Thermodynamically Equivalent Configurations for Thermally Coupled Distillation

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In this work a systematic way of generating all the thermodynamically equivalent structures for a given sequence of separation task in N-1 distillation columns (N is the number of components to be separated) is presented first. The problem is represented through a set of symbolic relations that can be written as propositional logic expressions. Alternatives from fully thermally coupled systems (only one reboiler and one condenser) to conventional columns (each column with a reboiler and a condenser), plus all the intermediate possibilities, are considered. Then all the thermodynamically equivalent structures that do not have control problems associated with the flow transfer among columns are generated. Again the problem is formulated as a set of symbolic relations written as propositional logic expressions. A formula with a detailed derivation to calculate the number of alternatives for a given sequence of tasks is also presented. Logic expressions in propositional logic form or translated into algebraic expressions in terms of binary variables can be integrated in the framework of Disjunctive Programming to extract the best solution for a given objective function. Several results are presented with systems involving mixtures of 4 and 5 components.

Introduction

It has been proved (Triantafyllou and Smith, 1992; Rudd, 1992; Agrawal and Fidkowski 1998a) that for a mixture of three components the Petlyuk configuration reduces the total vapor flow by 10 to 50% as compared to conventional systems using direct and indirect split arrangements. It seems that this reduction in total vapor flow rate is common to all the fully thermally coupled systems with more than three components. The lower vapor flow rate is of interest in the design of distillation systems because of the lower energy consumption, the smaller diameters required for the distillation columns, and the smaller heat exchangers needed for reboiling and condensing. Furthermore, the system requires only two heat exchangers while other systems need three or more. However, the increase in the number of sections in thermally coupled distillation systems can also increase the capital costs. Between these two extreme possibilities (conventional columns and thermally coupled distillation systems) there are all the intermediate partially thermal linked columns using an intermediate number of heat exchangers and sections. It is likely that the optimum design may belong to this class of systems.

The fully thermally coupled distillation systems are rarely used in industry, and it is only recently that there has been renewed interest in them. Difficulties in the control are probably the main reason. Let us consider, for example, the Peltlyuk configuration of Figure 1b. In this configuration the vapor has to be transferred back, from the main or product column to the prefractionator, and forth, from the prefractionator to the main column. In this condition the vapor flow rate has a difficult control, because neither of the columns

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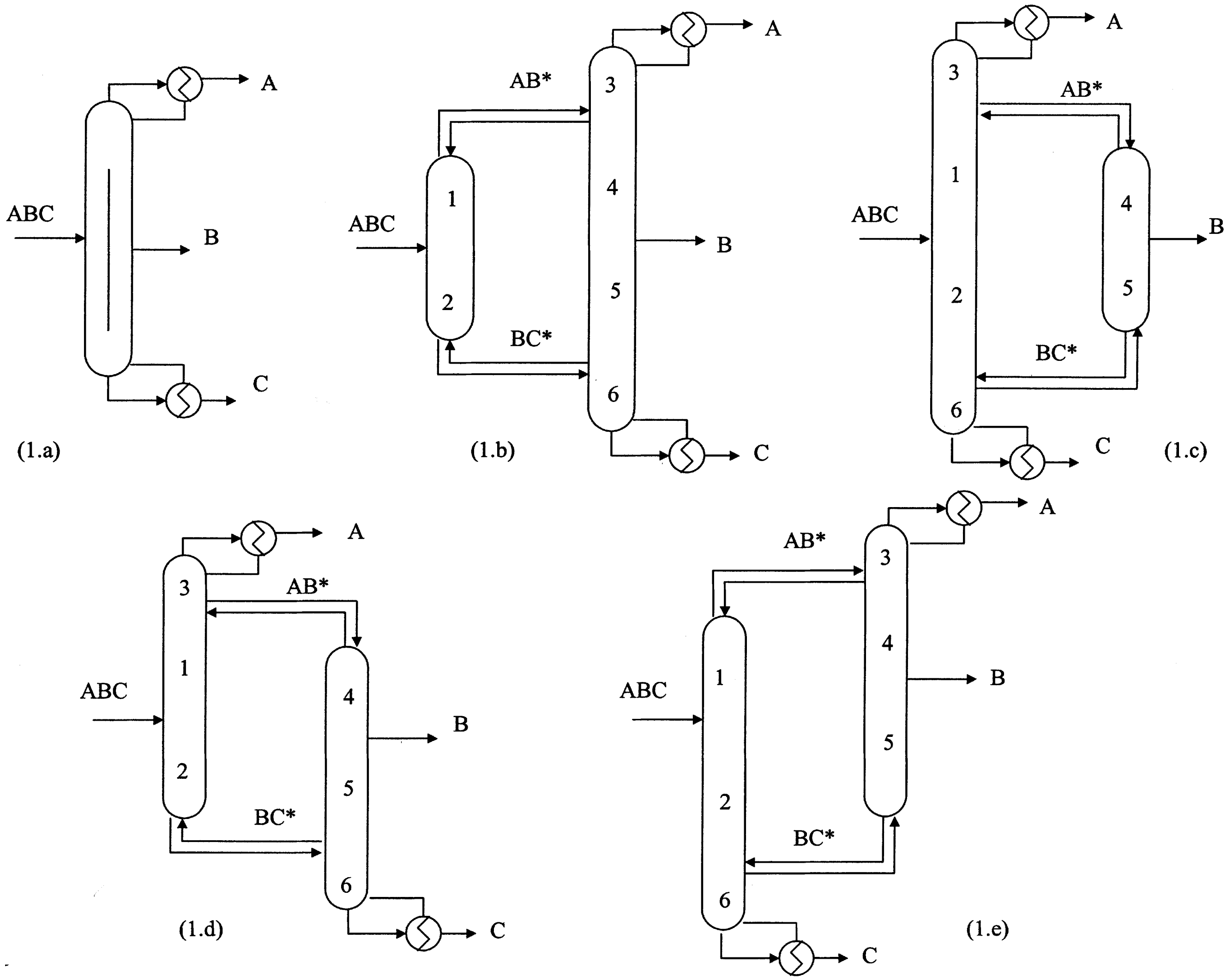


Figure 1. Divided wall column and the thermodynamically equivalent configurations for a mixture of three components.

has a higher or lower pressure than the other column. Therefore, the bottom part of the prefractionator has to be at lower pressure than the lower section of the main column, and the pressure in the upper section of the main column has to be lower than the pressure in the upper section of the prefractionator.

Kaibel (1988), Smith and Linnhoff (1988), and later Agrawal and Fidkowski (1998b) showed that the Peltluyk configuration can be rearranged to different column structures, all of them thermodynamically equivalent (temperatures, flows, compositions, and so on, are the same in all configurations). Consider again Figure 1. While the configuration presented in Figure 1c has the same controllability problems as Figure 1b, configurations Figures 1d and 1e do not have those problems. In these cases, the vapor flows from column to prefractionator or from prefractionator to column, but not both simultaneously. Therefore, one column can be maintained at higher pressure than the other, and the vapor flow can be controlled by a single valve in the transfer vapor line, while liquid can be transferred either through static head or by using liquid pumps (Agrawal, 1999).

In this work we first present a systematic way of generating all the possible rearrangements for a given sequence of separation tasks, and show how it can be integrated with the model in order to select the best arrangement of columns. Then, we will show how the previous model can be modified to select only among the arrangements that are easier to operate.

The rest of the article is divided into three main sections. In the first one a brief outline of how to generate superstructures is presented. Although this is not the objective of this article, this section is necessary for the full understanding of the other two parts. The second part deals with the generation of all the alternatives for the rearrangement of a given sequence of tasks in N-1 thermally coupled columns, and how to extract the optimal solution from among all the alternatives. The third part is a modification of the second one in order to generate only arrangements that are "easier to control."

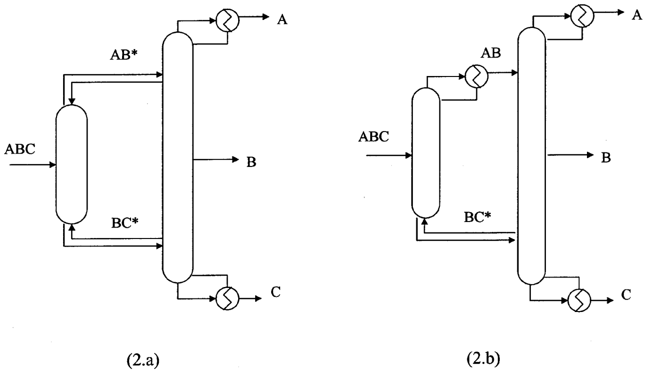


Figure 2. Separation of three components: (a) Peltlyuk configuration; (b) a heat exchanger is introduced associated to state *AB* (both configurations are not thermodynamically equivalent).

Some basic concepts and definitions

We introduce the definitions of column section, task, and state, as well as some common concepts that will be used throughout the article. A column (separation) section is defined to be a portion of a distillation column that is not interrupted by entering or exiting streams or heat flows (Hohmann 1982). States are defined as all physical and chemical properties of a stream. They can be quantitative like temperature, pressure, or composition, or qualitative like phase (liquid or vapor) (Papalexandri and Pistikopoulos, 1996; Yeomans and Grosmann, 1999). For example in a mixture of three components *ABC* all the possible states are *ABC, AB, BC, A, B, C* (defined, in this case, only as a function of their composition, although more characteristics can be introduced if necessary). A task is defined as the physical or chemical transformations between adjacent states (Yeomans and Grossmann, 1999). Some possible tasks for the previous example are *A/BC* (separate *A* from *BC*) or *AB/BC* (separate *AB* from *BC*).

For the sake of simplicity, in some parts of the article the states will be defined only as a function of composition. However, in general it is necessary to distinguish between states that involve a heat exchanger or states without one (that is, states formed by a single stream, or states formed by two streams: vapor and liquid). Consider, for example, Figure 2. If the states are defined only as a function of their composition, both separation sequences would be *AB/BC* – *A/B* – *B/C*. It is clear, however, that the heat exchanger associated with state *AB* makes these two sequences different. Therefore, the states that do not involve a heat exchanger will be denoted with the symbol (*). Both separations are then, *AB**/*BC** –*A/B* –B/*C*; *AB/BC**– *A/B*– *B/C*.

In order to illustrate the different aspects addressed in this article, consider the separation of a mixture of four components, *ABCD*. It is well known that for separating this type of mixture into their pure components we can use three conventional columns (in general $N-1$ conventional columns, N being the number of streams—in this case components— that leave the system). Therefore, with only conventional columns we require six heat exchangers—2(N-1)—and six column sections (rectifying and stripping section in each column). It is also well known that we can perform the separation using five different sequences (Thomson and King, 1972) (Figures 3.1, 3.5, 3.7, 3.9, and 3.11).

At this point it is useful to distinguish between heat exchangers associated with intermediate states and heat exchangers associated with a state (stream) that leaves the system (in our example, pure components). For example, in Figure 2 heat exchangers associated with state *AB* belong to class I, and heat exchangers associated with states *A* and *C* belong to class II.

Beginning with a sequence of conventional columns it is possible to remove exchangers of class I without changing the number of column sections (Figure 3.1 to 3.12). The heat exchanger is replaced by a vapor stream and a liquid stream. In other words, removing an intermediate heat exchanger produces a thermal coupling between columns. However, if we want to remove any heat exchanger of class II, we have to increase by two the number of column sections by the heat exchanger that is removed (Agrawal, 1996). See Figure 3.13 to 3.30. It is possible to remove all the heat exchangers except, of course, those associated with components *A* and *D* (Figure 3.25 to 3.30). The number of column sections will increase up to 10 ($4N-6$). However it is possible to continue increasing the number of column sections until 12 -$N(N-1)$ Agrawal (1996). See Figures 3.31 to 3.32. A systematic way of generating all of these sequences was presented by Caballero and Grossmann (2001) and will be outlined in the section Generation of Thermally Linked Sequences of this article. In Figure 3, except for the first sequence, only the cases of maximum thermal coupling and no thermal coupling are presented. However, any intermediate state could involve a thermal link or a heat exchanger. Taking this into account for the case of a 4 = component mixture there are "148" nonthermodynamically equivalent separation sequences using all of them 3 –(N-1) columns (20 sequences with 6 sections, 48 with 8 sections, 48 with 10 sections, and 32 with 12 sections).

