A Matrix Method for Multicomponent Distillation Sequences

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We describe a simple-to-use "matrix" method for obtaining all the basic distillation configurations and additional thermally coupled configurations that separate a zeotropic multicomponent feed into essentially pure product streams. This provides an opportunity to rank-list the configurations for a given application subject to criteria of interest. The only information needed to generate the configurations is the number of components in the feed. We have successfully enumerated all the configurations for feeds containing up to eight components. The method can also be used to generate nondistillation and hybrid separation configurations, and even easy-to-retrofit configurations. We illustrate the use of this method by applying it to the highly energy-intensive problem of petroleum crude distillation. We have identified more than 70 new configurations that could potentially have lower heat duty than the existing configuration. A significant number of these could reduce the heat demand by nearly 50%. *AIChE J,* 56: 1759–1775, 2010
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Introduction

Industrial mixtures to be separated often have more than two components. To separate a multicomponent mixture into more than two product streams each enriched in one of the components, we usually cannot use a single separation unit. Instead, we need a sequence or configuration of separation units. As distillation is usually the preferred separation process in the chemical industry, several attempts have been made to draw all possible distillation sequences for a multicomponent separation. The earliest attempt probably dates back to Lockhart, 1947[1]—and this problem continues to be addressed even in the present.

The problem can be stated as follows: Given an n-component zeotropic mixture to be separated into n essentially pure product streams, we want to find the distillation sequence that best satisfies one or more of our desired goals. The goals can be minimization of total heat duty, capital cost and total cost, heat integration with the rest of the process, etc.

A distillation configuration to separate an n-component feed into n pure product streams can be classified as having less than $(n - 1)$ distillation columns, exactly $(n - 1)$ distillation columns, or more than $(n - 1)$ distillation columns. Distillation configurations with less than $(n - 1)$ columns are attractive for very limited cases of feed compositions and relative volatilities of components. Therefore, we shall only consider distillation configurations with at least $(n - 1)$ distillation columns, which can further be classified as either basic or nonbasic distillation configurations. Basic distillation configurations are those that use exactly "$n - 1$" distillation columns and produce n product streams, each enriched in one of the components.[2] Each distillation column uses one reboiler and one condenser. Nonbasic configurations are those that use more than "$n - 1$" distillation columns to achieve the same task.[3] These configurations tend to have more capital cost than basic distillation configurations because of the additional columns and their associated condensers and reboilers. Through extensive calculations for a

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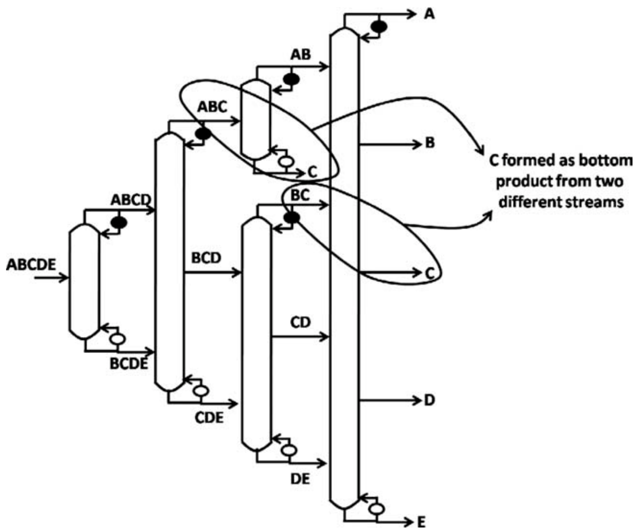

Figure 1. An example of a nonbasic configuration that uses five distillation columns for separation of a five-component feed mixture.

four-component feed mixture, Giridhar and Agrawal[3] found that the operating cost for nonbasic configurations in terms of total heat duty is never less than that of optimal basic configurations. Therefore, optimal basic configurations are expected to be less expensive to build and operate than a given nonbasic configuration. Nonbasic configurations containing *n* or more columns can thus be omitted from the search space (the complete set of all feasible distillation sequences), thereby considerably reducing the size of the search space.[3]

When a feed stream is split into product streams by a separation unit, the split can be classified as either a "sharp" split or a "nonsharp (sloppy)" split. In sharp splits, the feed stream is split into product streams that have no, or at most acceptably small amounts of, overlapping components. On the other hand, nonsharp splits allow components of intermediate volatility to distribute in significant amounts between product streams. If a mixture ABC is the feed, it can "sharp" split to only A/BC or AB/C, whereas it can also "nonsharp" split to AB/BC. In a sharp-split distillation configuration, every split is a sharp split. In a nonsharp-split configuration, at least one of the splits must be nonsharp. All sharp-split configurations are basic distillation configurations, and nonsharp-split configurations could be either basic or nonbasic. Some distillation configurations have "divided wall" distillation columns that incorporate two or more columns into a single shell. For our purposes, divided wall columns will not be treated as single distillation columns.

Also, in an actual sharp-split distillation, it will be very difficult to drive the concentrations of other components in a stream to exactly zero. For sharp-split A/BC, stream BC will have components B and C in significant amounts, whereas component A will not be completely absent, but will be present in at most some acceptably small amount. Throughout this article, we shall treat A as the most volatile component in any mixture, with volatility decreasing in alphabetical order.

An example of a nonbasic configuration for a five-component feed is shown in Figure 1. It uses five distillation columns, and it produces C as the bottom product from two different streams, ABC and BC. This forces the use of an additional distillation column. In fact, whenever a stream is produced as the top product from two different streams or as the bottom product from two different streams, we would have to use a nonbasic configuration. In Figure 1 and throughout the remainder of this article, filled circles are used to represent condensers, and nonfilled circles are used to represent reboilers.

All the basic distillation configurations to separate a three-component feed into three essentially pure product streams are shown in Figure 2. These are basic configurations because they use two distillation columns each with only one reboiler and one condenser. The configurations of Figures 2a,b are sharp-split configurations, whereas Figure 2c shows a nonsharp-split configuration.

We refer to streams that are transferred between distillation columns in a configuration as intermediate product streams or submixtures. These are the streams that are present outside distillation columns in a configuration, except the main feed stream and the final product streams. In Figure 2 for instance, streams AB and BC are submixtures. A submixture's reboiler can be replaced by transfer of a vapor stream between distillation columns, or its condenser can be replaced by transfer of a liquid stream between distillation columns (Figure 3). These are referred to as thermal coupling links between distillation columns. Some configurations with thermal coupling have been known for a while.[4,5] Figure 3a shows an example of a configuration with thermal coupling, which is derived from the distillation configuration of Figure 2a. In this configuration, stream ABC is the feed, streams A, B, and C are the final products, and submixture stream BC's reboiler has been replaced by a thermal coupling link between the two distillation columns. Figure 3b shows another example of a thermally coupled configuration,

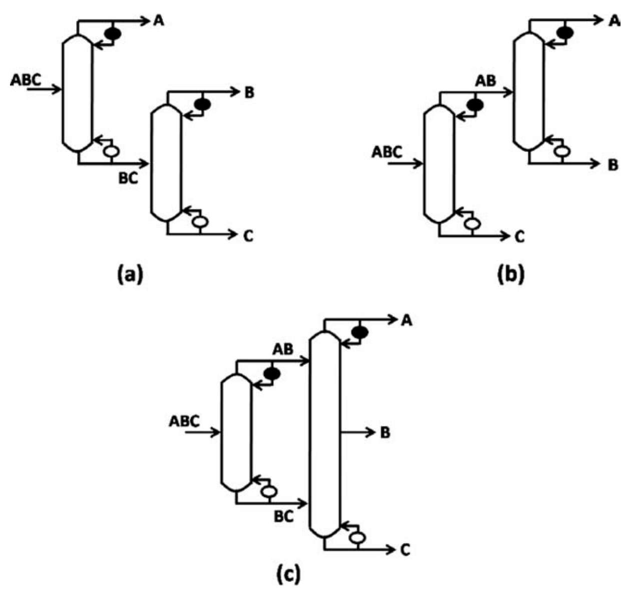

Figure 2. Basic distillation configurations for a three-component feed mixture.

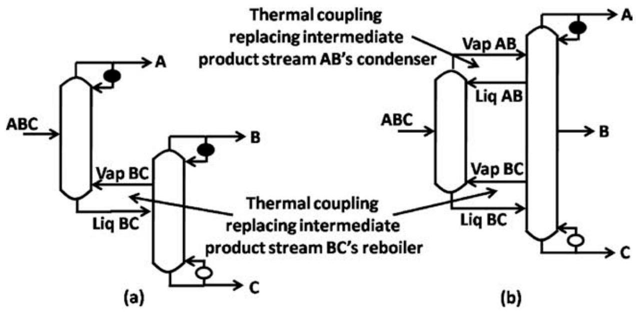

Figure 3. Thermally coupled analogs of the basic configurations shown in Figures 2a,c, respectively.

and it is derived from the basic configuration of Figure 2c. In this manner, we can obtain additional thermally coupled distillation configurations from any corresponding basic configuration. These additional configurations with thermal coupling often have lower total vapor duty than the corresponding basic configurations. Hence, these additional configurations should also be included in a useful search space.

The additional thermally coupled configurations derived from a basic configuration can be classified as having either partial or complete thermal coupling. A thermally coupled configuration derived from any basic configuration with thermal coupling at all the reboilers and condensers that can be replaced is referred to as having complete thermal coupling. On the other hand, if some of the intermediate product stream's reboilers or condensers are still present in the thermally coupled configuration, the new configuration has partial thermal coupling.

For any n-component feed, there always exists only one basic distillation configuration, which produces n final products and transfers $(n + 1)(n - 2)/2$ intermediate products. This distillation configuration thus involves the transfers of all possible intermediate products, and it uses $n(n - 1)$ distillation sections, thereby giving it the highest number of column sections among all the basic configurations. This configuration deserves special attention because with complete thermal coupling, it always has the lowest minimum total vapor duty for performing an n-component separation.[6,7] This is a configuration with "full" thermal coupling for separating a given n-component feed.

