An MILP Formulation for Heat-Integrated Distillation Sequence Synthesis

In this paper the problem of distillation sequence synthesis with heat integration is formulated and solved as a mixed integer linear superstructure optimization problem. Allowing both conventional and multieffect distillation, two alternatives for generating a superstructure are shown, then a formulation is presented which approximates the synthesis problem as a mixed integer linear programming (MILP) problem. Examples illustrate the method. Reported computer times suggest that earlier algorithmic methods for discovering the better sequences among those allowed by the problem formulation, and without the option of multieffecting, are an order of magnitude slower than this approach.

M. J. ANDRECOVICH and A. W. WESTERBERG

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

SCOPE

Several earlier papers have considered the synthesis of heat-integrated distillation sequences. The problem is to find the optimal column sequence to separate a multicomponent mixture into several, often pure-component, products. Column pressures are selected to increase the temperature level of certain columns so the heat available from their condensers can be used as the heat needed by the reboilers of other columns. The number of alternative sequences of columns one can choose from is large. For example 14 different column sequences exist for separating a five-component mixture into five pure-component products. For each sequence many ways exist to heat-integrate the columns contained within it. If we then add the possibility of accomplishing each separation task (we define a task to be the separation possible within a single column) in more than a single column to permit the opportunities for multieffect solutions, the number of alternatives increases manyfold more. How does one find the better solutions for problems of this type?

Earlier related work on synthesizing heat-integrated distillation column sequences (reviewed in Nishida et al., 1981, and in Andrecovich and Westerberg, 1984) is based largely on the use of heuristics and tree search algorithms. The resulting algorithms have been computationally intense with the exception of a recent but more restricted branch and bound algorithm by Andrecovich and Westerberg, the only other work to have considered multieffect solutions.

Another approach often suggested for process synthesis is to create a superstructure within which are embedded numerous structural alternatives. By optimizing, it is hoped that the superstructure will be reduced to the substructure representing the optimal solution for the problem. This approach is computationally enormous if rigorous models are used. Such models will likely also contain numerous local optima. Papoulias and Grossmann (1983) show how to develop approximate models for the superstructure approach using mixed integer linear models. Such models are computationally more tractable and because an efficient enumeration scheme is possible, do not get caught by local optima.

Earlier work (Cerda, 1980; Cerda et al., 1983; Papoulias, 1982; Papoulias and Grossmann, 1983) has shown that one can represent the minimum utility heat integration problem as a linear programming problem. We show here that this model can be coupled linearly to the total material flows in a distillation sequence superstructure, and thus we can develop an approximate model of linear material flow with minimum utility use and solve as a linear programming problem for this class of problem. Realistic economic models, however, force one to add a fixed charge for including a unit within the final solution. Otherwise the solution contains a little bit of far too many units. (Obviously a fixed charge allows one to better model the six-tenths power capital cost model for columns.) A mixed integer linear model results which is readily solved using generally available codes.

CONCLUSIONS AND SIGNIFICANCE

We have presented a mixed integer linear programming formulation for the synthesis of sequences of heat integrated distillation columns. We have presented two ways to generate the needed superstructure model and then given the needed mathematical formulation of the approximate mixed integer linear programming model required. The method is computationally fast. It also permits solutions to include tasks which are multieffected if they are economically preferred. Finally, we observe that such a model can become part of a larger model to synthesize complete process flowsheets, such as described in the work of Papoulias and Grossmann (1983).

Correspondence concerning this paper should be addressed to A. W. Westerberg.
M. J. Andrecovich is currently with BOC Group Tech Center, Murray Hill, NJ 07974.

INTRODUCTION

The first section of this paper describes the generation of a superstructure for the problem of heat-integrated distillation sequence synthesis. Next the formulation and solution of a mixed integer linear program (MILP) which describes this superstructure is discussed. The third section describes the solution of two example problems using this method. The final section discusses the advantages and disadvantages of this method.

GENERATION OF A SUPERSTRUCTURE

The ideal superstructure which can be generated for any given problem possesses two important properties: It contains every feasible solution, and it can be easily searched. For many problems, however, it is not possible to develop a superstructure which satisfies both criteria. An all-inclusive superstructure may contain so many alternative solutions that it is impossible to search them all, even with the most efficient computer techniques. On the other hand, the superstructure may contain few alternatives but the effort required to evaluate each alternative may be prohibitively large.

Fortunately, very good solutions to the problems as posed can often be found with much less effort than would be required to find and verify the optimal solution. These solutions are found by limiting the number of solutions within the superstructure or by limiting the search effort. The number of alternative solutions in a superstructure can be limited by eliminating many alternatives which are predetermined to be either suboptimal or closely related to alternatives which remain. It is often possible to search this reduced superstructure when it is not possible to search the all-inclusive superstructure. The search effort can be limited by restricting the search to only a prespecified number of alternatives or by restricting the amount of computer time used in searching. The best solution found in these restricted searches is used as the best solution to the problem. It is important in these limited searches to find a good feasible solution early in the search so that a feasible solution exists when the search is terminated. Heuristic methods such as the "greedy method" (Ibaraki, 1976) have been developed to find a feasible solution quickly. This feasible solution can be used to trim the remaining search. These two methods, reducing the size of the superstructure and limiting the search effort, can also be combined. The main drawback of these methods is that they do not examine all feasible solutions, so the optimal solution found may not be the optimal solution to the original problem. Very good solutions are often found, but usually no estimate can be made of how far the solution obtained using these incomplete search methods is from the optimum solution.

The ideal superstructure for heat-integrated distillation sequence synthesis should contain all feasible distillation sequences, all feasible heat integration structures within these sequences, and all feasible operating conditions for any column within the superstructure. Including all possible distillation sequences is easy if only simple, sharp distillation columns are used and if only pure products are desired. The number of such sequences, NS, is given by (Thompson and King, 1972) $NS = [2(NC-1)]!/[NC!\,(NC-1)!]$. The opportunities for heat exchange and the column operating conditions are interrelated, so it is not possible to determine them independently. In addition, the column operating conditions are continuous variables which must be made discrete to include them in a finite superstructure. Each of these points will be discussed in further detail in the succeeding sections.

A superstructure of distillation columns is constructed from single distillation tasks. These single tasks can be combined to form distillation sequences and the sequences can be combined to form a superstructure. Describing the distillation tasks and sequences which can be used for a given problem is easy if only simple, sharp distillation columns are used and if only pure products are desired. The list splitting technique of Hendry and Hughes (1972) is used. The single distillation tasks for a five-component example and the distillation sequences they form are illustrated in Figure 1. If nonsharp distillation columns are used or if the desired products are mixtures, describing the distillation tasks and sequencies is more difficult. The technique of Hendry and Hughes can be used as a guideline, but the engineer must add nonsharp distillation columns where appropriate and remove any columns which are inappropriate (e.g. which separate two components that form one mixed product).

Once the distillation tasks and their connection in distillation sequences have been determined, it is still necessary to specify the number of columns performing each distillation task and their operating conditions. The important operating variables in each column are the reflux ratio, the degree of feed vaporization, and the column pressure. Choosing the optimum reflux ratio, which also determines the number of trays if the product purity is specified, is a compromise between the increased utility cost associated with higher reflux ratios, the increased capital cost associated with the higher vapor flow rates at higher reflux ratios, and the decreased capital cost associated with the fewer trays required at higher reflux ratios. The economically optimum reflux ratio usually lies in the range of 1.1 to 1.25 times the minimum reflux ratio. However, since the cost function is flat for a short range above the optimum, the optimum reflux ratio is often assumed to be 1.2 to 1.3 times the minimum reflux ratio (King, 1971). In this study it will be assumed that the optimal ratio is always 1.2 times the minimum reflux ratio. Also in this study the degree of feed vaporization is assumed to be the same in all columns. The feed to each column is assumed to be saturated liquid.

Determining the number of columns for each distillation task to include in the superstructure and the pressure at which to operate each of them is difficult since these choices determine the opportunities for heat integration. One way to handle this problem is to determine arbitrarily the number of columns of each type and their pressure levels. This can be done by including several columns which operate at arbitrarily chosen pressures between ambient pressure and the critical pressure of the distillate. For example,

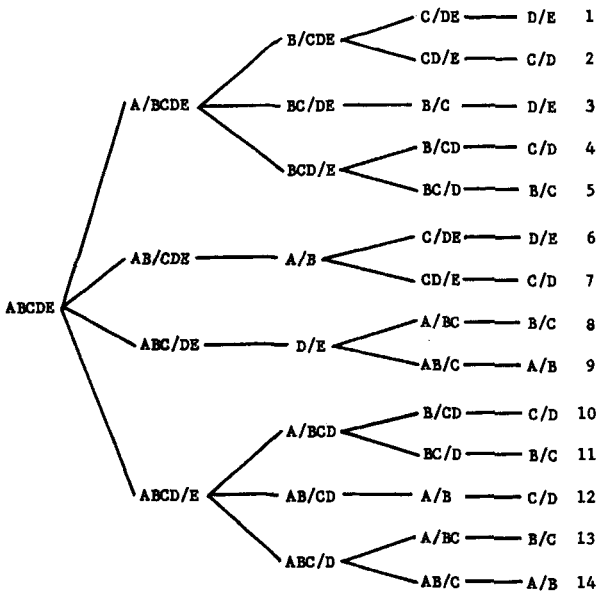

Figure 1. The distillation sequences for a five-component feed stream.

column pressures could be chosen at prespecified intervals of 10, 20, or 1,000 kPa or could be chosen randomly. The opportunities for heat integration in such a superstructure can be determined by calculating the condenser and reboiler temperatures for each column. Heat integration is feasible if the condenser temperature of one column is greater than ΔT_{min} above the reboiler temperature of another.