When we use only conventional columns, there is a correspondence between separation tasks and columns (a given column performs a single separation task). However, this is not true for fully or partially thermally coupled systems. For these systems it is important to differentiate between the sequence of tasks and the specific sequence of columns that performs the separation. Consider again Figure 1b to 1e. This figure shows different arrangements of two columns, but the sequence of tasks is the same. For example, it is possible to go from the configuration in Figure 1b to 1e by changing section 6 from column 2 to column 1, but flows, temperatures, and pressures of the sections remain unchanged. These configurations are "thermodynamically equivalent." This concept can be generalized to N component mixtures. Note that in these cases a single column can perform more than one separation task. All the rearrangements of these sequences of tasks in N-1 columns will be addressed in the section Arrangements of Fully Thermally Coupled Distillation Columns for a Given Separation Sequence. Some particular cases, like those presented in Figure 4—divided wall columns, special separations with less than N-1 columns, and systems with more than N-1 columns — will not be considered.

For some thermally coupled systems, the same operational and control problems seem to arise as in the case of the Peltlyuk configuration. However, there always exists a thermodynamically equivalent structure that is "easier to control." A systematic way of generating all these easier-to-control configurations is presented in the section Arrangements of Easier to Control Thermally Coupled Distillation Columns for a Given Separation. Although a proof is not presented, results of that section suggest the following postulate:

Postulate 1. Given a sequence of tasks for separating a mixture of N components that do not form azeotropes and that produces a partially or fully thermally coupled configuration, there is always a thermodynamically equivalent sequence—usually more than one—in which it is possible to transfer the vapor flow streams from columns at higher to lower pressures.

Agrawal (1996) gave a set of heuristics for drawing by hand fully thermally coupled systems for an N = component mixture using from $4N$-6 to $N(N$-1) column sections. Caballero and Grossmann (2001) using the STN formalism of Yeomans and Grossmann (1999) proposed a systematic way of generating and modeling a superstructure for general distillation systems from conventional columns to fully thermally coupled systems, including all the intermediate structures. Two aspects are important in that work. First, there is no assump-

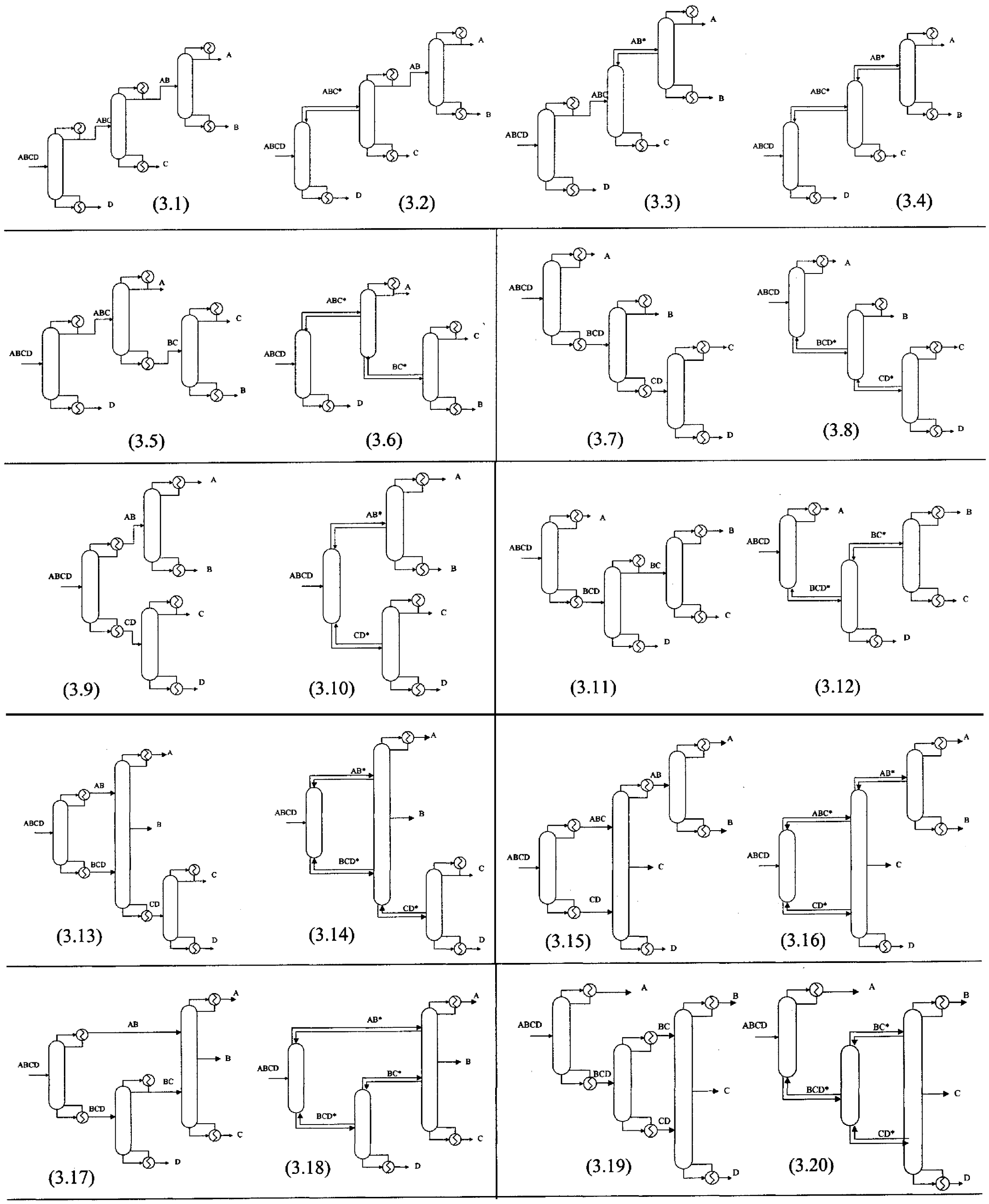


Figure 3. Sequences of three columns for separating a four component mixture (see text).

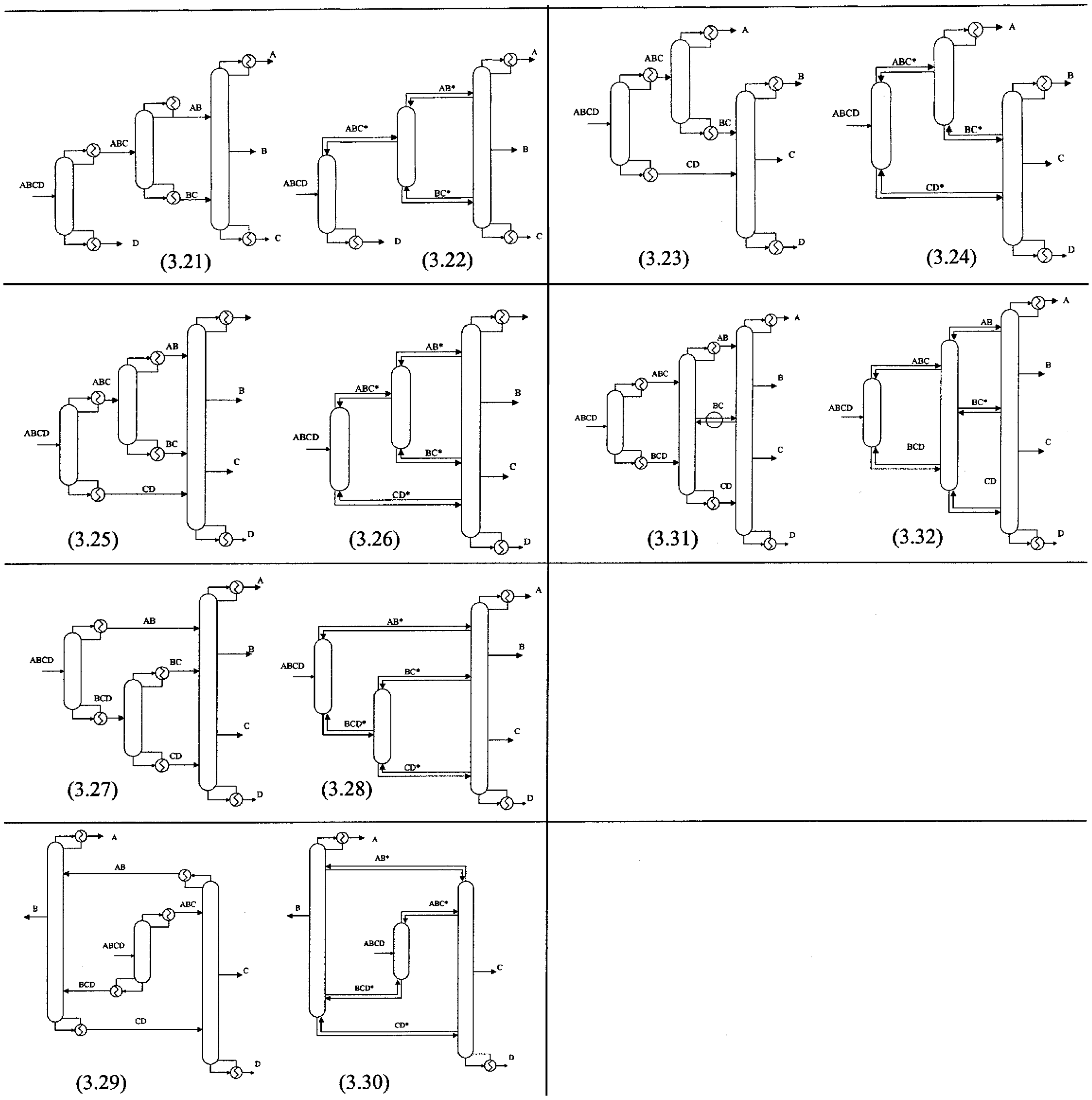


Figure 3. (Continued) Sequences of three columns for separating a four component mixture (see text).

tion about the particular arrangement of columns (recall that for partially or fully thermally coupled systems there is a large number of possible rearrangements for a given sequence of tasks), and second, it allows the systematic model of the system independent of the level of detail (shortcut, aggregated, or rigorous).

However, as pointed out before, two important questions must be addressed in order to synthesize sequences of fully or partially thermally coupled distillation systems. The first one can be stated as follows: Given a sequence of separation tasks, find the optimal arrangement of sections in N-1 thermally coupled columns. This includes a procedure for generating all the possible arrangements and extracting the optimal one. The second is to find the optimal arrangement that is *easier to control*, in which it is possible to uniformly fix the pressure of each column in a way that the vapor always flows from higher to lower pressures.

These two questions were also addressed by Agrawal (1999, 2000). He proposed a series of rules (or steps) for generating all of the possible arrangements by hand. However, this author did not present a systematic way for extracting the best arrangement. More operable configurations were generated by moving sections among columns, for which this author proposed a series of rules for generating these structures from a given base structure. The major difficulty with this approach is that there is not a single base structure for a sequence of tasks, and it is unclear how to extract the best rearrangement from among all the possible alternatives.

Generation of Thermally Linked Sequences

Here we present a brief outline of how to generate superstructures for fully thermally coupled systems and how to model such a superstructure in order to extract the optimal configuration. Further details can be found in Caballero and Grossmann (2001).

The problem to be solved in this part can be stated as follows: a mixture of N components, which do not form azeotropes, is given. The objective is to find the best sequence of separation tasks among all the possible sequences in order to obtain N streams of pure components. As was indicated earlier, this part has two steps: generating the superstructure and modeling it in order to extract the best configuration.

Generating a superstructure is straightforward using the State Task Network formalism of Yeomans and Grossmann (1999). An example of a 4-component mixture is shown in Figure 5a. An aggregated superstructure for the four components is shown in Figure 5b (Caballero and Grossmann, 1999).

Caballero and Grossmann (2001) have shown that a set of five rules are enough for extracting all the feasible sequences from these superstructures. These rules can be expressed in mathematical form using propositional logic in terms of Boolean variables, and then, if desired, translated to an equivalent set of algebraic inequalities in terms of binary variables.

