Studies in Chemical Process Design and Synthesis:

Part V: A Simple Heuristic Method for Systematic Synthesis of Initial Sequences for Multicomponent Separations

A simple heuristic method for the systematic synthesis of initial sequences for multicomponent separations is proposed and applied to a number of synthesis problems which have been solved previously using other methods. Based on reported costs, it is shown that the initial sequences synthesized for the test problems by the new heuristic method are cheaper than those obtained by other ordered heuristic methods. These initial sequences are also either identical to or at most a few percents higher in costs than those optimum sequences obtained by other algorithmic, heuristic-algorithmic and heuristic-evolutionary methods. The new method is straightforward to apply by hand and it does not require any mathematical background and computational skill from the user.

V. M. NADGIR

Department of Chemical Engineering
Auburn University
Auburn University, AL 36849

and

Y. A. LIU

Department of Chemical Engineering
Virginia Polytechnic Institute and
State University
Blacksburg, VA 24061

SCOPE

An important process design problem is the systematic synthesis of multicomponent separation sequences which is concerned with the systematic selection of the method and sequence for separating a multicomponent mixture into several products of relatively pure species. The general techniques which have been developed for solving the separation sequencing problem have included: algorithmic approaches involving some established optimization principles (e.g., Hendry and Hughes, 1972); heuristic methods based on the use of rules of thumb (e.g., Rudd et al., 1973, pp. 155–208); evolutionary strategies wherein improvements are systematically made to an initially created separation sequence (e.g., Stephanopoulos and Westerberg, 1976); and thermodynamic methods involving applications of thermodynamic principles (e.g., Hohmann et al., 1980). In some situations, two or more of these techniques have been used together in the synthesis (e.g., Seader and Westerberg, 1977). A review of previous studies on multicomponent separation sequencing can be found in Nishida et al. (1981).

A disadvantage of many existing algorithmic and evolutionary techniques as reviewed by Nishida et al. is that their applications require special mathematical background and computational skill from the user. Although heuristic rules to guide the order of separation sequencing have long been available, many of the known heuristics contradict or overlap others; and procedures to resolve these conflicts have not been adequately developed. Recently, some success has been reported on the applications of certain heuristics together with evolutionary strategies for multicomponent separation sequencing (Seader and Westerberg, 1977; Nath and Motard, 1981).

In this work, a simple heuristic method for the systematic synthesis of initial sequences for multicomponent separations is proposed. A comparison of the new heuristic method with other recent methods, particularly the heuristic-evolutionary methods by Seader and Westerberg (1977) and by Nath and Motard (1981), is presented. A number of illustrative examples are given to demonstrate the simplicity and effectiveness of the proposed method.

CONCLUSIONS AND SIGNIFICANCE

This work proposes and demonstrates a simple heuristic method for the systematic synthesis of initial sequences of multicomponent separations. The new method involves the sequential applications of the following seven heuristics: (1) favor ordinary distillation and remove mass separating agent first; (2) avoid vacuum distillation and refrigeration; (3) favor the smallest product set; (4) remove corrosive and hazardous components first; (5) perform difficult separations last; (6) remove the most plentiful component first; and (7) favor equimolal (50/50) split. The first two heuristics decide the separation methods to be used. The next three heuristics give guidelines about the forbidden splits due to product specifications, as well as the essential first and last separations. The last two heuristics are used to synthesize the actual initial separation sequences, with the help of an auxiliary sequencing parameter, called the coefficient of ease of separation (*CES*) (Eq. 1).

The new heuristic method has been applied to a number of synthesis problems which have been solved previously using other methods. Based on reported costs for the illustrative examples presented, it is shown that the initial sequences synthesized by the new heuristic method are cheaper than those obtained by other ordered heuristic methods of Seader and Westerberg (1977) and of Nath and Motard (1981). These initial sequences are also either identical to or at most a few percents higher in costs than those optimum sequences obtained by other algorithmic (e.g., Hendry and Hughes, 1972), heuristic-algorithmic (e.g., Thompson and King, 1972), and heuristic-evolutionary (Seader and Westerberg, 1977; Nath and Motard, 1981) methods.

The new heuristic method offers the advantages of simplicity

Correspondence concerning this paper should be addressed to Y. A. Liu.

and effectiveness, coupled with good performance. Further, the new method is straightforward to apply by hand and it does not require any mathematical background and computational skill from the user.

INTRODUCTION

Multicomponent separation systems are found in widespread use in the chemical and petroleum industries. An important process design problem in multicomponent separations is the separation sequencing, which is concerned with the selection of the optimal method and sequence for the separation. This problem is often solved by first arranging the components in the mixture to be separated in some ranked lists of appropriate physical and/or chemical properties such as relative volatility or solubility in water. The resulting ranked list for each property gives the component name and its property ranking relative to other components in the mixture. This list also allows a separation step to be considered as a list splitting operation, in which components above a certain property value are separated from components below that value.

The generation of a separation sequence then involves the selection of an appropriate property list and the choice of separation keys within the list. For example, sequences for separating a three-component mixture of A, B and C into pure components by two ordinary distillation columns can be found by first arranging the components in a ranked list of relative volatility from most volatile (A) to least volatile (C), and then examining the different splits in the two columns. One possible sequence involves making the split A/BC in the first column followed by the split B/C in the second column. Another possible sequence corresponds to making the split AB/C in the first column and the split A/B in the second column. Some excellent discussion of the background information related to the separation sequencing problem can be found in the textbooks by Rudd et al. (1973, pp. 155–208 and 288–295), King (1980, pp. 710–720), and Henley and Seader (1981, pp. 527–555). A review of previous studies on the subject can be found in Nishida et al. (1981).

A SIMPLE HEURISTIC METHOD

Essentially all of the reported heuristic rules for separation sequencing can be broadly classified into four categories: (1) *method heuristics* (designated as M heuristics) which favor the use of certain separation methods under given problem specifications; (2) *design heuristics* (designated as D heuristics) which favor specific separation sequences with certain desirable properties; (3) *species heuristics* (designated as S heuristics) which are based on the property differences between the species to be separated; and (4) *composition heuristics* (designated as C heuristics) which are related to the effects of feed and product compositions on separation costs. Note that these classifications of heuristics are the same as those described by Tedder (1975), except that the method heuristics as defined here are broader than Tedder's "state" heuristics.