The fully thermally coupled configuration for a three-component feed—shown in Figure 3b—obviously has complete thermal coupling. The configuration in Figure 3a also has complete thermal coupling, but it is not the fully thermally coupled configuration for a three-component feed, because it does not involve transfer of submixture AB, and it does not use six sections. In fact, the configuration of Figure 3b is the only fully thermally coupled configuration for a three-component feed. Similarly, there always exists only one fully thermally coupled configuration for any n-component feed ($n > 2$). They are often referred to as Petlyuk configurations. These are obviously nonsharp-split configurations, and they use the maximum number of column sections among basic distillation configurations. However, it is possible to have other non-Petlyuk configurations with only one reboiler and one condenser containing less than $n(n - 1)$ sections but at least $4n - 6$ sections.[8]

On the other hand, sharp-split configurations use the minimum number of distillation sections among basic distillation configurations. This is because a sharp-split configuration uses $(n - 1)$ distillation columns, with each column having exactly one rectifying section and exactly one stripping section (like the configurations of Figures 2a,b). Hence, they use the minimum $2(n - 1)$ distillation sections, produce n final product streams, and transfer the minimum $(n - 2)$ submixture streams from one distillation column to another. In all basic configurations, the number of column sections thus varies from $2(n - 1)$ to $n(n - 1)$, and the number of submixture streams varies from $(n - 2)$ to $(n + 1)(n - 2)/2$.

There have been considerable attempts in the past to elucidate all the basic configurations and their associated thermally coupled versions for an n-component feed. Thompson and King[9] calculated the number of sharp-split configurations for a given n-component feed but did not calculate the number of nonsharp-split configurations. Petlyuk et al.[5] provided some nonsharp-split basic configurations, but their main thrust was fully thermally coupled distillation configurations to reduce the total heat duty. Some later work to include nonsharp distillation configurations also focused on thermally coupled configurations.[8,10]

Rong et al.[11] and Fidkowski[12] presented formulations to generate both basic and nonbasic configurations, but these are burdened by a rapid increase in the number of nonbasic configurations with the number of components in the feed stream.

Agrawal presented a formulation to generate only the basic configurations and all the associated thermally coupled configurations for a multicomponent mixture containing any n number of components.[2] In this formulation, the generation of configurations is indicated by deciding the presence or absence of reboilers and condensers with the intermediate boiling product streams. When compared with the common practice of generating configurations starting at the feed end, a unique aspect of this method is that the configurations are generated starting at the product end. The formulation used the fact that if a stream is produced from a distillation column, then the feed mixture that produces the product must exist in the upstream network. The method also ensured that every submixture transferred between the columns produced the product streams without allowing disappearance of any component present in the submixture. Caballero and Grossmann[13] presented a formulation to find the $(n - 1)$ column configuration with the lowest cost that also included all the heat integration possibilities in the search space. By their method, they were successful in listing the number of basic distillation configurations for up to six components in the feed. Recently, Giridhar and Agrawal[14] provided an alternative algorithm to generate the search space of all feasible basic distillation configurations and create a rank-list of these configurations based on total heat duty.

Because of the rapid increase in the size of the search space with the number of components in the feed, none of the prior efforts attempted to elucidate a complete search space of distillation configurations for more than six components in the feed. Also, all the above approaches were strongly dependent on the number of components in the feed, whereby the authors had to manually list all possible streams and separation tasks depending on the number of components in the feed and then provide mathematical constraints linking them.

In this work, we propose a new matrix-based method to generate the search space of all basic configurations for any number of components in the feed. It is quite simple and can be easily used for higher number of components in the feed. In this method, we write variables in a matrix form to use physical insights in the problem that help to simplify the problem. The method can easily incorporate the additional configurations with thermal coupling in the search space. It is also useful for other network synthesis problems involving nondistillation separation devices.

Method for Generating the Search Space of Basic Distillation Configurations

The matrix method is a six-step method to generate the complete search space of basic distillation configurations for any n-component zeotropic feed mixture containing components A, B, C, … etc. In this mixture, A is the most volatile component and volatility decreases sequentially in alphabetical order.

Let us first observe that for a given feed stream, only certain product streams can be derived from the top and bottom of a distillation column. For instance, a stream BCDE can lead to only stream BCD, BC, or B as the top product and stream CDE, DE, or E as the bottom product. For this to hold true, we have assumed that in a distillation column, any product stream has at least one less component than the feed stream. Recall that in our nomenclature, when a stream like BC is produced from BCDE, it does not necessarily mean that D and E are totally absent from BC, but when present, their concentrations are acceptably small and they eventually show up as acceptable impurities in the "pure" product streams B and C from further distillation of BC.

Step 1

The first step of our method is to identify the predominant number of components that need to be separated as product streams from a feed stream. This is available from the problem definition and is the only information needed to generate the complete search space. For instance, if we are given a five-component feed to be separated into five pure products each enriched in one of the components, we identify the predominant number of components as $n = 5$.

Step 2

The second step is to generate an $n \times n$ upper triangular matrix. For our example case of the five-component feed mixture, it implies generation of a 5×5 matrix.

In the matrix, all the elements below the diagonal are assigned a value of zero (i.e., $x_{i,j} = 0\ \forall i > j$) and all the elements in the upper triangular part correspond to unique streams. Suppose the components are numbered in decreasing order of their volatility, i.e., let A = component number 1, B = component number 2, and so on. In any row "i" of the matrix, for all $j \geq i$, let the first component of the stream

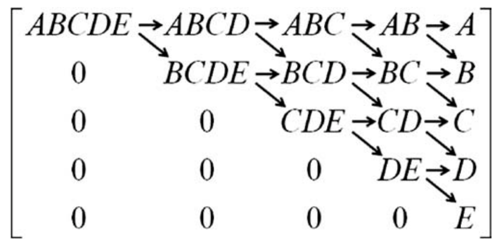

Figure 4. Matrix for a five-component feed mixture.

corresponding to each element in that row be component number "i." Also, in any column "j" of the matrix, let the number of components in the stream corresponding to each element of that column be "$n + 1 - j$" for $i \leq j$. Hence, we obtain the following equations:

$$\text{First component for mixtures in row "}i\text{"} = \text{Component "}i\text{"} \tag{1}$$

$$\text{Number of components for mixtures in column "}j\text{"} = n + 1 - j. \tag{2}$$

Therefore, to obtain the stream corresponding to any (i,j), we can calculate the number of components in the mixture using $n + 1 - j$, and then write the ith component first followed by subsequent heavier components till the total number of components in that mixture is included. For instance, in Figure 4, when (row, column) = (3,4), then row = 3 implies that the mixture will contain component number 3, i.e., component C, as the lightest component, and column = 4 implies that the mixture has two components (since $5 + 1 - 4 = 2$). As the next heaviest component is D, the mixture corresponding to the third row and the fourth column is binary mixture CD. Therefore, for the five-component feed stream, we thus get the streams ABCDE, ABCD, BCDE, ABC, BCD, CDE, AB, BC, CD, DE, A, B, C, D, and E at the locations shown in Figure 4. This is a simple way of generating all feasible mixtures given the number of components in the feed.

Listing the streams in this manner within the matrix helps us apply physical insights. If we choose any stream in the matrix, except the final products, and move horizontally to the right, all the streams that we encounter on this path are candidate top products from the distillation of the chosen stream. And when we move diagonally down and to the right, all the streams that we encounter on this path are candidate bottom distillation products. The streams that are not encountered cannot be top and bottom products of the chosen stream.

Similarly, once we pick any stream, except the main feed, and move horizontally to the left, or diagonally up and to the left, all the streams that we encounter on these paths are candidate feeds which upon distillation can produce the picked product stream. No other stream that is not on these paths can produce the picked product stream. For instance, in Figure 4, if we start from stream BCD and move horizontally to the right, we can identify streams BC and B as candidate top products. Similarly, if we move diagonally down and to the right, we can identify streams CD and D as candidate bottom products. Also, if we move horizontally to the

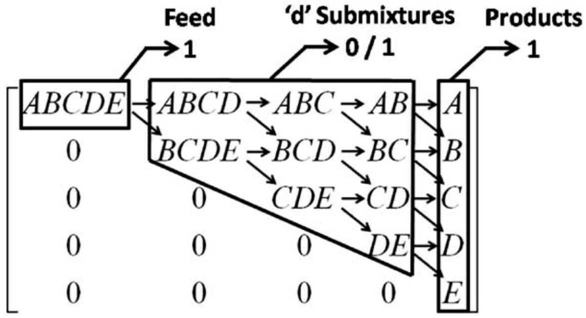

Figure 5. Identification of the "*d*" submixtures in a matrix.

left and diagonally up and to the left, we can identify streams ABCD and BCDE as candidate feeds to a distillation column to possibly produce stream BCD. None of the streams of the matrix which are not on these paths can be possible feeds or products of stream BCD. Thus, we have an $n \times n$ upper triangular matrix with favorable properties that have physical interpretations.

Step 3

The next step is to classify an element of the matrix as corresponding to either the main feed stream, a submixture stream, or a final product stream.

This is quite simple, because an $n \times n$ matrix contains $n(n+1)/2$ total elements in the upper triangular portion, of which the main feed stream is in the first column of the matrix, the "n" final product streams are in the final column, and the "d" submixture streams are in the intermediate columns. The equation that gives the number of submixture streams is thus:

$$d = [n(n+1)/2] - n - 1. \quad (3)$$

For instance, for the matrix shown in Figure 4, we have $5(5+1)/2 - 5 - 1 = 9$ submixtures in columns 2 to 4, namely streams ABCD, BCDE, ABC, BCD, CDE, AB, BC, CD, and DE, in addition to the main feed stream ABCDE in column 1 and the final product streams in column 5. These streams are boxed in Figure 5.