We define the following index sets, which we will use later in the mathematical formulation of the logical set of rules for extracting the feasible sequences of the superstructure. (We will illustrate it for the case of a mixture of four components)

$$\text{TASKS} = \{t|t \text{ is a given task}\}$$

$$\text{e.g.}\quad \text{TASK} = \{A/BCD; AB/BCD; AB/CD; ABC/CD; ABC/D; A/BC; AB/BC; AB/C; B/CD; BC/CD; BC/D; A/B; B/C; C/D\}$$

$$\text{STATES} = \{s|s \text{ is a state}\}$$

$$\text{e.g. STATES} = \{ABCD; ABC; BCD; AB; BC; CD; A, B, C, D\}$$

$$\text{ISTATE} = \{m|m \text{ is an intermediate state}\}$$

$$\text{e.g.}\quad \text{ISTATE} = \{ABC, BCD, AB, BC, CD\}$$

$$\text{COMP} = \{i| \text{ is a pure component}\}$$

$$\text{e.g.}\quad \text{COMP} = \{A, B, C; D\}$$

$$TS_s = \{\text{tasks t that the state s is able to produce}\}$$

$$\text{e.g.}\quad TS_{ABCD} = \{A/BCD; AB/BCD; AB/CD; ABC/CD; ABC/BCD\ ABC/D\}$$

$$TS_{ABC} = \{A/BC; AB/BC; AB/C\}$$

$$ST_s = \{\text{task t that are able to produce state s}\}$$

$$\text{e.g.}\quad ST_{ABC} = \{ABC/BCD; ABC/CD; ABC/D\}$$

$$ST_{BCD} = \{A/BCD; AB/BCD; ABC/BCD\}$$

$$ST_{AB} = \{AB/BCD; AB/CD; AB/C; AB/BC\}$$

$$ST_{BC} = \{A/BC; AB/BC; BC/CD; B/CD$$

$$ST_{CD} = \{AB/CD; ABC/CD; B/CD; BC/CD\}$$

According to our previous definition, a task will be performed by two column sections. For example, the task *AB/CD* will produce the state *AB* through a column section and the state *CD* through another column section. Therefore, a task can be regarded as a pseudocolumn. Similar to conventional columns, we call the upper section of a task the rectifying section, and the lower section of a task the stripping section.

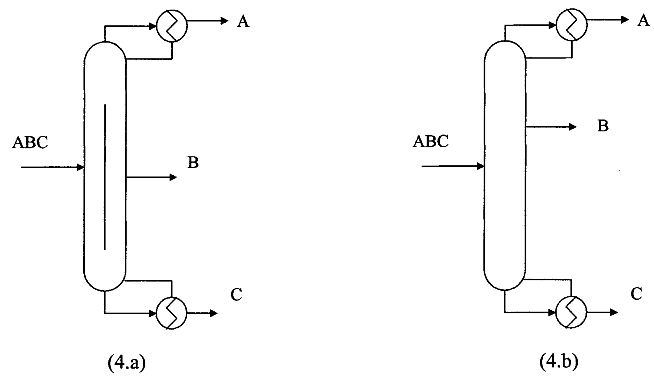


Figure 4. (a) Divided wall column; (b) column with a lateral extraction.

Taking this into account, we also define the following index sets

$$RECT_s = \{\text{task t that produces state s by a rectifying section}\}$$

$$RECT_{ABC} = \{ABC/CD, ABC/D; ABC/BCD\}$$

$$RECT_{AB} = \{AB/BCD; AB/BC; AB/C\}$$

$$RECT_{BC} = \{BC/CD\}$$

$$STRIP_s = \{\text{task t that produces state s by a stripping section}\}$$

$$STRIP_{BCD} = \{A/BCD; AB/BCD; ABC/BCD\}$$

$$STRIP_{BC} = \{AB/BC; A/BC\}$$

$$STRIP_{CD} = \{AB/CD; ABC/CD; B/CD; BC/CD\}$$

$$PREC_i = \{\text{task t that produce pure product i through a rectifying section}\}$$

$$PREC_A = \{A/BCD; A/BC; A/B\}$$

$$PREC_B = \{B/CD; B/C\}$$

$$PREC_C = \{C/D\}$$

$$PSTR_i = \{\text{task t that produce pure product i through a stripping section}\}$$

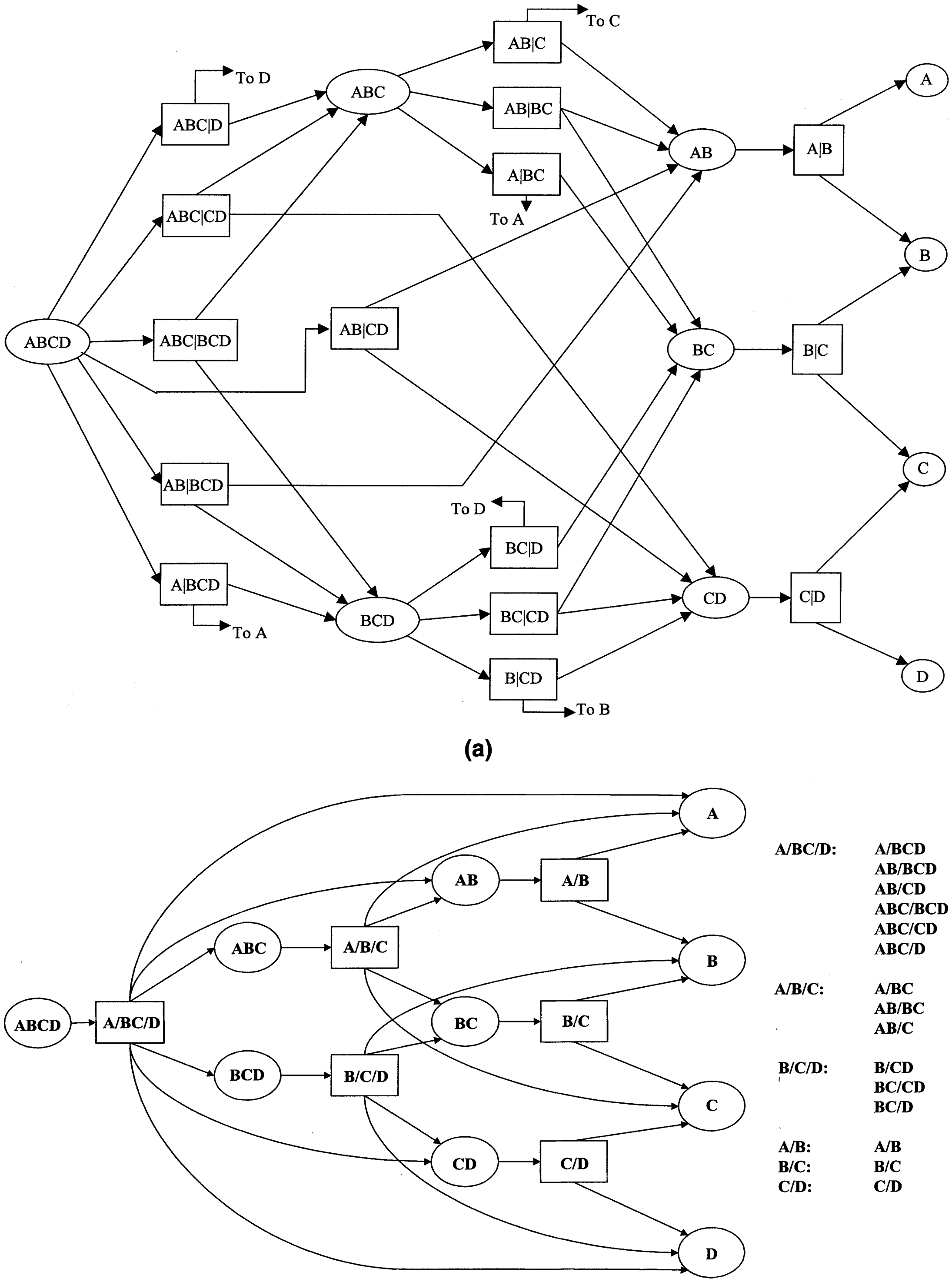
$$PSTR_B = \{A/B\}$$

$$PSTR_C = \{AB/C; B/C\}$$

$$PSTR_D = \{ABC/D; BC/D; C/D\}$$

Next we define the Boolean variable Y_t such that the variables are True if the Task t is selected in the configuration, and False otherwise. The Boolean variables, Wi, are defined to take the value True if there is a heat exchanger associated with the pure product i. The five rules for extracting the feasible configurations and the equivalent logical proposition follow:

1. A given state s can give rise to at most one task. For example, the state *ABC* could produce the tasks *A/BC*,



(b)

Figure 5. (a) STN superstructure for a 5-component mixture; (b) aggregated superstructure.

AB/BC, or *AB/C*, but only one of those will be selected in a given sequence

$$\underset{t \in ST_s}{\veebar} Y_t \veebar Z \quad s \in \text{STATES} \tag{1}$$

where Z is an auxiliary variable that means "do not choose any of the options of the set ST_s."

2. A given state (except products) can be produced at most by one or two contributions: one must come from a rectifying section of a task, and the other from a stripping section of a

task

$$\underline{\vee}_{t \in PREC_s} \quad Y_t \underline{\vee} Zs \in \text{STATES}$$

$$\underline{\vee}_{t \in PSTR_s} \quad Y_t \underline{\vee} Zs \in \text{STATES} \qquad (2)$$

Again Z is an auxiliary Boolean variable that means that none of the other options in the disjunction is selected.

3. The lightest and heaviest products are produced by only one contribution. However, an intermediate product, i, can be produced by one or two contributions

$$\bigvee_{t \in (PREC_i \cup PSTR_i)} Y_t \quad i \in \text{COMP}$$

$$\underline{\vee}_{t \in PREC_i} \quad Y_t \underline{\vee} Z \quad i \in \text{COMP}$$

$$\underline{\vee}_{t \in PSTR_i} \quad Y_t \underline{\vee} Z \quad i \in \text{COMP} \qquad (3)$$

The first of the previous three equations ensures that product i is produced by at least one task, while the other two equations ensure that product i is produced by the contributions of at most one rectifying section and at most one stripping section.

3.1. If the intermediate product is produced by two contributions, one must come from a stripping section of a task and the other for a rectifying section, and no heat exchanger is associated to this product

$$\left(\bigvee_{t \in PREC_i} Y_t\right) \wedge \left(\bigvee_{t \in PSTR_i} Y_t\right) \Rightarrow W_i \quad i \in \text{COMP} \qquad (4)$$

3.2. If the intermediate product is produced by only one contribution, the heat exchanger associated with this product must be selected

$$\neg\left(\bigvee_{t \in PREC_i} Y_t\right) \Rightarrow W_i \quad i \in \text{COMP}$$

$$\neg\left(\bigvee_{t \in PSTR_i} Y_t\right) \Rightarrow W_i \quad i \in \text{COMP} \qquad (5)$$

4. Connectivity is simply the mathematical (logical) expression of the relations between tasks in the superstructure

$$Y_t \Rightarrow \bigvee_{k \in TS_s} Y_k \quad \forall t \in ST_s, \quad s \in \text{STATES}$$

$$Y_t \Rightarrow \bigvee_{k \vee ST_S} Y_k \quad \forall t \in TS_S, \quad s \in \text{STATES} \qquad (6)$$

5. The minimum number of tasks is $N-1+N-E=2N$-1-E (E is the number of heat exchangers of class II heat exchangers associated to states leaving the system) $2 \leq E \leq N$. It is interesting design systems with the minimum number of tasks (or column sections); however, for fully thermally coupled systems, it is possible to obtain schemes with up to $N(N$-1) sections (an intermediate state produced by more than one task). The classic superstructure proposed by Sargent and Gaminibandara (1976) uses $N(N$-1) column sections, therefore, we also considered this possibility

$$\sum_{t \in \text{TASK}} Y_t = N - 1 - \sum_{i \in \text{COMP}} W_i \qquad (7)$$

For fully thermally coupled systems, the previous equation is only a lower bound.

