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A Synthesis Method for Multicomponent Distillation Sequences with Fewer Columns

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An easy-to-use matrix-based method for the systematic synthesis of distillation configurations using less than n-1 columns to separate any zeotropic n-component feed into n product streams is described. The method is easily extended to obtain additional thermally coupled configurations. The only information needed to generate the configurations is the number of components in the feed, or equivalently, the number of distinct composition final product streams. We have successfully enumerated configurations for feeds containing up to eight components. This has resulted in a large number of hitherto unknown configurations even for four-component separations. Some of the novel configurations generated using the method have substantially lower heat duty than the previously known fewer column configurations for a four-component feed separation. Therefore, it is essential to include these novel configurations in the search space to find the optimal distillation configuration with fewer columns for a given application. © 2011 American Institute of Chemical Engineers *AIChE J*, 58: 2479–2494, 2012
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Introduction

Distillation is the most common separation technique accounting for around 95% of all separations in the chemical process industries.[1] Most of the industrial mixtures contain more than two components to be separated into a corresponding number of product streams, each enriched in one of the components. Such a multicomponent separation task cannot usually be achieved efficiently in a single distillation column. This necessitates sequencing of distillation columns to achieve the separation of multicomponent mixtures.

A sequence of distillation columns is referred to as a distillation configuration. Depending on the number of distillation columns in a configuration, a distillation configuration to separate an *n*-component feed into *n* product streams can be classified as a more than $n-1$ columns configuration, an exactly $n-1$ columns configuration or a less than $n-1$ columns configuration. We refer to a more than $n-1$ columns configuration as a *plus-column* configuration, an exactly $n-1$ columns configuration as a *regular-column* configuration and a less than $n-1$ columns configuration as a *subcolumn* configuration.

Some regular-column configurations are characterized by the following three features: (1) mixtures containing the same group of components are transferred only once from one distillation column to another, (2) each final product stream is produced from a single location in the configuration, and (3) the main feed stream and all the transfer streams are split into exactly two product streams. These configurations are referred to as basic distillation configurations.[2] On the other hand, plus-column configurations that violate the first two distinguishing features and satisfy the third distinguishing feature of basic configurations have been referred to as nonbasic configurations.[2] Nonbasic configurations tend to have more operating cost than the optimal basic configuration.[3] Moreover, since nonbasic configurations have additional distillation columns, they also tend to have more capital cost than basic configurations. Such configurations are eliminated from the search space, significantly reducing the size of the search space.[3]

Most of the systematic synthesis approaches in literature have focused on generating basic distillation configurations. Some researchers have described superstructure flowsheets that contain feasible distillation configurations.[4,5] An alternate to the superstructure approach is to mathematically synthesize individual distillation configurations. Agrawal[2] presented the first systematic rule-based algorithm to generate the set consisting of only the basic configurations. Caballero and Grossmann[6,7] and Giridhar and Agrawal[8] built on the observations proposed by Agrawal[2,5] and presented alternate mathematical formulations to generate the complete search space of basic distillation configurations. In both these formulations, the computational effort required to generate the complete set of configurations increases rapidly with the number of components in the feed. Ivakpour and Kasiri[9] proposed a formulation in which distillation configurations are represented mathematically as upper triangular matrices. In this formulation, distillation configurations are generated through all possible heat exchanger assignments as suggested earlier by Agrawal.[2] This approach of Ivakpour and Kasiri[9]

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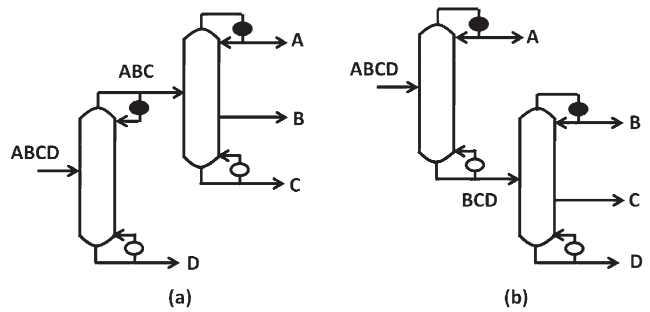

Figure 1. Examples of distillation configurations that use two distillation columns to separate a four-component mixture.[11,12]

is also computationally intensive, especially for larger number of components in the feed. Shah and Agrawal[10] independently proposed an alternate easy-to-use mathematical matrix-based formulation in which distillation configurations are generated by exploring all possible instances of the presence or absence of transfer streams. The matrix approach of Shah and Agrawal[10] is computationally efficient as reflected in the fact that for the first time even for an eight component mixture, the exact number of basic configurations and their thermally coupled analogs ranging in millions and billions could be easily elucidated.

We extend the matrix approach of Shah and Agrawal[10] for the synthesis of subcolumn configurations that can separate any n-component zeotropic mixture into n-product streams. Subcolumn distillation configurations use fewer distillation columns than basic configurations and thus tend to have less capital cost. However, subcolumn configurations usually have higher operating costs than the optimal basic configuration due to increased heat duty, especially for making high purity products. Therefore, for feed mixtures where the heat duties of subcolumn configurations are comparable to or lower than that of optimal basic configurations, it would be preferable to build the configurations with fewer columns.

Figure 1 shows some known subcolumn distillation configurations for separating a four-component feed mixture.[11,12] In these configurations, the volatilities of components decrease in alphabetical order. Therefore, *A* is the most volatile component followed by *B* and so on. Reboilers are represented by nonfilled circles and condensers are represented by filled circles. Furthermore, in streams such as *ABC*, it is not implied that other components such as *D* are completely absent. They may be present in acceptably small amounts. We refer to streams that are transferred between distillation columns as submixtures. For instance, in Figure 1a, stream *ABC* is a submixture and in Figure 1b, stream *BCD* is a submixture.

A split is defined as the act of separating a feed mixture into product streams of different compositions. The split of a feed stream can be classified as either a sharp split or a nonsharp (sloppy) split. In sharp splits, the product streams obtained by splitting a feed stream have no, or at most acceptably small levels of, overlapping components. In nonsharp splits, overlapping of components in significant amounts among the separated streams is allowed. For instance, the split *ABCD* – *AB*/*CD* is a sharp split as the product streams have no (or acceptably small levels of) overlapping components. On the other hand, the split *ABCD* – *ABC*/*BCD* is a nonsharp split with overlapping components being component *B* and component *C*. In sharp split configurations, each and every split is sharp and in nonsharp split configurations, at least one split is nonsharp. Therefore both the configurations of Figure 1 are sharp split sequences.

If a feed is split into more than two product streams, at least one of the product streams is a side-stream product. We refer to these splits as side-splits. For instance, splits *ABC* – *A*/*B*/*C* (Figure 1a) and *BCD* – *B*/*C*/*D* (Figure 1b) are side-splits with streams *B* and *C* as side-stream products of the two splits respectively.

Some other subcolumn configurations are due to Brugma.[13] He invented a subcolumn configuration that uses two columns to separate a four-component feed with liquid transfers between columns. The configuration shown in Figure 2a is identical to the configuration invented by Brugma,[13] except that the liquid transfer of stream *AB* is replaced by vapor transfer since such a vapor transfer would reduce the total heat duty of the configuration.

Thermal coupling between distillation columns is known to reduce the heat duty of distillation configurations.[14–16] A thermally coupled configuration can have partial or complete thermal coupling.[2] If all reboilers and condensers associated with submixtures in a configuration are replaced with thermal coupling links, then the derived thermally coupled configuration is said to have complete thermal coupling. If at least one of the reboilers or condensers associated with submixtures is retained in a thermally coupled configuration, then the configuration is referred to as having partial thermal coupling. Cahn and Di Miceli[17] introduced complete thermal coupling (Figure 2b) in the configuration in Figure 2a. We demonstrate that our synthesis method accounts for these known subcolumn distillation configurations, while also providing a large number of hitherto unknown configurations.