In a given distillation configuration, not all the possible submixtures are necessarily transferred between distillation columns. For example, there are $d = 3(3+1)/2 - 3 - 1 = 2$ submixtures, namely AB and BC, for a three-component feed. However, in Figure 2a, submixture AB is not transferred between distillation columns, and in Figure 2b, submixture BC is not transferred between distillation columns, whereas in Figure 2c both submixtures are transferred. These three configurations all have the main feed stream and the final product streams. They are different only because they transfer different submixtures. The presence or absence of such transfer submixture streams thus uniquely defines a basic distillation configuration. This is the basic principle behind the matrix method.

Hence, assume that a unique matrix corresponds to a unique basic distillation configuration. Also assume the presence of submixtures in this matrix only if the submixtures are transferred from one distillation column to another in a distillation configuration. Therefore, in the next few steps, we will provide an easy mathematical framework to indicate the presence of only certain submixtures in a matrix so that we obtain a corresponding basic configuration.

Step 4

In the fourth step, we create matrices representing all possible combinations of the presence and absence of submixtures. We first assign binary integer values (0 or 1) to each element in the upper triangular portion of the matrix. If the element of the matrix takes a value of 1, it implies that the corresponding stream is present in the distillation configuration; if the element takes a value of 0, it implies that the corresponding stream is absent in the configuration. The presence or absence of a stream refers to its occurrence outside the distillation columns of the configuration. The presence of a stream means it is either a submixture which is transferred between distillation columns, or it is the main feed stream, or it is one of the final product streams. However, in any distillation configuration, the main feed stream and the n-product streams (i.e., the (1,1) element and all the elements in the nth column of the matrix) have to exist. Therefore, those binary integer variables are forced to take a value of 1. We are left with "d" degrees of freedom from Eq. 3, which could take values of 0 or 1 and these correspond to the submixtures (like those boxed in Figure 5). Therefore, we have to generate and examine 2^d 0-1 upper triangular matrices, each of which corresponds to a candidate basic configuration. Many of these matrices are physically infeasible; the others correspond to feasible basic distillation configurations, and together they form the complete search space.

Step 5

In the fifth step, we want to eliminate physically infeasible configurations from the 2^d candidates. To do this, we use two physical facts: (a) except the main feed stream, any stream that exists in a distillation configuration must be produced by another stream, and (b) in the absence of chemical reactions, all components that enter a distillation column must also leave the distillation column. However, for our 2^d matrices, we actually implement three checks:

Check 1. For every stream that exists in a matrix (except the main feed stream), ensure that at least one corresponding stream that can act as its feed also exists within the matrix.

Check 2. Disallow physically impossible splits (for instance, a split-like BCDE forming B and DE, which is not allowed because C has disappeared in the split).

Check 3. Ensure that at least $n - 2$ submixtures are transferred.

Checks 1 and 2 have been derived from observations listed by Agrawal.[2,8] Caballero and Grossmann[13] have provided mathematical constraints that apply these checks for their state-task network representation. The first two checks, which correspond directly to the physical facts, are sufficient to ensure feasibility. The third check only helps us to speed up the process of eliminating infeasible sequences[13] and arises out of the following observation: we are interested in basic distillation configurations, and hence we have exactly

$(n - 1)$ distillation columns in a configuration. Each distillation column must necessarily have a top and a bottom product, while it may or may not have some sidedraw product streams. Each distillation column thus has at least two product streams. We have a minimum of $2(n - 1)$ product streams in a configuration, of which n are final product streams. This gives us the minimum number of intermediate product streams (or submixtures) as $2(n - 1) - n = (n - 2)$. In fact, the configurations that have exactly $(n - 2)$ out of the maximum possible "d" submixtures being transferred are the sharp-split configurations. It is clearly not possible to have a feasible basic distillation configuration that transfers less than $(n - 2)$ submixture streams.

The three checks can be easily implemented in a computer program for any n-component mixture because of the favorable properties of the matrix representation. Let $x_{i,j} = 0/1$ be the binary variables associated with the (i,j)th elements of the matrix. We shall now develop mathematical constraints to implement these physical checks.

Suppose we want to apply Check 1 for the (p,q)th element of the matrix. The check should of course be enforced only if the (p,q)th element has a binary value of 1 (i.e., the corresponding submixture is transferred between distillation columns in the configuration). Therefore, if $x_{p,q} = 1$, we must ensure that it is formed from at least one feasible feed—at least one of the (p,q)th element's candidate feed streams should also have a binary value of 1. Recall that the candidate feed streams of the (p,q)th stream can lie only on paths horizontally to the left and diagonally up and to the left from the (p,q)th element of the matrix. At least one of the binary variables on these paths must have a value of 1, so the first check simply results in:

$$\sum_{\substack{j=p \\ p \neq q}}^{q-1} x_{p,j} + \sum_{\substack{i=1 \\ p \neq 1}}^{p-1} x_{i,i+q-p} \geq x_{p,q} \forall q > 1, \forall p \leq q. \tag{4}$$

In Eq. 4, the first summation term on the LHS is the sum of all the binary variables on a horizontal path to the left of the (p,q)th element. For this term, $p \neq q$ ensures that we do not move in the lower triangular portion of the matrix. The second summation term on the LHS is the sum of all the binary variables on a diagonal path moving up and to the left of the (p,q)th element. Here, $p \neq 1$ ensures that we do not move out of the matrix.

If the RHS, $x_{p,q}$, takes a value of 0, all these LHS binary variables are free to be either 0 or 1, and the constraint does nothing to ensure the presence of the candidate feed streams. This is desired, as we do not need to ensure presence of feeds for a product stream that does not exist in the configuration. On the other hand, if the RHS takes a value of 1, at least one of the binary variables on the LHS is forced to take a value of 1, thereby ensuring that at least one of the feasible feed streams exists to produce the (p,q)th stream under consideration. Obviously, this constraint is not applied to the main feed stream, thereby giving us $q > 1$. Also, it is applied for all other streams in the matrix, but with $p \leq q$, to disregard elements of the matrix below the diagonal which have been assigned values of 0 while generating the $n \times n$ matrix. Equation 4 thus provides a mathematical constraint for applying Check 1.

Next, for applying Check 3, we use the following constraint:

$$\sum_{j=2}^{n-1} \sum_{i=1}^{j} x_{i,j} \geq (n-2). \tag{5}$$

Here, the LHS involves all the "d" binary variables corresponding to the submixture streams, and the inequality ensures that at least $(n - 2)$ of these submixture streams are transferred.

Therefore, Checks 1 and 3 result in simple linear inequality constraints. It is not as straightforward to obtain such constraints for Check 2, but Check 2 can still be implemented easily because of the matrix representation of a configuration.

To implement Check 2, we start from the main feed stream (in column 1 of the matrix) and go up to all the ternary streams (in column $n - 2$ of the matrix). This is because there is never disappearance of components in the distillation of any binary stream as all the final products are always present in any configuration. Therefore, we systematically apply Check 2 to all the elements that have values of 1, in the upper triangular portion of columns 1 through $(n - 2)$ of the matrix. If an element located in say column j–and thus containing $(n + 1 - j)$ components as seen from Eq. 2—has a binary value of 1, then from that element we march horizontally to the right and diagonally down to the right, until we hit the next "1" on each path. These are respectively identified as the top and bottom product of the stream we started from, as all preceding streams, if any, on these horizontal and diagonal paths have binary values of 0, thereby implying that they do not exist outside distillation columns in the distillation configuration. The respective matrix column locations j_1 and j_2 of these products give us the number of components in the top and bottom products as $(n + 1 - j_1)$ and $(n + 1 - j_2)$. The sum of these should be at least as much as the number of components $(n + 1 - j)$ in the stream under consideration.

Check 2 can be implemented in other ways as well. One possible alternative is to identify the first component of the top and bottom products as component number i_1 and component number i_2, respectively. This can be done by identifying the rows of the matrix in which these elements are located. The last component of the top product will then be component number $\bar{i}_1 = i_1 + (n+1-j_1) - 1 = (i_1 + n - j_1)$. We then obtain $i_2 - \bar{i}_1 \leq 1$ as the constraint to ensure Check 2. If this difference exactly equals 1, it can easily be seen that it corresponds to a sharp split (i.e., no overlapping components); if the difference is 0, then we have one overlapping component; if the difference is -1, then we have two overlapping components, and so on. Obviously, if the difference is greater than 1, it implies disappearance of components.

Check 2 can be converted to linear inequality constraints like Eqs. 4 and 5 corresponding to Checks 1 and 3, but this would require some additional binary decision variables and some additional linear constraints. One possible way to do this conversion is to define additional binary variables for all

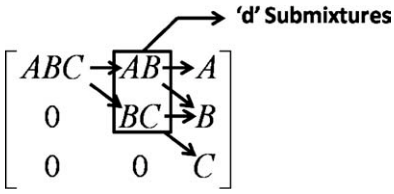

Figure 6. Matrix for a three-component feed mixture.

Submixtures are boxed.

the edges in a supernetwork[14] and specify constraints linking the edges to the matrix elements. These constraints will be similar to the connectivity constraints that link states and tasks in the method of Caballero and Grossmann.[13]

Instead of Checks 1 and 2, which are sufficient to obtain feasible configurations, one can also use any other checks as long as they perform the job of filtering out the infeasible candidate configurations. We presented Checks 1 and 2 because they have simple physical interpretations and their implementation can be easily automated, even though they may not be the strongest set of equations for a mixed integer nonlinear programming problem (MINLP) formulation to find the optimum configuration.

