techniques.

The combined heuristic design-decomposition approach to process synthesis was applied to the invention of chemical process flow sheets by Siirola, Powers, and Rudd (1971). Beginning with the chemical reaction scheme by which raw materials are transformed into desired products, Siirola (1971) stated that "if the approximate conditions of temperature and pressure for each reaction are specified by the chemist, as well as the conditions of purity required for each reactor feed and the approximate composition of each reactor effluent, the design problem can be effectively decomposed at each reactor." Such a problem decomposition produces as subproblems the various chemical reactors and the unsynthesized processing equipment required to support these reactors. The reactor support system may include such operations as preparation of feed material for each reactor, product separation, purification, and possible finishing of the reaction products.

Synthesis of the reactor-support system begins with the specification of all material routing throughout the process. This level of decomposition is carried out using heuristic scoring functions in conjunction with a linear programming allocation routine. All possible routings of components from points in the process where they are known to exist (raw material sources, reactor effluents) to points where they are required (product streams, reactor feeds) are evaluated according to a set of process heuristics and are assigned scores which attempt to quantify the desirability of carrying out each particular routing. The overall material flow is then selected using the linear programming routine to maximize the sum of the scores from all routings, subject to material balance constraints. This is equivalent to deciding the manner in which the reactor support system (as yet unsynthesized) will meet tear constraints on the various reactors, and may result in breaking down this support system into smaller subsystems such as a feed preparation system, a product purification system, etc. Synthesis of these smaller systems is the next step in the overall design.

Each of the support subsystems must meet certain tear constraints at reactor inlet streams and product exit streams. Meeting these constraints involves changes in temperature, pressure, and composition for streams originating at raw material inlets and reactor outlets. The sequence and manner in which these changes are made for each subsystem are again selected on the basis of process heuristics which specify the required temperature changing, pressure changing, and separation tasks to be carried out.

The final synthesis step is the specification of appropriate equipment for these tasks. However, this synthesis step does not conform to the general design decomposition pattern; rather than simply designating the appropriate piece of equipment for each task, Siirola, Powers, and Rudd (1971) recognized that because of the sequential way in which synthesis was performed, considerable savings could be achieved by integration of certain processing tasks such as heating and cooling, and so they attempted to detect such task combinations prior to selecting the process equipment.

Initial results from this study appear to be quite favorable. For example, Powers (1972) describes an application to a process for the manufacture of monochlorodecane by direct chlorination. However, it is evident that the final process depends heavily upon the nature of the heuristic rules and scoring functions which are used at the various decomposition levels. As pointed out by Masso and Rudd (1969), really satisfactory heuristics have yet to be developed for chemical process design, and so the practical solution of complex design problems using heuristic techniques is fairly limited at this time. Nevertheless, the advantages of such techniques, particularly in combination with some type of problem decomposition approach, should be stressed as they can offer reasonable solutions to quite general, complicated problems without a great deal of calculation, and consequently may be quite satisfactory for establishing initial design concepts.

Synthesis by Direct Optimization

Ichikawa and co-workers have attempted to apply well-known techniques of optimization and mathematical programming to the synthesis of chemical processes. The approach has been to imbed all possible process flow sheets into one combined flow sheet by defining all the interconnections which might exist between various pieces of equipment or systems of equipment. Split fractions with values between zero and one are assigned to all such interconnections and the solution to the synthesis problem is obtained by determining optimal split fractions for each interconnection along with optimal design parameters for each piece of equipment or system of equipment. Values of one or zero indicate whether pairs of equipment are connected or disconnected, respectively.

Based on this characterization of the synthesis problem, Umeda, Hirai, and Ichikawa (1972) used a direct search technique to synthesize a processing system which included two CSTRs, two simple distillation columns and several heat exchangers. Nineteen decision variables were selected to represent equipment interconnections and design variables, and apparently no problems were encountered in obtaining a solution. Recognizing that a major drawback to this approach to synthesis is the high dimensionality of decision variables for optimization, Umeda and Ichikawa (1972) proposed that the technique of Brosilow and Lasden (1965) be used to decompose the optimization problem. This approach was illustrated, as described later, by application to a simple two-reactor system. Results indicated that estimation of the Lagrange multipliers during optimization could cause difficulties and that final optimization of the derived processing system was necessary.

Irrespective of the nature of the optimization technique used to determine optimal split fractions, this approach to process synthesis is based on representation of discrete variables by continuous ones, and it is questionable whether the solution to the continuous optimization problem necessarily corresponds to the solution to the mixed integer problem. It is well known that rounding-off continuous solutions to integer programming problems can lead to nonoptimal solutions. Furthermore, it would also seem quite likely that a continuous optimization approach to the synthesis problem could lead to the determination of local optima in the form of solutions with optimal design and operating variables for nonoptimal structures. The combinatorial nature of the synthesis problem would indicate that continuous optimization solutions would be highly dependent upon the starting values for split fractions and design variables, with a high probability of local optima resulting from unfortunate choices of these. In

view of these difficulties and recognizing that generation of a general flow sheet to include all possible processing concepts is in itself a complicated problem, it would appear that the direct application of optimization techniques to process synthesis is restricted to fairly small problems and may not offer any great advantages in the general synthesis of chemical processes.

Well-established optimization techniques have also been applied to portions of the general chemical process synthesis problem such as heat-exchange networks and sequences of component separation equipment. These techniques include linear programming, dynamic programming, nonlinear programming, branch-and-bound, etc. The use of these techniques is discussed below in the section on applications.

Evolutionary Synthesis

Evolutionary synthesis refers to the synthesis of new processes by modification of previously generated processes. King, Gantz, and Barnes (1972) applied this technique as a succession of alterations involving identification of that portion of the most recent process which could be changed to greatest advantage, followed by generation of the appropriate change for that portion of the process and by an analysis of the new process. As discussed below, they applied their technique to the synthesis of a demethanizer tower in an ethylene plant in which successive improvements were sought to reduce the loss of ethylene to overhead tail gas. No theoretical guidance was given for the selection of process modifications; these were drawn from considerable engineering experience in the particular processing area. A second example detailed the use of the computer in implementing evolutionary synthesis logic for the design of a methane liquefaction process. Taking as design objective the minimization of energy consumption per unit quantity of methane liquified, King et al. (1972) selected the process element to be modified heuristically on the basis of a comparison of losses of available energy of the streams passing through each element. A second level of heuristics then guided the selection of new elements to replace the element to be modified. Further heuristic procedures were used to adapt the altered process to the remaining elements in the previous process. Synthesis of a satisfactory processing scheme was successful using this approach; however, it was recognized that the example was a well-known processing problem and that the principal value of this heuristic evolutionary synthesis technique might well be more in "helping the design engineer to structure his thinking better, rather than in the prospect of an ultimate totally computerized synthesizer."