In what follows, a simple ordered heuristic method for the systematic synthesis of initial sequences for multicomponent separations is proposed. This method involves the systematic use of the following seven heuristics, which are to be applied one by one in their given order. If any heuristic is not important in, or not applicable to, the synthesis problem, the next one in the method is considered.

1. *Heuristic M1* (*Favor Ordinary Distillation and Remove Mass Separating Agent First*). (a) All other things being equal, favor separation methods using only energy separating agents (e.g., ordinary distillation), and avoid using separation methods (e.g., extractive distillation) which require the use of species not normally present in the processing, i.e., the mass separating agent (*MSA*) (Rudd et al., 1973, pp. 174–181). However, if the separation factor or relative volatility of the key components $\alpha_{LK,HK} < 1.05$ (Van Winkle, 1967, p. 381; Seader, and Westerberg, 1977) to 1.10 (Nath and Motard, 1981), the use of ordinary distillation is not recommended. A *MSA* may be used, provided that it improves the relative volatility between the key components. (b) When a *MSA* is used, remove it in the separator immediately following the one into which it is used (Hendry and Hughes, 1972; Rudd et al., 1973, pp. 174–180; Seader and Westerberg, 1977).

2. *Heuristic M2* (*Avoid Vacuum Distillation and Refrigeration*). All other things being equal, avoid excursions in temperature and pressure, but aim higher rather than lower (Rudd et al., 1973, pp. 182–183). If vacuum operation of ordinary distillation is required, liquid-liquid extraction with various solvents might be considered. If refrigeration is required (e.g., for separating materials of low boiling points with high relative volatilities as distillate products), cheaper alternatives to distillation such as absorption might be considered (Sounders, 1964; Seader and Westerberg, 1977; Nath and Motard, 1981).

3. *Heuristic D1* (*Favor Smallest Product Set*). Favor sequences which yield the minimum necessary number of products. Equivalently, avoid sequences that separate components which should ultimately be in the same product (Thompson and King, 1972; King, 1980, p. 720). In other words, when multicomponent products are specified, favor sequences that produce these products directly or with a minimum of blending unless relative volatilities are appreciably lower than those for a sequence which requires additional separators and blending (Seader and Westerberg, 1977; Henley and Seader, 1981, p. 541).

4. *Heuristic S1* (*Remove Corrosive and Hazardous Components First*). Remove corrosive and hazardous materials first (Rudd et al., 1973, p. 170).

5. *Heuristic S2* (*Perform Difficult Separations Last*). All other things being equal, perform the difficult separations last (Harbert, 1957; Rudd et al., 1973, pp. 171–174). In particular, separations where relative volatities of the key components are close to unity should be performed in the absence of nonkey components. In other words, try to select sequences which do not cause nonkey components to be present in separations where the key components are close together in relative volatility or separation factor (Heaven, 1969; King, 1980, p. 715).

6. *Heuristic C1* (*Remove Most Plentiful Component First*). A product composing a large fraction of the feed should be separated first, provided that the separation factor or relative volatility is reasonable for the separation (Nishimura and Hiraizumi, 1971; Rudd et al., 1973, pp. 167–169; King, 1980, p. 715).

7. *Heuristic C2* (*Favor 50/50 Split*). If component compositions do not vary widely, sequences which give a more nearly 50/50 or equimolal split of the feed between distillate (D) and bottoms (B) products should be favored, provided that the separation factor or relative volatility is reasonable for the split (Harbert, 1957; Heaven, 1969; King, 1980, p. 715). If it is difficult to judge which split is closest to 50/50 and with a reasonable separation factor or relative volatility, perform the split with the highest value of *the coefficient of ease of separation* (*CES*) first.

The coefficient of ease of separation (*CES*) as proposed in heuristic $C2$ is defined as

$$CES = f \times \Delta \tag{1}$$

where f = the ratio of the molal flow rates of products (distillate and bottoms) B/D or D/B, depending on which of the two ratios, B/D and D/B, is smaller than or equal to unity; and $\Delta = \Delta T$ = boiling point difference between the two components to be separated, or $\Delta = (\alpha - 1) \times 100$ with α being the relative volatility or separation factor of the two components to be separated.

TABLE 1. COMPARISON OF THREE SYNTHESIS METHODS FOR SEPARATION SEQUENCING

Ordered Heuristic Method (This Work)	Heuristic-Evolutionary Method (Seader and Westerberg, 1977)	Heuristic-Evolutionary Method (Nath and Motard, 1981)
1. Step 1: To decide the separation methods to be used (a) heuristics *M*1a and *M*1b (favor ordinary distillation and remove mass separating agent first). (b) heuristic *M*2 (avoid vacuum distillation and refrigeration). 2. Step 2: To give guidelines about the forbidden splits due to product specifications (a) heuristic *D*1 (favor smallest product set). 3. Step 3: To give guidelines about the essential first separations (a) heuristic *S*1 (remove corrosive and hazardous component first). 4. Step 4: To give guidelines about the essential last separations (a) heuristic *S*2 (perform difficult separations last). 5. Step 5: To synthesize the actual initial separation sequence (a) heuristic *C*1 (remove most plentiful component first). (b) heuristic *C*2 (favor 50/50 split) (c) auxiliary sequencing parameter: *CES* (coefficient of ease of separation) defined by Eq. 1.	1. Step 1: To decide the separation methods to be used (a) heuristic *M*1a (favor ordinary distillation). (b) heuristic *M*2 (avoid vacuum distillation and refrigeration). 2. Step 2: To synthesize the actual initial separation sequence (a) heuristic S3a (perform easy separations first). (b) heuristic *C*1 (remove most plentiful component first). (c) heuristic *D*2 (direct sequence rule). (d) auxiliary sequencing parameters: relative volatility or separation factor α and feed molar percentage. 3. Steps 1a and 2a: To be applied together with Steps 1 and 2 when separations with mass separating agents are considered (a) heuristic *M*1b (remove mass separating agent first). (b) heuristic *D*1 (favor smallest product set). 4. Step 3: local evolutionary improvement of the initial separation sequence.	1. Step 1: To give guidelines about the forbidden splits due to product specifications (a) heuristic *D*1 (favor smallest product set). 2. Step 2: To decide the separation methods to be used (a) heuristic *M*1a (favor ordinary distillation) (b) heuristic *M*1c (remove mass separating agent properly). (c) heuristic *M*2a (avoid vacuum distillation) (d) auxiliary design heuristics: set the split fractions of the key components to the prespecified values; and set the operating reflux ratio to 1.3 times the minimum ratio for each distillation column. 3. Step 3: To synthesize the actual initial separation sequence (a) heuristic S3b (perform easy separations first). (b) Auxiliary sequencing parameter: *CDS* (coefficient of difficulty of separation) defined by Eq. 3. 4. Step 4: global evolutionary improvement of the initial separation sequence

In applying the new method to separation sequencing, method heuristics *M*1 and *M*2 first decide the separation methods to be used. Design heuristic *D*1 and species heuristics *S*1 and *S*2 then give guidelines about the forbidden splits due to product specifications, as well as the essential first and last separations. Finally, the actual initial sequences are synthesized by using composition heuristics C1 and C2 with the help of the coefficient of ease of separation (*CES*).