Any candidate matrix that satisfies all the checks is retained as a feasible basic distillation configuration. All other candidate matrices are discarded. Once we apply these checks for each of the 2^d candidate matrices, we are left with all feasible basic distillation configurations, each of which is obtained in the form of a 0-1 upper triangular matrix. However, we can also use the following alternative to obtain all the feasible basic distillation configurations: we generate linear inequality constraints corresponding to Checks 1, 2, and 3 and reformulate the problem as an integer programming (IP) problem, which can be solved to give a feasible solution (a basic distillation configuration). This basic configuration will now correspond to a vector of decision variables instead of a matrix of decision variables. We can then add a cut (a linear constraint) to eliminate this solution (configuration) and resolve the IP problem to arrive at another feasible solution. We can keep repeating this until there are no more feasible solutions. However, it is much faster and simpler to enumerate all the 2^d candidate matrices and quickly apply our checks to each.

The equations and procedure corresponding to the checks may look complicated, but they are just mathematical expressions of intuitive steps. Let us illustrate the use of all the steps of the method so far by generating a complete search space of basic configurations for a three-component feed. Although this is a simple case, the use of the checks should become clear.

Step 1 assigns $n = 3$ for the three-component feed stream. In Step 2, we declare a 3×3 matrix with all elements assigned zeros below the diagonal. For each of the remaining elements corresponding to say (i,j) with $i \leq j$, we can easily calculate the first component using Eq. 1, and the number of components of each element using Eq. 2, to obtain a description of the stream corresponding to that element. For example, for the element (1,1), the first component as given by Eq. 1 is component number 1, i.e., A, and the number of components as given by Eq. 2 is $3 + 1 - 1 = 3$. The stream corresponding to this element is stream ABC. We can repeat this procedure for each element to give us the matrix shown in Figure 6.

Step 3 makes us identify the two submixtures boxed in Figure 6 (since from Eq. 3, we get $d = 3(3 + 1)/2 - 3 - 1 = 2$). These elements of the matrix can thus take values of either 0 or 1. The other four elements are forced to take values of 1 as they correspond to the main feed stream and the three final product streams. Then, as per Step 4, we generate the $2^d = 2^2 = 4$ candidate matrices shown in Figure 7.

Step 5 involves implementing the checks to eliminate infeasible configurations. Check 1 is to be applied for all the elements except the (1,1) element corresponding to the main feed stream. Applying Eq. 4 for the (1,2) element, we obtain:

$$p = 1, q = 2 \Rightarrow x_{1,1} \geq x_{1,2}, \tag{4a}$$

which ensures a feed for the (1,2) element if it exists. Physically, it ensures that if stream AB is present in the configuration, then stream ABC, which is the only possible feed of stream AB, must also be present in the configuration.

Similarly, we get:

$$p = 2, q = 2 \Rightarrow x_{1,1} \geq x_{2,2} \tag{4b}$$

$$p = 1, q = 3 \Rightarrow x_{1,1} + x_{1,2} \geq x_{1,3} \tag{4c}$$

$$p = 2, q = 3 \Rightarrow x_{2,2} + x_{1,2} \geq x_{2,3} \tag{4d}$$

$$p = 3, q = 3 \Rightarrow x_{1,1} + x_{2,2} \geq x_{3,3}. \tag{4e}$$

Equation 4b ensures that if stream BC is present in the configuration, then stream ABC, which again, is the only possible feed of stream BC, must also be present in the configuration. The LHS of Eq. 4a comes from the first term on the LHS of Eq. 4, which corresponds to horizontal movement to the left, and the second term of Eq. 4 which corresponds to diagonal movement up and to the left makes no contribution. Also, the LHS of Eq. 4b comes from the second term on the LHS of Eq. 4 (diagonal movement) with the first term of Eq. 4 (horizontal movement) making no contribution. Equations 4a and 4b thus apply Check 1 to both the elements ($p = 1$ and $p = 2$) of column $q = 2$.

Similarly, Eqs. 4c, 4d, and 4e correspond to the three elements of the third column of the matrix: streams A, B, and C, respectively. For stream A, Eq. 4c ensures that at least one of the streams ABC or AB is present. The terms on the LHS of Eq. 4c correspond to horizontal movement, and there are no terms corresponding to diagonal movement, as there

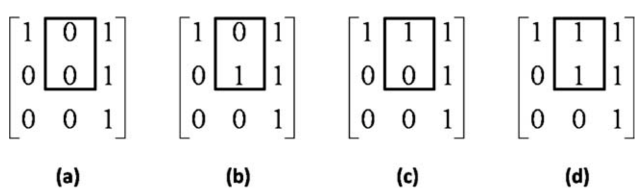

Figure 7. The $2^d = 4$ candidate matrices for a three-component feed mixture.

is no movement possible diagonally up and to the left of the (1,3) element. In the general *n*-component case, such an equation for the lightest final product will always be redundant as the main feed stream is always present, thereby always ensuring the presence of a feasible feed stream.

Next, Eq. 4e ensures that at least one of streams ABC or BC is present, to produce stream C, as stream C can be produced by no other stream. These terms on the LHS of Eq. 4e correspond to diagonal movement, as now there is no movement possible horizontally. Once again, such an equation for the heaviest final product will also always be redundant.

In fact, we will also obtain redundant equations for any submixture that contains the lightest component or the heaviest component of any *n*-component mixture, because the presence of the main feed stream always guarantees a feasible feed for any such submixture. Simply stated, we need not apply Check 1 for all the elements located in the first row of the matrix and in the diagonal of the matrix.

For stream B, Eq. 4d ensures that at least one of streams BC or AB must be present in the configuration. Note that there are no other streams that can produce stream B. Stream BC corresponds to the first term on the LHS of Eq. 4, and it arises due to horizontal movement to the left from stream B. Similarly, stream AB corresponds to the second term on the LHS of Eq. 4, and it arises due to diagonal movement up and to the left from stream B.

The matrices of Figures 7b–d satisfy all Eqs. 4a–4e, whereas the matrix of Figure 7a violates Eq. 4d. Therefore, the matrix of Figure 7a corresponds to an infeasible candidate configuration and can be eliminated. This is because in this candidate configuration, none of the candidate feed streams (AB or BC) is present to form B.

Ideally, once a candidate matrix does not satisfy any equation, we can discard it straightaway without applying any further equations to it. On the other hand, in order to be a basic configuration, a matrix must satisfy all the equations and checks. In this case, further equations and checks only need to be applied to the matrices of Figures 7b–d. However, to illustrate their use, we will still apply all the other equations and checks to all the candidate matrices.

Check 2 involves starting from column 1 of the matrix and proceeding till column $(n - 2) = 1$ (in this case). For feeds that have more than three components, we would have to consider more columns of the matrix, and not just the first column like in this example. Here, we apply Check 2 only for the single element present in column 1. This (1,1) element has a value of 1, so as per Check 2, we move horizontally to the right until we encounter the next 1. This gives us the top product. Similarly, we move diagonally to the right and identify the bottom product. Therefore, for the matrix of Figure 7a, elements (1,3) and (3,3) are identified as the products. They both have $3 + 1 - 3 = 1$ component each, as obtained from Eq. 2. The sum of their number of components is 2, which is lesser than the number of components in the (1,1) element of the matrix (as it has $3 + 1 - 1 = 3$ components as obtained from Eq. 2 again). Hence, the matrix of Figure 7a is once more identified as being infeasible, but this time because component B does not appear in either the distillate and the bottom product stream in spite of being present in the feed stream, and it has thus "disappeared" during the split. Simply stated, here ABC has split to A and C, which is physically impossible. If we repeat this procedure with the remaining matrices, we find that they correspond to feasible configurations.

This concludes the application of Checks 1 and 2. The matrices of Figures 7b–d are thus the complete set of basic configurations for a three-component feed.

Let us now illustrate the use of Check 3 to quickly eliminate infeasible configurations. Check 3 provides us with $x_{1,2} + x_{2,2} \geq 1$, which just states that we should have at least $(n - 2) = 1$ submixture. Hence, the inequality ensures that at least one of the submixture streams AB or BC must be present in the configuration. Once again, we see that the matrix of Figure 7a is infeasible, whereas the other three are feasible. The first two checks are in fact sufficient for obtaining feasible configurations, but this third check always provides us with a single simple inequality constraint, which can eliminate some of the infeasible configurations without even having to go through the first two checks.

Step 6

Now we have a feasible set of 0-1 upper triangular matrices representing the search space for basic distillation configurations. From these 0-1 matrices, we can easily deduce all the information necessary to "draw" a configuration, and this will be the sixth and the final step of our method. Step 6 simply provides a user of the method or an optimization solver with the locations of the various splits in the $(n - 1)$ distillation columns of a basic configuration. The procedure to do this is as follows: (i) replace the 1s in the matrix by the corresponding streams, (ii) identify all the splits in the matrix, (iii) stack the splits appropriately in distillation columns, and (iv) assign distillation column numbers.

Consider the matrix shown in Figure 8a. After replacing the 0-1 elements by the appropriate streams, we get the matrix shown in Figure 8b. We then start from the main feed stream ABCDE, and move horizontally to the right, and identify stream ABCD as the top product (this is the next "1" on a horizontal path), and we move diagonally down to the right and similarly identify BCDE as the bottom product. Therefore, the first split is ABCDE – ABCD/BCDE. We repeat the procedure with stream ABCD and get the second split as ABCD – ABC/BCD. Similarly, we also get the following splits: BCDE – BCD/CDE, ABC – AB/BC, BCD – BC/CD, CDE – CD/DE, AB – A/B, BC – B/C, CD – C/D, and DE – D/E. These splits are depicted in Figure 8c. Note that we are depicting these splits as pseudo-distillation columns. Here, we do not allow a stream to be formed as the top product from two different streams or as the bottom product from two different streams. This implicitly avoids generating nonbasic configurations.