A second method for determining the number of columns and their pressures is first to specify the desired heat integration possibilities, then determine the number of columns and their pressures so that these opportunities are realized. This is the case in multieffect distillation. The first column usually operates at 100 kPa or at a pressure high enough that the distillate vapor can be condensed using available cold utilities, whichever is greater. The reboiler temperature of this column can be determined once the pressure is fixed. The condenser temperature of the second column in a multieffect distillation must be at least ΔT_{min} above the reboiler temperature of the first column so that the condenser of the second column can provide the heat required in the reboiler of the first. The pressure in the second column is chosen to produce this condenser temperature. Additional effects can be included by increasing the pressure in each succeeding column so that its condenser temperature is ΔT_{min} above the reboiler temperature of the previous column.

In developing a superstructure for the synthesis of heat-integrated distillation sequences, the number of columns of each type and the pressure in each of them is determined by specifying the opportunities for heat integration between distillation columns. These opportunities for heat exchange depend on several factors. The most important are the column temperatures and pressures that determine which heat exchanges are feasible. Fixing the column pressure for a given separation also fixes the condenser and reboiler temperatures. Similarly, fixing the condenser or reboiler temperature fixes the pressure. In general, at higher pressures the reboiler and condenser temperatures for a given separation are higher than they would be at a lower pressure. Another important factor in determining the opportunities for heat exchange between columns is the minimum temperature difference allowed for heat exchange, ΔT_{min}. Smaller values of ΔT_{min} mean that smaller pressure differences between columns provide the same opportunities for heat integration which would require larger pressure differences if ΔT_{min} were higher.

The utilities which are available and the critical properties of the components determine the range of operating pressures for each column type. No column can operate at such a high pressure that no available hot utility can be used to reboil the bottoms or at such a low pressure that no available cold utility can be used to condense the distillate. No column may operate with a reboiler temperature above the critical temperature of the bottoms or a condenser temperature above the critical temperature of the distillate. These criteria determine the range of feasible operating pressures for each distillation task. If the lower pressure limit is below 100 kPa, a pressure of 100 kPa is often used as the lower limit since operating under vacuum is expensive.

A superstructure for the synthesis of heat-integrated distillation sequences can be generated by first determining the column types which should be included in the superstructure and then connecting these columns to form distillation sequences. The number of each column type which should be included and the pressure in each column are determined by considering the opportunities for heat integration. One procedure for generating such a superstructure is described in the following algorithm.

Reboiler/Condenser Temperature Algorithm

1. The allowable temperature range for all columns is set by the temperature of the available utilities and by the minimum temperature difference allowed between streams exchanging heat, ΔT_{min}. All columns must operate with condenser and reboiler temperatures between $T_{hot,max} - \Delta T_{min}$ and $T_{cold,min} + \Delta T_{min}$, where $T_{hot,max}$ is the temperature of the highest temperature hot utility available, and $T_{cold,min}$ is the temperature of the lowest temperature cold utility available.

2. The engineer must specify a set of distillation tasks which will be used in the superstructure. For each distillation task he must specify the composition of the feed, the key components for the separation, and the recovery of the keys in the distillate or bottoms. For sharp separations (separations where each entering component leaves in only one product stream) the method of ranking components in decreasing order of relative volatility developed by Hendry and Hughes (1972) is useful. If sharp separators cannot or should not be used, the engineer must use his judgment to include all distillation tasks which will produce feasible solutions.

3. Determine the composition of the distillate and bottoms streams in each column. For sharp separators these compositions have been specified by setting the recoveries of adjacent keys. For nonsharp separators or separators which split between nonadjacent keys, it may be necessary to perform shortcut distillation calculations to obtain these values.

4. Find the minimum operating pressure for each distillation task. For each task this is the bubble pressure at the distillate composition and at the coldest possible condenser temperature, $T_{cold,min} + \Delta T_{min}$. The reboiler temperature for each task at this pressure is obtained by calculating the dew point temperature at the bottoms composition. At this point, include one column of each distillation task in the superstructure and set its pressure at the pressure just calculated.

5. Find the lowest reboiler temperature of all columns in the superstructure. Define a new condenser temperature, $T_{cond,new}$ equal to $T_{reb,min} + \Delta T_{min}$. This is the minimum temperature at which the condenser of any other column could transfer heat to this reboiler. Add to the superstructure one column of each type with a condenser operating at $T_{cond,new}$.

6. Calculate the reboiler temperature for all columns added in step 5. This can be done in two ways:

(a) The first and simplest method is to assume that the temperature difference between the condenser and reboiler in a distillation column is constant and not affected by column operating conditions. The temperature difference between the reboiler and condenser calculated in step 4, ΔT_{RC}, can be used to calculate the reboiler temperature in any column if the condenser temperature is known.

$$T_{reb} = T_{cond} + \Delta T_{RC} \tag{1}$$

(b) The second method accounts for the fact that the temperature difference across a distillation column is a function of the column operating conditions. By performing calculations for example problems it has been found that the temperature difference between the reboiler and condenser in a distillation task increases linearly with temperature. This is illustrated in Figure 2. The variation in the temperature difference for any distillation task can be obtained by calculating the differences at two different temperatures (pressures) and fitting a straight line between the points. The temperature difference across any column of this type can then be obtained from the condenser temperature according to

$$\Delta T_{RC} = K_1 + K_2 * T_{cond} \tag{2}$$

where K_1 and K_2 are the intercept and slope of the line relating the condenser temperature and the temperature difference across a column. The reboiler temperature can again be obtained by Eq. 1.

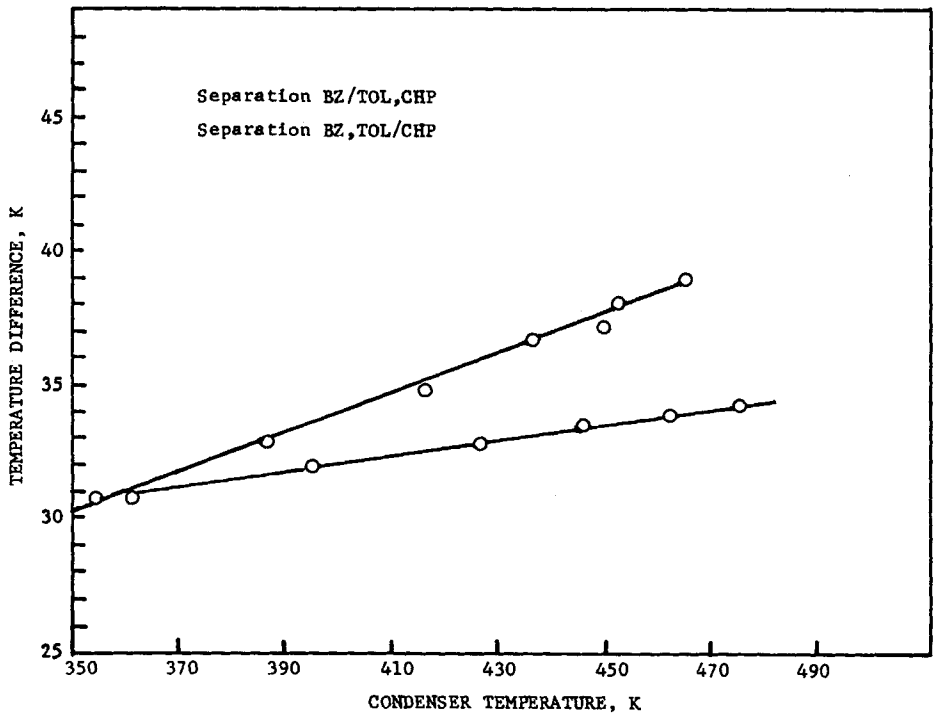

Figure 2. Variation of column ΔT with column operating temperature.

7. If the reboiler temperature for any column is outside the range determined in step 1, omit that column from the superstructure and omit distillation tasks of that type from consideration in further steps.

8. Remove the reboiler temperature found in step 5 from further steps and repeat steps 5 to 7 until no further columns can be added to the superstructure.

9. The pressure in each column can now be determined by performing either a bubble pressure calculation at the condenser conditions or a dew pressure calculation at the reboiler conditions. If the pressure in any column is greater than the critical pressure of either the distillate or bottoms stream, remove that column from the superstructure.

10. At this point the superstructure is complete. The number of columns of each distillation task has been determined, and the reboiler temperatures, condenser temperatures, and column pressures have been specified for each column.