Since CES involves the relative volatility or separation factor for the two components to be separated, the application of the new heuristic method depends indirectly on the separation temperature and pressure. A good correlation for the optimum overhead pressure (P_D) as a function of the normal feed bubble point (T_{FB}) in ordinary distillation has been presented by Tedder and Rudd (1978) as follows:

$$ln\ P_D = 1{,}751/(T_{FB} + 273) - 6.777 \quad (2)$$

In Eq. 2, P_D is in MPa and T_{FB} in °C such that $0.007 < P_D \leq 6.89$ and $-72°C < T < 699°C$. Also, a systematic procedure for specifying other operating pressures in ordinary distillation can be found in Henley and Seader (1981, pp. 432–434).

COMPARISON WITH RECENT HEURISTIC-EVOLUTIONARY METHODS

Table 1 compares the new heuristic method with two recent heuristic-evolutionary methods by Seader and Westerberg (1977) and by Nath and Motard (1981). The latter methods involve the following steps: (a) generating an initial separation sequence by using an ordered heuristic method; (b) developing a set of evolutionary rules to modify the initial separation sequence; (c) adopting a strategy to systematically apply the evolutionary rules; and (d) comparing the initial and modified separation sequences by some performance criteria such as separation costs.

The following additional heuristics have been included in the two heuristic-evolutionary methods summarized in Table 1 for the generation of initial separation sequences.

8. *Heuristic M1c* (*Remove Mass Separating Agent Properly*). A separation method using a mass separating agent (*MSA*) cannot be used to isolate another *MSA* (Nath and Motard, 1981).

9. *Heuristic D2* (*Direct Sequence Rule*). During distillation, when neither the relative volatility nor the molar percentage in the feed varies widely, remove the components one by one as distillate products. The resulting sequence is commonly known as the direct sequence, in which the optimum operating pressure tends to be highest in the first separator and is reduced in each subsequent separator (Lockhart, 1947; Harbert, 1957; Heaven, 1969; Rudd et al., 1973, pp. 183–184; Seader and Westerberg, 1977; King, 1980, p. 715).

10. *Heuristic S3a* (*Perform Easy Separations First*). Favor easy separations first. Specifically, arrange the components to be separated according to their relative volatilities or separation factors in an ordered list. When the adjacent relative volatilities of the ordered components in the feed vary widely, sequence the splits in the order of decreasing adjacent relative volatility (Seader and Westerberg, 1977).

11. *Heuristic S3b* (*Perform Easy Separations First*). Favor easy separations first. Specifically, arrange the separations to split the feed into the distillate and bottoms products in an increasing order of the coefficient of difficulty of separation (*CDS*) defined by

$$CDS = \frac{\log \dfrac{sp_{LK}}{1 - sp_{LK}} \cdot \dfrac{sp_{HK}}{1 - sp_{HK}}}{\log \alpha_{LK,HK}} \cdot \frac{D}{D+B} \cdot \left\{1 + \left|\frac{D-B}{D+B}\right|\right\} \quad (3)$$

where sp_{LK} and sp_{HK} are, respectively, the split fractions of the light and heavy key components in the distillate and bottoms products; *D* and *B* are, respectively, the molal flow rates of the distillate and bottoms; and $\alpha_{LK,HK}$ is the relative volatility of the key components (Nath and Motard, 1981).

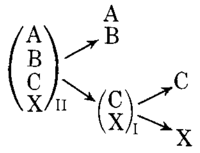
A number of differences among the three methods listed in Table 1 for the synthesis of initial separation sequences can be noted as follows:

(1) In steps 1 and 2 of the ordered heuristic method by Nath and Motard (1981), heuristic *D*1 (favor smallest product set) overrides heuristic *M*1a (favor ordinary distillation). A consequence of retaining the smallest product set is that sometimes a separation method using a mass separating agent is chosen instead of ordinary distillation. For example, consider the separation of a three-component mixture of *A*, *B*, and *C* into two products of (*A*,*B*) and *C*

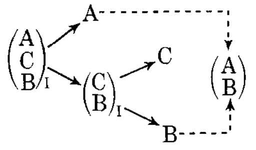
by ordinary distillation (method I) and extractive distillation with solvent X (method II), as described by Nath (1977). The rank lists (RL) of decreasing adjacent relative volatility corresponding to these two separation methods are given by:

$$RL(I)\text{: } A\ C\ B \qquad RL(II)\text{: } A\ B\ C\ X$$

The initial separation sequence resulting from applying steps 1 and 2 of the method by Nath and Motard is:



By contrast, the initial sequence obtained by applying steps 1 and 2 of the method presented in this work and that by Seader and Westerberg (1977) is:



This sequence favors the use of ordinary distillation (heuristic $M1a$), instead of having the smallest product set (heuristic $D1$). Also, in step 2 of the method by Nath and Motard, the heuristic of avoiding refrigeration has not been included.

(2) The incorporation of steps 3 and 4 in the method presented in this work represents an important difference of the new method from those by Seader and Westerberg and by Nath and Motard. These steps identify the essential first and last separations by using heuristics $S1$ (remove corrosive and hazardous components first) and $S2$ (perform difficult separations last) prior to the actual synthesis of the initial separation sequence. Also, the specific heuristics and auxiliary separation sequencing parameters used by each method for this synthesis of initial separation sequences are not identical, as shown in Table 1.