A somewhat more theoretical approach to evolutionary synthesis was taken by McGalliard and Westerberg (1972), who applied Lasdon's (1970) dual feasible decomposition method for process optimization in a procedure designed to determine whether a feasible modification to a given feasible structure would appreciate system return without having to optimize the entire modified system. The method is based on the generation of dual and primal bounds for the objective function of the modified system and makes use of information on the values of state and adjoint (Lagrange multipliers) variables obtained from previously optimized structures. McGalliard and Westerberg (1972) presented a strategy, based on primal and dual bounding information, by which the best of several proposed modifications could be selected in the synthesis of a process flow sheet, and applied this strategy to several heat-exchange network problems, as discussed below, with satisfactory results.

It should be pointed out that both of the above evolutionary synthesis procedures are necessarily local in their effect. Processes synthesized by these means depend heavily upon the initial assumed processing concept, and so it might be reasonable to apply evolutionary synthesis as a means of improving upon a scheme which has been generated by some more general synthesis technique.

APPLICATIONS OF PROCESS SYNTHESIS TECHNIQUES

The process synthesis techniques described above have been applied to a number of homogeneously and heterogeneously structured processes including heat and general energy exchange networks, sequences of component separation devices, chemical reactor networks, and to entire chemical processes. Important progress has been made and several of the synthesis methods can now be utilized effectively to solve industrial problems.

Heat-Exchange Networks

One essentially homogeneous subproblem of the general process synthesis problem concerns the synthesis of networks of heat exchangers, furnaces, heaters, coolers, condensers, and/or vaporizers to satisfy heating and cooling requirements for a set of process streams. Design of such integrated heat-recovery networks is a complex combinatorial problem involving pairing and sequencing of the exchanging streams. Very often a multitude of feasible configurations exist. For example, Lee, Masso, and Rudd (1970) cite an example involving two process streams to be heated and two process streams to be cooled in which 4200 ways exist for processing the four streams. For most processes, effective integration of heating and cooling requirements offers a significant economic return and, therefore, a great incentive exists to determine process heat-exchange networks in an optimal fashion.

Early investigators dealt with fixed equipment arrangements. Ten Broeck (1944) utilized the calculus to determine the optimal heat-transfer-rate allocation for a battery of three heat exchangers and three auxiliary coolers. Westbrook (1961) presented a more general optimization approach based on dynamic programming and illustrated its application by considering a train of five heat exchangers, five coolers, and a furnace. Fan and Wang (1964) applied the discrete maximum principle to a similar specified network arrangement. While the methods of these three studies could be used to determine the optimal heat-exchange networks, a trial-and-error approach would be required wherein various equipment arrangements proposed by the engineer would be evaluated.

The first systematic approach to the synthesis problem was that of Hwa (1965) in which separable programming (Miller, 1963) was used to eliminate unprofitable exchangers from a single network containing, hopefully, all of the various alternative configurations. In view of the enormous number of possible configurations and difficulties involved in combining them into one model, this approach did not provide any great advantage. Kesler and Parker (1969) considered the selection of the optimal configuration as a network problem in which the nodes represented the various types of heat transfer devices cited above, and the arcs represented the hot and cold streams of the process and auxiliary heating and/or cooling media. Heat transfer coefficients were assumed, thus avoiding detailed considerations of heat-exchanger design. They began by fracturing each stream in the process into heat elements or exchangelets small enough to express the problem objective function with sufficient accuracy as a linear function. A modified version of the assignment problem was used to

select feasible heat-exchange matches between hot and cold stream elements and groups of elements, and linear programming was used to suggest improvements in process configuration. Kesler and Parker claimed that their algorithm could be proven intuitively to lead to a global optimum in a finite number of steps. However, as Hwa (1965) pointed out, the nonlinearities in the equations are hyperbolic in nature, and so convexity of the objective function is therefore not always guaranteed. This may lead to convergence on one of the local optima.

Kesler and Parker illustrated their technique in two examples where the exchangelets were relatively coarse in size (from 1/6 to 1/3 of the stream heat transfer duty). Even so, the main matrices of heat exchange possibilities are large. For reasonably small size exchangelets, their procedure would seem to be of limited utility.

Masso and Rudd (1969) applied their heuristic structuring techniques, as described in an earlier section above, to three heat-exchanger-network problems. In each case, the objective was to structure a system capable of performing the specified heating and cooling tasks at minimum annual costs. One example involved five process streams, while the other two involved seven process streams. The examples were simplified to the extent that overall heat transfer coefficients and maximum allowable temperature approaches were assigned fixed values and all exchangers were operated as simple countercurrent-flow devices. They assumed that auxiliary heating and cooling should be used only when heat-exchange matches are not possible. They manipulated only full process streams. The tear variables selected and adjusted were intermediate stream temperatures. They utilized five rules for selecting the heat-exchange matches. In one of the examples, the difference between the cost of the optimal arrangement and other arrangements was a factor of as large as approximately three.

Lee, Masso, and Rudd (1970) developed an alternative solution procedure based on the branch-and-bound approach to constrained optimization as surveyed recently by Lawler and Wood (1966). In common with dynamic programming, the branch-and-bound technique involves an intelligent structured search of the space of all feasible solutions. Briefly, the procedure as developed by Lee et al. for application to heat-exchange networks consists of a relaxation of the network feasibility requirements to allow multiple use of process streams. Networks subsequently constructed are then ranked according to the value of the objective function (for example, annual cost) and the highest ranked network which does not violate network feasibility requirements is the optimal design, subject to certain limits on cyclicity of information flow. Computational experience with this method indicated that it was much easier to implement than competitive methods and, in addition, provided a guarantee of optimality. However, as with most of the other methods, the size of the problem which could be handled was limited by the enormous number of possible stream matchings. Nevertheless, sample problems involving 4, 5, and 6 process streams and corresponding ways of processing the streams equal to 4200, 3×10^6, and 1×10^{11}, respectively, were solved quite readily.