The next step consists of writing the equations of the model. The advantage of using the STN formalism together with generalized disjunctive programming (GDP) (Turkay and Grossmann, 1996) is that the formal structure of the model is always the same (see, for example, Yeoamans and Grossmann, 1999; Caballero and Grossmann, 1999). Conceptually the model can be written as follows

(P1)

$$\min f(x)$$
s.t.

$$\begin{bmatrix} Y_t \\ h_t(x)=0 \\ g_t(x) \leq 0 \end{bmatrix} \underline{\vee} \begin{bmatrix} \neg Y_t \\ B^t x = 0 \end{bmatrix} \quad \forall t \in \text{TASK}$$

$$\begin{bmatrix} W_i \\ p_i(x)=0 \end{bmatrix} \underline{\vee} \begin{bmatrix} \neg W_i \\ B^i x = 0 \end{bmatrix} \quad \forall i \in \text{COMP}$$

$$\begin{bmatrix} K_k \\ \Psi_k^1(x)=0 \end{bmatrix} \underline{\vee} \begin{bmatrix} \neg K_k \\ \Psi_k^2(x)=0 \end{bmatrix} \quad \forall k \in \text{ISTATE}$$

$$\Omega(Y_t, W_i) = True\, Y_t = \{\text{True}, \text{False}\}$$

In the preceding model x is a vector of continuous variables, and $f(x)$ is the objective function. The equations and constraints that are activated when task t is performed are given by $h_t(x)=0$ and $g_t(x) \leq 0$. The equations $p_i(x)=0$ include energy balances, mass balances, and costs that must be used if a heat exchanger associated to a pure product appears in the configuration ($W_i = \text{True}$). The sets of equations $\Psi_k^1(x)=0$ and $\Psi_k^2(x)=0$ are only connectivity equations that are different if there is ($K_k = \text{True}$) or is not ($K_k = \text{False}$) an intermediate heat exchanger. Figure 6 shows the model. This disjunctive representation has the advantage that it can be used with shortcut, aggregated, approximated, or rigorous models, depending on the level of detail that is desired. Further details about the model and solution can be found in Caballero and Grossmann (2001).

Arrangements of Fully Thermally Coupled Distillation Columns for a Given Separation Sequence

The problem addressed in this section can be stated as follows. Given a sequence of tasks for separating a mixture of N components in N pure streams. The objective is to obtain all of the possible thermodynamically equivalent arrangements of column sections in N-1 thermally coupled distillation

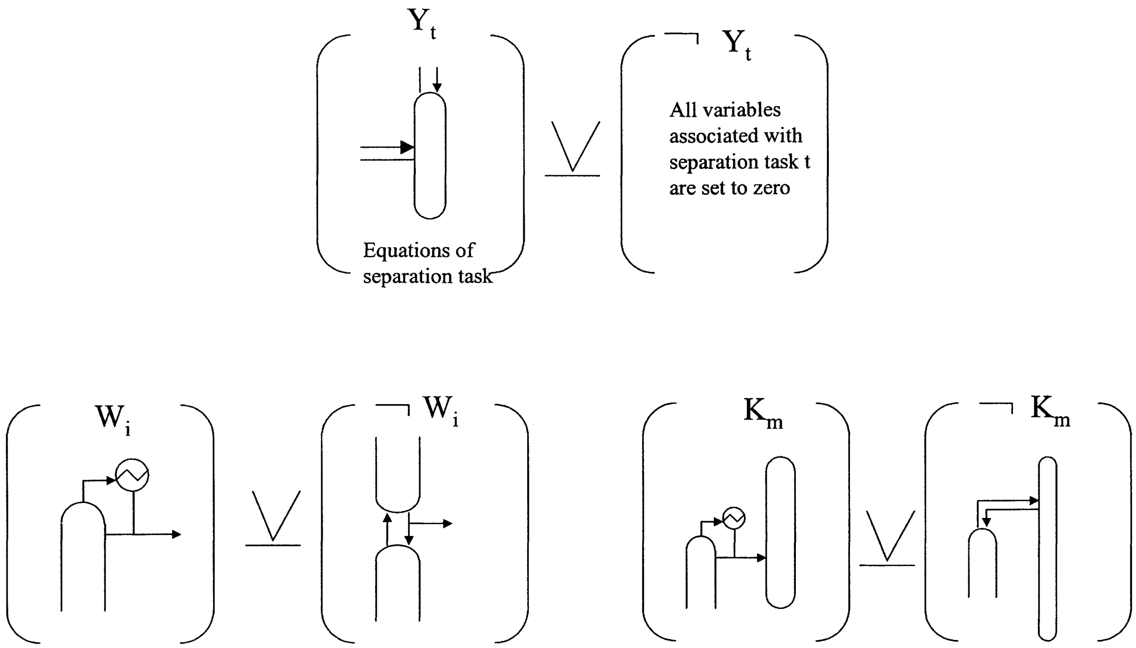


Figure 6. Conceptual representation of the disjunctive model for designing thermally coupled distillation sequences.

columns. No wall-dividing columns are considered. Figure 1 illustrates the objective of systematically finding all the arrangements for a mixture of three components .

The starting point is a sequence of feasible tasks (and states) to perform a given separation (a solution of the problem outlined in an earlier section). As an illustrative example consider the feasible sequence of tasks and states presented in Figure 7. This example corresponds to the separation of a mixture of five components using 20 column sections *N*(*N*-1). (Note that one possible rearrangement is that proposed by Sargent and Gaminibandara, 1976).

The problem of finding all of the possible rearrangements in *N*-1 columns can be reduced to solving a set of logical relationships among tasks and states. These relationships can be stated as logical propositions in terms of Boolean variables, and these in turn can be translated into a set of linear algebraic relationships in terms of binary variables to formulate the problem as a mixed integer linear program (MILP) that corresponds to an extended assignment problem (assigning a section to a given column with some extra constraints in order to assure the feasibility).

We introduce the following index sets (sequence of states and tasks of Figure 7 will be used to illustrate the procedure)

$$\text{SECTION} = \{j | j \text{ is a column section}\}$$

$$\text{COLUMN} = \{c | c \text{ is a column}\}$$

$$\text{STATE} = \{e | e \text{ is a state}\}$$

$$\text{PURE} = \{n | n \text{ is a pure product}\}$$

$$\text{REC}_e = \{\text{sections that give rise to state e coming from a rectifying section}\}$$

$$\text{STR}_e = \{\text{sections that give rise to state e coming from a stripping section}\}$$

$$\text{T\_REC}_e = \{\text{rectifying section produced by state e}\}$$

$$\text{T\_STR}_e = \{\text{stripping section produced by state e}\}$$

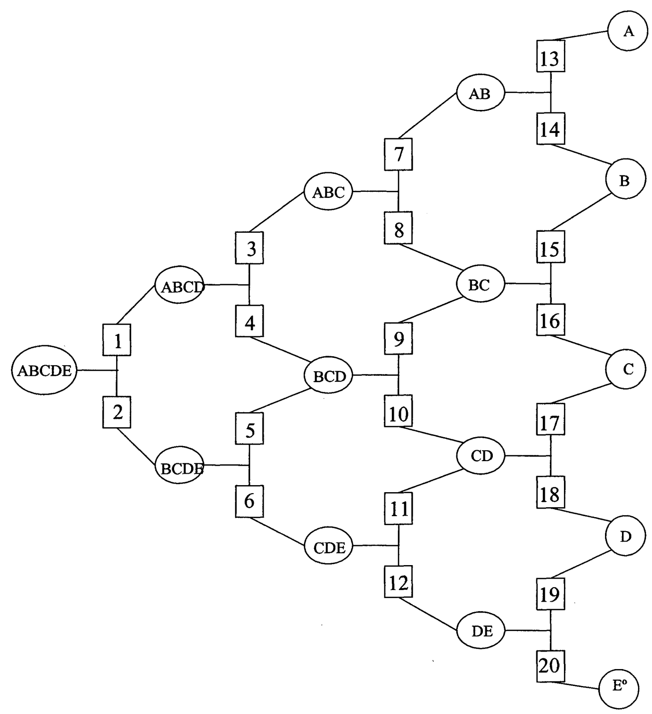


Figure 7. Sequence of tasks for a mixture of 5 components with 20 column sections.

Note that the set PURE can be considered a subset of STATE, if desired.

For the sequence presented in Figure 7, we have the following:

$$\text{SECTION} = \{\text{j1,j2,j3,}\ldots\text{,j20}\}$$

$$\text{COLUMN} = \{\text{c1,c2,c3,c4}\}$$

$$\text{STATE} = \{\text{ABCDE, ABCD, BCDE, ABC, BCD, CDE, AB, BC, CD, DE}\}$$

$$\text{PURE} = \{\text{A,B,C,D,E}\}$$

$$\text{REC}_{\text{ABCD}} = \{\text{j1}\};\ \text{REC}_{\text{ABC}} = \{\text{j3}\};\ \text{REC}_{\text{AB}} = \{\text{j7}\};\ \text{REC}_{\text{BCD}} = \{\text{j5}\};\ \text{REC}_{\text{BC}} = \{\text{j9}\};\text{REC}_{\text{CD}}\{\text{j11}\}$$

$$\text{STR}_{\text{BCDE}} = \{\text{j2}\},\text{STR}_{\text{CDE}} = \{\text{j6}\};\text{STR}_{\text{DE}} = \{\text{j12}\};\text{STR}_{\text{BCD}} = \{\text{j4}\};\ \text{STR}_{\text{CD}} = \{\text{j10}\};\ \text{STR}_{\text{BC}} = \{\text{j8}\}$$

We now define the Boolean variable $P_{j,c}$ that takes the value of True if the section j is assigned to the column c, and False otherwise.

The following set of logical relationships defines completely the problem:

1. Each column has at least one section

$$\bigvee_{j \in \text{SECTION}} P_{j,c} \quad \forall c \in \text{COLUMN} \tag{8}$$

2. A given section can only be assigned to one column

$$\underline{\bigvee}_{c \in \text{COLUMN}} P_{j,c} \quad \forall j \in \text{SECTION} \tag{9}$$

These first two relations correspond to those of the assignment problem.

3. Connectivity relationships.

3.1 There are no heat exchangers associated with state e. These relations are derived directly from the structure of tasks and states. It can be stated as follows: Among all the sections that reach or exit from a given state, only two, a rectifying section and a stripping section, must be assigned to a given column, c

$$\begin{aligned}
&P_{r,c} \Rightarrow P_{l,c} \vee P_{s,c} \quad r \in T_REC_e; l \in T_STR_e;\ s \in REC_e;\ \forall c \in \text{COLUMN}\\
&P_{l,c} \Rightarrow P_{r,c} \vee P_{s,c} \quad r \in T_REC_e; l \in T_STR_e; s \in STR_e;\ \forall c \in \text{COLUMN}\\
&P_{r,c} \Rightarrow P_{l,c} \vee P_{s,c} \quad r \in REC_e; l \in T_REC_e; s \in STR_e;\ \forall c \in \text{COLUMN}\\
&P_{l,c} \Rightarrow P_{r,c} \vee P_{s,c} \quad r \in T_STR_e; l \in STR_e; s \in REC_e;\ \forall c \in \text{COLUMN}
\end{aligned} \tag{10}$$

$$\begin{aligned}
&\neg P_{r,c} \vee \neg P_{l,c} \vee \neg P_{s,c} \quad r \in REC_e; l \in T_REC_e;\ s \in T_STR_e; \forall c \in \text{COLUMN}\\
&\neg P_{r,c} \vee \neg P_{l,c} \vee \neg P_{s,c} \quad r \in STR_e; l \in T_REC_e;\ s \in T_STR_e; \forall c \in \text{COLUMN}
\end{aligned} \tag{11}$$

The first four relations (Eqs. 10) are feasibility relations among sections for a given state. For example, consider the BCD state in Figure 7. In this particular case the four previous equations are as follows

$$\left.\begin{aligned}
P_{j9,c} &\Rightarrow P_{j10,c} \vee P_{j5,c}\\
P_{j10,c} &\Rightarrow P_{j9,c} \vee P_{j4,c}\\
P_{j4,c} &\Rightarrow P_{j5,c} \vee P_{j10,c}\\
P_{j5,c} &\Rightarrow P_{j4,c} \vee P_{j9,c}
\end{aligned}\right\} \quad \forall c \in \text{COLUMN} \tag{12}$$

The other two equations (Eqs. 11) assure that only two of those sections are assigned to column c. For example, in the BCD state, those equations are as follows

$$\left.\begin{aligned}
\neg P_{j4,c} \vee \neg P_{j9,c} \vee \neg P_{j10,c}\\
\neg P_{j5,c} \vee \neg P_{j9,c} \vee \neg P_{j10,c}
\end{aligned}\right\} \quad \forall c \in \text{COLUMN}. \tag{13}$$

3.2. There is a heat exchanger associated with state e. In this case, the stripping and rectifying sections (T_REC_e. T_STR_e) of state e must be in the same column

$$P_{r,c} \Leftrightarrow P_{s,c} \quad r \in T\_REC_e, \quad s \in STR_e \quad \forall c \in \text{COLUMN} \tag{14}$$

4. The two sections produced by the feed state must be assigned to the same column

$$P_{j1,c} \Leftrightarrow P_{j2,c} \tag{15}$$

In some special cases this last relation can be relaxed (the feed is introduced by the top or the bottom of a column).