This algorithm can be illustrated by application to the following example problems.

Example Problem 1

Component	x_F
A. Benzene	0.60
B. Toluene	0.30
C. Cycloheptane	0.10

Feed flow = 250 kgmol/hr
ΔT_{min} = 10 K
Desired Products: Pure Benzene
Pure Toluene
Pure Cycloheptane
Recovery in all columns is specified to be 98%.

Utilities	Temp. K	Cost $/10^6 kJ
1. Cooling Water (20 K Rise)	305	0.159
2. 101.4 kPa Steam	373	1.075
3. 446.4 kPa Steam	421	2.807
4. 1,138 kPa Steam	459	3.980
5. 4,243 kPa Steam	527	5.333

Example Problem 2

Component	x_F
A. Propane	0.05
B. Isobutane	0.15
C. *n*-Butane	0.25
D. Isopentane	0.20
E. *n*-Pentane	0.35

Feed flow = 907.2 kgmol/hr
ΔT_{min} = 10 K
Desired Products: Pure Propane
Pure Isobutane
Pure *n*-Butane
Pure Isopentane
Pure *n*-Pentane
Recovery in all columns is specified to be 98%.

Utilities	Temp. K	Cost $/10^6 kJ
1. Cooling Water (20 K Rise)	305	0.159
2. 172 kPa Steam	389	1.653
3. 414 kPa Steam	418	2.709
4. 1,138 kPa Steam	459	3.980
5. 2,862 kPa Steam	504	4.885

TABLE 1. EFFECT OF TEMPERATURE DIFFERENCE ON NUMBER OF COLUMNS IN A SUPERSTRUCTURE GENERATED USING THE REBOILER/CONDENSER TEMPERATURE METHOD

Example Problem	Min. Temp. Difference between Successive Reboiler temp. 0 K	5 K	10 K	20 K
1	180	50	29	17
2	>2,000	479	282	159

A major shortcoming of this algorithm is that the superstructure it generates has a very large number of columns. For a five-component problem the superstructure generated may contain columns operating at temperatures which differ by only one or two degrees for the same separation. One way to alleviate this problem is to require that the reboiler temperature chosen in step 5 differ from the reboiler temperature chosen in the previous iteration by an arbitrary temperature difference. A difference of 5, 10, or 20 K might be used. How this change affects the number of columns in the superstructure is shown in Table 1.

Another approach which can be used to generate a superstructure is to characterize each distillation task by a temperature difference which reflects both the difference between reboiler and condenser temperatures and the minimum temperature difference allowed for heat exchange. The characteristic temperature for each column is obtained from:

$$\Delta T_{\text{char},i} = T_{\text{reb},i} - T_{\text{cond},i} + \Delta T_{hx,i} \tag{3}$$

where the column reboiler and condenser temperatures are calculated at arbitrary conditions. The minimum operating pressure for the task may be used, but a value which better reflects the variation of this temperature difference with pressure can be obtained by evaluating the temperature difference at a mid-range pressure or by using the arithmetic average of values obtained at low and high pressures. The temperature difference for heat exchange, $\Delta T_{hx,i}$, is variable. For each column it is chosen so that the characteristic temperature of that column is an integer multiple of the lowest characteristic temperature in the system. A superstructure of distillation columns is then generated by including columns of each distillation task operating with condensers at the lowest feasible temperature and at temperatures which are integer multiples of the minimum characteristic temperature above this minimum feasible temperature. Since the characteristic temperature of each column is a multiple of this minimum characteristic temperature, columns will begin to stack upon themselves after a few columns have been generated. The following algorithm describes how to generate a superstructure using this method.

Characteristic Temperature Algorithm

1. The allowable temperature range for all columns is set by the temperature of the available utilities and by the minimum temperature difference allowed between streams exchanging heat, ΔT_{min}. All columns must operate with condenser and reboiler temperatures between $T_{\text{hot,max}} - \Delta T_{\text{min}}/2$ and $T_{\text{cold,min}} + \Delta T_{\text{min}}/2$, where $T_{\text{hot,max}}$ is the temperature of the highest temperature hot utility available, and $T_{\text{cold,min}}$ is the temperature of the lowest temperature cold utility available.

2. Repeat steps 2 and 3 of the reboiler/condenser temperature algorithm, above.

3. For each distillation task determine the difference between the reboiler and condenser temperatures which will be used to characterize that task. This can be obtained by calculating this difference at the lowest pressure at which the column could operate in a given problem, or by calculating this difference at the lowest and the highest feasible operating pressures and averaging them.

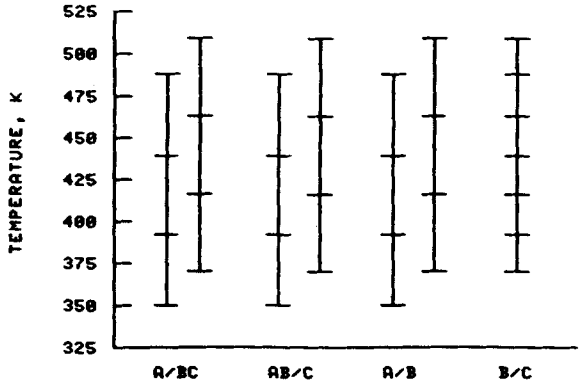

Figure 3. Superstructure for example problem 1 generated using the characteristic temperature method.

4. The temperature difference for heat exchange in each column is chosen so that the characteristic temperature for each column defined in Eq. 3 is an integer multiple of the minimum characteristic temperature in the system. The minimum characteristic temperature is initially obtained by adding ΔT_{min} to the smallest temperature difference calculated in step 3. The characteristic temperature for the other columns is chosen so that

$$\Delta T_{\text{char},i} = I * \Delta T_{\text{char,min}} \tag{4}$$

where I is an integer chosen such that

$$\Delta T_{\text{char},i} - \Delta T_{RC,i} \geq \Delta T_{\text{min}} \tag{5}$$

The minimum characteristic temperature may be increased so that Eqs. 4 and 5 can be satisfied for all distillation tasks.

5. The superstructure can now be generated. For each distillation task begin at the lowest feasible condenser temperature for that task (this temperature should be adjusted so that it is some integer multiple of the minimum characteristic temperature above the minimum allowable condenser temperature defined by the available utilities). Add one column of that type with a condenser at this temperature and add columns with condenser temperatures $\Delta T_{\text{char,min}}$, $2\Delta T_{\text{char,min}}$, $3\Delta T_{\text{char,min}}$, etc. above that. The limit to the number of columns of each type included in the superstructure is the maximum temperature of the hot utilities available. If the reboiler temperature of any column exceeds the maximum temperature calculated in step 1, eliminate that column from the superstructure and do not add any more columns of that type to the superstructure. The reboiler temperature of any column of type i is calculated by

$$\Delta T_{\text{reb}} = \Delta T_{\text{cond}} + \Delta T_{\text{char},i} \tag{6}$$

6. The pressure in each column can now be determined by performing either a bubble pressure calculation at the condenser conditions or a dew pressure calculation at the reboiler conditions. If the pressure in any column is greater than the critical pressure of either the distillate or bottoms stream, remove that column from the superstructure.

7. At this point the superstructure is complete. The number of columns of each distillation task has been determined and the reboiler temperatures, condenser temperatures, and column pressures have been specified for each column.

The superstructures generated by this algorithm for the example problems described previously are shown in Figures 3 and 4. The superstructure for example problem 1 contains 24 columns, and

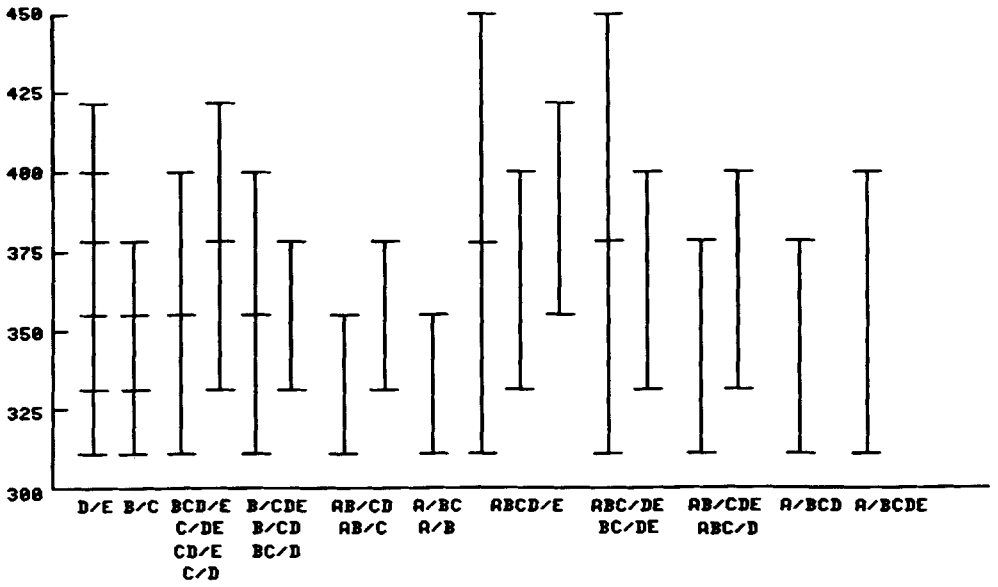

Figure 4. Superstructure for example problem 2 generated using the characteristic temperature method.

the superstructure for example problem 2 contains 55 columns. These numbers can be compared with the numbers in Table 1 which show the size of the superstructures for these example problems generated using the reboiler/condenser temperature method. This algorithm generates a superstructure which maintains much of the structure developed using the reboiler/condenser temperature method, but with many fewer columns. For this reason we suggest use of the characteristic temperature method rather than the reboiler/condenser temperature method.