(3) The last difference among the three methods is related to the evolutionary step. The method presented in this work is a purely heuristic procedure and does not include an evolutionary step. Both the methods by Seader and Westerberg and by Nath and Motard include an evolutionary step, but with a different evolutionary strategy. In the method by Seader and Westerberg, a local evolution of the initial separation sequence is adopted; while in the method by Nath and Motard, a global evolutionary strategy is used. This difference between the two evolutionary strategies may be illustrated by considering the change to be made to replace the separation method for a specific separation task in an initial sequence. The method of Seader and Westerberg does not change the initial downstream structure (the separation methods for the remaining separation tasks located downstream of the initial sequence). By contrast, the method of Nath and Motard completely eliminates the initial downstream structure and synthesizes a new sequence by heuristics.

ILLUSTRATIVE EXAMPLES

Example 1: Separation of Light Paraffins by Ordinary Distillation

Consider the separation of a mixture of five light hydrocarbons into pure components by ordinary distillation studied by Heaven (1969). The feed mixture is:

Species	Fraction	Relative Volatility[a] α	CES
A:Propane	0.05		
		2.00	5.26
B:i-Butane	0.15		
		1.33	8.25
C:n-Butane	0.25		
		2.40	114.50
D:i-Pentane	0.20		
		1.25	13.46
E:n-Pentane	0.35		

[a] At 37.8°C and 1.72 MPa.

The separation sequencing by the new heuristic method is done as follows.

(1) Heuristics $M1$ and $M2$: Use ordinary distillation with refrigeration at high pressure.

(2) Heuristics $D1$ and $S1$: Not applicable.

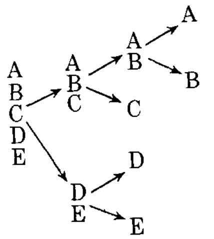
(3) Heuristic $S2$: Split D/E has the smallest relative volatility of $\alpha = 1.25$ compared to those of other splits. Therefore, it should be done last in the absence of A, B and C.

(4) Heuristic $C1$: Although E is a large fraction of the feed, it should not be separated first. This follows because the new heuristic method is an ordered one, and the preceding heuristic $S2$ overrides the present heuristic $C1$.

(5) Heuristic $C2$: For separating $ABCDE$, split ABC/DE is done first since it has the largest CES of 114.50. To separate ABC, the possible splits are A/BC and AB/C. By comparing the CESs for the splits,

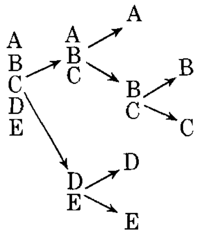
	A/BC	AB/C
f	0.05/0.40	0.20/0.25
$(\alpha - 1) \times 100$	100	33
CES	12.5	26.4

it can be seen that AB/C is preferred over A/BC. Thus, the resulting sequence, which performs split D/E last, is:



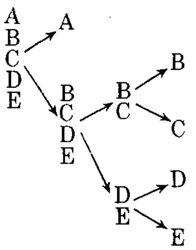
(sequence 1a)

The second sequence can be systematically found by considering the alternative splits in separating ABC, while retaining the best initial split (according to CES) ABC/DE and also performing the difficult split D/E last. Specifically, if split A/BC with the second largest CES of 12.5 for separating ABC is done first, instead of AB/C as in sequence 1a, a second sequence is found as follows:



(sequence 1b)

Since other sequences with an initial split ABC/DE which satisfy the constraints imposed by heuristics $M1$ to $C1$ do not exist, the third sequence is to be found by examining the alternative initial splits for separating $ABCDE$. Although initial splits $ABCD/E$ and AB/CDE have the second and third largest CES of 13.46 and 8.25, respectively, these splits are not recommended because of the constraint of performing the relatively difficult separations D/E and B/C last according to heuristic $S2$. Therefore, the third sequence is found by choosing split $A/BCDE$ first. The resulting sequence which performs splits B/C and D/E last, is:



(sequence 1c)

Table 2 compares the different sequences synthesized by the three methods listed in Table 1, based on the detailed design and costing reported by Heaven (1969). Sequence 1a corresponds to

TABLE 2. COMPARISON OF REPORTED SEQUENCES FOR EXAMPLE 1

Sequence	Cost $/yr[a]	Ordered Heuristic Method, This Work	Heuristic-Evolutionary Methods: Seader & Westerberg (1977)	Heuristic-Evolutionary Methods: Nath & Motard (1981)[c]
1a	858,780 (Best)	Initial Sequence	Final Sequence (Best)	
1b	863,580 (0.56%)[b]	2nd Sequence	Initial Sequence	
1c	871,460 (1.49%)	3rd Sequence		Initial and Final Sequence (Best)

[a] As reported by Heaven (1969).
[b] (0.56%) means that the cost of $863,580/yr is 0.56% above the minimum cost of $858,780/yr for the best sequence reported by Heaven (1969).
[c] As reported in Nath (1977).

the best sequence reported by Heaven (1969) using heuristics and by Seader and Westerberg using their heuristic-evolutionary method. Sequence 1b corresponds to a nearly optimal sequence with only 0.56% higher cost than sequence 1a. Sequences 1a and 1b also correspond to two best sequences reported by Rathore, et al. (1974) and by Gomez and Seader (1976), using optimization methods.

Example 2: Separation of Products from Thermal Cracking of Hydrocarbons

Consider the multicomponent separations involved in the large-scale thermal cracking of hydrocarbons to manufacture ethylene and propylene (Rudd, et al., 1973, pp. 183–185; King, 1980, pp. 708–710). The feed mixture is:

Species	Moles/Hr	Normal Boiling Point, T°C	ΔT	CES
A:Hydrogen	18	−253		
			92	23.0
B:Methane	5	−161		
			57	19.6
C:Ethylene	24	−104		
			16	14.6
D:Ethane	15	−88		
			40	18.1
E:Propylene	14	−48		
			6	1.1
F:Propane	6	−42		
			41	4.0
G:Heavies	8	−1		

It is desired to separate the feed into the following six products: AB, C, D, E, F and G. The separation sequencing by the new heuristic method can be done as follows.

(1) Heuristics $M1$ and $M2$: Use ordinary distillation with refrigeration at high pressure.

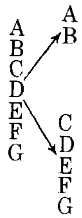
(2) Heuristic $D1$: Avoid splitting AB as it is a single product.

(3) Heuristic $S1$: Not applicable.

(4) Heuristic $S2$: Performs splits C/D and E/F last due to their small ΔT of 6 to 16°C.

(5) Heuristic $C1$: Not applicable.