5. For the products of intermediate volatility that leave the system, the rectifying and stripping sections must be assigned to the same column. Of course, this relationship will hold only if there is no heat exchanger associated with that pure product

$$P_{j,c} \Leftrightarrow P_{l,c} \qquad j \in REC_e; l \in STR_e; e \in PURE; \forall c \in COLUMN \tag{16}$$

Following the example in Figure 7, these relations are as follows

$$\left.\begin{aligned}
P_{j14,c} &\Leftrightarrow P_{j15,c}\\
P_{j16,c} &\Leftrightarrow P_{j17,c}\\
P_{j18,c} &\Leftrightarrow P_{j19,c}
\end{aligned}\right\} \quad \forall c \in \text{COLUMN} \tag{17}$$

The previous set of logical relationships include all of the possible configurations in N-1 columns. However, the problem is degenerate in the sense that it is possible to get equivalent solutions, but with different column assignments. For example, a possible solution for the set of sections of Figure 7 is

Column 1:$j1,j2$

Column 2:$j3,j4,j5,j6$

Column 3:$j7,j8,j9,j10,j11,j12$

Column 4:$j13,j14,j15,j16,j17,j18,j19,j20$

It is easy to check that if the assignments of any two columns are changed the sequence is exactly the same. However, from a mathematical point of view they are different solutions of the problem. To avoid this problem there are two alternatives. The first consists of adding a new set of logical constraints in order to avoid the degenerate solutions. However, this approach produces a large number of constraints that increase exponentially with the number of components. The second approach consists of fixing some assignments *a priori*. In our example, we can fix the assignments section 1 to column 1; section 4 to column 2; section 9 to column 3. It is easy to check that, say sections 1 and 4, cannot be assigned to the same column (if we add to the previous problem a constraint that forces sections 1 and 4 to belong to the same column, the problem becomes infeasible).

Finally, there are some constraints that are not strictly needed but help to solve the problem. Consider again Figure 7. It is clear from this representation that sections j7 and j14 cannot be simultaneously assigned to the same column. The same happens to sections j3, j8, and j15, and so on. Therefore, defining a new index set

$$\text{ROW}_j\{\text{column sections in the same row}\}$$

it is possible to write the following constraints

$$\underline{\bigvee}_{j \in \text{ROW}} P_{j,c} \underline{\vee} Z \quad c \in \text{COLUMN} \tag{18}$$

where Z is again a dummy Boolean variable that only means that no assignment is made.

Another interesting point is knowing what the total number of thermodynamically equivalent configurations is for a given sequence of tasks. Agrawal (2000) presented a procedure for calculating the number of sequences, but it appears that not all configurations are considered for systems with more than four components. The following formula predicts the total number of rearrangements

$$NC = 2^{(NT - NHE_I - 1)} \tag{19}$$

where NC is the total number of configurations, NT is the number of separation tasks, and NHE_I is the number of heat exchangers of class I (associated to nonpure products intermediate states) in the structure. A detailed derivation of Eq. 19 is shown in the Appendix. Some special cases are of interest. First, when only two heat exchangers are used and the number of column sections is $N(N\text{-}1)$ (as in Figure 7), the total number of configurations can be related to the number of components to be separated (N)

$$NC = 2^{(N(N-1)/2 - 1)} \tag{20}$$

And second, if we want only sequences of tasks using the minimum number of sections, then the total number of possible rearrangements as a function of the total number of components is

$$NC = 2^{((4N-6)/2 - NHE + 1)} \tag{21}$$

where NHE is the total number of heat exchangers.

Table 1 shows the results of some representative cases. Note that for the example presented in Figure 7 the total number of possible rearrangements is 512.

Examples 1 and 2

Due to the obvious lack of space, the 512 alternatives of the example cannot be presented here. However as an intro-

Table 1. Number of Thermodynamically Equivalent Configurations

Number of Components	Total Number of Heat Exchangers	Thermodynamically Equivalent Sequences (minimum-number of column sections)	Thermodynamically Equivalent Sequences $N(N+1)$ Column Sections
3	4	1	
3	3	2	
3	2	4	4
4	6	1	
4	5	2	
4	4	4	
4	3	8	
4	2	16	32
5	8	1	
5	6	4	
5	4	16	
5	2	64	512
6	10	1	
6	8	4	
6	6	16	
6	4	64	
6	2	256	16,384
10	18	1	
10	12	64	
10	8	1,024	
10	4	16,384	
10	2	65,536	$1.7592 \cdot 10^{13}$

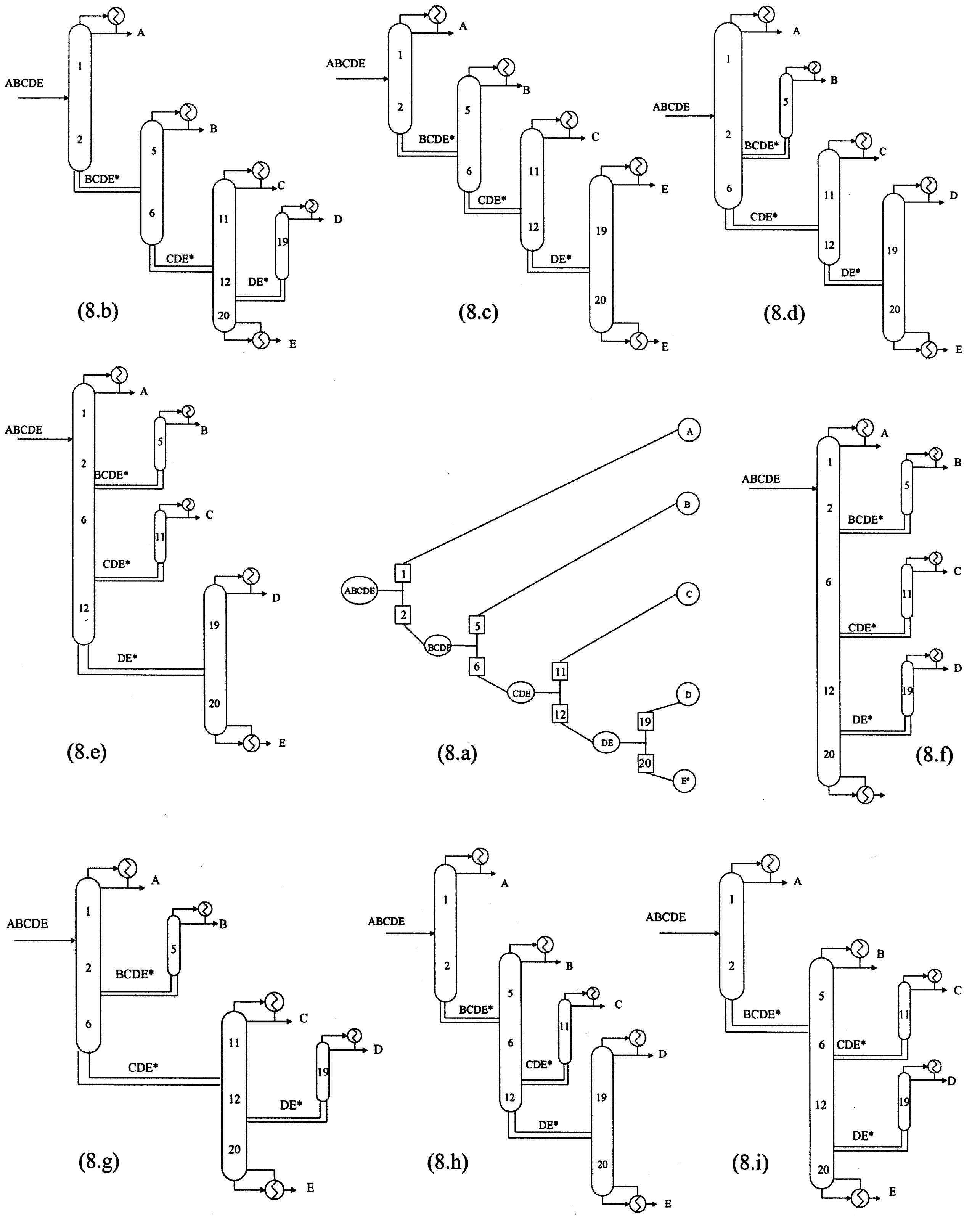


Figure 8. The 8 alternatives for the separation of a mixture of 5 components using 5 heat exchangers (according to 8.a).

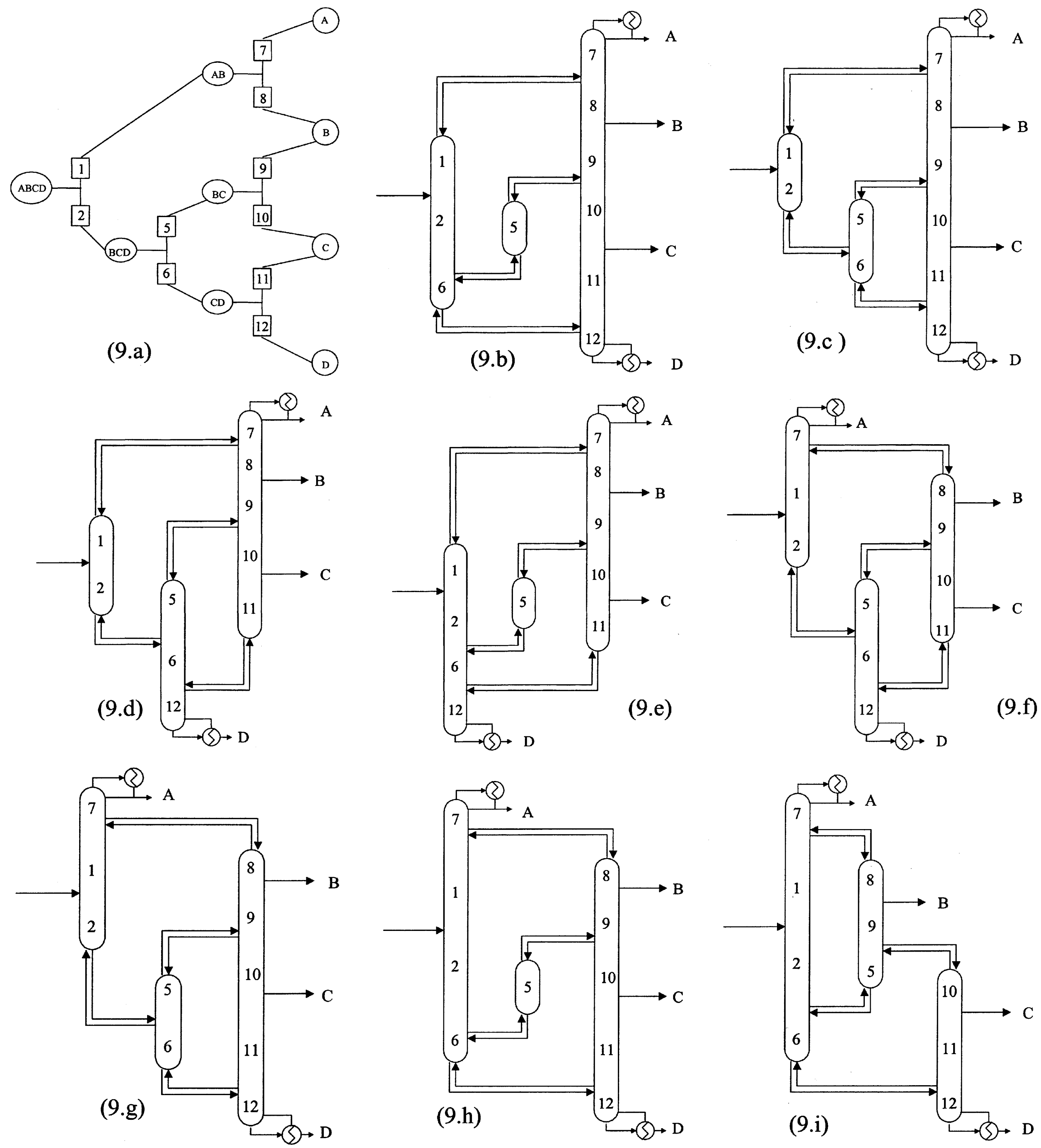


Figure 9. The 16 possible rearrangements of a mixture of 4 components for the sequence of tasks given in 9.a, using the minimum number of column sections.

duction, Figure 8 presents the eight alternatives for the separation of a mixture of five components using five heat exchangers, and Figure 9 shows the 16 possible rearrangements of a mixture of four components for a sequence of tasks using the minimum number of column sections.