FORMULATION AND SOLUTION OF THE MIXED INTEGER LINEAR PROGRAM (MILP)

In the previous section two methods have been described for generating superstructures for the problem of heat integrated distillation sequence synthesis. This section describes the formulation of a mixed integer linear program which represents this superstructure and the heat integration possibilities within it.

The MILP Objective Function

The first step in formulating a mixed integer linear program for this problem is devising an objective function which can be used to compare different alternatives. An obvious choice for the objective function is the project cost, which is made up of equipment costs and operating costs. The major equipment costs in a distillation system are those of the distillation columns themselves and of the heat exchangers used as condensers and reboilers. The major operating cost is the cost of the utilities used. The objective function should account for both of these costs.

The cost of a distillation column is a function of many variables. Among these are the feed flow rate to the column, the reflux ratio, the number of trays, the column temperature, the column pressure, and the physical properties of the feed components. Most of these parameters are fixed implicitly or explicitly by the superstructure generating algorithms of the previous section. The pressures and temperatures of each column are specified by these algorithms, and from them the minimum reflux ratio and minimum number of trays can be calculated (Fenske, 1932; Underwood, 1946, 1948). The operating reflux ratio is chosen to be 1.2 times the minimum as discussed earlier. This also determines the number of trays for a specified product purity (Gilliland, 1940; Erbar and Maddox, 1961). The only unspecified variable is the feed flow rate, which depends on the distillation system.

The cost of a distillation column increases with increasing feed flow if all other variables remain unchanged. This increase is not linear but reflects the six-tenths factor rule (Peters and Timmerhaus, 1968) commonly used to scale equipment costs. This rule is

$$\text{Cost}_2 = (\text{Capacity}_2/\text{Capacity}_1)^{0.6} * \text{Cost}_1 \qquad (7)$$

A linear approximation to this nonlinear behavior is required for this problem since a linear program cannot handle nonlinear

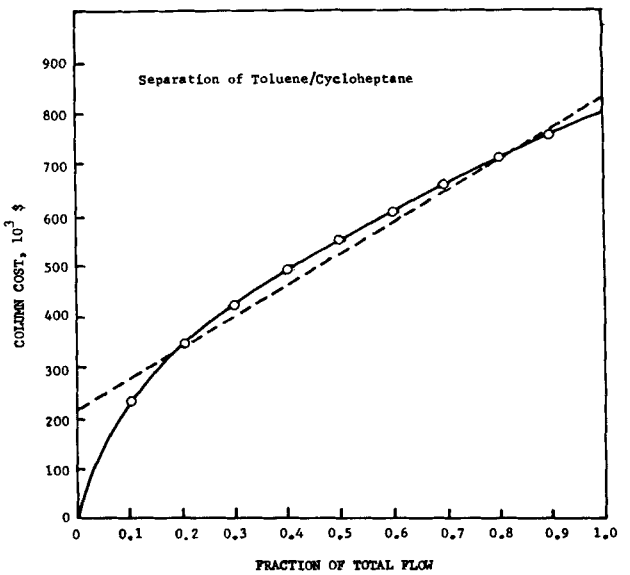

Figure 5. Column cost as a function of feed flow rate.

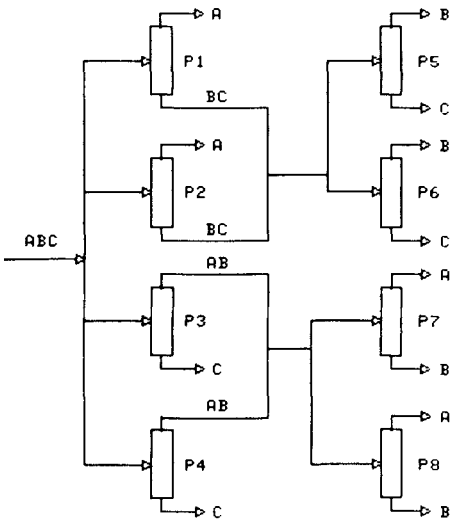

Figure 6. A small superstructure.

functions. The cost of one of the distillation columns from example problem 1 is shown as a function of the feed flow rate in Figure 5. This cost can be approximated by a function of the form

$$\text{Cost} = FC + V * F \tag{8}$$

where FC is a fixed cost associated with the column and V is the slope of the line relating the column cost to the feed flow rate, F. There are many values of FC and V which could adequately describe the actual nonlinear cost function, but in this study a line passing through the column costs at 20 and 80% of the maximum capacity is used. This is also shown in Figure 5. This particular function does not predict the actual column cost at low flow rates very well, but predicts the costs at high flow rates quite accurately. This type of behavior is desired in this problem since conventional distillation sequences within the superstructure would operate with columns at the maximum flow conditions, and multieffect systems usually split the flow evenly between two or three columns.

The utility costs of a distillation system include the costs of the hot utilities used in the reboilers and the cold utilities used in the condensers. The cost per unit of energy supplied or removed by each utility is given in the problem specifications, but the amount of each utility used is a function of the distillation sequence and the heat exchanger network.

The objective function for the mixed integer linear program can be formulated as $VC = (\text{CAC}/\alpha) + \beta(OPC)$, where CAC is the installed capital cost and OPC is the annual operating cost. The parameter α is the payout time for capital investment and β corrects the operating costs for the income tax rate. In this paper, unless otherwise noted the payout time is assumed to be 2.5 years and β is 0.52.

The venture cost for a distillation superstructure can be formulated as

$$VC = \frac{1}{\alpha} \sum_{k \in COLS} [FC_k + V_k F_k] + \beta \sum_{j \in HU} C_{HU,j} Q_H(j) + \beta \sum_{i \in CU} C_{CU,i} Q_C(i) \tag{9}$$

where $COLS$ is the set of all columns in the superstructure, HU is the set of all hot utilities, and CU is the set of all cold utilities. The first term is the capital cost of the distillation columns and the other terms are the costs of the hot and cold utilities which are required.

This objective function is subject to constraints of three types. Material balance constraints describe the permissible routes by which material may flow from one column in the superstructure to another. Some heat integration constraints describe the amount of heat associated with each reboiler and condenser while others describe the feasible heat exchange possibilities. The third type of constraints, which will be called integrality constraints, insure consistency between the continuous and integer variables.

Material Balance Constraints

Material balance constraints relate material flows into and out of columns in the superstructure. Each column separates its feed into two product streams whose amounts are related to the feed flow by

$$\begin{aligned} D_k &= \xi_D F_k \\ B_k &= \xi_B F_k = (1 - \xi_D) F_k \end{aligned} \tag{10}$$

where ξ_D is the fraction of the feed to column k which leaves in the distillate and ξ_B is the fraction which leaves in the bottoms. Interconnections between distillation columns are also described by material balance constraints. Consider the structure shown in Figure 6. The feed streams to many columns in this structure are product streams from other columns. For each intermediate product of two or more components in this structure, a constraint is written which states that the amount of each intermediate product produced by columns in the structure must equal the amount of that intermediate product fed to columns which further separate the product. That is

$$\sum_{i \in PS_m} \xi_i F_i - \sum_{j \in FS_m} F_j = 0 \qquad m \in IP \tag{11}$$

where PS_m is the set of all columns which produce a given intermediate product m as distillate or bottoms, FS_m is the set of all columns having intermediate product m as feed, F is the total flow rate to a column, IP is the set of all intermediate products, and ξ is the split fraction relating distillate or bottoms flows to feed flows. This constraint is written for each intermediate product. A similar expression is necessary for the feed to the distillation system:

$$\sum_{k \in FS_F} F_k = F_{\text{TOT}} \tag{12}$$

The total feed to the system must equal the sum of the feeds to all columns which process some portion of the feed stream.

Heat Integration Constraints

Once a superstructure has been developed the problem of heat integration within that superstructure is very similar to the heat exchanger network synthesis problem. The hot and cold streams are the condenser streams, reboiler streams, and the utilities. The temperature levels for the reboiler and condenser streams are fixed by specifying the column pressures, and the temperature level of each utility is given in the problem specification. The amount of heat associated with each reboiler and condenser however, is not known. Since all other column operating conditions are fixed in the development of the superstructure, the heat duty in each condenser and reboiler is only a function of the column feed flow rate. These duties are directly proportional to the feed flow.

$$Q_k = Q_{\text{ref},k} \frac{F_K}{F_{\text{ref},k}} \tag{13}$$

where $Q_{\text{ref},k}$ is the condenser or reboiler duty of column k with a feed flow rate of $F_{\text{ref},k}$, and Q_k is the reboiler or condenser duty for that column with a feed flow rate of F_k. In this study the maximum possible feed flow in a particular column is used to evaluate the reference heat duty for that column.