Next note that splits which make a common stream (product) must belong to the same distillation column to have a basic configuration that uses exactly $(n - 1)$ distillation columns. These common streams are withdrawn as sidedraws, and their associated reboilers and condensers are eliminated. Hence, splits ABCD – ABC/BCD and BCDE – BCD/CDE belong to the same distillation column as they both make BCD, which will be produced as a sidedraw. Similarly, splits ABC – AB/BC and BCD – BC/CD belong to the same

$$\text{(a)} \begin{bmatrix} 1 & 1 & 1 & 1 & 1 \\ 0 & 1 & 1 & 1 & 1 \\ 0 & 0 & 1 & 1 & 1 \\ 0 & 0 & 0 & 1 & 1 \\ 0 & 0 & 0 & 0 & 1 \end{bmatrix} \qquad \text{(b)} \begin{bmatrix} ABCDE & ABCD & ABC & AB & A \\ 0 & BCDE & BCD & BC & B \\ 0 & 0 & CDE & CD & C \\ 0 & 0 & 0 & DE & D \\ 0 & 0 & 0 & 0 & E \end{bmatrix}$$

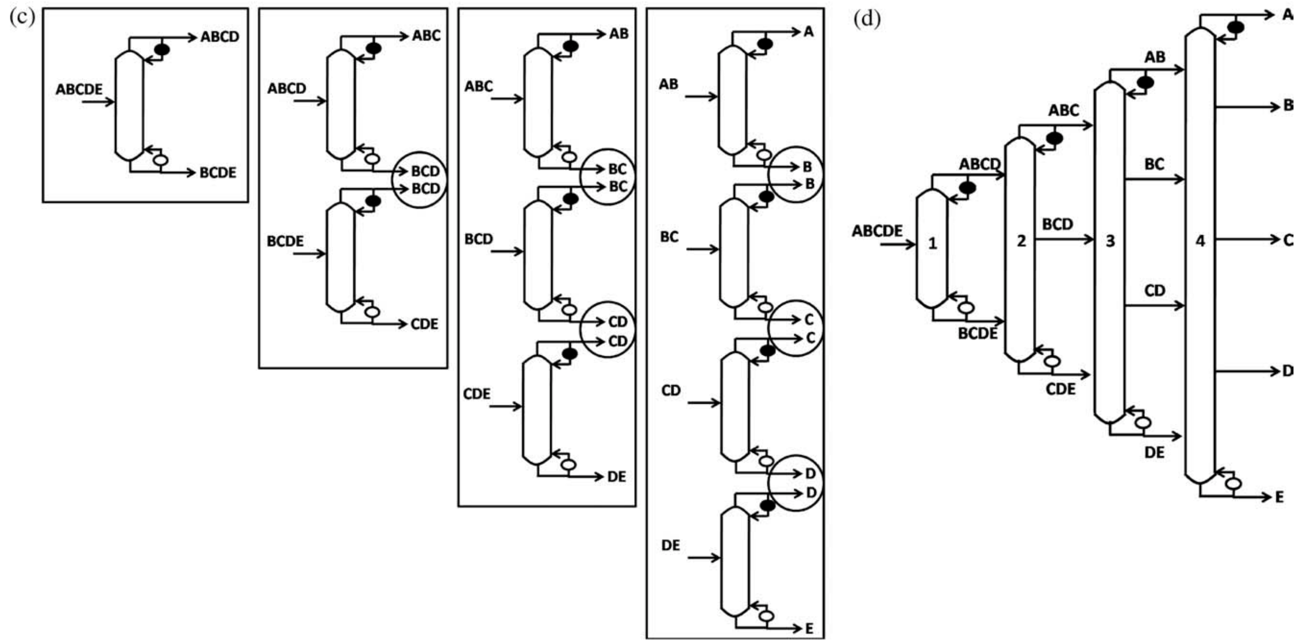

Figure 8. (a) A feasible 0-1 matrix; (b) Replacing 0-1s by appropriate streams in the matrix of Figure 8a; (c) Listing all possible splits and then grouping the splits that belong to the same distillation column; (d) Assigning distillation column numbers.

distillation column as they both make BC, and split CDE – CD/DE also belongs to this distillation column as it makes CD in common with the split BCD – BC/CD. In the same way, splits AB – A/B, BC – B/C, CD – C/D, and DE – D/E belong to the same distillation column. These groupings are illustrated by the boxes in Figure 8c. A consequence of this procedure is that to produce any stream on a horizontal path to the right, we must use a condenser. Similarly, to produce any stream on a diagonal path to the right, we must use a reboiler. Further, if the stream is produced by both a reboiler and a condenser, the reboiler and condenser are eliminated and the stream is then produced as a sidedraw.

Finally, we assign distillation column numbers to the splits. Therefore, split ABCDE – ABCD/BCDE belongs to, say, "distillation column 1" shown in Figure 8d, splits ABCD – ABC/BCD and BCDE – BCD/CDE both belong to, say, "distillation column 2" of Figure 8d, splits ABC – AB/BC, BCD – BC/CD, and CDE – CD/DE all belong to "distillation column 3," and finally splits AB – A/B, BC – B/C, CD – C/D, and DE – D/E all belong to "distillation column 4" of Figure 8d; we thus obtain the completed distillation configuration shown in Figure 8d. In this way, any 0-1 matrix can be converted easily to a distillation configuration figure. Within a distillation column that contains more than one split, a split whose feed has a lighter first component must be placed above a split whose feed has a heavier first component. Similarly, we can obtain the distillation configurations of Figure 2 from the matrices of Figures 7b–d.

Our interpretation of the matrix shown in Figure 8a led us to a basic configuration. We could have interpreted it differently to give a nonbasic configuration like the one shown in Figure 1 for instance, as this distillation configuration also corresponds to the same matrix given in Figure 8a where all possible submixtures are present. Another example where the application of the procedure of Step 6 leads to the satellite column configuration is provided in the Supporting Information.[8]

Finally, the method can be easily modified to obtain search spaces that contain special types of basic configurations only, as dictated by physical needs of an actual problem. For example, to obtain only the sharp splits, the inequality of Eq. 5 is replaced by an equality. Consider another example. If for a five-component feed ABCDE, say component C needs to be produced from a condenser. Such a requirement may be necessary if the purity of product C needs to be strictly controlled. Then, we can add equations that assign submixtures ABC and BC as both 0, i.e., $x_{1,3} = x_{2,4} = 0$.

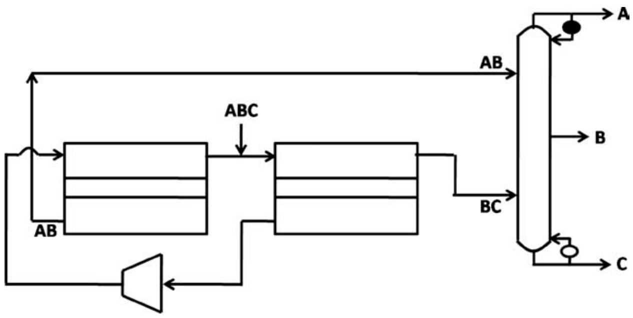

Figure 9. A hybrid separation sequence.

Method for Generating a Search Space Including Nondistillation Separation Devices

The aforementioned method can be used for any sequence synthesis; it is not limited to distillation network synthesis alone. To do this, we implement Step 1 to Step 5 as described earlier, but Step 6 is implemented a little differently. Each feasible matrix obtained after Step 5 now corresponds to more than one separation configuration, unlike the case where we consider distillation alone.

To briefly illustrate this procedure with a simple example, suppose that we can also use membrane cascades as separation units in addition to distillation columns. This means that we now have $N_{\text{options}} = 2$ unit operations as separation choices.

After Step 5, we obtain several feasible matrices. For each feasible matrix, we list all the splits corresponding to it, as described in Step 6. Suppose, we obtain s splits from a feasible matrix. Note that each of these splits is composed of two half-splits; therefore, the total number of half-splits corresponding to a matrix is $2s$. These correspond to rectifying and stripping sections for a distillation-based separation configuration. However, now any of the half-splits could also be a membrane-based separation task. Hence, the total number of basic configurations (including new hybrid schemes) that one can derive from the set of splits corresponding to each feasible matrix is $(N_{\text{options}})^{2s}$. This is because each half-split can be a different unit operation.

As an example, the hybrid scheme shown in Figure 9 uses membrane cascades for the half-splits of ABC – AB and ABC – BC, whereas it uses distillation for all the other half-splits: AB – A, AB – B, BC – B, and BC – C. Such schemes can be incorporated in a suitable optimization framework to arrive at a truly optimal sequence for any multicomponent separation problem.

For the sequence in Figure 9, we have assumed that A is the most volatile and the most permeable component, with volatility and permeability both decreasing in alphabetical order. In general, this may not be true and we have to accordingly modify the method.

Also, note that when we considered $N_{\text{options}} = 1$ unit operation, like distillation alone, then from each matrix we obtained $(1)^{2s} = 1$ basic configuration. Therefore, each matrix was always corresponding to exactly one basic configuration.

Method for Generating the Search Space of Thermally Coupled Configurations

We shall return our attention to only distillation throughout the remainder of this article. It is generally useful to include distillation configurations with thermal coupling in a search space. Also, each of the basic configurations that we obtain is uniquely characterized by the presence or absence of submixture streams. The submixture streams that are "present" in a configuration have either an associated reboiler or an associated condenser or are produced as sidedraws. The submixture streams with associated reboilers and condensers are candidate streams for thermal coupling, because these reboilers or condensers can be eliminated and replaced by a thermal coupling link. Depending on how many of the candidate streams have thermal coupling, the resulting new configuration is classified as having partial or complete thermal coupling.

For all the basic configurations that can be derived by the earlier described approach, we can look at the matrix corresponding to each configuration and identify the submixtures that are produced as sidedraws. This procedure was already described in Step 6 of the method, when we identified the splits that belong to the same distillation column. Sidedraw streams are simply the products that are formed from two different splits. All the remaining submixture streams that exist in the matrix necessarily have associated reboilers or condensers and are thus candidate streams for thermal coupling. Therefore, each of these N_{cand} candidate streams has one more 0-1 type decision associated with it, regarding whether or not it has thermal coupling. The case where none of the candidate streams has thermal coupling is the basic configuration we began with. Therefore, we can go through all the $2^{N_{\text{cand}}} - 1$ additional combinations to obtain all the thermally coupled configurations.