A technique somewhat similar to that of Kesler and Parker (1969) was developed by Kobayashi, Umeda, and Ichikawa (1971). By making the rather restrictive assumption of equality of all heat-exchange duties (including auxiliary heaters and coolers), and allowing exit process stream temperatures to assume values somewhat different than specified, Kobayashi et al. were able to formulate the problem as one of optimal assignment in linear programming. The solution is obtained iteratively. The calculations are initialized by first assuming the total heat-exchange duty and the number of heat exchangers. The optimal network is then determined by linear programming. The complex method is then used to obtain the optimum total heat-exchange duty for the optimal network. This two-level procedure is iterated until the same network is indicated on successive iterations. Despite the assumption of equality of all heat-exchange duties, in a practical example involving exchange between a crude oil and its distillation products, Kobayashi et al. achieved a more optimum but more complex network than that commonly used in conventional commercial plants. Stream splitting was allowed in the example.

Nishida, Kobayashi, and Ichikawa (1971) considered the same problem as Kobayashi et al. (1971) and utilized the same iterative algorithm to solve for the optimal system except that, as discussed below, they substituted a new graphical procedure for the linear programming technique described above. As in the earlier work, they decomposed the heat-exchange network into an interior subsystem containing all the process heat exchangers and an exterior subsystem consisting of only auxiliary heaters and coolers. The latter were utilized only after heat exchange among streams was accomplished. Because an analytical solution was sought, the following simplifying assumptions were made for mathematical convenience: (1) the cost of the interior subsystem was approximated by a linear function of the heat-transfer areas, (2) the use of countercurrent shell-and-tube exchangers, (3) no phase changes for process streams, and (4) equal values of the effective overall heat-transfer coefficient for all exchangers. The objective was to find the interior process structure and allocation of heat duty within that structure so as to minimize the total heat-transfer area for a specified total heat duty and process stream properties. In the general case, multiple heat ex-

Table 2. Applications of Synthesis Techniques to Chemical Processes

Structures	References
A. Homogeneous:	
Heat-exchanger networks	Hwa (1965), Kesler and Parker (1969), Masso and Rudd (1969), Lee et al. (1970), Kobayashi et al. (1971), Nishida et al (1971), McGalliard and Westerberg (1972).
Multicomponent distillation separation sequences	Lockhart (1947), Harbert (1957), Rod and Marek (1959), Petlyuk et al. (1965), Nishimura and Hiraizumi (1971), King (1971), Hendry and Hughes (1972).
Reactor networks	Ichikawa and Fan (1972), Umeda and Ichikawa (1972).
B. Heterogeneous:	
Energy-transfer networks	King et al. (1972), Menzies and Johnson (1972).
Selection and sequencing of separation processes	Siirola and Rudd (1971), Thompson and King (1972), Powers (1972).
Entire chemical processes	Siirola et al. (1971), Siirola and Rudd (1971), Umeda et al. (1972), Powers (1972), Ichikawa and Fan (1972), Goto and Matsubara (1972), Rudd et al. (1973).

change and splitting were allowed for a given process stream. They utilized a heat content diagram on which streams were represented by hot and cold blocks, whose areas were equal to the heat-transfer rates. The following theorems were utilized to manipulate the heat content diagram so as to achieve an optimal interior structure (unfortunately, a global optimum was not guaranteed):

1. The optimal structure is of the countercurrent flow type;
2. On the heat content diagram, the hot blocks and cold blocks are matched consecutively in decreasing order of stream temperature;
3. The exchanger output temperature of a hot stream being cooled is never lower than the exchanger output temperature for the next hottest cold stream; and
4. If the total heat duty is smaller than both the total amount of heat removed from the hot streams and the total amount of heat added to the cold streams, the higher-temperature portion of the hot blocks and the lower-temperature portion of the cold streams are to be used to exchange heat. As an example, Nishida et al. (1971) solved almost the same problem as Kobayashi et al. (1971) but obtained a different optimal structure. Despite the many assumptions necessary to develop their analytical approach, Nishida et al. did obtain a better network than that commonly employed in commercial plants.

The evolutionary synthesis approach of McGalliard and Westerberg (1972), as described earlier, was demonstrated by them on the same three heat-exchange-network synthesis problems used as examples by Lee et al. (1970). As initial feasible structures, all process streams were heated or cooled by auxiliary units. For two of the examples, the optimum structures determined were identical to Lee et al. However, for the most complex example, involving six process streams, McGalliard and Westerberg obtained a different optimal structure with a slightly higher annual cost than that obtained by Lee et al.

Energy Transfer Networks

More general energy-transfer networks employ not only heat exchange but also expansion, compression, and/or the use of refrigerants for cooling. The systematic synthesis of such complex networks has received only very limited attention despite the great economic incentive in industrial applications.

As mentioned earlier, King, Gantz, and Barnes (1972) demonstrated strategies of systematic evolutionary process synthesis with two examples involving energy-transfer networks. The first example involved a demethanizer column that offered considerable potential for cost reduction because of the normally high operating pressure and extensive use of refrigeration. Starting with an initial feasible structure involving ordinary distillation with a refrigerated partial condenser, their technique led a final synthesis with half the annual operating cost. The final scheme involved tail-gas autorefrigeration, feed prechilling with multiple column entry points, and use of recycle propane to reduce ethylene loss. It is interesting to note that the two best synthesis schemes they generated were considerable improvements over all other schemes and were sufficiently novel to warrant the preparation of patent applications. In a second example, they sought to synthesize a methane liquefaction process corresponding to the minimum energy consumption per unit quantity of methane liquefied. Heuristics for guiding the evolution of the synthesis were implemented entirely on a digital computer. The optimal scheme generated was rather complex involving heat exchangers, refrigerated coolers, expansion valves, compressors, phase separators, and mixers. Again, the optimal structure represented considerable savings in energy over a simple initially assumed scheme.

Menzies and Johnson (1972a, b) described a flexible modular program system for the synthesis of optimal energy recovery networks to meet both stream temperature and pressure specifications. Their technique is based on the branch-and-bound method of Lee et al. (1970) but incorporated heuristics to prescreen prospective stream matches for exchange. This heuristic prescreening increased considerably the size of the problem which could be solved using the branch-and-bound method. In two examples, they applied their technique to the synthesis of energy-recovery networks for both high- and low-pressure ethylene plants. The sequences of distillation columns were fixed in both cases. They permitted countercurrent heat exchange, polytropic single-stage compression, adiabatic (valve) expansion, and adiabatic stream mixing/splitting. For the high-pressure case, the configuration of the resulting optimal energy-recovery network was essentially identical to that of an existing commercial plant. For the low-pressure case, they developed a more optimal configuration than had appeared previously in the literature.