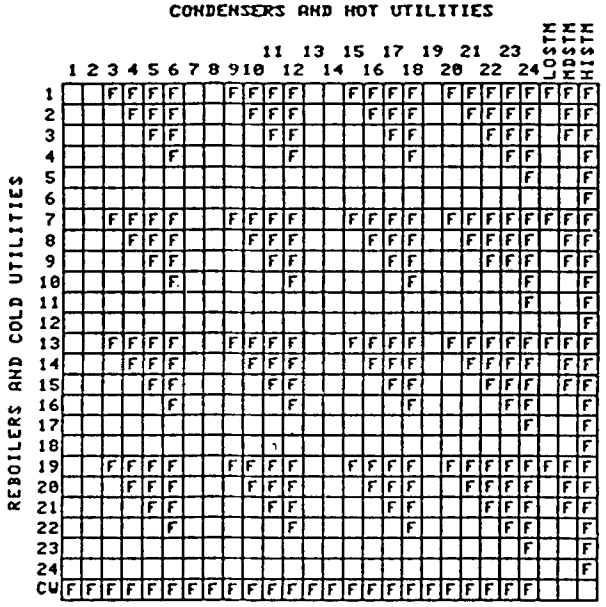

Figure 7. Stream matrix for the superstructure of example problem 1 generated using the characteristic temperature method.

It is also assumed in this study that the heat duties in the condenser and reboiler of each column are equal. This assumption can be justified by considering an energy balance around a single distillation column. Heat enters the column in the reboiler and with the feed stream; heat leaves in the condenser and with the distillate and bottoms products. The distillate and bottoms products are normally saturated liquids. If it is further assumed that the feed stream is also a saturated liquid (as is the case in this study), the net effect of these streams on the overall energy balance is only the difference in sensible heat between the product streams and the feed streams. The reboiler and condenser heat loads involve latent heat effects which are much larger than the sensible heat effects, particularly at high reflux ratios. If the sensible heat effects are ignored entirely, the energy balance states that the condenser and reboiler duties must be equal.

The feasible heat exchange possibilities within the superstructure for heat-integrated distillation sequence synthesis can be described using methods developed for heat exchanger network synthesis: formulation as a transportation problem in linear programming (Cerda et al., 1983) or as a transshipment problem in linear programming (Papoulias, 1982).

Transportation Problem Formulation

The transportation formulation for describing the feasible heat exchanges within a superstructure for heat-integrated distillation sequence synthesis is quite simple. The problem can be represented using the stream matrix developed by Pho and Lapidus (1973). This is illustrated in Figure 7. All of the condensers and hot utilities are listed across the top of the matrix and reboilers and cold utilities are listed along the side. Each row of the matrix is associated with a single reboiler or cold utility and each column is associated with a single condenser or hot utility. This is a simplification of the partitioning procedure developed by Cerda et al. (1983) which is made possible by the assumption that the condenser and reboiler streams exchange heat at a single temperature rather than over a temperature range as is usually the case in heat exchanger network synthesis. This assumption is certainly true for condensing or reboiling single-component product streams but is not true for multicomponent product streams. The boiling range of multicomponent product streams is neglected in this study and such streams are assumed to receive or transfer heat at their bubble point temperature. This temperature is chosen because all the heat rejected in a condenser is rejected at or above the bubble point temperature, and because in a kettle type reboiler all the heat is absorbed near the bubble temperature of the bottoms.

The possibilities for heat exchange within the superstructure can be determined from the superstructure and can be represented using the stream matrix. If the condenser temperature of one column is greater than the reboiler temperature of another by more than $\Delta T_{\min}$, then heat exchange between that condenser and reboiler is feasible and the letter F should be placed at the intersection of the row and column in the stream matrix corresponding to this match. Cells within the matrix corresponding to infeasible heat exchanges are left blank. In superstructures generated using the characteristic temperature method, the criterion for feasible heat exchange is that the condenser temperature of one column is greater than or equal to the reboiler temperature of another since $\Delta T_{\min}$ has been included in the characteristic temperature.

The amount of heat available in each condenser and reboiler is a function of the feed flow to the column

$$Q_k = Q_{\text{cond},k} = Q_{\text{reb},k} = K_k F_k \tag{14}$$

where K_k is a constant which can be obtained from Eq. 13. If the heat duty is greater than zero, Q_k units of heat must be rejected in the condenser and must be added in the reboiler. The heat from the condenser can be transferred to the reboiler of any other column with which a feasible match exists or to any feasible cold utility. Heat must be supplied to the reboiler from feasible condenser streams or hot utilities. For each condenser or reboiler the amount of heat transferred must equal the amount of heat which is available. These criteria can be written as:

$$\sum_{i \in FM_{\cdot j}} q(ij) = Q_H(j) \qquad j \in HS \tag{15}$$

$$\sum_{j \in FM_{i \cdot}} q(ij) = Q_C(i) \qquad i \in CS \tag{16}$$

where $q(ij)$ is the amount of heat transferred from condenser or hot utility j to reboiler or cold utility i; $FM_{\cdot j}$ is the set of feasible matches between reboilers or cold utilities and condenser or hot utility j; $FM_{i \cdot}$ is the set of feasible matches between condensers or hot utilities and reboiler or cold utility i; $Q_C(i)$ or $Q_H(j)$ is the amount of heat associated with each reboiler, condenser, or utility stream; HS is the set of all hot streams, process or utility; and CS is the set of all cold streams, process or utility. This transportation method of describing the heat integration requires $2NS + NHU + NCU$ constraints involving $\sigma(NS + NHU)(NS + NCU)$ variables where σ is the number of feasible heat matches divided by the total number of heat matches which are possible.

The amount of cold utility i which is required can be obtained from:

$$Q_C(i) = \sum_{j \in FM_{i \cdot}} q(ij) \qquad i \in CU \tag{17}$$

and the amount of hot utility j from:

$$Q_H(j) = \sum_{i \in FM_{\cdot j}} q(ij) \qquad j \in HU \tag{18}$$

These expressions for $Q_C(i)$ and $Q_H(j)$ can be substituted for the energy use terms in the objective function, Eq. 9.

Heat exchanger costs can also be included with the transportation formulation of the heat integration. This is done by relating the cost of the heat exchanger for each feasible match in Eqs. 15 and 16 to the amount of heat exchanged in that match. The cost of a heat exchanger is related to its area by the six-tenths factor rule given in Eq. 7. The area is calculated from

$$A = \frac{Q}{U\Delta T_{LM}} \tag{19}$$

where Q is the amount of heat transferred, U is the heat transfer coefficient for the match, and ΔT_{LM} is the log mean temperature difference between the streams in the match. The heat transfer coefficient can be estimated for each match, and ΔT_{LM} can be determined for each match from the temperatures in the superstructure. The heat exchanger area is then only a function of the amount of heat transferred in a particular match. The relation between the cost and the area is actually nonlinear, but in this analysis it is assumed that cost is directly proportional to area:

$$C_{HX,ij} = \frac{A_{ij}}{A_{\text{ref}}} C_{HX,\text{ref}} \tag{20}$$

where $C_{HX,\text{ref}}$ is the cost of a heat exchanger of area A_{ref}. The cost of a heat exchanger in this method is

$$\text{Cost} = C_{HX,ij}q(ij) = \frac{C_{HX,\text{ref}}}{A_{\text{ref}}U\Delta T_{LM}} q(ij) \tag{21}$$

All of the heat exchanger costs can be included in the formulation by adding the following terms to the objective function:

$$CAC_{HX} = \frac{1}{\alpha} \sum_{i \in CS} \sum_{j \in FM_i} C_{HX,ij}q(ij) \tag{22}$$

The Transshipment Model

The transshipment model does not explicitly describe the heat exchange between individual streams. The problem is partitioned into temperature intervals, and energy balances are written around each interval. This formulation gives the minimum amount of hot and cold utilities required for a given problem but does not define the heat exchanger network. In most cases, however, a heat exchanger network can be constructed by inspection. The advantage of this formulation over the transportation formulation is that fewer variables and constraints are required.

The rules used to determine the temperature intervals are (Papoulias, 1982):

1. Reduce the temperature of each hot utility by $\Delta T_{\min}$. If the reboiler/condenser temperature method has been used to generate the superstructure, also reduce the condenser temperature of each column by $\Delta T_{\min}$. If the characteristic temperature method has been used, do not adjust the temperatures since they have already been corrected for $\Delta T_{\min}$.

2. Place the reduced temperatures of all condensers and hot utilities as well as the temperatures of the reboilers and cold utilities on a list and arrange the temperatures on this list in decreasing order. This list defines the temperature intervals. Each condenser, reboiler, and utility stream is assigned to an interval according to its temperature. Condenser temperatures should be included in the interval just below where they appear on the list, and reboiler temperatures should be included in the interval just above the temperature at which they appear.