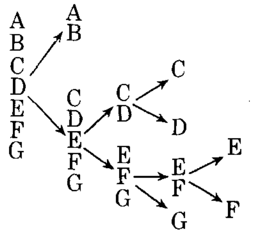
(6) Heuristic $C2$: For separating $ABCDEFG$, the best split is $AB/CDEFG$ which has the largest CES of 19.6 and also retains AB as a single product:



For separating $CDEFG$, splits C/D and E/F are performed last so that the remaining splits to be chosen are CD/EFG and $CDEF/G$. Split CD/EFG is done first since it has a larger CES of 28.7:

	CD/EFG	$CDEF/G$
f	28/39	8/59
ΔT	40	41
CES	28.7	5.6

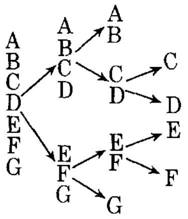
The resulting sequence, which performs splits C/D and E/F last, is:



(sequence 2a)

This is exactly the same as the one presently being practiced in the industry (Rudd et al., 1973, p. 185; King, 1980, p. 718).

The second sequence can be obtained by making the split $ABCD/EFG$ first (which has the second largest CES of 18.1 in splitting $ABCDEFG$ into two products) and performing the difficult splits C/D and E/F last as in the above sequence. This second sequence is:



(sequence 2b)

This sequence is used by the industry in the thermal cracking of naphthas (King, 1980, p. 718).

Example 3A: Separation of Light Olefins and Paraffins by Ordinary Distillation

Consider the separation of a mixture of light olefins and paraffins by ordinary distillation. The feed mixture as originally presented by Thompson and King (1972) is:

Species	Mole Fraction	Relative Volatility[a] α	CES
A:Ethane	0.20		
		3.50	62.5
B:Propylene	0.15		
		1.20	10.7
C:Propane	0.20		
		2.70	139.1
D:1-Butene	0.15		
		1.21	9.0
E:n-Butane	0.15		
		3.00	35.3
F:n-Pentane	0.15		

[a] At 37.8°C and 0.1 MPa.

It is desired to find the sequences for separating the feed into pure components, namely, A, B, C, D, E and F (Seader and Westerberg, 1977; Nath, 1977). The separation sequencing by the new heuristic method is done as follows.

(1) Heuristics $M1$ and $M2$: Use ordinary distillation with refrigeration at high pressure.

(2) Heuristics $D1$ and $S1$: Not applicable.

(3) Heuristic $S2$: Splits B/C and D/E are relatively difficult separations due to their small values of relative volatility, α = 1.20–1.21. Thus, these splits should be done last.

(4) Heuristic $C1$: Not applicable since none of the components represents a larger fraction of the feed.

(5) Heuristic $C2$: ABC/DEF represents a 55/45 split between the distillate and bottoms and has a reasonable value of relative volatility of α = 2.70 along with the largest value of CES of 139.1. Therefore, this split should be done first:

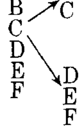
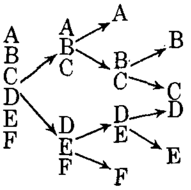


TABLE 3. COMPARISON OF REPORTED SEQUENCES FOR EXAMPLE 3A

Sequence	Ordered Heuristic Method, This Work	Heuristic-Evolutionary Methods: Seader & Westerberg (1977)	Nath & Motard (1981)[b]
3a	Initial Sequence	Final Sequence $1,153,000/yr (Best)	2nd Sequence $685,189/yr (8.7%)
3b	2nd Sequence	2nd Sequence $1,213,000/yr (5.2%)[a]	Initial Sequence $748,178/yr (18.6%)
3c			Final Sequence $630,454/yr (Best)
3d			3rd Sequence $805,105/yr (27.7%)
3e		Initial Sequence $1,234,000/yr (7.0%)	

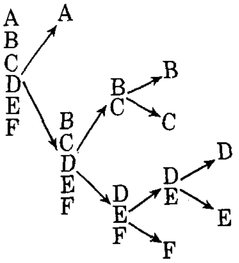
[a] (5.2%) means that the cost of $1,213,000/yr is 5.2% above the minimum cost of $1,153,000/yr for the best sequence reported in the same reference.
[b] As reported in Nath (1977).

The resulting sequence, which splits B/C and D/E last according to heuristic $S2$, is:



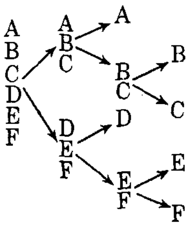
(sequence 3a)

The second sequence can be obtained by making split $A/BCDEF$ first (which has the second largest CES of 62.5 in splitting $ABCDEF$ into two products) and performing the difficult splits B/C and D/E last as in the above initial sequence. This sequence is:

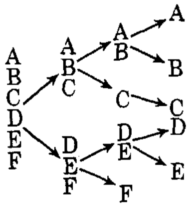


(sequence 3b)

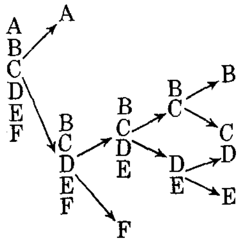
Table 3 compares the sequences synthesized by the three methods summarized in Table 1. The additional sequences (3c to 3e) listed in the table are as follows (Seader and Westerberg, 1977; Nath, 1977):



(sequence 3c)



(sequence 3d)



(sequence 3e)

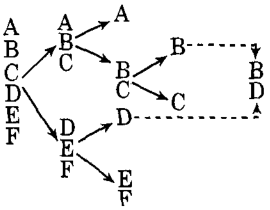
The costs tabulated in Table 3 and in subsequent examples should be compared on a relative basis, as different annual costs were reported for the same sequences by different investigators. It is apparent that different assumptions and/or methods for design calculations and equipment costing might have been employed by different investigators.

Table 3 shows that sequence 3a is cheaper than those obtained by the ordered heuristic methods by Seader and Westerberg and by Nath and Motard, namely, sequences 3e and 3b, respectively. Sequences 3a and 3b also correspond to the best and second best sequences obtained by the heuristic-evolutionary method by Seader and Westerberg. By applying a global evolutionary strategy to modify the initial sequence (sequence 3b), Nath (1977) has reported a cheaper final sequence (sequence 3c), which has not been obtained by the new heuristic method and by the heuristic-evolutionary method of Seader and Westerberg. The latter authors and Gomez and Seader (1976) suggested that the failure to obtain sequence 3c was due to the small, but important, effect of component F (n-pentane) of the relative volatility for D/E (1-butane/n-butane). Finally, Table 3 shows that there is no guarantee that the application of the global evolutionary strategy of Nath and Motard will always improve the initial and subsequent sequences. This is evident by noting that the third sequence (sequence 3d) is more expensive than the second sequence (sequence 3a) during the evolutionary synthesis by the method of Nath and Motard.