For separating a three-component feed into three product streams, Wright[14] invented a divided wall column which uses a single column shell with a vertical partition to combine two thermally coupled columns. Kaibel[18] extended the application of such columns with vertical partitions to feeds with four or more number of components (Figure 2c). The configurations of Figures 2b, c are equivalent from the perspective of energy consumption. Divided wall columns thus essentially combine two or more thermally coupled columns into a single shell.[19,20] In this article, we limit our synthesis framework to thermally coupled subcolumn configurations with multiple columns. If needed, the equivalent divided wall versions of these configurations can be easily generated.

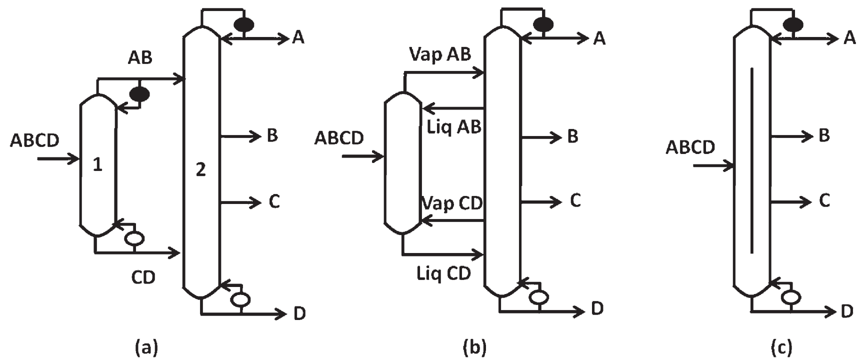

Figure 2. Some known configurations using less than $n-1$ columns (a) Brugma configuration[13] (b) Cahn and Di Miceli configuration[17] (c) Kaibel configuration.[18]

The potential of the subcolumn configurations to provide economic benefits has been demonstrated for ternary mixtures.[21,22] Kim and Wankat[11] were the first to compare different sharp split subcolumn configurations for separating a four-component mixture. Errico et al.[12] provided a systematic algorithm to generate sharp split subcolumn configurations with and without thermal coupling for any number of components in the feed. Until recently, there has been no attempt to include nonsharp split subcolumn configurations in the search space. Ivakpour and Kasiri[9] proposed elimination of distillation columns which do not produce final products in the basic configurations synthesized by their matrix formulation as a means to obtain both sharp as well as nonsharp split subcolumn configurations. This strategy does not always lead to physically feasible configurations. There is clearly a need for a method that will produce the feasible search space consisting of both sharp and nonsharp split subcolumn configurations.

Our goal is to develop a simple and generalizable method to systematically generate the search space of subcolumn configurations for any number of components in the feed. We extend the matrix-based approach of Shah and Agrawal[10] by adding two additional steps to include the set of subcolumn configurations in the existing feasible search space of basic distillation configurations.

Method to Generate the Search Space of Distillation Configurations

The matrix-based method of Shah and Agrawal[10] is a six-step method to generate the complete search space of basic regular-column configurations. We now describe an eight-step matrix method that encompasses the earlier six-step matrix-based method and extends it by adding two additional Steps 7 and 8 to generate subcolumn configurations with less than $n-1$ columns.

In the matrix method, the n-component feed mixture comprises of components A, B, C, etc. wherein component A is the most volatile component in the feed and the volatility decreases sequentially in alphabetical order. In the distillation of any feed stream, it is assumed that any product stream has at least one less component than the feed stream. For instance, a feed stream CDE can only produce two or more of the streams CD, DE, C, D and E as its product streams. Also, our method generates configurations where a final product stream enriched in one of the components is produced at only one location and submixture streams containing the same group of components are also transferred only once from one distillation column to another. These are two features that the generated subcolumn configurations will have in common with the basic distillation configurations studied extensively in the literature.

Detailed description of Steps 1 – 6 of the matrix method can be found in the earlier work;[10] here we will describe them briefly with emphasis on the new Steps 7 and 8.

Step 1

The first step of our method is to identify the predominant number of components that need to be separated into product streams from a given feed stream. This is the only information needed to generate the search space and is available from the problem definition. For instance, if we are given a four-component feed mixture to be separated into four products, then we identify the predominant number of components in the feed as $n = 4$.

Also, when a feed contains more than n components, but some of the components of close volatilities are lumped together to produce only n product streams, each with its distinct composition, then for the purpose of this problem we treat the feed mixture as an n-component mixture. In such a case, one or more of A, B, C, etc. may represent not single components but lumped mixtures.

Step 2

In the second step of our method, we generate an $n \times n$ upper triangular matrix. The upper triangular matrix elements correspond to unique streams whereas all the matrix elements below the diagonal are assigned a value of zero. For our four-component example, this step implies generation of a 4×4 upper triangular matrix as shown in Figure 3.

The arrangement of the streams in this manner within a matrix allows use of physical insights. If we choose any stream in the matrix, except the final products, and move horizontally to the right, all streams that we encounter on the path are candidate top distillation products. Similarly, all streams that we encounter on the diagonal path to the right are candidate bottom distillation products of the chosen stream. Thus, the structure of the $n \times n$ upper triangular matrix enables us to easily determine the top and bottom products for any given stream.

Step 3

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

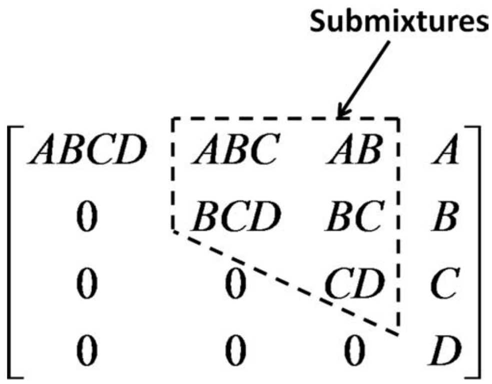

Figure 3. Matrix for four-component feed mixture.

The $n \times n$ matrix contains $n(n+1)/2$ elements in the upper triangular portion. The main feed stream is located in the first column of the matrix and the n final product streams are located in the last column of the matrix. The remaining streams in the intermediate columns of the matrix are the d submixtures where $d = n(n+1)/2-1-n$. For our example of $n = 4$, there are $d = 4(4+1)/2-1-4 = 5$ submixtures, namely streams *ABC*, *BCD*, *AB*, *BC* and *CD* (Figure 3).

In our matrix approach, we utilize the fact that the presence or absence of submixture streams describes a distillation configuration. For instance, both the distillation configurations in Figure 1 have the main feed stream and the four final product streams. However, Figure 1a is defined by the presence of submixture *ABC* and the absence of submixtures *BCD*, *AB*, *BC* and *CD*; while Figure 1b is defined by the presence of submixture *BCD* and the absence of the other submixtures.

Step 4

In this step, we create all the possible matrices that indicate the presence or absence of submixtures in a configuration. We assign binary integer variables, 0 or 1, to each element in the upper triangular portion of the matrix. A matrix element with a value of 1 indicates that the corresponding stream is present in the configuration and an element with a value of 0 implies the absence of the corresponding stream in the configuration.

In a distillation configuration, the main feed stream and the n final product streams will always be present and their corresponding locations in the matrix (i.e., the (1,1) element and all the elements in the nth column of the matrix) will always take a value of 1. However, the d submixtures may or may not be present in a configuration and each of these can take values of either 0 or 1. Thus, we can systematically generate 2^d candidate matrices with all the possible 0 – 1 combinations of the d submixtures. For our four-component feed mixture example in Figure 3, we have $d = 5$ giving us $2^5 = 32$ candidate matrices shown in Figure 4.