An upper bound on the number of intervals is:

$$NI \le 2NS + NHU + NCU - 1 \tag{23}$$

The number of intervals for a superstructure generated using the reboiler/condenser method will be close to the maximum, but the number of intervals for a superstructure generated using the characteristic temperature method will be much smaller than the maximum. This is because in superstructures generated using this method, reboiler and condenser temperatures are repeated.

The temperature intervals have been constructed so that it is always thermodynamically possible for the hot streams in an interval to transfer heat to the cold streams in the same interval or to cold streams in any lower temperature interval. An energy balance can be written around each interval m:

$$Q_{m-1,m} + \sum_{j \in HS_m} Q_H(j) - \sum_{i \in CS_m} Q_C(i) - Q_{m,m+1} = 0 \tag{24}$$

The terms $Q_{m-1,m}$ and $Q_{m,m+1}$ are residual terms representing the flow of heat to interval m from interval $m-1$ and to interval $m+1$ from interval m. These residual flows are necessary since the amount of heat which enters an interval in the hot streams may not balance the amount of heat required by the cold streams. These residual flows transfer the heat between intervals to balance the heating and cooling required in the entire problem.

The amount of each utility required is the minimum amount which will allow all of the interval energy balances to be satisfied. These amounts are determined explicitly in this formulation, unlike the transportation formulation.

Heat exchanger costs can also be included using the transshipment formulation of the heat integration. If all of the heating and cooling in a distillation system were supplied by utilities, the total amount of heat transferred would be

$$Q_{\text{TOT}} = \sum_{k \in COLS} (Q_{\text{cond},k} + Q_{\text{reb},k}) = 2 \sum_{k \in COLS} Q_k \tag{25}$$

if the reboiler and condenser duties for each column are equal. The amount of heat which is exchanged in condenser/reboiler matches can be determined by subtracting the total amount of hot and cold utilities required from the total amount of heating and cooling required.

$$Q_{R/C} = 2 \sum_{k \in COLS} Q_k - \sum_{j \in HU} Q_H(j) - \sum_{i \in CU} Q_C(i) \tag{26}$$

This equation accounts for the heat in a reboiler/condenser match twice, once as an amount of heat generated and once as an amount of heat absorbed. The actual amount of heat transferred in reboiler/condenser matches for a given system is half the amount calculated in Eq. 26. The total heat transferred in a distillation system is the sum of the heat transferred to cold utilities, the heat transferred from hot utilities, and the heat transferred in reboiler/condenser matches.

$$\begin{aligned} Q_{\text{TOT}} &= \sum_{i \in CU} Q_C(i) + \sum_{j \in HU} Q_H(j) \\ &\quad + 0.5\left[2 \sum_{k \in COLS} Q_k - \sum_{i \in CU} Q_C(j) + \sum_{j \in HU} Q_H(j)\right] \\ &= \sum_{k \in COLS} Q_k + 0.5\left[\sum_{i \in CU} Q_C(i) + \sum_{j \in HU} Q_H(j)\right] \end{aligned} \tag{27}$$

If the reboiler and condenser duties in each column are equal, the amount of hot utilities required is equal to the amount of cold utilities required. The amount of heat transferred in a distillation system is then

$$Q_{\text{TOT}} = \sum_{k \in COLS} Q_k + \sum_{i \in CU} Q_C(i) = \sum_{k \in COLS} Q_k + \sum_{j \in HU} Q_H(j) \tag{28}$$

The cost of the heat exchangers can be estimated using Eq. 21. The heat transfer coefficient and log mean temperature difference must be chosen to characterize the total heat exchange in the system rather than in a single exchanger. One good choice for the heat transfer coefficient is the arithmetic average of a reboiler and condenser heat transfer coefficient which are characteristic of the system. An upper bound on the heat exchanger costs could be obtained by using $\Delta T_{\min}$ for the log mean temperature difference, but a larger value would provide more realistic results. In this study the quantity $2\Delta T_{\min}$ will be used as the log mean temperature difference to estimate heat exchanger costs for the transshipment heat integration formulation.

Integrality Constraints

Neither the material balance nor the heat integration constraints account for the fact that each alternative distillation system consists of only a few distillation columns in the superstructure. Integer variables and constraints must be added to account for this. A binary variable is associated with each column in the superstructure. These variables indicate which columns are present in a particular distillation system. A binary variable with a value of one indicates that the column associated with that variable appears in the solution, a binary variable with a value of zero indicates that it does not.

These binary variables must be included in the objective function to account properly for the capital cost of a distillation sequence. According to Eq. 8 the cost of a distillation column is: Cost = $FC + V * F$, where FC is a fixed cost and V is the increase in cost for each unit of feed flow. The fixed charge in each column must be multiplied by the binary variable for that column so that the fixed charge is only included in the value of the objective function if that column is present in a particular solution.

It is also necessary to relate the value of continuous variables to the value of the corresponding binary variables. Specifically, the feed flow rate and heat duty associated with any column which is not present in a particular solution should be set to zero. This can be accomplished by adding the following constraint for each column in the superstructure:

$$F_k - U_k y_k \leq 0 \tag{29}$$

where U_k is an upper bound on the feed flow rate to column k. If the binary variable y_k is set to one, this constraint becomes

$$F_k \leq U_k \tag{30}$$

If the binary variable is set to zero, the feed flow rate will be set to zero. If the feed flow is zero, the heat duty will also be zero since the duty is proportional to the feed flow rate.

THE COMPLETE MILP FORMULATION

A complete mixed integer linear program can now be formulated for the problem of distillation sequence synthesis with heat integration. Two versions are possible; one uses the transportation problem model for the heat integration, the other uses the transshipment model. These two formulations will now be presented.

MILP Formulation with the Transportation Model for Heat Integration

$$\text{MIN}\ \frac{1}{\alpha}\sum_{k\in COLS}[FC_k y_k + V_k F_k] + \frac{1}{\alpha}\sum_{i\in CS}\sum_{j\in FM_{i\bullet}} C_{HX,ij}q(ij) + \beta\sum_{i\in CU} C_{CU,i}\sum_{j\in FM_{i\bullet}} q(ij) + \beta\sum_{j\in HU} C_{HU,j}\sum_{i\in FM_{\bullet j}} q(ij)$$

such that

$$\sum_{k\in FS_F} F_k = F_{\text{TOT}}$$

$$\sum_{i\in PS_m}\xi_i F_i - \sum_{j\in FS_m} F_j = 0 \qquad m \in IP$$

$$Q_k - K_k F_k = 0 \qquad k \in COLS$$

$$\sum_{i\in FM_{\bullet j}} q(ij) = Q_H(j) \qquad j \in HS$$

$$\sum_{j\in FM_{i\bullet}} q(ij) = Q_C(i) \qquad i \in CS$$

$$F_k - U_k y_k \leq 0 \qquad k \in COLS$$

$$y_k = 0 \text{ or } 1 \qquad k \in COLS$$

The decision variables in this formulation are the binary variables, y_k, the flow rates to each column, F_k, and the amount of heat transferred between each hot and cold stream, $q(ij)$.

MILP Formulation with the Transshipment Model for Heat Integration

$$\text{MIN}\ \frac{1}{\alpha}\sum_{k\in COLS}[FC_k y_k + V_k F_k] + \frac{C_{HX,sys}}{\alpha}\left[\sum_{k\in COLS} Q_k + \sum_{i\in CU} Q_C(i)\right] + \beta\sum_{i\in CU} C_{CU,i}Q_C(i) + \beta\sum_{j\in HU} C_{HU,j}Q_H(j)$$

such that

$$\sum_{k\in FS_F} F_k = F_{\text{TOT}}$$

$$\sum_{i\in PS_m}\xi_i F_i - \sum_{j\in FS_m} F_j = 0 \qquad m \in IP$$

$$Q_k - K_k F_k = 0 \qquad k \in COLS$$

$$Q_{m-1,m} + \sum_{j\in HS_m} Q_H(j) - \sum_{i\in CS_m} Q_C(i) - Q_{m,m+1} = 0 \qquad m = 1,2,\ldots,NI$$

$$F_k - U_k y_k \leq 0 \qquad k \in COLS$$

$$y_k = 0 \text{ or } 1 \qquad k \in COLS$$

The decision variables for this formulation are the binary variables, y_k, the flow rates to each column, F_k, the amount of each utility required, $Q_H(j)$ and $Q_C(i)$, and the residual heat transferred between intervals, $Q_{m,m+1}$ and $Q_{m-1,m}$.

SOLUTION OF THE MIXED INTEGER LINEAR PROGRAM

Either of the MILP formulations just described can be solved using commercial software systems. The MILP's for the examples to be presented in this section have been solved using LINDO (Schrage, 1981), an interactive linear programming and integer programming solution code.

The solution process can be accelerated by establishing a good feasible solution early in the MILP search, or even before initiating the search. A good solution can often be obtained by examining the solution to the linear program which results from relaxing the integer constraints on the MILP. From the columns present in this relaxed solution, choose a feasible distillation sequence which contains the minimum number of colums required to effect the desired separations. The integer solution corresponding to this choice is often a very good bound on the problem solution. This bound can be included in the problem formulation by adding a constraint which states that the value of the objective function must be less than the value of the objective function corresponding to this predetermined solution.