Example 3B: Separation of Light Olefins and Paraffins by Ordinary Distillation

This example has the same six-component feed mixture as in Example 3A, and it is desired to find the sequences for separating the feed into the following products: A, BD, C and EF (Thompson and King, 1972). The separation sequencing can be done by using heuristic D1 and making minor changes in step (4) in the solution to Example 3A.

(4) Heuristic C2: As in the separation sequencing for Example 3A, the split ABC/DEF is performed first. The resulting products, ABC and DEF, are separated as follows. To separate ABC, the difficult split B/C is done last as in the case for Example 3A. To separate DEF, the only recommended split according to heuristic D1 is D/EF since EF is a single produce. The other desired product BD can be obtained by blending B and D from the preceding separations. The resulting sequence is:



(sequence 3a′)

The second sequence can be systematically found by examining the alternative splits in separating ABC, while retaining the best initial split (according to CES) ABC/DEF for separating $ABCDEF$ and also the split D/EF which gives EF as a single product. Thus, if the second best split according to CES in separating ABC, namely, AB/C, is done first instead of A/BC:

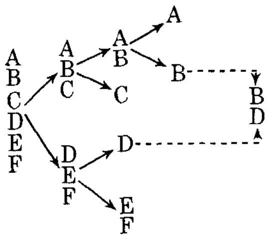
	A/BC	AB/C
f:	20/35	20/35
$(\alpha - 1) \times 100$:	250	20
CES:	142.8	11.4

TABLE 4. COMPARISON OF REPORTED SEQUENCES FOR EXAMPLE 3B

Sequence	Ordered Heuristic Method, This Work	Heuristic-Evolutionary Method (Nath & Motard, 1981)[a]	Heuristic-Algorithmic Method (Thompson & King, 1972)	Algorithmic Method (Westerberg & Stephanopoulous, 1975)
3a′	Initial Sequence	Final Sequence $600,395/yr (Best)	Final Sequence $694,000/yr (Best)	Best Sequence $602,760/yr (Best)
3b′	2nd Sequence			2nd Best Sequence $642,068/yr (6.5%)
3c′	3rd Sequence	Initial Sequence $663,385/yr (10.5%)	2nd Best Sequence $760,000/yr (9.5%)	3rd Best Sequence $685,632 (13.8%)

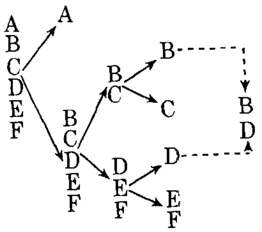
[a] As reported in Nath (1977).

the resulting sequence is:



(sequence 3b′)

Since no other sequences with an initial split ABC/DEF which satisfy the constraints imposed by heuristics $M1$ to $C1$ are available, the third sequence is to be found by considering the alternative initial splits for separating $ABCDEF$. Specifically, if the second best split (according to CES) in separating $ABCDEF$, namely, $A/BCDEF$, is chosen instead of ABC/DEF, the resulting sequence is:



(sequence 3c′)

Table 4 compares the sequences synthesized by several methods, including the new heuristic method and the heuristic-evolutionary method by Nath and Motard. It is significant to note from Table 4 that sequences 3a′, 3b′ and 3c′ exactly correspond to the best, second best and third best sequences, respectively, obtained by Westerberg and Stephanopoulos (1975) using an optimization method. Sequence 3a′ is identical to the best sequence obtained by the heuristic-evolutionary method by Nath and Motard and by the heuristic-algorithmic method by Thompson and King (1972). Also, sequence 3a′ is cheaper than the initial sequence obtained by the ordered heuristic method by Nath and Motard.

Example 4: n-Butylene Purification by Ordinary and Extractive Distillation

Consider the multicomponent separations in the industrial purification of n-Butylene (Hendry and Hughes, 1972). The feed mixture is:

Species	mol %	Relative Volatility[a] $(\alpha)_I$	$(\alpha)_{II}$	$(CES)_I$	$(CES)_{II}$
A:Propane	1.47				
		2.45		2.163	
B:1-Butene	14.75				
		1.18	1.17	3.485	3.29
C:n-Butane	50.30				
		1.03	1.70	1.510	35.25
D:trans-Butene-2	15.62				
E:cis-Butene-2	11.96				
		2.50		9.406	
F:n-Pentane	5.90				

[a] $(\alpha)_I$ = adjacent relative volatility at 65.6°C and 1.03 MPa for separation method I, ordinary distillation; $(\alpha)_{II}$ = adjacent relative volatility of 65.6°C and 1.03 MPa for separation method II, extractive distillation.

The rank lists (RL) of decreasing adjacent relative volatility corresponding to separation methods I and II are given by:

$RL(I)$: $ABCDEF$ $\quad RL(II)$: $ACBDEF$

The desired products of the separation are the following: A, C, BDE and F. The separation sequencing by the new heuristic method can be done as follows.

(1) Heuristic $M1$: Use extractive distillation for split C/DE and ordinary distillation for all other splits.

(2) Heuristic $M2$: Use low temperature and ambient to moderate pressure.

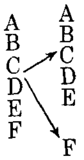
(3) Heuristic $D1$: Avoid splitting DE as both D and E are in the same product, and blend together B and DE to obtain a multicomponent product BDE.

(4) Heuristic $S1$: Not applicable.

(5) Heuristic $S2$: Since split C/DE is difficult and requires extractive distillation, it should be performed last in the absence of A, B and F.

(6) Heuristic $C1$: Although C is a large fraction of the feed, it should not be separated first due to the preceding heuristic $S2$. Further, it is preferred to carry out the extractive distillation for splitting C/DE at the end of the sequence. This will avoid having the mass separating agent as a possible contaminant in the intermediate separations of the sequence.