A basic feature of solutions to design subproblems such as the energy-exchange network problems described here and in the previous section is that they serve to extend the limit of available technology in a manner that assists in the solution of larger synthesis problems. Once a reliable method has been developed for the solution of some specific subproblem, that method may be used as a building block in process synthesis because a designer may then specify process subtasks in terms of the solution to one or more subproblems rather than in terms of the elements which make up the solutions to the subproblems. For example, rather than being forced to specify some particular heat-exchange configuration to meet fixed process heating and cooling requirements, the designer need only specify that the requirements be met optimally. He is then free to direct his attention to other aspects of the process—in particular, the aspects that result in determination of process heating and cooling requirements. In terms of the process design decomposition theory outlined earlier, solutions to synthesis subproblems have the effect of reducing the extent to which X must be decomposed so as to reach the region of available technology; that is, so as to satisfy Equation (4).

Multicomponent Distillation Separation Sequences

Multicomponent separation problems commonly arise as subproblems in general process synthesis as a result of a problem decomposition whereby the amounts and destination of several components at some point in the process are set (that is, as tear variables). An example would be the specification of component flow rates in a reactor effluent and the designation of the points in the process (reactor inlets, product streams, waste streams, etc.) to which each component was to be sent. The resulting multicomponent separation problem is concerned with the optimal selection and sequencing of separation equipment (distillation columns, extractors, dryers, screens, etc.) so as to meet the specified process requirements. Several such multicomponent separation problems may exist within a process which may include, for example, feed preparation, reactant recovery and product separation, product finishing, and waste-treatment sections so that design of a satisfactory process is often very dependent upon design of optimal separation sequences.

As discussed in the next section, choice of the appropriate separation operations and their sequencing is a complicated combinatorial problem. However, in many processes,

ordinary distillation using energy-separating agents only is feasible and frequently is the most economically attractive means of separating more-or-less pure components, or groups of components from a multicomponent mixture (Oliver, 1966; King, 1971). The design of individual distillation columns has received much attention in the literature, but only a few studies have been reported that deal with the determination of the optimal sequence of distillation columns. Although the number of possible separation-operation sequences for a homogeneous process system is greatly reduced from the general heterogeneous process system where all possible separation methods are considered, the problem may still be a formidable one when a large number of components must be separated.

As discussed by King (1971), if the mixture contains only components A, B, and C, two distillation operations are necessary and two different sequences of the two distillation operations are possible. With A, B, and C in the order of decreasing volatility, the direct sequence consists of separating A from B and C in the first operation and then separating B from C in the second distillation. The so-called "indirect" sequence proceeds by first separating C from A and B, followed by the separation of A from B. As the number of components to be separated increases, the number of possible sequences increases rapidly. If R components are separated into R products, then $R - 1$ ordinary distillation operations are required, assuming that each receives a single feed and produces single overhead and bottoms products. If S_R denotes the number of different possible sequences, then the following recurrence relation may be developed (for example, King, 1971) from which S_R may be obtained as a function of R:

$$S_R = \sum_{j=1}^{R-1} S_j S_{R-j} \qquad (14)$$

Starting with $S_1 = S_2 = 1$, the values for S_R shown in Table 3 may be generated. As seen, beyond just a few components, it may be impractical to consider in detail all possible sequences.

In an attempt to overcome these combinatorial difficulties, several investigators have recommended the use of heuristic techniques in determining optimal sequencing of distillation columns. Even though these heuristics, as presented below, often conflict, they do have value in that when the number of components to be separated is large, the number of sequences that should be examined in detail can be reduced considerably.

The earliest heuristic known to be applied in practical process design was the simple rule that the components should be removed one-by-one as overhead products from a series of distillation columns. This is often referred to as the direct sequence rule and results usually in a reduction in the operating pressure as the process flow progresses through the series of columns. By utilizing approximate distillation design methods in conjunction with economic analyses, Lockhart (1947) examined sequences for three-product systems commonly encountered in the processing of natural gasoline. He concluded that direct sequence was not optimum when the least volatile component was the predominant constituent of the feed. In this case, he recommended the use of the indirect sequence.

Harbert (1957) presented the principle that the preferred sequence is the one that requires the minimum total heat duty because this factor dominates the economic evaluation. This led to the statement of his two rules, (1) the most difficult separation(s) should be performed with the minimum quantity of feed (that is, placed last in the sequence), and (2) a sequence should be sought which

Table 3. Number of Possible Column Sequences S_R for Separating R Components into R Products (Heaven, 1969)

No. of components, R	No. of possible column sequences, S_R
2	1
3	2
4	5
5	14
6	42
7	132
8	429
9	1,430
10	4,862
11	16,796

favors nearly equimolal division of each column feed into distillate and bottoms products. The latter rule was believed to be the strongest of the two and the one most ignored in design practice at that time.

The first quantitative approach to the optimum distillation sequence problem was taken by Rod and Marek (1959). Their criterion of optimality was the total column vapor flows in the sequence on the assumption that reboiler operating costs were predominant. They verified this proposition only for the benzene-toluene-ethylbenzene system at atmospheric pressure. This system does not represent difficult separations. Using a short-cut design procedure, they were able to develop their criteria in terms of relatively simple algebraic equations containing only the relative volatilities and feed compositions. The direct sequence was used as the standard sequence for comparison. They restricted the application of their technique to only ternary and quaternary cases. Although they developed no general heuristics from their treatment, their criteria appear to be consistent with the findings of Lockhart (1947).

Nishimura and Hiraizumi (1971) expressed the economic function in terms of both the tower volumes and reboiler duties. Again, using a short-cut design procedure, they developed equations for comparing the economics of the possible sequences. From a study of the equations, they stated two sequence-selection rules: (1) For a feed having an approximately equimolal composition in all components and approximately equal relative volatilities between successive ordered components, the direct sequence is preferred, and (2) for a feed that is predominant in one component, it should be the first product distilled out in the sequence. Thus, they verified in a more quantitative and general fashion, the ideas presented by the earlier investigators. Nishimura and Hiraizumi also considered briefly the case where two components of the multicomponent mixture are very close in relative volatility.

Recently, in a fairly comprehensive investigation of optimum sequencing of distillation columns, Heaven (1969), as quoted by King (1971), identified the following four general sequence-selection heuristics based chiefly on energy consumption considerations. Heaven paid particular attention to difficult separations:

1. Direct sequences which remove the components one by one as distillates are generally favored when the remaining heuristics given below do not apply.
2. "Sequences which give a more nearly equimolal division of the feed between the distillate and bottoms product should be favored."
3. Separations where the relative volatility of two adjacent components is close to unity should be performed in the absence of the other components. That is, such a sep-

aration or separations should be reserved until last in the sequence.