Solution of Example Problems

Example problems 1 and 2 described earlier have been solved using the methods presented in this section. The superstructures developed for these problems have been described earlier. The MILP used to solve these two problems will be generated using the

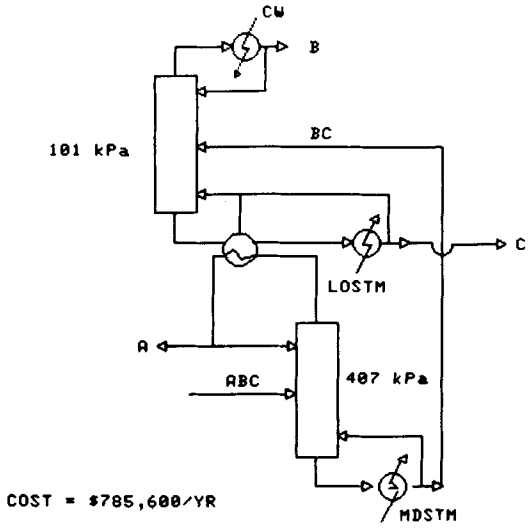

Figure 8. Solution to example problem 1 using MILP method.

superstructure obtained by the characteristic temperature method; these superstructures are shown in Figures 3 and 4.

The operating conditions in each column were obtained using the PROCESS flowsheeting system (Simulation Sciences, 1982). For each distillation task in each problem simulations were run at several pressures between 101.4 and 1,520.3 kPa. Using this data, a linear fit of the column temperature difference as a function of condenser temperature and of the column heat duty as a function of the condenser temperature was obtained for each distillation task. Column pressures were obtained from the condenser temperatures by linear interpolation between data points obtained from the simulations.

The costs for each column were obtained using the method of Guthrie (1969). A cost index of 266.1 (September, 1980) was used to adjust the costs reported in his paper (cost index 115.7 in mid-1968) to current conditions. The reference heat exchanger costs which are required were also evaluated using the method of Guthrie.

Example problem 1 was solved using both the transportation and transshipment formulations for the heat integration. The MILP using the transportation formulation has 24 binary variables, 296 continuous variables, and 105 constraints. Solution of this MILP using a good feasible solution as a bound required 40.6 seconds of

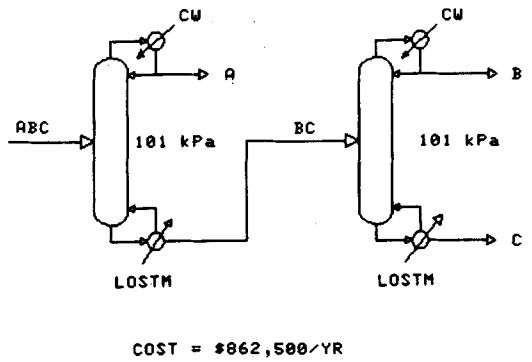

Figure 9. Best solution to example problem 1 without heat integration.

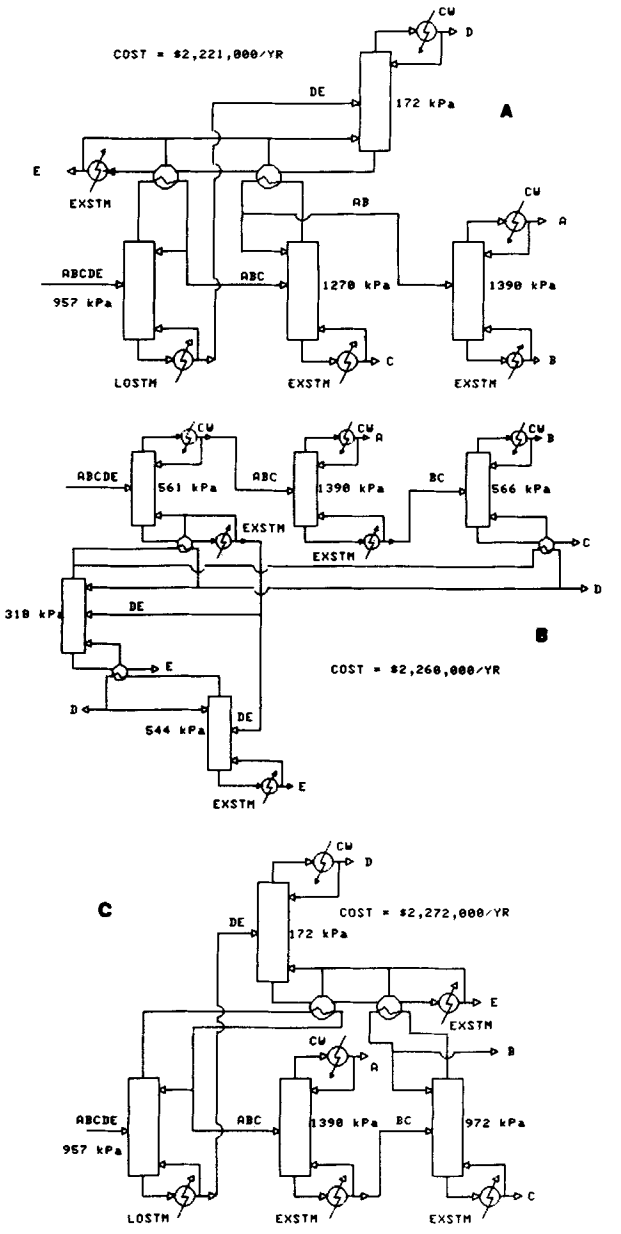

Figure 10. Solutions to example problem 2 using MILP method.

DEC-20 CPU time using LINDO. The MILP formulation of this same problem using the transshipment formulation for the heat integration has 24 binary variables, 64 continuous variables, and 66 constraints. Solution of this formulation using the same bound as in the transportation formulation required only 13.2 seconds of DEC-20 CPU time using LINDO.

The same solution is found using both formulations. This solution is shown in Figure 8. The cost of this solution using the transshipment objective function is $785,600/yr; the cost using the transportation formulation is $794,400/yr. The cost of this solution with a detailed application of Guthrie's method is $883,400/yr. These solutions can be compared to the best solution without heat integration, shown in Figure 9. The cost for this sequence from the MILP is $862,500/yr, and from a more detailed costing is $920,800/yr.

Example problem 2, which is a problem studied by other in-

TABLE 2. COMPARISON OF SOLUTIONS TO EXAMPLE PROBLEM 2

Sequence	Cost, 10^3 \$/yr
MILP Solution 1	2,221
Morari Solution 2	
MILP Solution 2	2,260
MILP Solution 3	2,272
Morari Solution 1	
Rathore Solution 1	2,313
Morari Solution 3	2,379
Rathore Solution 2	2,413
Rathore Solution 3	2,480
Sophos Solution	3,011

vestigators (Rathore et al., 1974; Sophos et al., 1978; Morari and Faith, 1980), was solved using only the transshipment formulation for the heat integration. The MILP formulation for this problem has 55 binary variables, 121 continuous variables, and 131 constraints. Solution of this MILP using a good solution as a bound required 74.2 seconds of DEC-20 CPU time. The cost of this solution using the MILP objective function is \$2,221,000/yr. The next

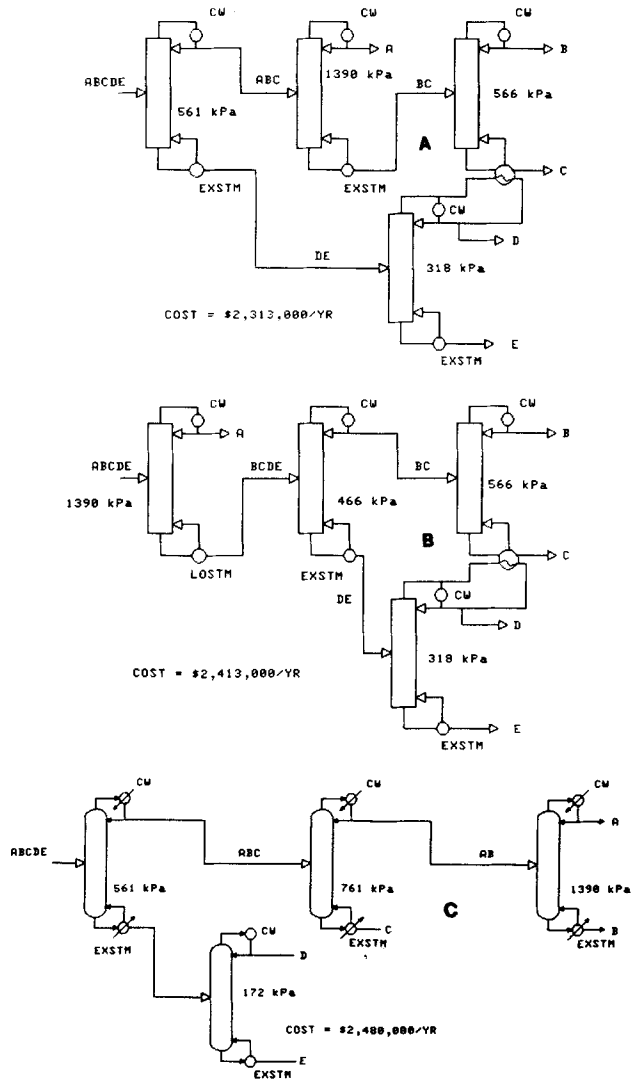

Figure 11. Rathore solutions to example problem 2.