Steps 5 and 6

Steps 5 and 6 of the method describe the set of rules to identify and then draw the feasible basic regular-column distillation configurations from the set of candidate matrices generated in Step 4. The procedure for these steps is discussed in detail by Shah and Agrawal[10] and we will not repeat them in this article. For the synthesis of the subcolumn configurations alone, Steps 5 and 6 are skipped and after Step 4 one directly goes to Steps 7 and 8.

Step 7

In this step, we deduce connectivity information required to identify configurations with less than $n-1$ columns from the 2^d matrices generated in Step 4. The outline of the procedure to do this is: (a) Replace the 1's in the candidate matrix by the corresponding streams, (b) Identify all the splits in the matrix, and (c) Assign distillation column numbers to the identified splits

Consider the matrix shown in Figure 5a. This matrix corresponds to candidate matrix (xxv) in Figure 4. After replacing the 1's in the matrix by the corresponding streams, we obtain the matrix shown in Figure 5b.

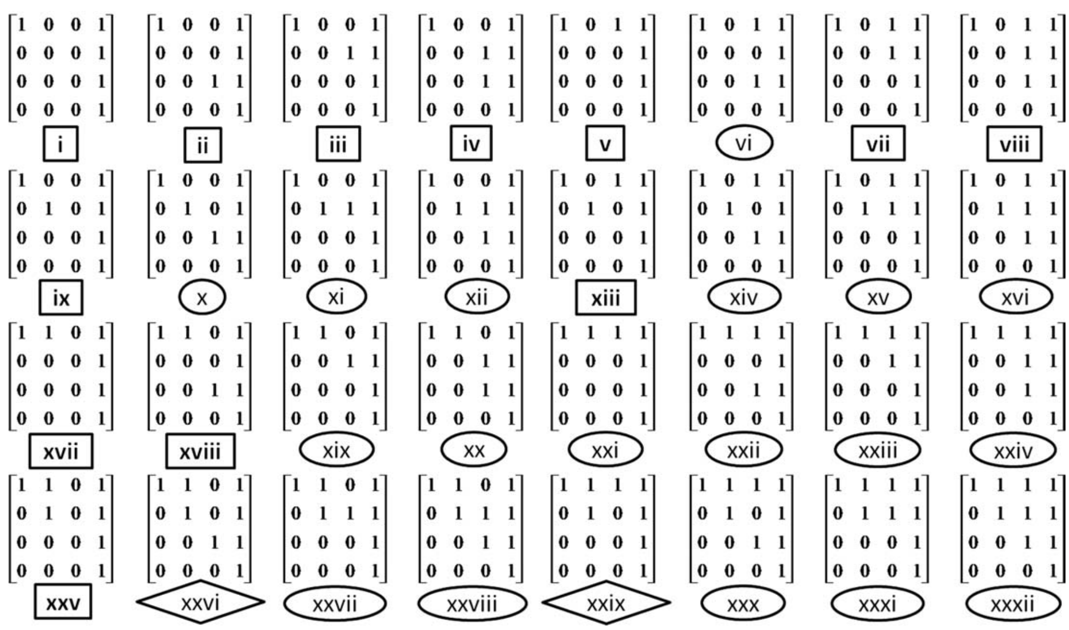

Figure 4. Candidate matrices for a four-component feed mixture (Rectangles: Correspond to subcolumn configurations; Ovals: Correspond to regular-column configurations; Diamonds: Correspond to infeasible configurations).

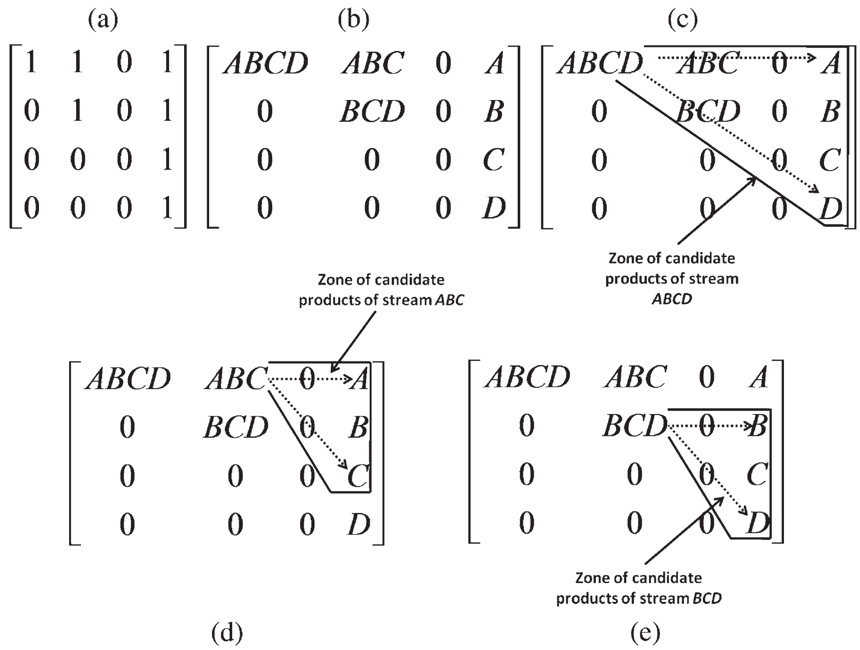

Figure 5. (a) A candidate four-component 0 – 1 matrix (Figure 4: xxv), (b) replacing 1's by appropriate streams (c) Illustration of zone containing candidate products of stream *ABCD*, (d) Zone containing candidate products of stream *ABC*, (e) zone containing candidate products of stream *BCD*.

Next, we identify all the splits in the matrix. As discussed in Step 2 of the method, if we choose any nonfinal product stream in the matrix, all streams that we encounter on the path horizontally to the right of the chosen stream are candidate top distillation products and all streams that we encounter on the diagonal path to the right are candidate bottom distillation products. This interpretation of the matrix was sufficient to determine the set of feasible basic regular-column configurations.

However, to generate the subcolumn configurations, we cannot limit our interpretation to simply the horizontal and diagonal paths from a nonfinal product stream in the matrix. We have to consider additional streams in the matrix that are not on the horizontal or the diagonal paths emanating from the stream being distilled. For this purpose, we first define a zone which is bounded by the horizontal, the diagonal and the final product column lines in the matrix. Thus in Figure 5c, when stream *ABCD* is to be distilled, the zone associated with it is defined by the lines *ABCD* to *A*, *ABCD* to *D* and the vertical column containing the final products. All the streams within the zone are considered as candidate products of the feed stream under consideration. In Figure 5c, it means that streams *ABC* and *A* are candidate top product streams, streams *BCD* and *D* are candidate bottom product streams, while the additional streams *B* and *C* contained within the zone are candidate side-streams.