Arrangements of Easier to Control Thermally Coupled Distillation Columns for a Given Separation Sequence

The problem addressed in this section is similar to the one presented in the previous section, but it includes an addi-

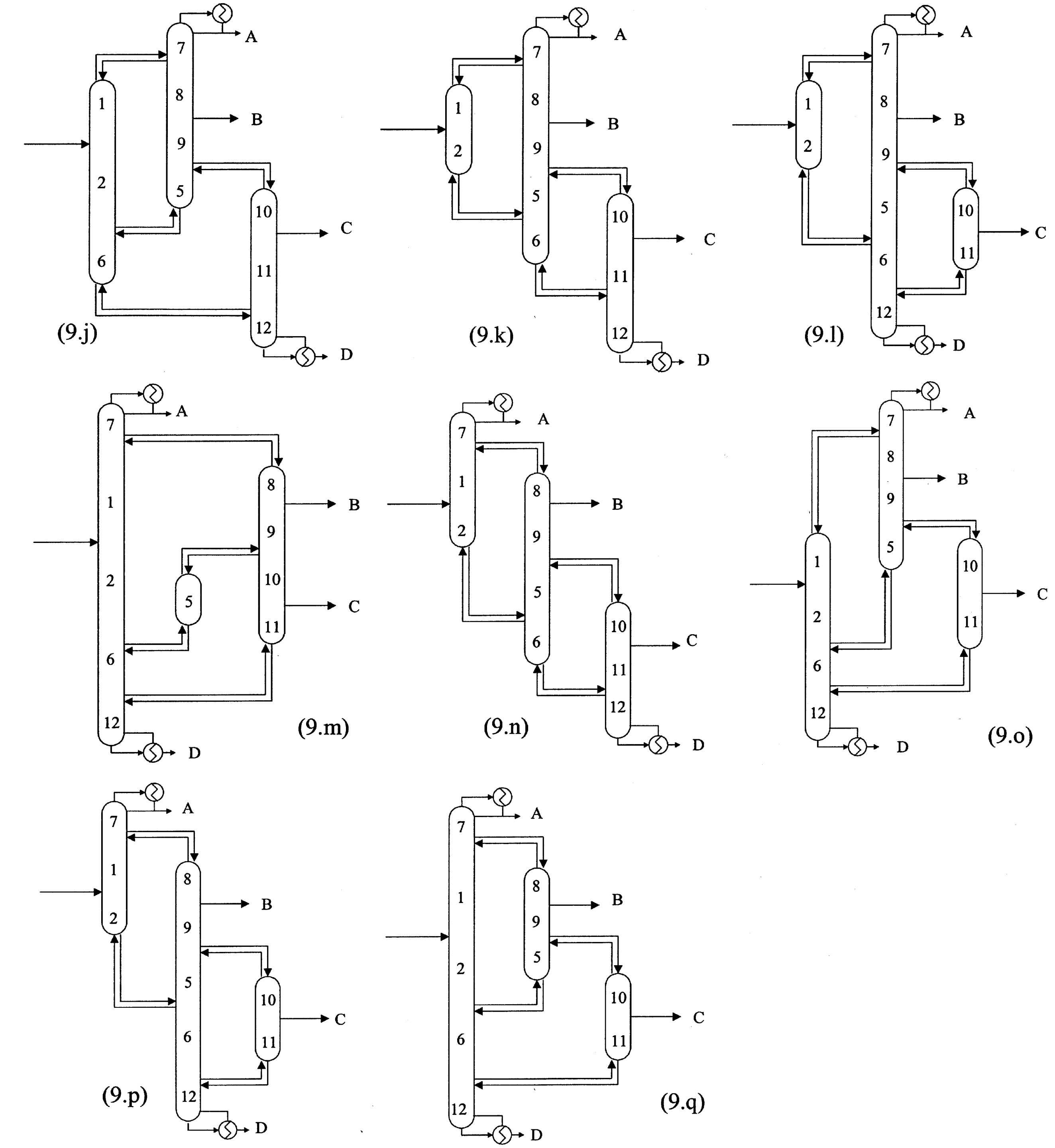


Figure 9. (Continued) The 16 possible rearrangements of a mixture of 4 components for the sequence of tasks given in 9.a, using the minimum number of column sections.

tional constraint: "The vapor should flow from higher to lower pressure columns."

Consider again Figure 1 from the Introduction. While the configurations presented in Figure 1b and 1c have control problems associated with the vapor transfer among columns, the configurations shown in Figures 1d and 1e do not have that problem. In these cases the vapor flows from column to prefractionator, or from prefractionator to column, but not both simultaneously. In this section we present a set of logical relationships that allows the generation of all these easier-to-control configurations.

Due to the fact that this case is an extension of the previous one, all the equations introduced for generating all thermodynamically equivalent configurations are also part of this model, except for those variables that are fixed to avoid degeneracy. However, an important modification is included. In this case: the set COLUMNS is an *ordered set* in which the first element (first column) corresponds to the column at lower pressure, and the rest of the elements are ordered in increasing pressures.

The new logical relationships are as follows:

1. There is a reboiler in the column at higher pressure and a condenser in the column at lower pressure

$$\left.\begin{array}{l}\underset{s\in \mathrm{PURE}_A}{\underline{\bigvee}} P_{s,c1}\\ \underset{s\in \mathrm{PURE}_X}{\underline{\bigvee}} P_{s,cq}\end{array}\right\} \begin{array}{l} c1,\, cq \in \text{COLUMN/first and last column in}\\ \text{the set, respectively, A most volatile}\\ \text{component: X less volatile component}\end{array} \tag{22}$$

2. Consider a state e that has no associated heat exchanger.

(2.1). If at the state e the flow only reaches from a rectifying section s, then if the rectifying section r and the stripping section l that exit from state e belong to the same column c, the section s must be assigned to a column cc at higher pressure

$$P_{r,c} \wedge P_{l,c} \Rightarrow P_{s,cc} \qquad \begin{array}{l} r \in T\_REC_e; l \in T\_STR_e; s \in REC_e \\ c, cc \in \text{COLUMN/ord(cc)} > ord(c) \end{array} \tag{23}$$

Figure (10a) illustrates this situation. Recall that the set COLUMN is an ordered set. Also, the term *ord* makes reference to the position (order) that a given element has in the set.

2.2. If at the state e the flow only reaches from a rectifying section s, then if the rectifying section r that exits from the state e and the rectifying section s belong to the same column c, the stripping section l that exits from state e must be assigned to a column at higher pressure

$$P_{r,c} \wedge P_{s,c} \Rightarrow P_{l,cc} \qquad r \in T\_REC_e; l \in T\_STR_e; s \in REC_e$$
$$c, cc \in \text{COLUMN/ ord}(cc) > \text{ord}(c) \tag{24}$$

Figure 10b illustrates this situation.

2.3. If at the state e the flow only reaches from a stripping section k, then if the rectifying section r and the stripping section l that exit from state e belong to the same column c, the section k must be assigned to a column cc at lower pressure

$$P_{r,c} \wedge P_{l,c} \Rightarrow P_{k,cc} \qquad k \in STR_e; r \in T\_REC_e; l \in T\_STR_e$$
$$c, cc \in \text{COLUMN/ ord}(cc) < \text{ord}(c) \tag{25}$$

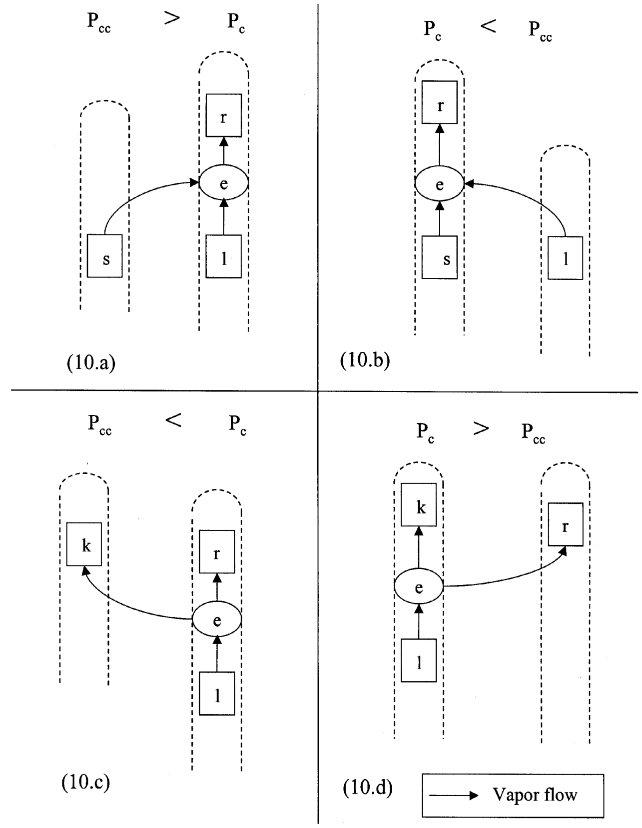


Figure 10. The logical relationships between columns presented in relations 2.1. to 2.4

Figure 10c illustrates this situation. 2.4. If at the state e the flow only reaches from a stripping section k, then if the stripping section l that exit from state e and the stripping section k belong to the same column c, the rectifying section r that exits from state e must be assigned to a column cc at lower pressure

$$P_{k,c} \wedge P_{l,c} \Rightarrow P_{r,cc} \qquad k \in STR_e; r \in T\_REC_e; L \in T\_STR_e$$
$$c, cc \in \text{COLUMN/ ord}(cc) < \text{ord}(c) \tag{26}$$

Figure 10d illustrates this situation.

If a state has a heat exchanger associated with it, these relationships do not apply, because no vapor flow transfer is needed. In those states in which more than one section reaches (systems with more than the minimum number of sections), the vapor flow always can be transferred from the column at high pressure.