4. "Separations involving very high specified recovery fractions should be reserved until last in the sequence." The last two heuristics are a restatement of Harbert's first rule, but in terms of the separate effects of relative volatility and degree of product purity. Heaven's above list appears to include the heuristics of all previous investigations.

More recently, Powers (1971) suggested the use of weighting factors to be applied to the heuristics for determining optimal column sequencing. The particular heuristics he considered were

1. The direct sequence is normally favored,
2. If one component is predominant in a feed, remove it first,
3. Leave the difficult separation(s) until last in the sequence, and
4. Remove any corrosive components early in the sequence. The weighting factors developed for combining the effects of the above four heuristics were tested by Powers on several industrial multicomponent separation problems with favorable results.

As an alternative approach to heuristics for reducing the number of column sequences that must be examined in detail to arrive at the optimum sequence, Hendry and Hughes (1972) effectively applied the technique of dynamic programming (Bellman, 1957) in conjunction with a list-processing operation that can be implemented on a digital computer. Because the approach of Hendry and Hughes is a general one that can be applied to both homogeneous and heterogeneous separation-process systems, a description of their method is deferred to the next section.

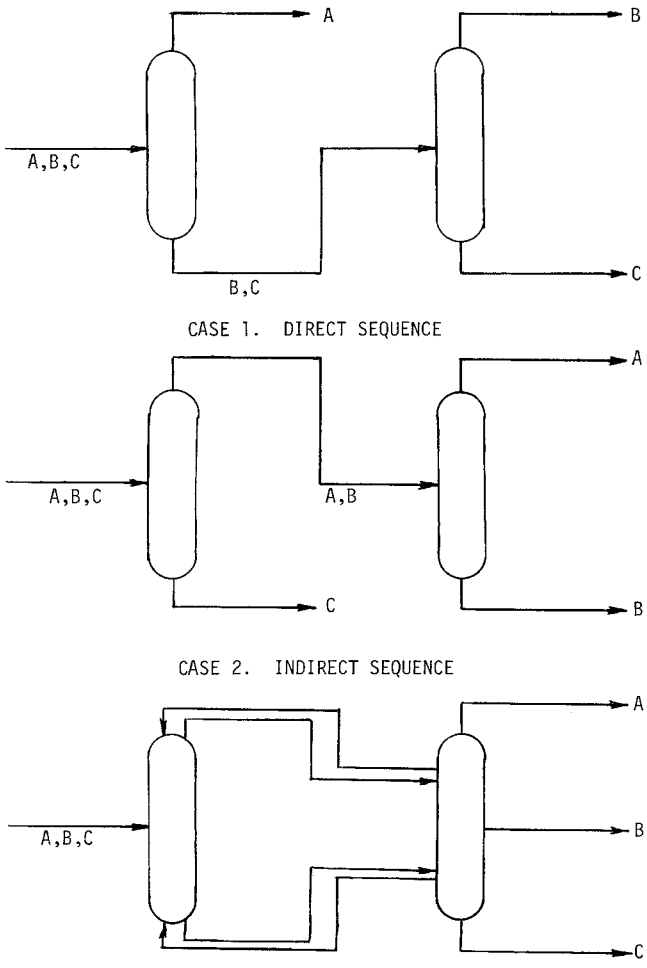

Fig. 3. Schemes for a 3-component distillation process.

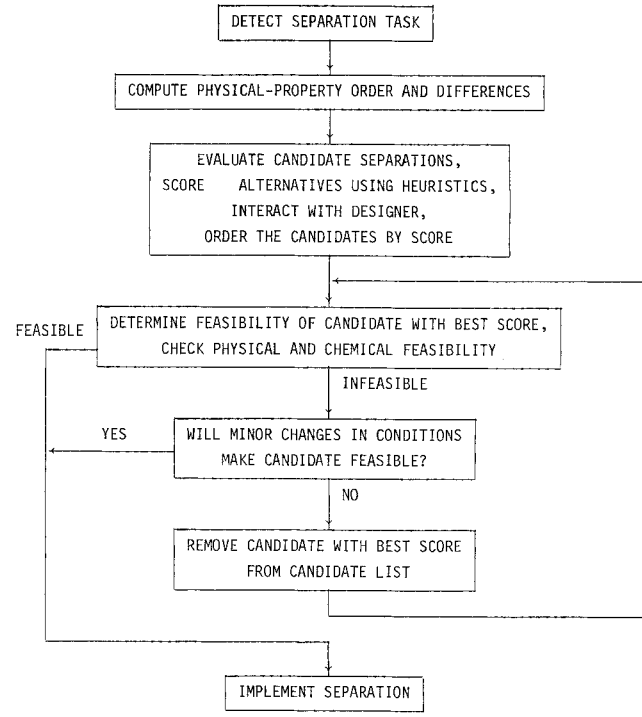

Fig. 4. AIDES separation-synthesis logic (Powers, 1971).

While other investigators considered only conventional distillation schemes for separating an R-component mixture into R products by $(R-1)$ two-sectional columns, each having a single condenser and a single reboiler, and without recycle among columns, Petlyuk, Platonov, and Slavinskii (1965) and Stupin and Lockhart (1971) did not make this restriction. Besides using the vapor traffic as an economic criterion, they also used a term which included the work of separation. They showed that for a symmetrical ternary feed, a sequence which more closely approaches a thermodynamically reversible distillation is preferred. Such a sequence is shown in Figure 3 as Case 3 compared to the conventional direct and indirect sequences shown as Cases 1 and 2, respectively. They also stated the advantages of intermediate condensers and reboilers and the use of combinations of pairs of rectification and stripping sections. As an example they considered the difficult separation of a mixture of xylenes. Their work indicates the need for process synthesis procedures which do not unduly restrict the process structure.

Selection and Sequencing of Separation Processes

The more general multicomponent separation problem in which several alternate types of separation techniques are considered was first approached by Siirola and Rudd (1971) and Powers (1972). Recognizing that the synthesis of separation sequences of heterogeneous structure was a basic subproblem in the overall synthesis of chemical processing systems, they sought to develop means by which the type and order of separation operations could be determined using heuristic techniques. Systematic procedures for generating separation schemes were set up as part of a general process synthesis program known as AIDES (Adaptive Initial Design Synthesizer). These procedures were based on the use of design heuristics for the selection of the most appropriate separation operation for a multicomponent mixture. Application of these heuristics to the original mixture generated new (smaller)

groups of components to which the heuristics were again applied. The process was carried out sequentially in this fashion until no further separations were required and the separation sequences (hopefully optimal) had been synthesized.