We eliminate some of the candidate product streams from the zone to enforce two of the distinguishing features of basic configurations, whereby any stream is transferred (or produced in case of the final products) only once in the distillation configuration. Each candidate product stream is examined sequentially starting from the submixture containing the highest number of components. Thus for the example in Figure 5, from the candidate top products of stream *ABCD* (i.e., streams *ABC* and *A*), stream *ABC* is the first stream (first "1" in Figure 5a) encountered on the horizontal path to the right of feed stream *ABCD*; hence it is identified as the top product of feed *ABCD*. Furthermore, from the product zone of stream *ABC* (Figure 5d), we see that streams *A, B*, and *C* form the list of candidate products of stream *ABC*. The assumption of a stream being transferred (or produced in case of final product streams) only once in the configuration implies that the candidate products of stream *ABC* should not be direct candidate products of stream *ABCD*. Else, the common candidate products of streams *ABCD* and *ABC* will be produced from more than one distillation column in the configuration, i.e., once from the column with stream *ABCD* as the feed and then from the column with stream *ABC* as the feed. Through this analysis we can conclude that streams *A*, *B,* and *C*, which are candidate products of stream *ABC*, should not be candidate products of stream *ABCD*. Similar analysis for the bottom product from the distillation column leads to stream *BCD* as the only product stream below the *ABCD* feed (Figure 5e). This gives us the first split as *ABCD* – *ABC*/*BCD*. A similar procedure is followed to identify all the splits for the remaining streams in the matrix to yield the associated configuration.

For instance, from the zone of stream *ABC* in Figure 5d, we can identify the top product as stream *A* and the bottom product as stream *C*. In the zone of candidate products of stream *ABC*, there is no stream that can produce stream *B* other than stream *ABC* itself. Therefore stream *B* is identified as a side-stream product of feed *ABC*. Thus, we obtain the second split as side-split, *ABC* – *A*/*B*/*C*. Similarly for stream *BCD* in Figure 5e, we obtain the split as *BCD* – *B*/*C*/*D*. Note that for each of these splits we have just one side-stream product. For splits involving larger number of components, the number of side-stream products from a feed stream could be more than one. It is convenient to think of

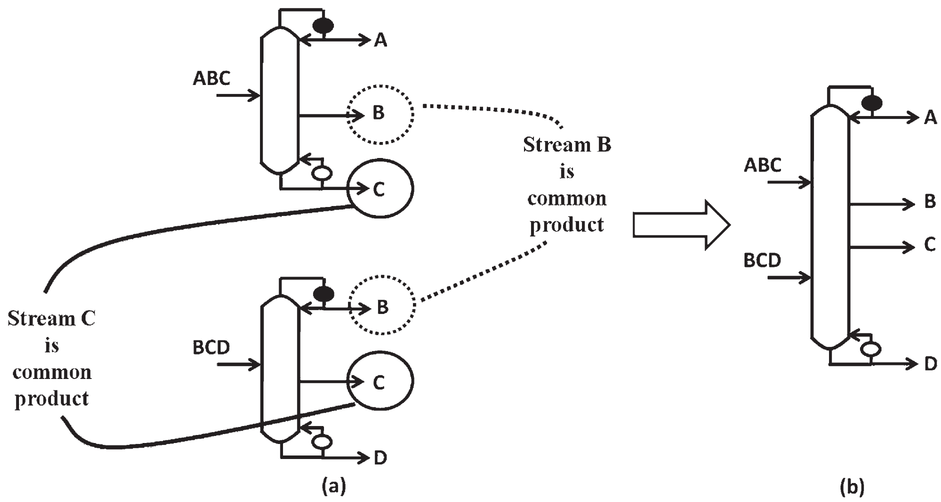

Figure 6. An illustration of grouping of splits producing common streams (a) Before grouping (b) After grouping.

splits as pseudo-distillation columns with a reboiler associated with the top product and a condenser associated with the bottom product.

To assign a distillation column number to each split, splits that produce common streams must be placed in the same distillation column. This ensures that any stream is transferred (or produced in case of final product streams) only once in the distillation sequence. The streams that are common to splits are withdrawn as side-streams and their associated reboilers and condensers, if any, are eliminated. For the candidate matrix of Figure 5a, the splits producing common streams are placed in the same distillation column as shown in Figure 6. In Figure 6a, split *ABC – A/B/C* produces stream *B* as a side-stream and stream *C* from the reboiler; while split *BCD – B/C/D* produces the common streams *B* and *C* from the condenser and as a side-stream, respectively. In this case, the condenser associated with stream *B* as well as reboiler associated with stream *C* are eliminated and both the splits are grouped in to one distillation column thus producing streams *B* and *C* as side-stream products (Figure 6b).

After grouping the splits making common streams, distillation column numbers are assigned to all the splits obtained from the matrix and a table containing this information is created. For example, for the matrix in Figure 5a, all the splits are summarized in Table 1. In this table, the feed split *ABCD – ABC/BCD* is assigned to distillation column "1". The remaining splits, *ABC – A/B/C* and *BCD – B/C/D*, must belong to the same column, say, distillation column "2". This completes the connectivity table having information of all splits and corresponding column numbers for the candidate matrix in Figure 5a.

We can similarly obtain the connectivity information of all splits in the distillation configuration for every candidate 0 – 1 matrix generated in Step 4.

Step 8

In this step, we eliminate matrices that do not lead to feasible subcolumn configurations from the 2^d candidates. To do this, we apply three checks to the connectivity tables obtained from each of the 2^d candidate configurations. If any of the checks is not satisfied, then the corresponding candidate matrix is eliminated from the search space of subcolumn configurations. The checks below are numbered in continuation with Checks 1, 2, and 3 described by Shah and Agrawal.[10]

Check 4. Ensure that at least one feed stream in the configuration produces a side-stream product, or in other words, the split of at least one feed stream should produce more than two product streams.

Check 5. Disallow configurations containing columns having the same stream as feed and product.

Check 6. Disallow configurations having two submixture streams transferred in opposite directions between two distillation columns wherein one of the submixtures is a product of the other submixture.

Let us first consider examples that illustrate each of the checks. Applying Step 7 to the two candidate matrices (xxxii) and (xxvi) of Figure 4 and to the five-component candidate matrix of Figure 7a gives us the connectivity information provided in Tables 2, 3, and 4, respectively.

In the connectivity table obtained from matrix (xxxii) of Figure 4 (Table 2), each feed stream produces exactly two product streams. Since none of the splits in the configuration is a side-split, Check 4 is violated. The presence of at least one side-split is a characteristic feature of subcolumn configurations. Hence the configuration obtained from matrix (xxxii) is not a feasible candidate for configurations with less than $n-1$ distillation columns and is eliminated from the search space of subcolumn configurations.

The splits in Table 3 for matrix (xxvi) of Figure 4 are depicted as pseudo-distillation columns in Figure 8. Splits *ABC – A/B/C* and *BCD – B/CD* belong to the same distillation column because they make the common product stream *B*, which will be produced as a side-stream. Also, since split *CD – C/D* makes the product stream *C* in common with split

Table 1. Splits in Distillation Configuration Corresponding to Matrix of Figure 5a

Split Number	Feed Stream	Product Streams: Top	Bottom	Side-Stream	Distillation Column Number
1	*ABCD*	*ABC*	*BCD*	–	1
2	*ABC*	*A*	*C*	*B*	2
3	*BCD*	*B*	*D*	*C*	2

(a)

$$\begin{bmatrix} 1 & 1 & 1 & 0 & 1 \\ 0 & 0 & 0 & 1 & 1 \\ 0 & 0 & 1 & 0 & 1 \\ 0 & 0 & 0 & 0 & 1 \\ 0 & 0 & 0 & 0 & 1 \end{bmatrix}$$

(b)

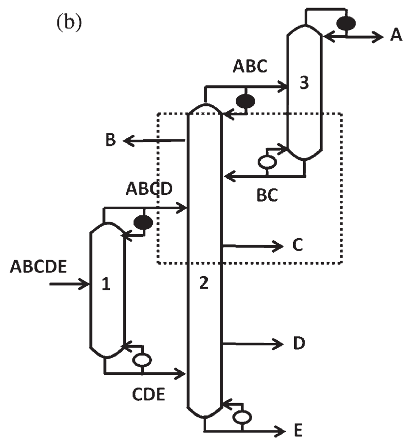

Figure 7. (a) A candidate five-component 0 – 1 matrix, (b) subcolumn configuration obtained from the matrix of (a).