To illustrate systematic introduction of thermal coupling, consider the configuration shown in Figure 10a. We have a five-component feed, and hence, from our corresponding 5×5 matrix, we obtain the possible submixtures as streams ABCD, BCDE, ABC, BCD, CDE, AB, BC, CD, and DE. Of these, only streams ABCD, BCD, AB, and BC are present in the configuration. None of these is produced as sidedraws, i.e., they are all produced from just one split each, and therefore they are all candidate streams for thermal coupling. We then go through the $2^4 - 1 = 15$ possible combinations to obtain additional thermally coupled configurations. These configurations range from having partial thermal coupling to complete thermal coupling. Some of these configurations are shown in Figure 10b.

Once the basic configurations and their thermally coupled versions are known, additional configurations using reboilers and condensers at intermediate column locations or with transfer streams between columns may be drawn.[15,16] These will add to the number of possibilities, and while we have not considered them in this article, they could be included when appropriate.

Results and Discussion

A summary of the method is shown in the block diagram of Figure 11. Using the method, we can enumerate all feasible basic distillation configurations and additional thermally coupled configurations for any n-component mixture. The number of distillation configurations for n up to eight is listed in Table 1. The basic configurations for three to eight

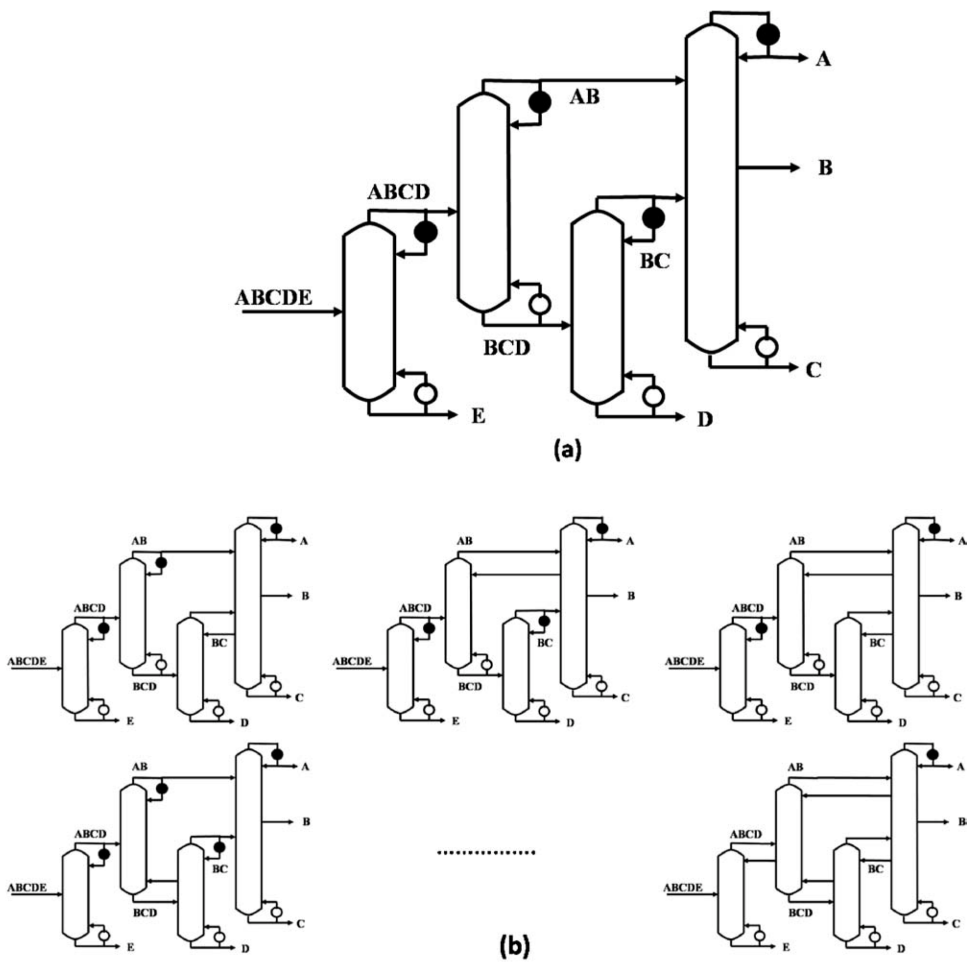

Figure 10. (a) A basic configuration and (b) thermally coupled configurations obtained from the basic configuration of Figure 10a, ranging from partial to complete thermal coupling.

components in the feed are provided as Supporting Information with this article.

Our method enables us to solve problems for n up to eight because of the following reasons:

The number of 0-1 elements of interest in our $n \times n$ matrix is the same as the number of all possible streams in a distillation network (seen through Step 2) and is equal to $n(n + 1)/2$. Of these, only the elements corresponding to submixtures provide degrees of freedom. Hence, the degrees of freedom are given by $d = [n(n + 1)/2] - n - 1 = O(n^2)$ in our matrix-based approach. Therefore, the degrees of freedom in our approach grow quadratically with the number of components in the feed.

On the other hand, in some earlier works, the degrees of freedom are the number of edges in a supernetwork, giving $d = (n - 1)(n)(n + 1)/3 = O(n^3)$.[14] Hence, we see that in this case, the degrees of freedom grow cubically with the number of components in the feed. The difference between these two approaches becomes even more pronounced because we generate 2^d candidate configurations to obtain the search space. Using our matrix-based approach, we can thus easily enumerate distillation configurations for a higher number of components in the feed using just a single computer.

Also, the favorable properties of the matrix representation facilitate automation of the enumeration process for any n-component feed. This helps us in generating the search space for any general number of components in the feed.

Our method thus presents a simple way to generate all the basic configurations that can perform a given multicomponent distillation task. Furthermore, each of the basic configurations results in additional configurations which range from partial to complete thermal coupling, and these can also be easily generated by our matrix-based method.

Performance evaluation

Now we have to search through the complete search space to find the configuration that best satisfies our desired goals. Among the different objective functions, we have chosen to evaluate the minimum total vapor duty using Underwood's equations.[17]

The total vapor duty is defined as the summation of all individual column vapor flow requirements—the summation of the vapor flows at all the reboilers in the configuration. The minimum total vapor duty simply means that the flows of various overlapping components in the submixtures of a configuration are adjusted to minimize the total vapor duty

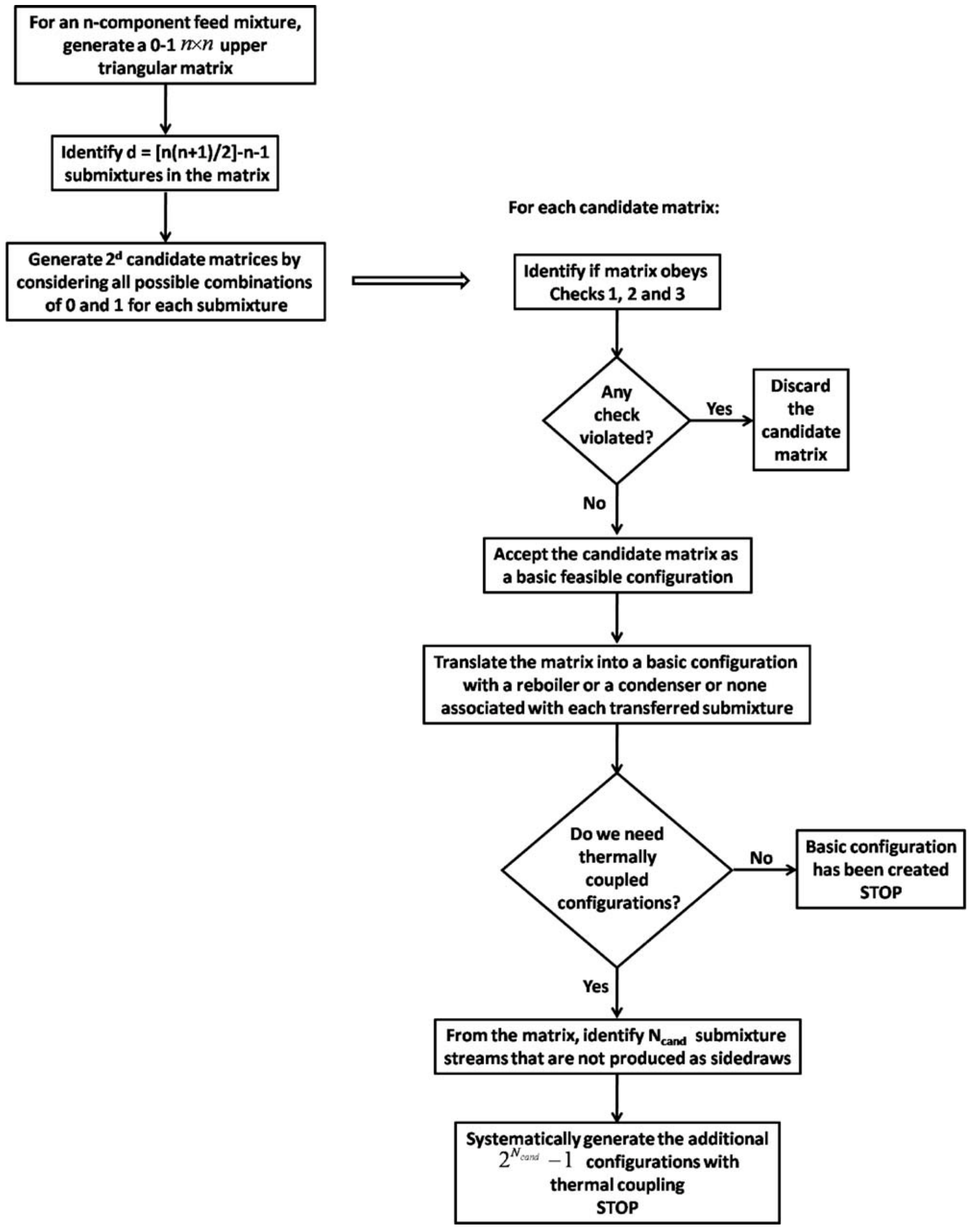

Figure 11. Block diagram showing key steps of method.