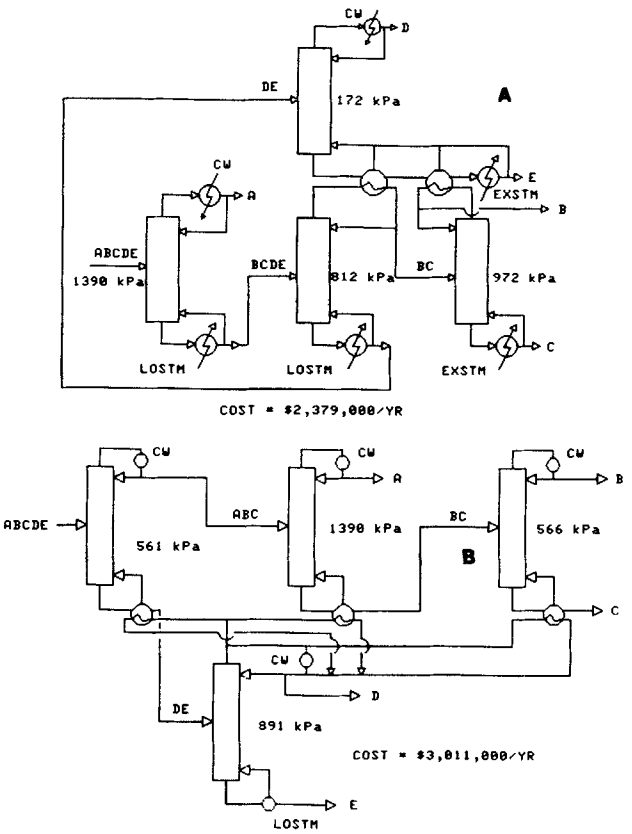

Figure 12. Sophos and Morari solutions to example problem 2.

best solution was found by adding an integer constraint to eliminate the optimal solution. The cost of this solution is \$2,260,000/yr. The third best solution has a cost of \$2,272,000/yr. These solutions are shown in Figure 10.

The distillation systems determined by the MILP method can be compared to the systems found by other investigators. The difficulty is that the costs of these sequences were originally determined using different economic assumptions than those used in this study. Rathore et al. and Morari and Faith, for example, used a 10-year payout time and assumed β was equal to one to obtain their costs. Sophos et al. assumed a payout time of 3.33 years and assumed β equal to 0.52. Another difference between the economic assumptions of these works and of the work in this study is the utility costs. In this work it is assumed that utilities are available only at discrete temperature levels with discrete costs. Rathore and Sophos have assumed that utilities are available at any temperature desired and that the cost of a utility is proportional to its temperature. For these reasons the costs of the sequences obtained by these investigators have been recalculated. The costs of their best solutions are obtained by calculating the cost of the same sequence with the same heat integration structure which is present in the superstructure. This can be done by constraining the integer variables in the MILP. The results are shown in Table 2. The distillation structures different from the MILP solutions are shown in Figures 11 and 12.

The Sophos solution suffers substantially because of differences in the utility costs. Raising the pressure in the D/E column high enough to reboil both the A/BC and ABC/DE columns requires that the more expensive low pressure steam be used in the D/E column reboiler rather than the cheaper exhaust steam. This increase in the cost of steam offsets the savings gained by the in-

creased heat integration. If continuous utility costs are used, the penalty for raising the pressure in the D/E column is much less, and this alternative is more attractive.

DISCUSSION

In this paper a new method has been presented for solving the problem of distillation sequence synthesis with heat integration. This method is an improvement over the previously developed methods for several reasons. The first is that it is faster than previous methods. Solution of the MILP for the Rathore problem required only one minute of CPU time compared with 26 minutes for other methods. The overhead for formulating the MILP must also be considered, but the flash calculations used to develop the superstructure and the shortcut distillation sizings required to formulate the MILP require only seconds of CPU time. A second advantage of this approach is that it includes multieffect distillation as a possibility. Most column types appear in the superstructure more than once, and if a multieffect distillation sequence is the least expensive solution to a given separation problem, this approach will find it. A third advantage of this method is that it can be used to include heat integrated distillation sequences in MILP formulations for the synthesis of total processing systems (Papoulias, 1982). The MILP formulation can also be easily modified to include integration with process streams.

The major drawback of this method is that it is limited to finding solutions which are included in the superstructure. The superstructures generated in this work have included only distillation columns performing simple, sharp splits. However, these superstructures contain many very good solutions as can be seen by comparing the solutions generated using this method to solutions obtained by other methods.

Further developments using this method are also possible. One extension of this work would be the development of a software package which ties together the disparate parts of this method. The engineer would only have to input the feed conditions and product requirements, and the program, interacting with the engineer, would generate the solution. Another extension of this work would be researching the application of a method of this type to other separation processes or to other unit operations.

ACKNOWLEDGMENT

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NOTATION

For clarity, column subscripts have been suppressed in the text for some of these variables.

α = payout time for capital investment
A_{ij} = heat exchanger area for heat transfer from heat source j to heat sink i
A_{ref} = reference heat exchanger area
B_k = flow of bottoms product from column k
$C_{CU,i}$ = annual cost per unit of heat flow to a cold utility for the cold utility
$C_{HU,j}$ = annual cost per unit of heat flow from a hot utility for the hot utility
$C_{HX,ij}$ = cost of heat exchanger area per unit of heat transfer for heat transfer from heat source j to heat sink i
$C_{HX,\text{ref}}$ = reference heat exchanger cost
CAC = installed capital cost for a process
CAC_{HX} = installed capital cost for all heat exchangers
$COLS$ = index set of all columns in a superstructure
Cost = annualized cost for a column
CS = index set of heat sinks
CU = index set of all cold utilities
D_k = flow of distillate product from column k
F_k = feed flow to column k
$F_{\text{ref},k}$ = reference feed flow to column k
F_{TOT} = total feed to process
FC_k = fixed charge cost for column k
$FM_{i,\bullet}$ = index set of all feasible matches between all heat sources and heat sink i
$FM_{\bullet,j}$ = index set of all feasible matches between hot source j and all heat sinks
FS_m = index set of all columns having (intermediate) product m as feed
HS = index set of all hot heat sources
HU = index set of all hot utilities
I = integer variable
IP = index set of all intermediate products
K_1, K_2 = constants
K_k = constant relating heat flow to feed flow for column k
NC = number of components in mixture
NCU = number of cold utilities
NI = number of temperature intervals
NHU = number of hot utilities
NS = number of different column sequences possible
OPC = annual operating cost for a process
PS_m = index set of all columns producing intermediate product m as either distillate or bottoms
Q = rate of heat flow
$Q_C(i)$ = rate of heat flow into cold sink i from a column condenser
$Q_H(j)$ = rate of heat flow from hot source j into a column reboiler
q_{ij} = rate of heat transfer from heat source j to heat sink i
Q_k = heat flow for column (assuming condenser and reboiler heat duties are about equal)
$Q_{m-1,m}$ = heat flow from temperature interval m-1 to m
$Q_{R/C}$ = total flow of heat between reboilers and condensers for a process
$Q_{\text{ref},k}$ = reference heat duty for column k
Q_{TOT} = total heat transferred in a distillation system
$T_{\text{cold,min}}$ = temperature of coldest available "cold" utility
$T_{\text{cond},i}$ = condenser temperature for column i
$T_{\text{cond,new}}$ = minimum new condenser temperature which could supply heat to an already included column when constructing a superstructure
$T_{\text{hot,max}}$ = temperature of hottest available "hot" utility
$T_{\text{reb},i}$ = reboiler temperature for column i
$T_{\text{reb,min}}$ = minimum reboiler temperature within the superstructure being constructed
U = heat transfer coefficient
U_k = upper bound on the feed flow to column k
V_k = coefficient for estimating variable cost for column k
VC = annualized cost for a process
y_k = binary variable (value 0 or 1 only)

Greek Letters

β = constant to convert annual operating cost to an annualized cost (to account for income tax) for a process

$\Delta T_{char,i}$ = characteristic temperature for a column; ΔT_{RC} plus ΔT for heat exchange for a column

$\Delta T_{char,min}$ = minimum characteristic temperature for all columns in structure

$\Delta T_{hx,i}$ = ΔT for heat exchange for column i

ΔT_{LM} = log mean temperature difference for heat exchange

ΔT_{min} = minimum allowable approach temperature for heat exchange

ΔT_{RC} = reboiler temperature minus condenser temperature for a column

σ = fraction of heat matches which are feasible for a superstructure

ξ_B = fraction of feed to column which exits in the bottoms product

ξ_D = fraction of feed to column which exits in the distillate product

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