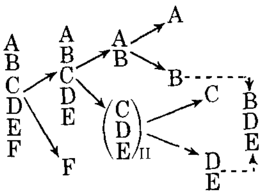
(7) Heuristic $C2$: For separating $ABCDEF$, split ABC/DEF is performed last so that the remaining splits to be chosen are $A/BCDEF$, $AB/CDEF$ and $ABCDE/F$. The latter split is chosen since it has the largest $(CES)_I$ of 9.406:



To separate $ABCDE$, the possible splits are $A/BCDE$ and AB/CDE. Since

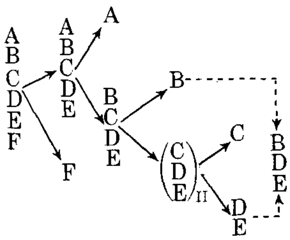
	$A/BCDE$	AB/CDE
f	1.47/92.63	16.22/77.88
$(\alpha - 1) \times 100$	145	18
CES	2.301	3.749

split AB/CDE is preferred over $A/BCDE$. The resulting sequence, which splits A/B and C/DE last, is as follows:



(sequence 4a)

A second sequence is obtained by splitting $A/BCDE$ first, instead of AB/CDE:



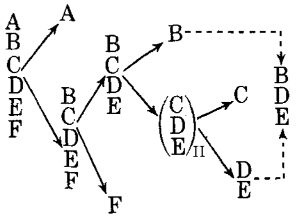
(sequence 4b)

TABLE 5. COMPARISON OF REPORTED SEQUENCES FOR EXAMPLE 4

Sequence	Ordered Heuristic Method, This Work	Algorithmic Method (Hendry and Hughes, 1972)[a]	Heuristic-Evolutionary Methods: Seader & Westerberg (1977)	Heuristic-Evolutionary Methods: Nath & Motard (1981)
4a	Initial Sequence	\$867,400/yr (0.8%)		
4b	2nd Sequence	\$878,200/yr (1.8%)		
4c	3rd Sequence	\$860,400/yr (Best)	Final Sequence \$860,400/yr (Best)	Final Sequence \$658,737/yr (Best)
4d	4th Sequence	\$878,000/yr (2.0%)	Initial Sequence \$878,000/yr (2.0%)	
4e	5th Sequence	\$872,400/yr (1.5%)	2nd Sequence \$872,400/yr (1.5%)	2nd Sequence \$669,844/yr (1.7%)
4f		\$1,095,600/yr (27.3%)		Initial Sequence \$1,171,322/yr (77.8%)

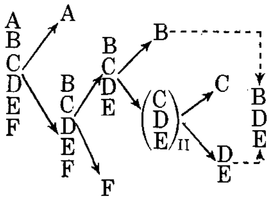
[a] As reported in Hendry (1972) and quoted in Henley and Seader (1981, p. 547).

As there are no other sequences with an initial split $ABCDE/F$ which satisfy the constraints imposed by heuristics $M1$ to $C1$, the third sequence is to be found by examining the alternative initial splits for separating $ABCDEF$. Thus, if the second best initial split for separating $ABCDEF$ by ordinary distillation, $AB/CDEF$, with the second largest $(CES)_I$ of 3.485, is done first, the resulting sequence, which splits A/B and C/DE last, is:

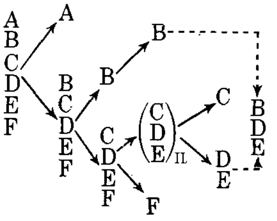


(sequence 4c)

Alternatively, if the third best initial split for separating $ABCDEF$ by ordinary distillation $A/BCDEF$ with the third largest $(CES)_I$ of 2.163 is performed first, two other sequences, which split C/DE last, can be found as follows:



(sequence 4d)

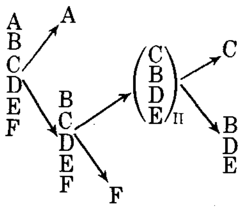


(sequence 4e)

Note that sequence 4d is better than sequence 4e according to the values of CES in separating $BCDEF$:

	$BCDE/F$	$B/CDEF$
f	5.90/92.62	14.75/83.77
$(\alpha - 1) \times 100$	84.06	24.85
CES	5.355	4.375

Table 5 compares the sequences synthesized by different methods for the present problem. The additional sequence (4f), which involves replacing the split B/C in sequence 4e by extractive distillation, is as follows:



(sequence 4f)

Table 5 shows that sequence 4a is cheaper than the initial sequences obtained by the ordered heuristic methods by Seader and Westerberg and by Nath and Motard, namely, sequences 4d and 4f, respectively. Based on the costs reported by Hendry (1972), sequence 4a has only a 0.8% higher cost than that for the best sequence (4c) synthesized by the heuristic-evolutionary methods by Seader and Westerberg and by Nath and Motard. Also, both the new heuristic method and the ordered heuristic method by Seader and Westerberg have generated much cheaper initial sequences (4a and 4d) than that (4f) by the ordered heuristic method by Nath and Motard.

CONCLUDING REMARKS

It is important to note that although heuristic $S1$ (remove corrosive and hazardous components first) has not been explicitly applied in the preceding examples, the use of this heuristic has been shown to be important in deciding the essential first splits in many separation sequencing problems in which feed mixtures contain corrosive and hazardous components. Typical example problems solved in this work have included the multicomponent separation sequencing in the manufacture of detergents from petroleum (Rudd et al., 1973, pp. 281–295). In this problem, it is essential to remove the corrosive hydrogen chloride from the mixture of chlorinated products and unreacted species (decane, monochlorodecane, dichlorodecane, chlorine and hydrogen chloride). For this reason, heuristic $S1$ has been included in the new heuristic method.

The preceding examples have shown that the initial sequences synthesized by the new heuristic method are cheaper than those generated by the ordered heuristic methods of Seader and Westerberg (1977) and of Nath and Motard (1981). These initial sequences are also either identical to or at most 2% higher in costs than those optimum sequences obtained by other algorithmic (e.g., Hendry and Hughes, 1972), heuristic-algorithmic (e.g., Thompson and King, 1972) and heuristic-evolutionary (Seader and Westerberg, 1977; Nath and Motard, 1981) methods. The only exception is Example 3A, for which Nath (1977) has reported a final sequence (sequence 3c) that has not been obtained by the new heuristic method and by the heuristic-evolutionary method of Seader and Westerberg. This final sequence is about 8.7% lower in cost than the initial sequence (sequence 3a) synthesized by the new heuristic method. An explanation for the failure to obtain sequence 3c suggested by Seader and Westerberg has been given previously in Example 3A.