(similar to minimum reflux calculations in the case of a binary distillation).

We could have used any other objective function like capital cost, total cost, etc., but we chose the minimum total vapor duty because operating cost usually dominates capital cost in distillation, and operating cost is proportional to the total heat duty, which is proportional to the minimum total vapor duty. This assumption holds especially well for large-scale distillation processes, and it allows us to do a quick screening instead of rigorously evaluating the total cost of each configuration. We then obtain the best few configurations, which can be examined in further detail.

To calculate the minimum total vapor duty of each configuration, we formulate a nonlinear programming problem (NLP), which calculates the flows of the various submixture streams in the configuration to minimize its total vapor duty. The details of this formulation are not included in this article. Special care must be taken for the convergence of the NLP solver to a globally minimum solution.

Table 1. Number of Distillation Configurations Grows Rapidly as the Number of Components in the Feed Increases

Number of Components in the Feed	Number of Basic Configurations	Number of Additional Configurations with Thermal Coupling
3	3	5
4	18	134
5	203	5925
6	4373	502,539
7	185,421	85,030,771
8	15,767,207	29,006,926,681

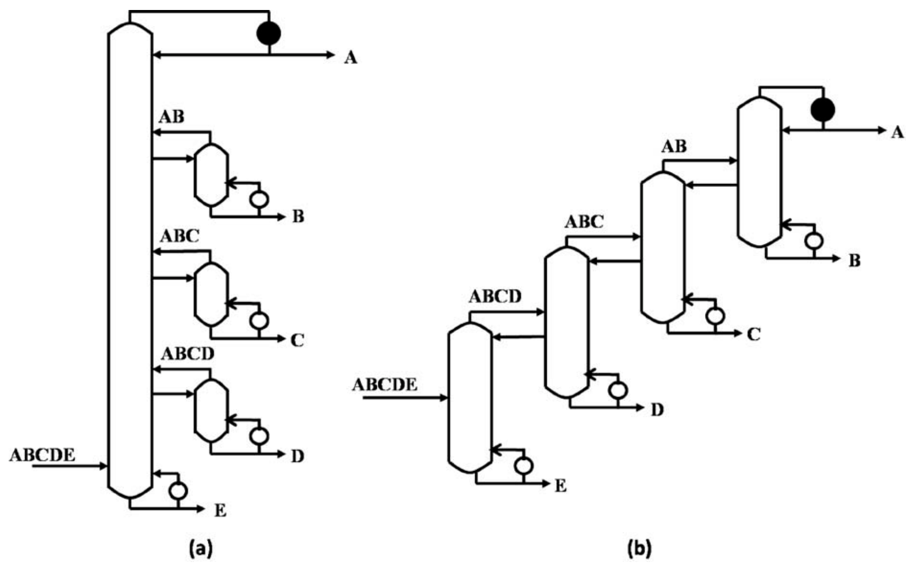

Figure 12. (a) Conventional configuration for petroleum crude distillation and (b) rearrangement to give equivalent indirect split configuration.

After estimating the minimum total vapor duty for each configuration by solving the corresponding NLP, we obtain a rank-list of all the distillation configurations in order of increasing vapor duty. We can thus choose the configuration that has the least vapor duty or examine in further detail all configurations that lie within, say, y% of this configuration with the least duty, where "y" can be any number chosen to meet a certain objective. Thus, we will then have solved the problem of finding the best distillation sequence(s) for the n-component separation.

Suppose we decide to discard all the basic configurations and their analogs with partial thermal coupling from the search space. Now we have a search space containing only the completely thermally coupled analogs of the basic configurations. Searching through this smaller space will give us the completely thermally coupled configuration with the lowest total vapor duty. However, it is incorrect to assume that the corresponding basic configuration will also have the lowest total vapor duty compared to all other basic configurations. The reason for this conclusion is that, even though it has been known that the fully thermally coupled configuration (Petlyuk-type configuration) always has lowest minimum total vapor duty for any n-component feed,[6,7] we have observed that the corresponding basic configuration can be outperformed by other basic configurations.[14] Therefore, we should be careful while devising strategies to reduce the search space as there is not a straightforward one-to-one correspondence between the rank lists of the basic configurations and their thermally coupled analogs.

In our approach, we do not solve the problem as an MINLP. In an MINLP approach, the optimization attempts to return the single best configuration that minimizes our objective function subject to all the constraints. However, many topologically different sequences have very similar minimum total vapor duty values, creating difficulties in convergence of the optimization method to a globally minimum sequence. To avoid getting trapped in local minima, we propose total enumeration of the search space, followed by solving an NLP for each configuration. We thus first propose solution of the IP by total enumeration using the matrix method, followed by solving an NLP for each feasible solution of the IP. This approach is highly amenable to parallel computing once the IP is solved, as the NLP of a basic configuration can be solved independently of all the NLPs of the other basic configurations.

An example

To illustrate the efficacy of our method, we apply it to the problem of petroleum crude distillation. Total world petroleum production in 2007 was ~81.5 million bbl of crude oil/day,[18] and petroleum crude distillation roughly consumes energy at the equivalent of 2% of the crude processed.[19] This works out to nearly 1.6 million bbl/day, thereby placing crude distillation among some of the most energy-intensive processes.

Petroleum crude ABCDE is typically distilled into the following five fractions: naphtha (A), kerosene (B), diesel (C), gas oil (D), and residue (E). Although refineries operating around the world process different crude oils, they usually still have the same distillation configuration that has been in use for more than 75 years.[19] This configuration consists of a main column with side strippers (Figure 12a). Appropriate rearrangement of the distillation column sections of the configuration shown in Figure 12a shows that it is equivalent to the indirect split sequence shown in Figure 12b.[20]

The distillation column sections in thermally coupled configurations can be rearranged to improve the intercolumn transfer of vapor streams.[21,22] All the rearranged sequences will operate with similar, if not the same, energy consumption. Even though it is possible to draw all possible rearranged sequences and include them in the search space, we do not think it is essential. For each basic configuration, only one set of thermally coupled configurations ranging from partial to complete thermal coupling is included in the optimization of total vapor duty. Only after a thermally coupled configuration has been identified to be of interest, the rearranged versions are explored for operability, manufacturability, etc.

As we choose to minimize the total vapor duty, the only information we need to provide is as follows: (1) the number of components in the feed, which is used in the matrix method to generate all feasible distillation configurations, (2) the feed composition and thermal quality, which is used in solving Underwood's equations, and (3) the relative volatilities of the components, which are also used in solving Underwood's equations. The relative volatility of each A, B, C, and D with respect to E is taken to be 45.3, 14.4, 4.7, and 2.0, respectively. A mixture containing 46.1% A, 19.5% B, 7.3% C, 11.4% D, and 15.7% E as mole fractions is used to represent a light petroleum crude, whereas a mixture containing 14.4% A, 9.3% B, 10.1% C, 3.9% D, and 62.3% E is used to represent a heavy petroleum crude. In both cases, 90% of E in the feed is taken to be liquid. The remainder of the feed is taken to be vapor. Hence, we have a two-phase feed to the distillation configuration. This completes the description of the example problem.

Before we present our simulation results, it is worth noting that the indirect split configuration of Figure 12a is used in a refinery with a number of modifications. Liquid pump-around loops are used to provide intermediate cooling to the main column. This reduces the load on the main condenser. Also, in place of reboilers, steam at different pressures is injected at the bottoms of the side strippers. Such differences certainly impact the operation of a configuration. However, in our simulations, we calculate the minimum total vapor duty for each configuration assuming conventional reboilers and condensers. It is on this basis that all the configurations are compared with each other.

As we assume petroleum crude to be a five-component mixture, we obtain 203 feasible basic distillation configurations (Table 1), and 5925 additional options that range from partial to complete thermal coupling including the currently used configuration in Figure 12b, giving a total of 6128 configurations. As will be seen later, these generally differ significantly in their vapor duties.

Case I—No Transfer of a Residue (E)-Containing Stream Between Distillation Columns. The heaviest component, residue (E), can create fouling type of problems in the distillation sequence. Therefore, it may not be desired to process E throughout the sequence; it may be necessary to separate it out early. This fixes the first split in the sequence to ABCDE – ABCD/E. To ensure this in our method, we fix the elements corresponding to streams BCDE, CDE, and DE to have values of 0 in the matrix, so that there are no residue (E)-containing streams being transferred between columns.

Now we obtain 18 basic configurations (instead of 203) to distill ABCD further, as it is now a four-component feed problem. Once again, note that the indirect split sequence without any thermal coupling is still just 1 of these 18 basic configurations. We want to analyze how the remaining 17 of these 18 configurations perform. Also, these 18 configurations consist of five sharp-split configurations and 13 non-sharp-split configurations.

In Table 2, we provide the heat duty for the indirect split basic configuration and its completely thermally coupled analog, in units of "minimum total molar vapor flow/molar flow of feed." Among the five sharp-split configurations, the

Table 2. Total Heat Duty of the Indirect Split Sequences in Units of "Minimum Total Molar Vapor Flow/Molar Flow of Feed" for Petroleum Crude Distillation

	Light Crude	Heavy Crude
Indirect split basic configuration	0.9996	1.0363
Indirect split configuration with complete thermal coupling	0.6813	0.8457

indirect configuration is found to have the lowest vapor duty, and this is also true for the thermally coupled versions of these configurations. This reaffirms the choice of the indirect split configuration for all these years. In fact, without any thermal coupling, none of the 17 basic configurations has lower minimum total vapor duty than the completely thermally coupled indirect split configuration—for both light and heavy crude. However, completely thermally coupled derivatives of 8 of 17 configurations have lower minimum total vapor duty for light crude distillation; and six of these eight configurations have lower minimum total vapor duty for heavy crude distillation.[23] Three of these eight configurations with their reductions in minimum total vapor duty for both light and heavy crude distillations are shown in Figure 13, the last of which shows the maximum reduction. We observe that the other two configurations also have potential to significantly reduce total vapor duty while not being drastically different from the currently used conventional configuration and could be good candidates for retrofitting in an existing plant.