ABC – *A*/*B*/*C*, split *CD* – *C*/*D* also belongs to the same distillation column. Therefore, splits *ABC* – *A*/*B*/*C*, *BCD* – *B*/*CD* and *CD* – *C*/*D* belong to the same distillation column "2" (Figure 8). This distillation column "2" has stream *CD* as its feed stream and also as its product stream which violates Check 5 and makes the configuration physically infeasible. Thus, the configuration obtained from the matrix (xxvi) of Figure 4 is eliminated from the search space even though it does not violate Check 4.

The distillation configuration corresponding to the five-component candidate matrix in Figure 7a and the connectivity in Table 4 shown in Figure 7b. This configuration has splits *ABCD* – *ABC*/*B*/*C*/*D* and *CDE* – *C*/*D*/*E* as side-splits and no submixture stream of the configuration is a feed and a product of the same distillation column. Thus, the configuration of Figure 7b satisfies Checks 4 and 5. However, the configuration has streams *ABC* and *BC* being transferred in opposite directions between distillation columns "2" and "3" with stream *BC* produced from stream *ABC* in column "3", thus violating Check 6.

On the other hand, the configuration obtained from the candidate matrix of Figure 5a and the corresponding Table 1 is a feasible four-component subcolumn configuration. Checks 4 – 6 can thus be applied to generate feasible subcolumn configurations from among all candidate matrices.

From the 2^d four-component candidate matrices, following Steps 1 – 6 of the method we find that the matrices denoted with oval symbols in Figure 4: (vi), (x)-(xii), (xiv)-(xvi), (xix)-(xxiv), (xxvii)-(xxviii), (xxx)-(xxxii) correspond to the complete search space of feasible basic distillation configurations that use exactly three distillation columns.[2] These configurations are regular-column basic configurations. Following Steps 1 – 4 and Steps 7 – 8, we find that the matrices in Figure 4: (i)-(v), (vii)-(ix), (xiii), (xvii)-(xviii), (xxv) correspond to feasible subcolumn distillation configurations that use less than three distillation columns. The two matrices denoted with diamonds in Figure 4: (xxvi), (xxix) result in infeasible distillation configurations and are eliminated from the search space of configurations with exactly $n-1$ columns or with less than $n-1$ columns.

The subcolumn distillation configurations for $n = 4$ are provided in Figure 9. Configurations (a) through (f) of Figure 9 are sharp split subcolumn distillation configurations. The configurations (g) through (l) of Figure 9 are nonsharp split subcolumn distillation configurations. Earlier attempts[11,12] have generated only sharp split subcolumn configurations and the nonsharp split subcolumn configurations synthesized by our matrix method were previously unknown.

Consider the distillation configurations in Figure 10. Both these configurations have the same sequence of splits, i.e., the first split is *ABCD* – *A*/*B*/*CD* in column "1", followed by the split *CD* – *C*/*D* in column "2". They only differ in the location of side-stream *B* relative to the location of feed stream *ABCD* in distillation column "1". From the synthesis perspective, both the configurations in Figure 10 are equivalent to the configuration shown in Figure 9d. Previous works[11,12] have considered such configurations with the same sequence of splits but differing in the location of side-streams as distinct configurations. However, we treat such configurations as one configuration only and the locations and the phases of the side-streams are decided through flowsheet optimization that will depend on feed composition, relative volatilities, etc.

It is worth mentioning that depending on the withdrawal location of side-streams relative to the feed location, the side-stream products in distillation columns with side-splits could be contaminated with heavier or lighter components.

Table 2. Splits in Distillation Configuration Corresponding to Matrix (xxxii) of Figure 4

Split Number	Feed Stream	Product Streams: Top	Bottom	Side-Stream	Distillation Column Number
1	*ABCD*	*ABC*	*BCD*	–	1
2	*ABC*	*AB*	*BC*	–	2
3	*BCD*	*BC*	*CD*	–	2
4	*AB*	*A*	*B*	–	3
5	*BC*	*B*	*C*	–	3
6	*CD*	*C*	*D*	–	3

Table 3. Splits in Distillation Configuration Corresponding to Matrix (xxvi) of Figure 4

Split Number	Feed Stream	Product Streams: Top	Bottom	Side-Stream	Distillation Column Number
1	*ABCD*	*ABC*	*BCD*	–	1
2	*ABC*	*A*	*C*	*B*	2
3	*BCD*	*B*	*CD*	–	2
4	*CD*	*C*	*D*	–	2

Table 4. Splits in the Distillation Configuration Corresponding to Matrix in Figure 7a

Split Number	Feed Stream	Product Streams Top	Bottom	Side-Stream	Distillation Column Number
1	*ABCDE*	*ABCD*	*CDE*	–	1
2	*ABCD*	*ABC*	*D*	–	2
3	*ABC*	*A*	*BC*	–	3
4	*CDE*	*C*	*E*	*D*	2
5	*BC*	*B*	*C*	–	2

For instance, in Figure 10a, side-stream *B* is drawn from distillation column "1" below location of feed stream *ABCD*. Below the feed location, the distillation column will contain not only component *B* but also components *C* and *D* in detectable amounts. The amount of component *B* in the side-stream can be maximized by increasing the boilup ratio in the column to a very high value consequently requiring large energy to produce pure side-stream product *B*. Similarly, other subcolumn distillation configurations in this article that have distillation columns with side-splits could also require large energy to make pure side-stream products.

It must be noted that for the four-component example, each of the thirty feasible matrices for a four-component feed mixture uniquely corresponds to either a configuration with exactly three distillation columns or a configuration with less than three distillation columns. A feasible matrix does not correspond simultaneously to a configuration with less than $n-1$ columns as well as a configuration with exactly $n-1$ columns. However, this is not true when we consider higher number of components in the feed. Consider the five-component matrix shown in Figure 11a. By applying Steps 5 and 6 of the method to the matrix in Figure 11a we generate the basic configuration shown in Figure 11b which has four distillation columns. On the other hand, by applying Steps 7 and 8 of the method, we generate the subcolumn configuration shown in Figure 11c. Thus, depending on the interpretation of a matrix, the same matrix can correspond to a basic configuration (with exactly $n-1$ columns) as well as a subcolumn configuration (with less than $n-1$ columns).

Method to Generate the Search Space of Thermally Coupled Configurations

Introduction of thermal coupling in a distillation configuration often tends to reduce its total heat duty requirement. Shah and Agrawal[10] have provided a procedure to obtain additional thermally coupled configurations by extending the matrix-based approach of basic configurations. A similar approach can be used to obtain the additional thermally coupled configurations with less than $n-1$ distillation columns. In this procedure, reboilers and condensers associated with submixtures in a configuration are eliminated and replaced with thermal coupling links. Therefore, each of the candidate submixture streams for thermal coupling can have additional 0 – 1 variables associated with it, indicating absence or presence of thermal coupling at its reboiler or condenser. Hence, from the total number of candidate submixture streams in a configuration, N_{cand}, we can obtain $2^{N_{\text{cand}}} - 1$ additional configurations having partial to complete thermal coupling. We can thus systematically obtain all additional thermally coupled configurations from the search space of subcolumn configurations generated using the matrix method.