The new heuristic method has also been tested with many other multicomponent separation sequencing problems reported in the literature. The specific synthesis problems solved by the new method have included essentially all of the problems described by Thompson and King (1972), Westerberg and Stephanopoulos (1975), Stephanopoulos and Westerberg (1976), Rodrigo and Seader (1976), Gomez and Seader (1976), Seader and Westerberg (1977), Nath (1977), and Nath and Motard (1981). Also, many test problems presented in the textbooks of Rudd et al. (1973, Chapter 5) and Henley and Seader (1981, Chapter 14) have been solved. The results from this work have shown essentially the same conclusion as observed in preceding examples.

Finally, it should be pointed out that the magnitude of cost differences for example problems, as shown in Tables 2 to 5, are relatively small. As was suggested by Tedder (1975), the magnitude of possible round-off errors (noises) resulting from the application of optimization techniques to separation sequencing problems with different initial solutions could often be greater than the cost differences indicated in these tables. Under such situations, it is important to select the best sequence from several initial sequences based on additional performance criteria other than the total cost such as the ease of startup and shutdown, the operating temperature and pressures relative to the critical points, the overall tower dimension, etc. The new heuristic method as presented in this work, provides a simple and effective procedure for the systematic synthesis of good initial sequences for carrying out such a multiobjective design optimization.

ACKNOWLEDGMENT

This work was supported in part by a grant from the Engineering Experiment Station, Auburn University. The authors would like to thank J. D. Seader of the University of Utah, R. L. Motard of Washington University, and D. W. Tedder of the Georgia Institute of Technology, for their helpful comments on this paper. Special appreciation is also extended to Motard for providing the authors with a copy of Nath's dissertation.

NOTATION

B = molal flow rate of bottoms product, mol/h
CES = coefficient of ease of separation defined by Eq. 1
CDS = coefficient of difficulty of separation defined by Eq. 3
D = molal flow rate of the overhead (distillate) product, mol/h
F = molal flow rate of the feed, mol/h
f = D/B or B/D such that $f < 1$
P_D = optimal overhead pressure, MPa
sp = split fraction of feed in a given product
T = normal boiling point of a component, °C
T_{FB} = normal bubble point of a feed stream, °C

Greek Letters

α = relative volatility of key components to be separated

Subscripts

LK = light key component
HK = heavy key component

Symbols

Δ = ΔT or $(\alpha - 1) \times 100$

LITERATURE CITED

Gomez, M. A., and J. D. Seader, "Separation Sequence Synthesis by a Predictor-Based Ordered Search," *AIChE J.*, **22**, 970 (1976).
Harbert, W. D., "Which Tower Goes Where?" *Pet. Ref.*, **36**, No. 3, 169 (1957).
Heaven, D. L., "Optimum Sequencing of Distillation Columns in Multicomponent Fractionation," M.S. Thesis, University of California, Berkeley (1969).
Hendry, J. E., "Computer Aided Synthesis of Optimal Multicomponent Separation Sequences," Ph.D. Thesis, University of Wisconsin, Madison, WI (1972).
Hendry, J. E., and R. R. Hughes, "Generating Separation Process Flowsheets," *Chem. Eng. Prog.*, **68**, No. 6, 71 (1972).
Henley, E. J., and J. D. Seader, *Equilibrium-Stage Separation Operations in Chemical Engineering*, Wiley, New York (1981).
Hohmann, E. C., M. T. Sander, and H. Dunford, "A New Approach to the Synthesis of Multicomponent Separation Schemes," AIChE National Meeting, Chicago, (Nov., 1980).
King, C. J., *Separation Processes*, 2nd Ed., McGraw-Hill, New York (1980).
Lockhart, F. J., "Multi-Column Distillation of Natural Gasoline," *Pet. Ref.*, **26**, No. 8, 105 (1947).
Nath, R., "Studies in the Synthesis of Separation Processes," Ph.D. Thesis, The University of Houston, Houston (1977).
Nath, R., and R. L. Motard, "Evolutionary Synthesis of Separation Processes," *AIChE J.*, **27**, 578 (1981).
Nishida, N., G. Stephenopoulos, and A. W. Westerberg, "A Review of Process Synthesis," *AIChE J.*, **27**, 321 (1981).
Nishimura, H., and Y. Hiraizumi, "Optimal System Pattern for Multicomponent Distillation Systems," *Int. Chem. Eng.*, **11**, 188 (1971).
Rathore, R. N. S., K. A. Van Wormer, and G. J. Powers, "Synthesis of Distillation Systems with Energy Integration," *AIChE J.*, **20**, 940 (1974).
Rodrigo, B. F. R., and J. D. Seader, "Synthesis of Separation Sequences by Ordered Branch Search," *AIChE J.*, **21**, 785 (1975).
Rudd, D. F., G. J. Powers, and J. J. Siirola, *Process Synthesis*, Prentice-Hall, Englewood Cliffs, NJ (1973).
Seader, J. D., and A. W. Westerberg, "A Combined Heuristic and Evolutionary Strategy for the Synthesis of Simple Separation Sequences," *AIChE J.*, **23**, 951 (1977).
Souders, M., "Countercurrent Separation Processes," *Chem. Eng. Prog.*, **60**, No. 2, 75 (1964).
Stephanopoulos, G., and A. W. Westerberg, "Studies in Process Synthesis—II. Evolutionary Synthesis of Optimal Process Flowsheets," *Chem. Eng. Sci.*, **31**, 195 (1976).
Tedder, D. W., "The Heuristic Synthesis and Topology of Optimal Distillation Networks," PhD Dissertation, University of Wisconsin, Madison, WI (1975).
Tedder, D. W., and D. F. Rudd, "Parametric Studies in Industrial Distillation. Part II: Heuristic Optimization," *AIChE J.*, **24**, 316 (1978).
Thompson, R. W., and C. J. King, "Systematic Synthesis of Separation Schemes," *AIChE J.*, **18**, 941 (1972).
Westerberg, A. W., and G. Stephenopoulous, "Studies in Chemical Process Synthesis. I: Branch and Bound Strategy with List Techniques for the Synthesis of Separation Schemes," *Chem. Eng. Sci.*, **30**, 963 (1975).
Van Winkle, M., *Distillation*, McGraw-Hill, New York (1967).

Manuscript received April 9, 1981; revision received August 5 and accepted December 14, 1982.