The logic used by AIDES in the selection of a separation technique and split point for a multicomponent mixture is shown in Figure 4. The first step is the generation of lists of the various components according to each of the physical and chemical properties to be considered as a possible basis for the separation, for example, components listed in increasing value of boiling point, melting point, solubility, etc. Then each candidate split point on each of these property lists is assigned a score indicating its desirability as a separation operation. The score is based on heuristics which are considered to reasonably guide separation selection. The magnitude of the property difference, the temperature and pressure for the separation, the amounts of the materials on either side of the split point and the corrosivities of the materials are some of the factors considered in the score. Relative importance of these factors is determined initially by intuition and may be altered to reflect experience from previous solutions. If the candidate separation with the most favorable score is feasible both physically (no undesirable phase behavior) and chemically (no undesirable reactions occurring during separation), or can be made so by minor changes in process conditions, then it is selected. Otherwise, alternate candidates are examined.

This procedure is, in essence, an attempt to overcome the problems associated with the use of several different heuristics for separation synthesis. The separation operation chosen at any point in the synthesis is the result of the combined effect of the various heuristics. Obviously, such a procedure depends heavily upon the nature of the heuristics that are included and also upon the weighting factors. It is quite likely that many optimal separation sequences could be found that would not conform to these particular heuristics and their respective weighting factors. In addition, the simple study of distillation sequences, as discussed in the previous section, offers no guidance as to means by which heuristics governing alternate separation techniques may be included. Indeed, for separation operations which have not been studied as thoroughly as distillation, detailed heuristics such as those presented earlier have not yet been developed. The problems associated with the selection and weighting of heuristics for the general-purpose design of separation sequences will require a great deal of further investigation if such heuristic techniques are to become a useful means for the synthesis of optimal separation processes.

A second approach to the general multicomponent separation problem has been proposed by Thompson and King (1972). While this approach is also heuristic in nature, it makes use of the prediction of actual costs for the separation of pairs of components using various separation techniques. The general strategy of this method is shown in Figure 5. The first step is the identification of a feasible set of products for the process. Here products are considered to be groups of one or more components with the same destination. Based on information concerning the destination of each component in the mixture and guided by a heuristic which states that the best process will have the least number of products, a product set is created which contains as few extra products as possible. Extra products result from the inability of the given separation operations to separate the required products without splitting groups of components bound for the same destination.

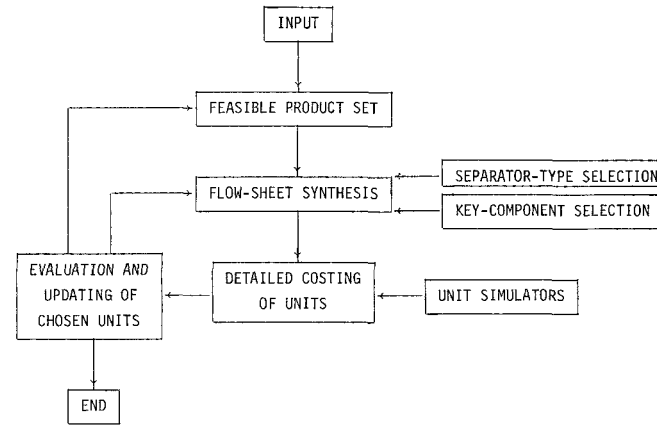

Fig. 5. Multicomponent separation-process synthesis strategy (Thompson and King, 1972).

Such extra products must subsequently be mixed in order to obtain the required products.

The next step is the generation of a separation sequence that will give these products. This is carried out using heuristic techniques in combination with a scheme for the prediction of separation costs. The prediction scheme is based on the simple formula:

$$\text{Estimated cost of separator} = (\beta_{ijk})\,(N_{ijk}) \qquad (15)$$

where N is proportional to the number of stages required to separate product i from product j by separator k under prespecified split fractions, and β is a weighting factor for that separator. Initial values of all β_{ijk} are arbitrarily set at one dollar, so as to encourage testing of various types of units. Values of the β_{ijk} are updated during synthesis, as described later. Synthesis of a flow sheet begins with an examination of the feed stream to the process and the insertion of an initial separator. Next, each of the separator effluent streams is examined for the need of further separation. The synthesis continues, ending when each of the final effluent streams contains only one of the products in the product set. Selection of a separator and key components at each stage is based on a heuristic requiring the cheapest of all possible candidate separations to be used first. Cost comparisons are made using Equation (15). Thompson and King note, however, that the selection could be based on any set of heuristics, possibly in a manner similar to that used by Rudd and co-workers as described above.

Once the separation sequence has been established, detailed simulation routines are used for sizing and costing of each of the separators involved. This information is used to update the weighting factors used in cost prediction:

$$\beta_{ijk} = \text{actual cost of unit}/N_{ijk} \qquad (16)$$

A new separation sequence is then produced based on the updated β_{ijk} values. The procedure is repeated until no lower-cost process can be found, whereupon a new heuristic which states that energy separating agents are to be favored over mass separating agents is envoked. Accordingly, if the lowest cost sequence contains mass separating agent operations which are required so that multicomponent products can be processed to their common destination without splitting, then the heuristic requiring a minimum of extra products is relaxed. A new product set which will allow the use of alternate energy separating agent operations is created by splitting multicomponent products, and the synthesis procedure is repeated.

Thompson and King point out that their procedure is experimental in nature, an "examination of one basic approach and some variations upon it." They describe several of the difficulties which have resulted from this approach and indicate that the general strategy will be altered "in ways suggested by the types of failures and inadequacies which are encountered."

While this approach undoubtedly has much to recommend it because it can apparently create reasonable processing schemes without a large consumption of computer time, in common with all heuristic approaches, it can create at best schemes with a high probability of optimality and is unquestionably liable to complete failure when confronted with a sufficiently complex and unusual situation.

In view of the extremely large savings which may result from structural changes in a process flow sheet (as compared with those from subsequent optimization of individual equipment items), it would be worthwhile to consider the possibility of general separation sequence synthesis by a more rigorous and direct method. Heuristic techniques were applied to the separation problem as a direct result of its combinatorial nature. The motivation was to avoid an exhaustive examination of the enormous number of different sequences which could possibly be used to accomplish the required separations. However, similar combinatorial problems have been solved without recourse to either heuristic techniques or exhaustive examination, a good example being the well-known "shortest-route problem." Here N nodes are present in an interconnected network, each node connected to one or more of the others. Given the length of each of these interconnections, the problem is to construct a route from node 1 to node N such that the sum of the interconnections between adjacent nodes on this route is a minimum. For highly-connected networks, the number of different routes which are possible rises quite rapidly with the number of nodes; yet this problem has several very efficient means of solution which do not require examination of all possible routes. The key features of this problem are the nodes and their individual connections to other nodes, and not the various ways these nodes may be arranged to form routes from node 1 to node N. In a similar fashion, the key features of the multicomponent separation problem are the smaller groups of components which may occur during synthesis and the means by which these groups may be further decomposed, and not the multitude of ways in which these decompositions may be arranged to form a feasible separation sequence.