Examples 3 and 4

As an example, Figure 11 shows the 19 thermodynamically equivalent easier-to-control configurations for a given sequence of separation tasks of five components using $4N-6$ column sections.The configurations in Figure 11 coincide with

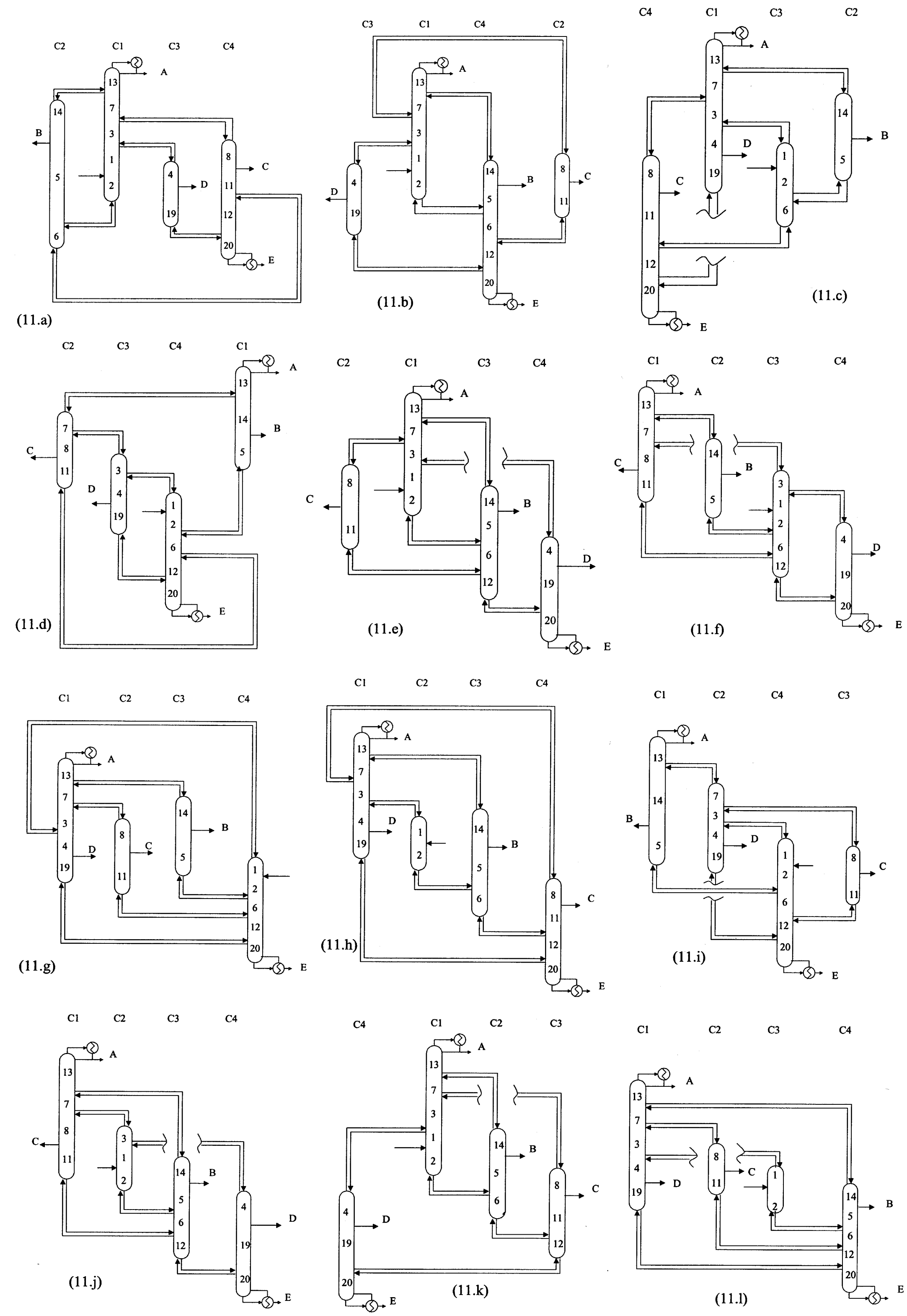


Figure 11. The 19 thermodynamically equivalent configurations that are "easier to control" for the sequence of tasks (11.t) in a mixture of 5 components using the minimum number of column sections.

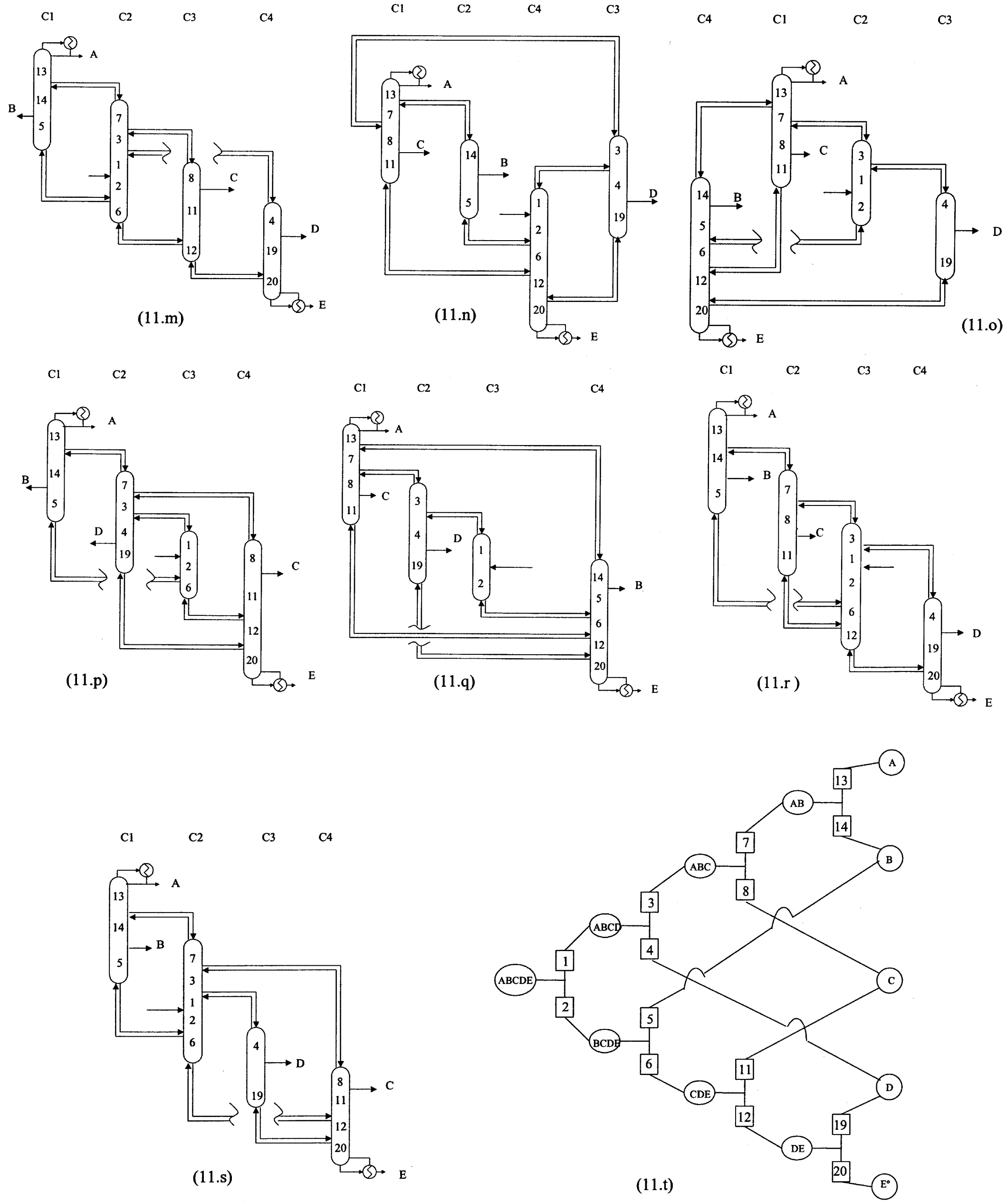


Figure 11. (Continued) The 19 thermodynamically equivalent configurations that are "easier to control" configurations for the sequence of tasks (11.t) in a mixture of 5 components using the minimum number of column sections.

Agrawal's work (2000, Figure 5) and validates the proposed methodology. Finally, as a summary example, Figure 12 shows all of the possible arrangements using two columns for separating a 3-component mixture. Figure 12 also includes all the thermodynamically equivalent schemes.

Example 5

This example illustrates how to integrate the logic expressions presented earlier with sizing and cost equations in order to synthesize an optimum configuration.

Consider the sequence of tasks in Table 2. This case corresponds to a 5-component system in a fully thermally coupled separation sequence using the minimum number of column sections (64 thermodynamically equivalent configurations). In this case, vapor and liquid flows and the number of theoretical trays in each section are known. With these data, minimum diameters of each of the column sections can be calculated using the procedure presented by Stichlamir and Fair (1998) (see Table 2).

Since we are comparing thermodynamically equivalent sequences, it is not necessary to include the cost of heating and

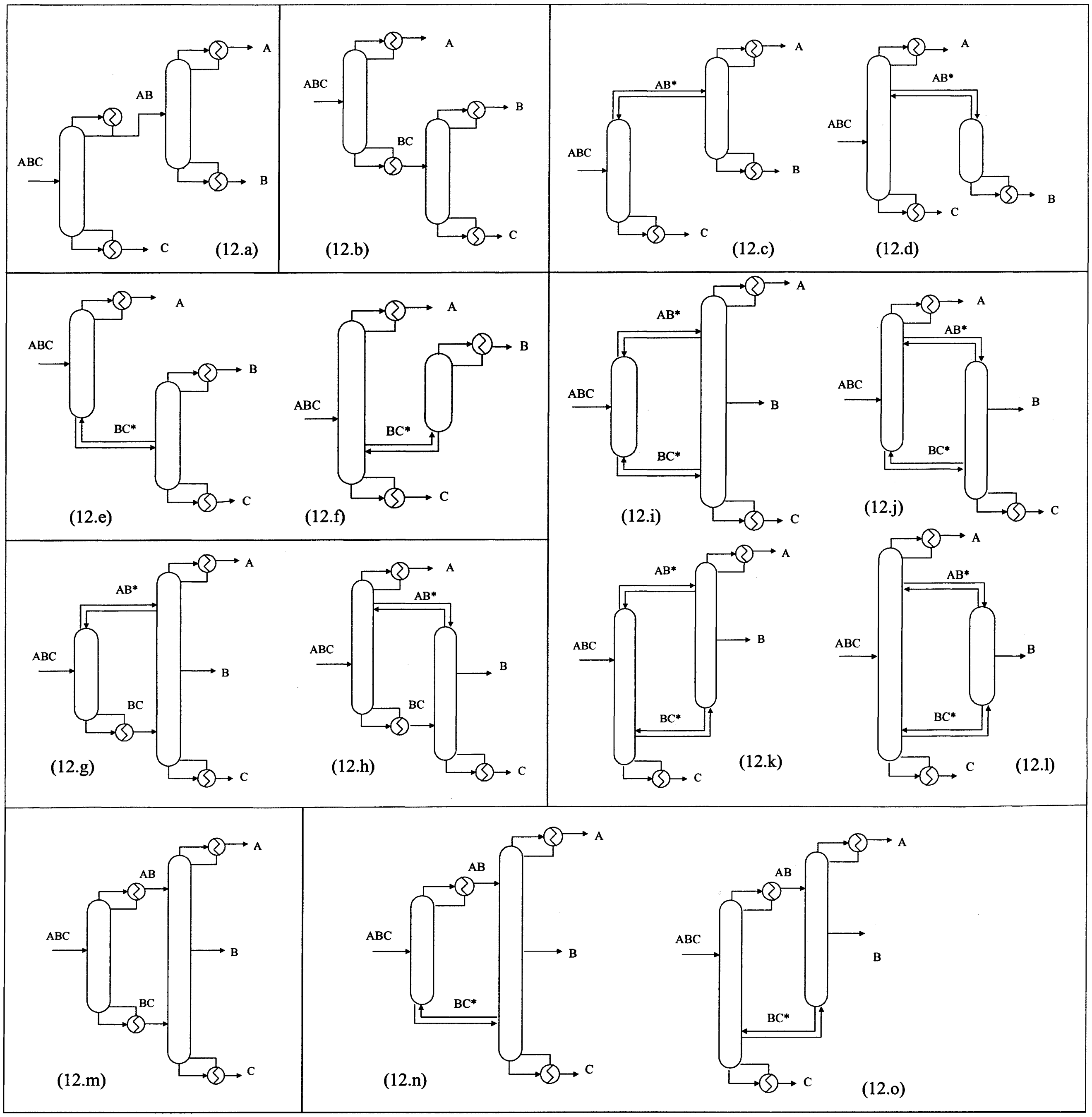


Figure 12. All of the possible rearrangements, in two columns, for a mixture of 3 components; all the thermodynamically equivalent schemes are included.

cooling duties, or cost associated with heat exchangers (reboiler and condenser).