To identify such easy-to-retrofit configurations, we force some of the submixture elements of the matrix to take values of either 0 or 1, depending on the nature of the similarities we want to enforce.

In a typical refinery processing 250,000 bbl/day, the proposed schemes could potentially save ~160,000 bbl/year for heavy crude distillation and ~354,000 bbl/year for light crude distillation. For a crude price of $ 50/bbl, it means a savings of ~8 million to 18 million dollars/year per refinery. Of course, if successfully implemented in all the refineries that collectively process 81.5 million bbl/day, the annual savings could potentially run in billions of dollars with worldwide crude oil savings in the range of 52–115 million bbl/year.

Case II—Optimization in the Complete Search Space. If we allow transfers of E-containing streams between distillation columns of the sequence, then instead of only 17, all the remaining 185 basic configurations and their thermally coupled derivatives become available for consideration.

Interestingly, when compared with the completely thermally coupled indirect split configuration, 63 of the remaining 185 basic configurations have lower minimum total vapor duty for the light crude distillation (up to ~31.7%), and 1 of these 63 basic configurations has lower minimum total vapor duty for the heavy crude distillation (by ~1.5%). Thus, it is possible to find basic configurations, i.e., configurations without any thermal coupling, which could perform significantly better than the currently used configuration with complete thermal coupling.

If we also allow thermal coupling in the 185 configurations, we can identify many more configurations that have

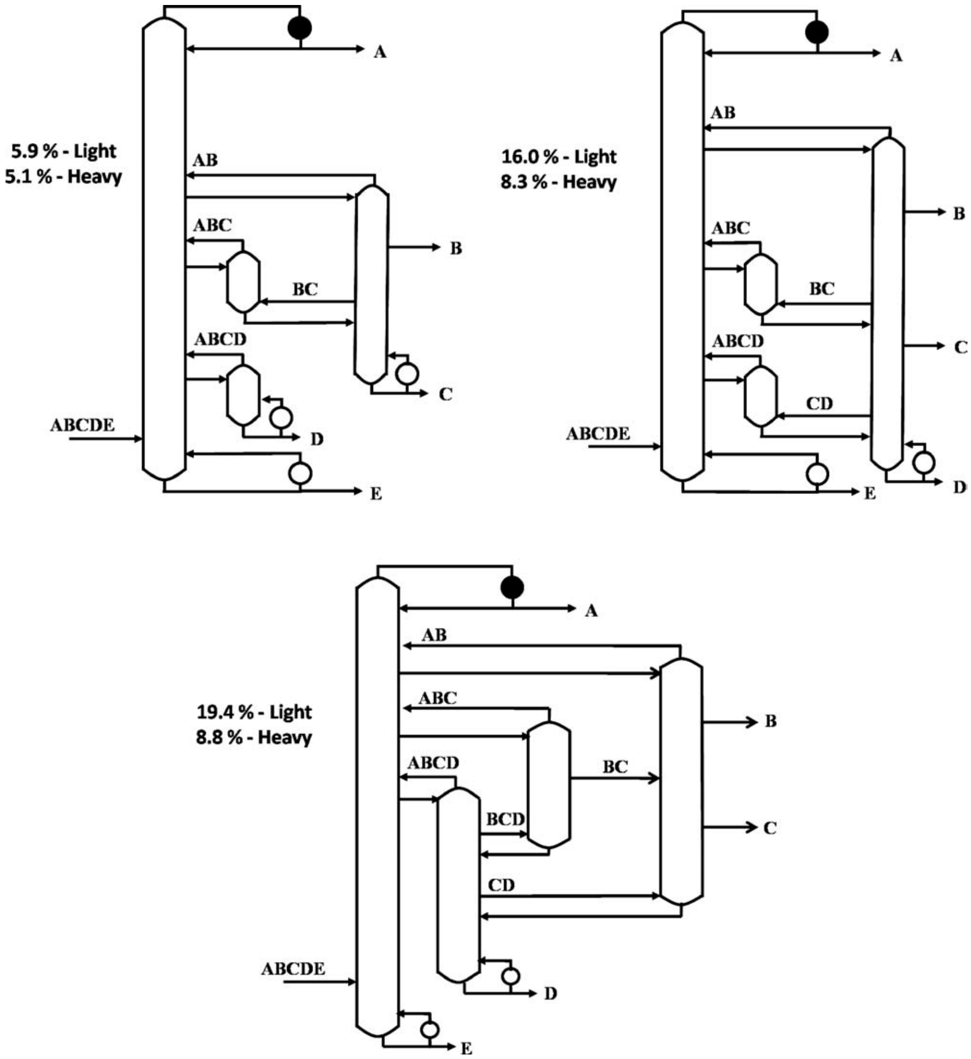

Figure 13. Some easy-to-retrofit configurations for petroleum crude distillation, with their reductions in total vapor duty requirement for light and heavy crude distillation.

lower heat demand.[24] In such a case, we observe that the best new configurations with complete thermal coupling reduce total vapor duty by ~48.2% for the light crude distillation and by ~17.1% for the heavy crude distillation. These numbers were 19.4 and 8.8%, respectively, in Case I when we constrained the problem with no transfer of residue (E)-containing streams between columns.

Hence, it is worth allowing some transfers of E-containing submixtures, but it is not necessary to include all such transfers for the maximum benefit. For instance, we present a configuration in Figure 14a with two (of the maximum possible three) E-containing submixture transfers and seven (of the maximum possible nine) total submixture transfers. This configuration shows 48.2% reduction in the minimum total vapor duty for the light crude distillation and 17.1% reduction for the heavy crude distillation. This is the maximum savings, as seen from the savings of the fully thermally coupled configuration for a five-component feed mixture. Hence, adding more E-containing submixture transfers or more total transfers provides no further improvement.

Similarly, for each of these derived configurations, it is often not needed to go all the way to complete thermal coupling, because partial thermal coupling can sometimes provide all the improvement; in such cases, subsequent introduction of thermal coupling provides no further significant improvement. This is seen from the configuration presented in Figure 14b, which has the same total vapor duty as the one in Figure 14a.

Once again, we observe that these new configurations can potentially reduce the vapor duty requirement by significant amounts. Again, in a typical refinery processing 250,000 bbl/day, the proposed schemes could potentially save ~310,000 bbl/year for heavy crude distillation and ~880,000 bbl/year for light crude distillation. For a crude price of $ 50/bbl, it means a savings of ~15 million to 44 million dollars/year per refinery. Again, if successfully implemented in all the

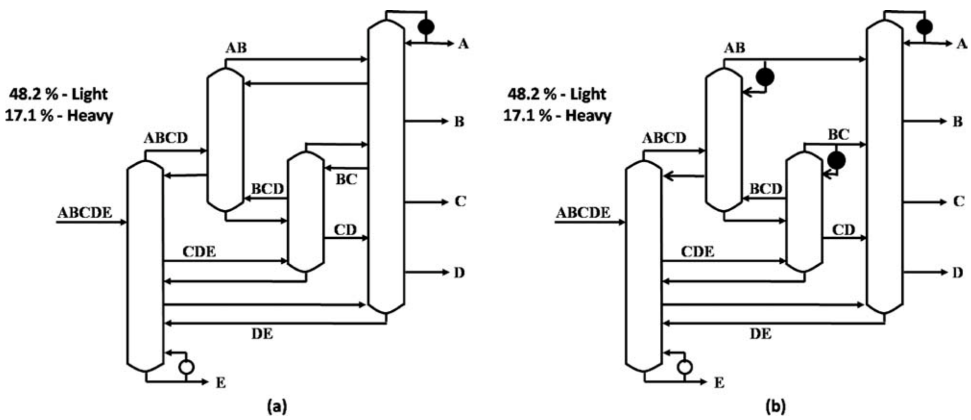

Figure 14. (a) One of the completely thermally coupled configurations with maximum reduction in heat duty for light as well as heavy crude distillation, and (b) one of its partially thermally coupled analogs with the same energy benefit.

refineries that collectively process 81.5 million bbl/day, the annual savings could potentially run into billions of dollars with worldwide crude oil savings in the range of 100–290 million bbl/year.

Halbouty[25] defines a giant oil field as one that has produced or will produce at least 100 million bbl of oil. Therefore, these numbers potentially translate to discovery of a new giant heavy oil field every year or to discovery of a new giant light oil field every 4 months.

The current distillation configuration operates with a number of heat integrations in a refinery that are fairly important in reducing the overall heat consumption within a refinery. Configurations that are more amenable to heat integration with hot or cold streams available elsewhere in the refinery could well prove to be more attractive in spite of a higher total vapor duty for a standalone operation. We do not account for such an effect in our simulation. It is hoped that the availability of a large number of configurations with much lower standalone vapor duties will provide us with an array of distillation configurations that are also likely to have similar heat integration advantages as the current indirect split configuration.

Conclusions

We introduced a novel and easy-to-use "matrix" method that generates a complete search space for multicomponent basic distillation configurations. We then easily enumerated the additional configurations with thermal coupling. The method is easily generalizable for any number of components in the feed. Importantly, the use of the method is not limited to distillation network synthesis alone.

Another useful aspect of the method is that we can identify attractive configurations that can be built in a grassroots plant, or we can also identify configurations that are easy-to-retrofit to an existing configuration.

Application of the method to the energy-intensive problem of petroleum crude distillation shows that there is tremendous scope for improvement.

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