From a consideration of separation operations as simple list-processing operations, an efficient and systematic method, that takes advantage of the sophisticated analysis procedures available for design and costing of separators was developed by Hendry and Hughes (1972) to synthesize optimal multicomponent separation sequences from the optimal sequences for smaller groups of components. The list processing representation of separation operations, upon which this systematic build-up of optimal sequences is based, is reviewed next.

Figure 6 poses a hypothetical four-component separation problem in which pure products are desired. Two property differences are to be exploited, property I and property II, each with a different ordering of the species. An enormously large number of ways exist in which the material can be separated. It is desired that the optimum way be found.

Any separation scheme is assembled from subgroup separation problems. A four-component separation is assembled from a variety of three-component-separation and two-component-separation schemes. The example sequence in Figure 6 consists of a two-component separation [(A, B) separated by property I], a three-component separation [(B, A, C) separated by property II], and a four-component separation [(A, B, C, D) separated by property I]. By examination of the property listings, the number of subgroup separations can be determined as three groups of three-component separation problems and four groups of two-component separations, as shown. These are the limited subgroups from which the optimum separation scheme can be assembled.

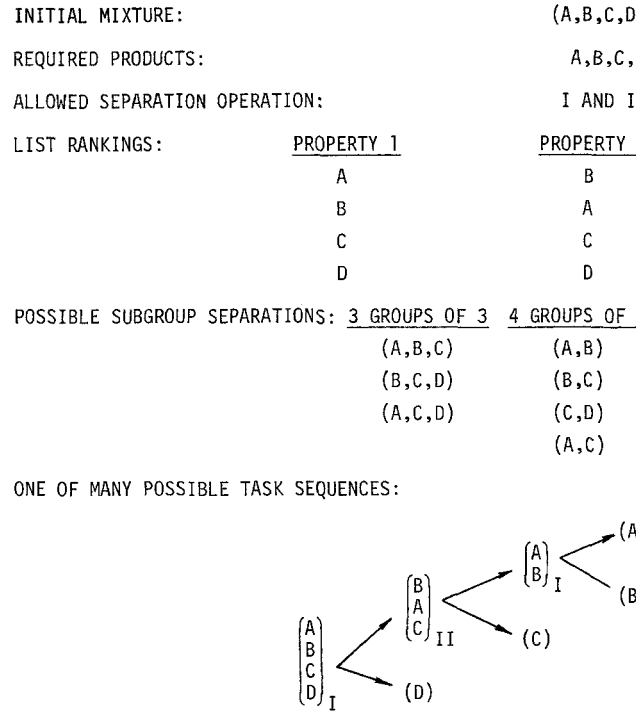

Fig. 6. Hypothetical four-component separation.

Each of the two-component subgroups are examined by computing a relevant cost function and, in each case, by computing the cost of separating the species by exploiting property I and property II. Since the two-component separations are last in any separation sequence, the least costly way of separation is selected for each two-component separation.

Next, the three three-component subgroup separations are considered. These can be broken down in a variety of ways. For example, (A, B, C) can be separated into A and (B, C) using property I or into B and (A, C) using property II or into (A, B) and C using property I or property II.

At this point, that separation scheme which leads to the lowest total cost (the sum of the cost of the three-component separation and the cost of the previously determined best two-component separation) is selected. For each three-component separation, the optimum sequence is chosen.

Finally, five ways exist in which the original four-component separation can be performed. For example, we can separate (A, B, C, D) using property II into (B, A) and (C, D) and then use the best way of separating (B, A) into A and B and the best way of separating (C, D) into C and D. Performing these calculations for all five schemes in which the original (A, B, C, D) group can be separated will lead to the lowest cost factor.

The Hendry-Hughes method may be represented in

terms of Rudd's (1968) design decomposition theory for general process synthesis. Let X be the set of task constraints directing a multicomponent mixture that must be separated (that is, X contains all component flow rates and required destinations and so defines the separation problem). Then X may be broken up into subtasks $\hat{S}$, S_I, and S_{II}, where $\hat{S}$ represents some initial split of X into two-component groups, S_I represents decomposition of the first of these groups to satisfy product destination requirements, and S_{II} represents a similar decomposition of the second group. Thus, the following relationships hold:

$$\begin{aligned}
&\hat{S} = \hat{X} \cup T,\\
&S_I = X_I \cup T,\\
&S_{II} = \hat{X}_{II} \cup T,\\
&X = \hat{X} \cup (X_I \cup X_{II}),\\
&\hat{X} \cap X_I = 0,\\
&\hat{X} \cap X_{II} = 0,\\
&\hat{X}_I \cap X_{II} = 0,
\end{aligned} \tag{17}$$

where T is the set of tear constraints (subgroup temperatures, pressures, etc.) that unite tasks $\hat{S}$, S_I, and S_{II} to accomplish the original task defined by X. Optimal-separation-sequence selection may be based on

$$O^*(X) = \underset{\substack{\text{split-point fraction}\\ \text{separation technique}}}{\text{Opt}} \left[\underset{T}{\text{Opt}} [O^*(\hat{S}) + O^*(S_I) + O^*(S_{II})] \right], \tag{18}$$

operating at optimal values of its design values. Equation (18) may then be applied to all S_k for which resulting S_i, S_j either are zero or correspond to previously determined two-component systems; and, consequently, $O^*(S_k)$ may be computed for all three-component systems. This recursive generation of optimal separation subsequences is actually an application of the principles of dynamic programming by Bellman (1957) to the synthesis problem and makes use of the fact that any decomposition of the original separation problem results in a finite set of separation subproblems that may be derived by use of the list-processing techniques outlined earlier.