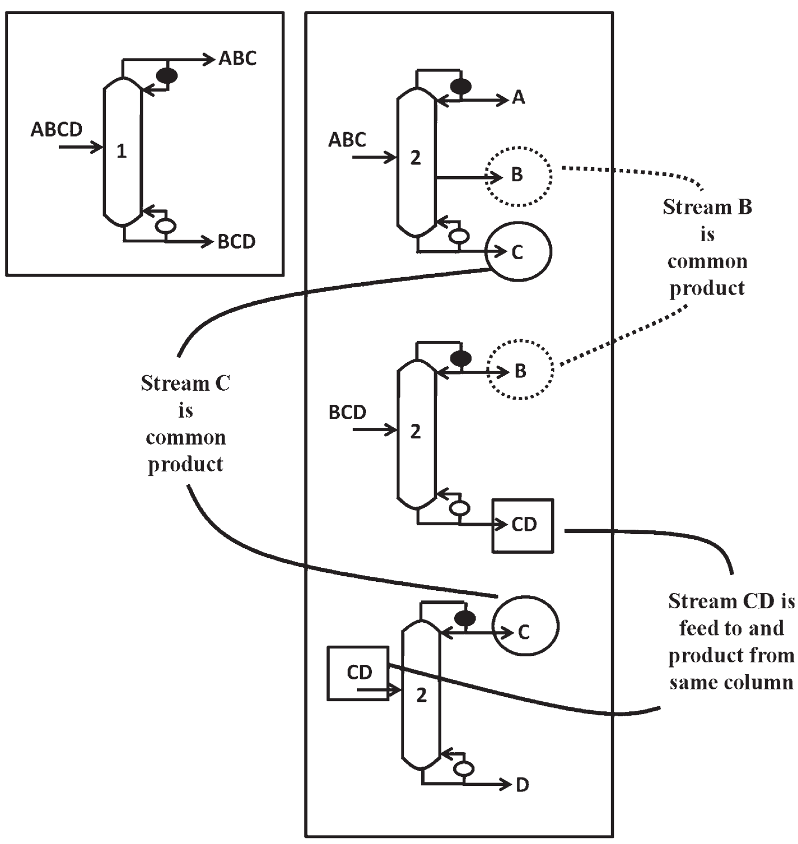

Figure 8. Listing and grouping of splits corresponding to candidate matrix (xxvi) of Figure 4.

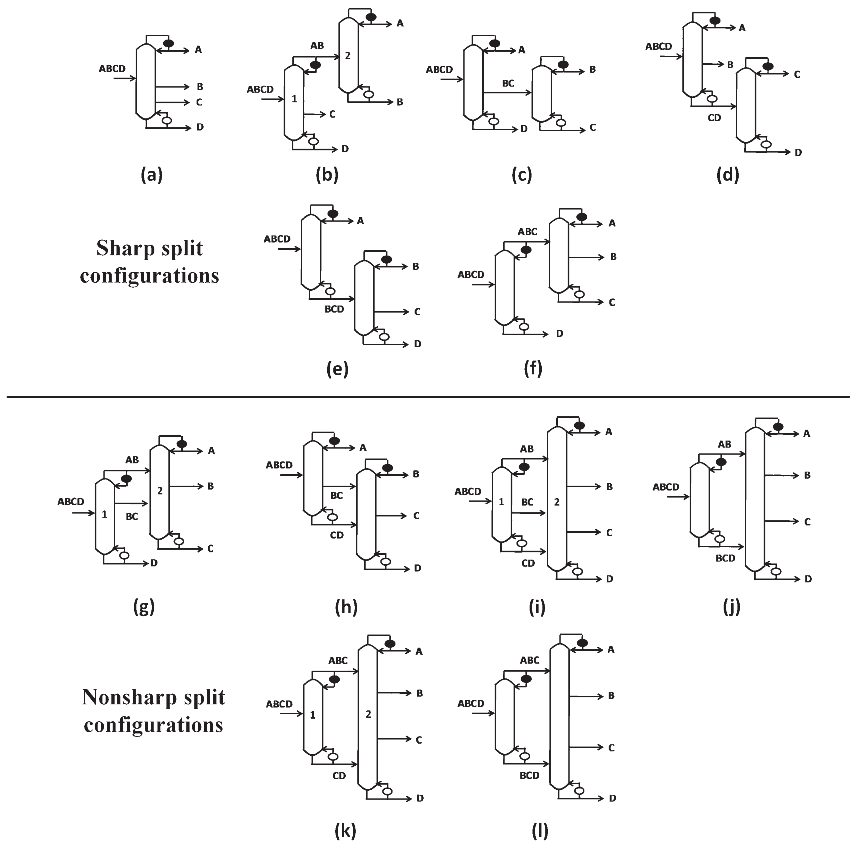

Figure 9. Search space of subcolumn configurations for four-component feed mixture.

Results and Discussion

Shah and Agrawal[10] presented a flowchart with the key steps of the matrix-based method to synthesize the search space of basic distillation configurations. We extend the flowchart to include the steps to generate the subcolumn distillation configurations. The composite flowchart is shown in Figure 12. The number of subcolumn configurations for up to eight components in the feed is listed in Table 5. The number of basic configurations for up to eight components in the feed has been provided in the earlier work.[10]

Our method thus provides a simple algorithm to generate the search space of subcolumn distillation configurations for any number of components in the feed thereby extending the algorithm for generating basic configurations.[10] The corresponding thermally coupled configurations having partial to complete thermal coupling can also be easily generated. We have not considered configurations having reboilers and condensers at intermediate column locations.[20,23] Such possibilities can also be included when appropriate.

Several previously unknown subcolumn configurations can be generated using the matrix method. However, there are other subcolumn configurations, such as the one shown in Figure 13, that are not generated by our proposed method. The search space of subcolumn configurations is thus not complete. To address this issue, we provide a more elaborate method in the Appendix to generate all distillation configurations with side-splits including subcolumn, regular-column and also plus-column configurations. For the purpose of this article, we focus on the subcolumn configurations generated by Steps 1 to 4 and Steps 7 and 8. We believe that the set generated here is large enough to provide a suitably optimal configuration for a given application. However, if required,

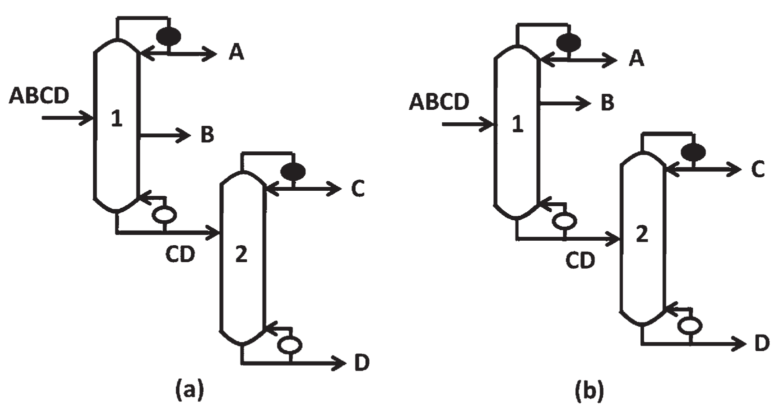

Figure 10. Possible instances of configuration in Figure 9d.