Due to the fact that only separation tasks are considered in a state task approach, and not actual columns, the total annual cost can be only approximately estimated. For example, if it is assumed that each separation task is a column. However, if we rearrange all these tasks in N-1 columns we obtain columns with different lengths and diameters depending on which tasks are grouped, therefore the total cost change. If the energy cost is very high, there will not be a large difference among the different configurations, but if the capital cost is also important, then some important differences can be found. It is not uncommon to find different configurations with very similar costs (among all the thermodynamically equivalent) that produce a degree of flexibility that can be used to select alternatives based on some other considerations. Interestingly, they could be included in the model simply by adding some constraints.

For cost estimation we have used the procedure proposed by Ulrich as presented in Turton et al. (1998).

Table 2. Data for Example 5. Separation of 5-Component Mixture

.A: *n*-pentane; *B*: *n*-hexane; *C*: n-hepthane; *D*: n-octane; *E*: n-nonane Feed 50 kmol/h. isomolar Separation Sequence: ABCD/BCDE, ABC/D, B/CDE, AB/C, C/DE, A/B, D/E Separation Task	Column Section	Diameter Section (DS, m)	Number of Trays in Section (NS)
ABCD/BCDE	S1	1.07	3
	S2	1.30	3
ABC/D	S3	1.02	13
	S4	0.35	13
B/CDE	S5	1.08	13
	S6	1.82	13
AB/C	S7	1.11	12
	S8	0.45	12
C/DE	S11	1.34	15
	S12	2.36	14
A/B	S13	1.21	10
	S14	0.55	10
D/E	S19	1.50	16
	S20	3.13	16
Optimal arrangement: Cost: $1,361,471			
Column 1: J13, J7, S8, S11 Column 2: J14, J5 Column 3: J3, J4, J19 Column 4: J1, J2, J6, J12, J20			

Mathematically the optimization model can be written as follows

P2:

$$
\left.\begin{array}{l}
\min \sum_{c \in \text{ COLUMNS}} (CBM_c + CBMI_c) \\
\text{s.t.} NT_c = \sum_{s \in \text{SECTION}} NS_s P_{s,c} \\
\begin{bmatrix} Y1_c \\ 7 \le NT_c \le 10 \\ Fq_c = 2 \end{bmatrix} \veebar \begin{bmatrix} Y2_c \\ 10 \le NT_c \le 20 \\ Fq_c = 1.5 \end{bmatrix} \veebar \begin{bmatrix} Y3_c \\ 20 \le NT_c \\ Fq_c = 1 \end{bmatrix} \\
CpI_c \ge CpIS_s p_{s,c} \; s \in \text{SECTION} \\
CBMI_c = 1.2 CpI_c NT_c Fq_c \\
H_c = 2 + 0.5 NT_c \\
D_c \ge DS_s p_{s,c} \; s \in \text{SECTION} \\
k1_c = 3.2104 + 04641 D_c - 0.0462 D_c^2 \\
k2_c = 0.6477 - 0.1029 D_c + 0.0101 D_c^2 \\
k3_c = 0.2476 - 0.0650 D_c + 0.00942 D_c^2 \\
log(Cp_c) = k1_c + k2_c log(H_c) + k3_c [log(H_c)]^2 \\
CBM_c = Cp_c (2.5 + 1.72) \\
\Omega(P_{s,c}) = \text{TRUE}
\end{array}\right\}
$$

$$\times \text{c} \in \text{COLUMNS}$$

$$CBM_c, CBMI_c, D_c, H_c, Fq_c, NT_c, Cp_c \ge 0;$$
$$Y1_c, Y2_c, Y3_c = \{\text{TRUE,FALSE}\}$$

where *CBM* and *CBMI* are the bare module costs for vessels and internals respectively; *Cp* and *CPI* are the purchased costs of the equipment, assuming ambient operating pressure and carbon steel construction; *NT* is the number of trays in a column or in a section (*NS*); D makes reference to the column diameter, calculated as the maximum of the diameters of individual column sections; *H* is the column height (assuming 50 cm as space between trays), plus 2 m for vapor disengagement at the top and liquid sump at the bottom; and *Fq* is a factor based in the number of trays in the vessel.

In problem P2, *Y1*, *Y2*, *Y3* are Boolean variables introduced to select the factor *Fq* as a function of the number of plates in the column. Note also that variable $p_{s,c}$ is a binary (0,1) variable that takes the value 1 if the Boolean variable $P_{s,c}$ takes the value True.

Finally, the last equation in problem P2 corresponds to the set of logic constraints previously cited in the article.

The optimal configuration is shown in Figure 13. Note that this solution is also a configuration that is easier to control.

Conclusions

In this article, we have presented a systematic way for generating all of the possible arrangements of a given sequence of separation tasks in *N*-1 distillation columns. The problem is represented through a set of symbolic relations that can be written in propositional logic form. These logic propositions completely determine the space of alternatives for arranging a sequence of tasks in fully or partially thermally coupled distillation systems using *N*-1 distillation columns. This space of alternatives includes from fully thermally coupled distillation systems (only one reboiler and one condenser) to conventional columns going through all the intermediate possibilities.

In order to avoid the control problems associated with transfer vapor flow from columns at high to lower pressures,

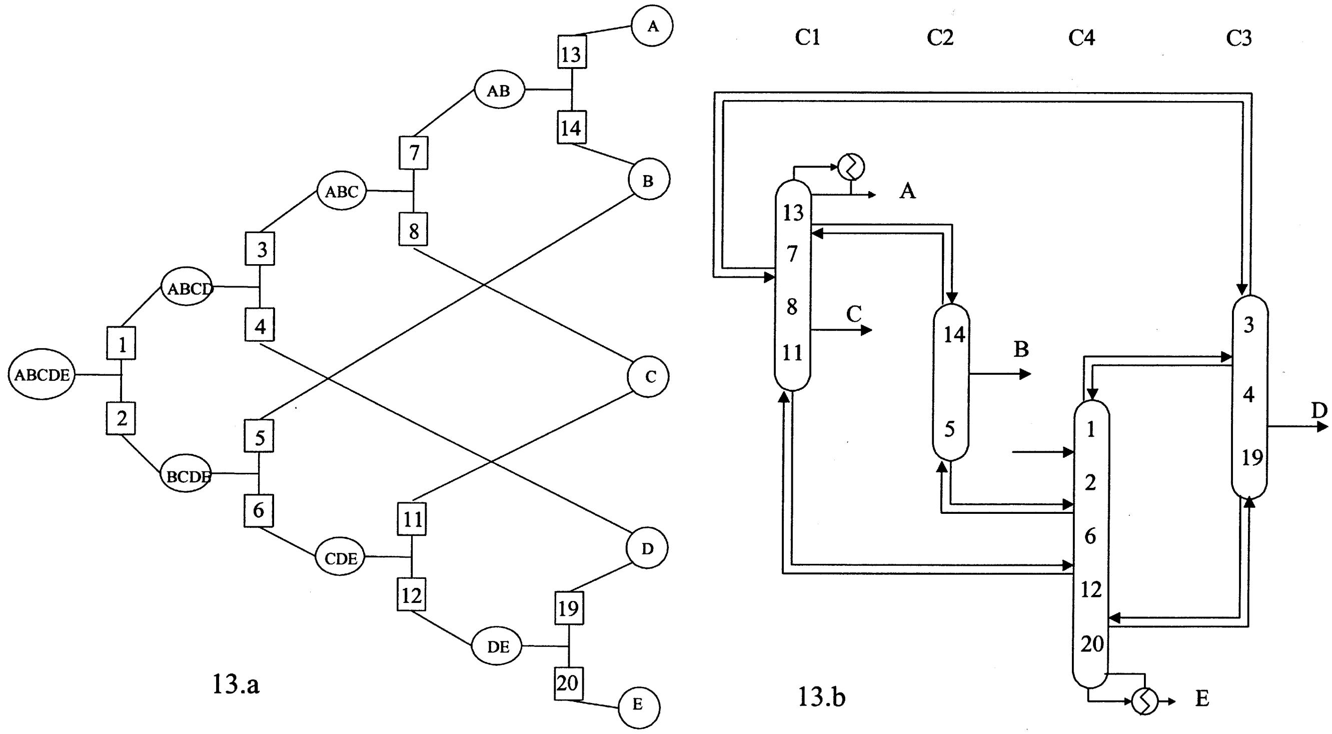


Figure 13. Optimal solution of Example 5.

a set of logic relations was added to the previous problem to generate only alternatives in which the vapor flow always goes from higher to lower pressures (easier-to-control configurations).

These logical relations can be used to generate alternatives, or they can be included in an optimization framework in order to obtain the best configuration, given some optimization criterion. In this context, disjunctive programming is especially well suited, because it allows inclusion of these sets of relations directly or translated into a set of algebraic equations in terms of binary variables.

Although it is not the objective of this work, it is interesting to note that partially or fully thermally coupled sequences are the starting point for the design of columns with divided walls. While for a 3-component mixture there is a single possibility, for mixtures with more than four components there is a large number of possibilities for merging two or more columns in column/s with internal wall/s. Different sequences of tasks, together with different thermodynamically equivalent rearrangement possibilities, open a new and promising set of alternatives that will be reported in a future article.

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Appendix A: Number of Thermodynamically Equivalent Schemes for a Given Sequence of Tasks

Consider, for example, the tasks presented in Figure 7. In this figure the boxes are column sections and the circles are tasks. Consider a state, for example, the state AB. If there is a heat exchanger associated to state AB it is clear that section 13 and section 7 must belong to different columns. However, if the state AB does not have a heat exchanger associated with it, sections 13 and 7 can be assigned to the same or to different columns. In fact there are two possibilities: section 13 and 7 belong to the same column, or section 13 and 7 belong to different columns. The same happens with states *ABC, ABCD, BCDE, CDE*, and *DE*.

State BC (internal sate) is somewhat different. If there is no heat exchanger, there are only two possibilities: sections 8 and 9 belong to one column and sections 15 and 16 belong to another column, or section 8 and 16 belong to one column and sections 9 and 15 belong to another column. The same happens to the other internal states (BCD and CD). This observation was previously reported by Agrawal (2000), and is the base of step 3 in his procedure for generating, by hand, thermodynamically equivalent configurations.

Sections 14 and 15 must belong to the same column. The product (state) B only produces one option. The same happens to product C, so sections 16 and 17 must belong to the same column, and to product D and sections 18 and 19. Again this observation was previously reported by Agrawal (2000), and it is step 1 in the previously mentioned procedure.

For any other sequence of tasks the same results are obtained. Whatever state is considered (except the initial state and those that leave the system — pure products), it produces two alternatives (provided that that state has no associated heat exchanger).

In general, a given state can be reached by only one column section (if we are using the minimum number of column sections), and produces two column sections. Two of these three sections belong to the same column (one stripping and one rectifying section), which produces two possibilities. If we are using more than the minimum number of column sections, some internal states can be reached by two column sections (a stripping and a rectifying section). Again, as was shown previously, only two possible arrangements are possible.

Therefore, the total number of thermodynamically equivalent configurations for a given sequence of task is given by

$$NC = 2^{\substack{\text{Number of} \\ \text{thermal links}}} = 2^{\substack{\text{number of intermediate states} \\ \text{without heat exchanger associated}}} \tag{A1}$$

The number of the intermediate states (all the states except the initial one and those that leave the system) is equal to the number of separation tasks (NT) minus one (that task is associated with the initial mixture), and minus the heat exchangers of class I (NHE_I). Thus

$$NC = 2^{(NT - NHE_I - 1)} \tag{A2}$$

Usually we are interested in designing systems that use the minimum number of column sections. Consider first the case of fully thermal coupling (only two heat exchangers). In this case, the number of tasks is $(4N - 6)/2$. A heat exchanger associated with a product that leaves the system (class II) reduces by one the number of tasks, and a heat exchanger of class I divided by 2 reduces the number of configurations.

Then, for a system using the minimum number of column sections, the number of configurations is

$$NC = 2^{((4N-6)/2)(NHE_{II}-2)-NHE_I-1)} = 2^{((4N-6)/2NHE+1)} \tag{A3}$$

where NHE_{II} is the number of heat exchangers of class II, and NHE is the total number of heat exchangers. Note that there are always at least two heat exchangers of class II, so these must be subtracted from the final number of heat exchangers.

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