When Equation (18) is written for any subproblem S_k, it becomes

$$O^*(S_k) = \underset{\substack{\text{split-point location}\\ \text{separation technique}}}{\text{Opt}} \left[\underset{T}{\text{Opt}} [O^*(\hat{S}_k) + O^*(S_i) + O^*(S_j)] \right], \tag{19}$$

where again $\hat{S}_k$, S_i, and S_j represent a decomposition of S_k. The interior optimization may also be written in terms of D_k, the design variables for the initial split of S_k. Equation (19) then becomes

$$O^*(S_k) = \underset{\substack{\text{split-point location}\\ \text{separation technique}}}{\text{Opt}} \left[\begin{array}{l} \underset{D_k}{\text{Opt}} [O[\hat{S}_k(D_k)] + O^*[S_i(T_i)] + O^*[S_j(T_j)]] \\ \text{Subject to: } T_i = f_i(D_k),\ T_j = f_j(D_k) \end{array} \right], \tag{20}$$

where f_i, f_j define the relationships that exist between T_i, T_j, and D_k. Thus, two-component optimal sequence synthesis, for which $O^*(S_i)$, $O^*(S_j)$ are zero, reduces to

$$(S_k) = \underset{\text{separation technique}}{\text{Opt}} \left[\underset{D_k}{\text{Opt}} [O[\hat{S}_k(D_k)]] \right]; \tag{21}$$

that is, selection of the best available separation technique.

It must be emphasized that the synthesis of optimal separation sequences by the Hendry-Hughes method is not simply an application of dynamic programming. In their method, efficient synthesis depends upon the use of dynamic programming in conjunction with a list-processing operation that takes advantage of the properties inherent in separation processes; that is, the splitting of groups of components into subgroups according to relative rankings on physical-property lists. Application of dynamic programming alone to this synthesis problem would necessitate consideration of a vector of state variables with dimensions at least as large as the number of components in the original mixture and would increase the required separator calculations to the point where direct examination of all possible sequences was a more efficient synthesis procedure. Consequently, the list-processing aspect of the synthesis technique has a great effect upon synthesis efficiency; and it is this simplification which makes a dynamic-programming approach to the problem possible.

Mitten and Nemhauser (1963) have discussed the general problem of synthesis of optimal multistage processes using dynamic-programming techniques and have indicated several means of easing the difficulties caused by large state spaces. Their discussion is of a general, theoretical nature; no specific examples are given. Direct application of the recursive relations derived in their work to the synthesis of general chemical-processing systems would appear unlikely, chiefly because of the large state spaces involved in such systems. However, it may well be that dynamic-programming techniques could be used in conjunction with procedures which take advantage of the nature of chemical-processing systems (such as, list splitting in separation systems) to aid in the synthesis of optimal processes.

Reactor Networks

Rather than using a single reactor to conduct a chemical reaction, advantages often exist for utilizing a network or sequence of reactors. For example, sequences of continuous-stirred-tank reactors (CSTR) arranged in series are commonly employed. As described by Aris (1964), the principle of dynamic programming can be employed effectively to determine the optimum number of reactor stages. Other, more complex arrangements are also effective in certain cases; for example, the combined use of CSTR and plug-flow reactors (PFR), as discussed by Aris (1969). Systematic methods of synthesizing complex reactor networks are just beginning to emerge.

Umeda and Ickikawa (1972) utilized their decomposition technique described earlier to determine the optimal arrangement of a single CSTR and/or a single PFR to conduct the irreversible second-order reaction $A \rightarrow B$. A general flow scheme permitted the two reactors to be connected in series and/or in parallel. The objective function was the equipment investment. For the numerical case studied, the optimal arrangement was a CSTR followed by a PFR.

Entire Chemical Processes

The development of techniques for the systematic synthesis of entire chemical processes, including reactors, com-

ponent and phase-separation equipment, energy-transfer devices, etc., is an extremely formidable problem. It has received the attention of Rudd and co-workers and of Ichikawa and co-workers using the methods of heuristics, direct search, and evolutionary search, as described earlier. 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 of process synthesis but also interacts with the designer to allow for the intuitive aspects of synthesis. Requiring only a minimum of input data, concerning mainly feed-stream specifications, product specifications, and the chemical-reaction path(s), AIDES has been successfully employed to produce alternate process flow schemes (not necessarily including the optimum) for the:

1. Manufacture of sulfuric acid from the off gases of a mineral processing plant,
2. manufacture of ethyl acetate from ethyl alcohol and air, and
3. manufacture of monochlorodecane by the direct chlorination of decane.

The AIDES program should prove to be a valuable design tool capable of suggesting novel processing schemes.

The direct optimization method of Umeda, Hirai, and Ichikawa (1972) is more difficult to apply than AIDES; but it can lead to the optimal system. All of the alternative processing schemes must be combined into a single integrated system. Their technique then results in the elimination of certain interconnecting streams in favor of others. In addition, certain items of equipment may be discarded to achieve the optimal system. They solved a typical process-design problem involving reaction and component separation steps with recycle. A large number of possible processing schemes were embedded in the original flow diagram, which contained 32 process streams and 24 equipment items. The optimal scheme was achieved after three minutes of computing time on an IBM 360/751 digital computer. Ten streams were eliminated, along with two heat-exchange devices.

An evolutionary-search approach was applied by Ichikawa and Fan (1972) to the synthesis of a simple process for producing B from A by a reaction path involving a single, first-order irreversible reaction, $A \rightarrow B$, to be conducted isothermally. The only equipment items available to the designer were a CSTR, a PFR, and an ideal component separator. Starting with an initial feasible process structure, consisting of a single CSTR, their procedure led to the optimal system as one involving a PFR followed by a separator with recycle of unreacted A back to the PFR.

Last, brief mention may be made of a recent paper by Goto and Matsubara (1972). They integrated three possible synthesis schemes for a process involving a first-order reversible reaction including:

1. a CSTR with extraction, settling, and two distillation columns;
2. a CSTR with extraction, settling, and one distillation column;
3. a CSTR with one distillation unit

into a single process flow diagram and then solved directly for the optimal arrangement under different conditions.

NOTATION

Operators

$\cup$ = or (inclusive or)
$\cap$ = and (at the same time)
$\subset$ = contained in

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Determination of Friction Factors for Pulsatile Flow of Water in Distensible Tubes

Theoretical models for computing velocity profiles and friction factors for sinusoidally pulsing laminar flow in linearly elastic tubes were derived from the Navier-Stokes equations, a modified Fanning equation, and the pressure-tube wall movement relationship. The theoretical results were checked experimentally for N_{Re} and $\lambda = \omega R^2/\nu$ values up to 2000 and 57, respectively. It was found that the theoretical friction factors predicted the experimental values to within less than 13%. Also the results showed that the energy losses in elastic tubes were greater than in rigid tubes.

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SCOPE

A study of pulsating flow in distensible tubes has received attention from investigators interested in research on the human circulatory system. Much effort has been directed towards describing blood flow satisfactorily. This requires a thorough knowledge of the tapering and branching of the blood vessels, mechanical properties of the ves-

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