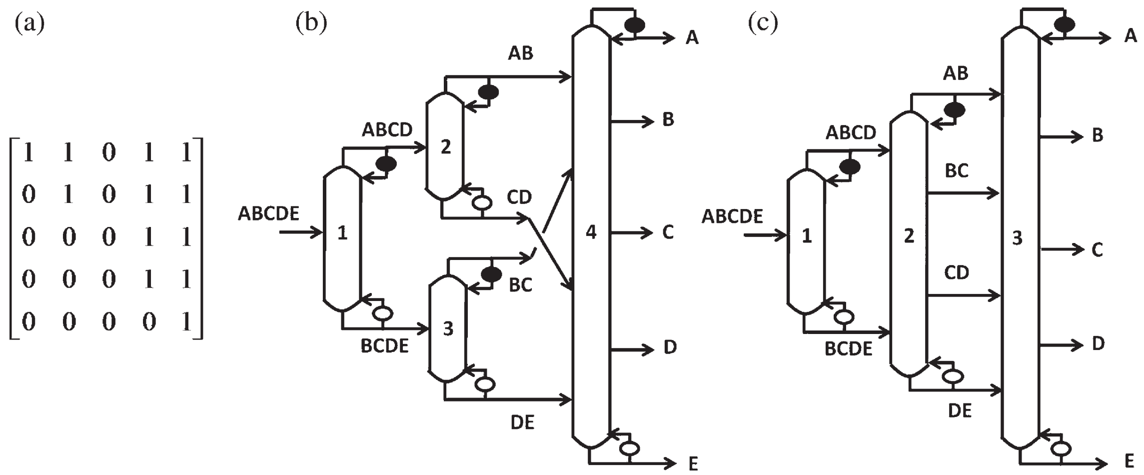

Figure 11. (a) Example of a candidate matrix for five-component feed mixture, (b) Distillation configuration with four columns obtained from the matrix of (a), (c) Distillation configuration with three columns obtained from the matrix of (a).

the complete set of side-split configurations can be generated by following Steps 1 – 4 of the matrix method along with new Steps 7A – 8A which are described in the Appendix and can be included in a search space.

Case Study

The efficacy of the matrix method is illustrated through a case study. Our focus is on comparing the heat duty requirements of the new four-component subcolumn configurations with the heat duty requirements of the previously known subcolumn and basic regular-column distillation configurations.

We consider separation of a four-component feed mixture comprising of 30% n-butane (*A*), 40% n-pentane (*B*), 25% n-hexane (*C*), and 5% n-octane (*D*) (in mole percent). Separation of this feed mixture using subcolumn distillation configurations has been studied by Kim and Wankat[11] and Errico et al.[12] They have compared the five sharp split configurations among the eighteen basic configurations[2] to the six sharp split configurations among the twelve subcolumn configurations shown in Figure 9.

In our case study, using the data provided by Errico et al.,[12] the feed is a 100 kmol/hr liquid stream at a temperature of 334.55 K. The required molar purities of the products are 99% each for n-butane, n-pentane and n-octane and 98% for n-hexane (mole percent). The distillation configurations are simulated using the stage-by-stage distillation model RADFRAC in Aspen Plus. The pressure of each distillation column is fixed to 3 atm and the Peng-Robinson equation of state is selected to calculate the appropriate thermodynamic properties. The total reboiler duty of a configuration is chosen as the objective function. For the simulations of subcolumn configurations, we treat relative locations of side-streams (such as side-stream *B* in Figure 10) as optimization variables.

All distillation columns are simulated with 100 stages. Such a large number of stages enables each column section to have more than enough stages ensuring that the simulation results are relatively insensitive to the exact tray locations for feeds and side-draws. Thus, only the column section that a side-stream product belongs to is selected by the optimization algorithm whereas the exact tray location within a column section is not optimized.

For basic configurations, all final product streams are produced as liquids since the objective of our problem is minimizing the total reboiler duty and producing them as vapor will unnecessarily increase the total reboiler duty of each distillation configuration. Furthermore, the transfer streams associated with reboilers are produced as liquids and the transfer streams associated with condensers are produced as vapors. Transfer side-streams can be produced as either single-phase liquid streams or single-phase vapor streams or two-phase liquid-vapor streams as decided by the optimization algorithm.

For the subcolumn configurations, any stream (including final product streams) associated with a reboiler is produced as liquid, any stream (excluding final product streams) associated with a condenser is produced as vapor. Any side-stream (including final product streams) is produced as either a single-phase liquid stream or a single-phase vapor stream or a two-phase liquid-vapor stream as determined by the relative location of the side-stream with respect to the feed through optimization. To minimize the total reboiler duty, it is advantageous to produce side-streams located above the feed location as liquids since the liquid in the rectifying section is richer in the intermediate components than the vapor.[21] Similarly it is advantageous to produce side-streams located below the feed location as vapor, since the vapor in the stripping section is richer in the intermediate components than the liquid.[21]

The vapor duty requirement of a distillation configuration is indicative of its heat duty requirement. Therefore, instead of simulating all eighteen basic configurations, we first optimized them with respect to their vapor duty requirements for the chosen feed conditions using Underwood's equations.[24] The best basic configuration identified through these calculations is presented in Figure 14 and this configuration was rigorously simulated and optimized using Aspen Plus. The stage numbers and the total reboiler duty of this configuration are provided in Table 6.

Among the six previously known sharp split subcolumn distillation configurations, the configuration of Figure 9b was

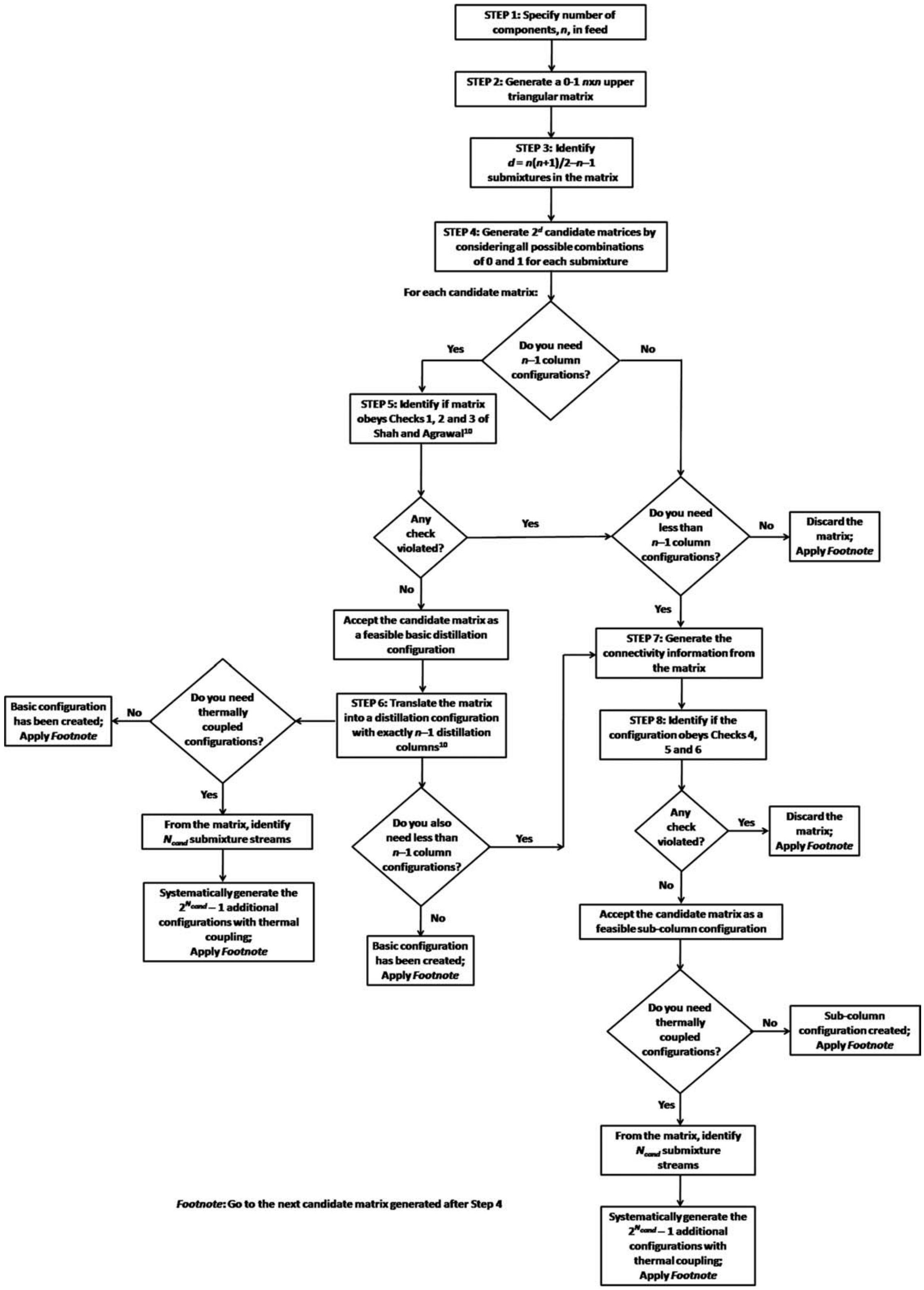

Figure 12. A flowchart showing key steps of the matrix method.

found to have the lowest total reboiler duty requirement. However if we also consider the nonsharp split subcolumn distillation configurations, the configuration of Figure 9i was found to have the least total reboiler duty requirement among all the subcolumn distillation configurations of Figure 9. The configuration of Figure 9g was found to have the highest total reboiler duty requirement among the nonsharp split subcolumn distillation configurations. The total reboiler duty requirements and stage numbers of these distillation configurations are also presented in Table 6.

From the simulation results presented in Table 6, it can be seen that the best sharp split subcolumn configuration

Table 5. Number of Subcolumn Distillation Configuration and Additional Thermally Coupled Configuration

Number of Components in the Feed	Number of Subcolumn Configurations without Thermal Coupling	Number of Additional Thermally Coupled Configurations
3	1	0
4	12	18
5	198	1279
6	5142	124,346
7	224,257	20,168,590
8	17,056,898	5,739,609,045

requires nearly six times more reboiler duty than the best basic configuration. Also, the best nonsharp split subcolumn configuration requires only 15% more reboiler duty than the best basic configuration. Since this configuration uses one less distillation column, on the basis of overall cost including both capital and energy consumption, it could prove to be a cheaper alternative for the distillation of the considered feed mixture. Moreover the worst nonsharp split subcolumn configuration requires significantly lower total reboiler duty than the best sharp split subcolumn configuration. Therefore, nonsharp split subcolumn distillation configurations should be considered in the search space of distillation configurations with fewer columns.

Are we Missing the Brugma Configuration[13]?

It can be observed that the subcolumn configurations in Figure 10 do not include the distillation configuration of Figure 2a proposed by Brugma.[13] For the considered case study, optimization of the configuration of Figure 9l provided flow rates where there is around 1% *C* in stream *ABC* and around 2% *B* in stream *BCD* (in mole percent). Therefore, among the configurations in Figure 9, the configuration of Figure 9l approaches towards the configuration of Figure 2a. Further, simulation of the configuration of Figure 2a shows that it has a slightly higher total reboiler duty than the configuration of Figure 9l (Table 7).

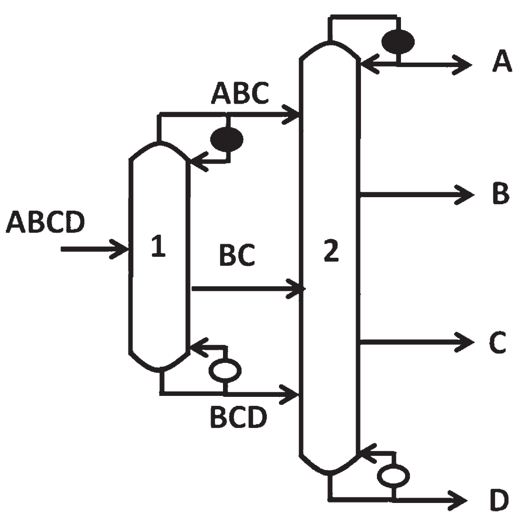

Figure 13. A configuration missing in the search space generated by Steps 1 – 8 of the matrix method.

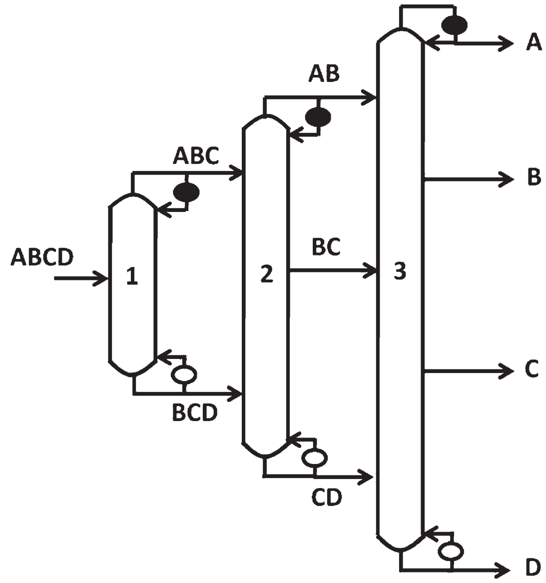

Figure 14. Basic configuration with lowest heat duty requirement.

It is possible that the distillation configuration of Figure 9l reduces exactly to the configuration of Figure 2a during an optimization of the process parameters. For instance, during optimization of the flow rates of streams *ABC* and *BCD* in Figure 9l to minimize the total heat duty, the flows might adjust such that component *C* is almost completely absent in stream *ABC* and component *B* is almost completely absent in stream *BCD* giving us the configuration shown in Figure 2a. Hence the configurations proposed by Brugma,[13] Cahn and Di Miceli[17] and Kaibel[18] should be treated as subsets of configuration of Figure 9l with or without appropriate thermal coupling links. In fact, the configurations of Figures 9j, k could also be considered as subsets of the configuration of Figure 9l. Figure 15 illustrates how some distillation configurations are merely optimization outcomes of other distillation configurations. With the inclusion of these configurations in the search space, it is no longer necessary to explicitly include Brugma's or its associated configurations of Figure 2 in the search space.

Purity Constraints on Subcolumn Configurations

Consider the sharp split subcolumn configurations in Figures 9a–f. The side-streams in these configurations are generally contaminated by the heavier or lighter components of the feed depending on the relative location at which they are produced with respect to the feed location in the column. These configurations are thus constrained by the purity requirements of the final product side-streams. Making these side-streams of very high purity could result in high reflux duties and hence high operating costs. This problem can also occur in the nonsharp split configurations shown in Figures 9g–h.

However, other subcolumn configurations such as the ones in Figures 9i-l are quite versatile in making products of any prespecified purity. While they can easily produce impure products with varying proportions of lighter and heavier components, they are also capable of producing all the products at any desired level of high purity. The ability to make products of arbitrarily high purity using fewer distillation columns is one of the main advantages of the Brugma configuration.[17] In such an extreme case, when all four products

Table 6. Energy Savings of Novel Distillation Configurations

	Best Sharp Split Subcolumn Configuration	Worst Nonsharp Split Subcolumn Configuration	Best Subcolumn Configuration	Best Basic Configuration
Search Space	Figure 9a–f	Figure 9g–1	Figure 9	All 18 configurations[2]
Figure	9b	9g	9i	14
Stage Numbers	**Column 1**	**Column 1**	**Column 1**	**Column 1**
	ABCD in – 50	*ABCD* in – 66	*ABCD* in – 33	*ABCD* in – 50
	C out – 75	*BC* out – 33	*BC* out – 66	
	Column 2	**Column 2**	**Column 2**	**Column 2**
	AB in – 50	*AB* in – 25	*AB* in – 25	*ABC* in – 25
		BC in – 75	*BC* in – 50	*BCD* in – 75
		BC out – 50	*CD* in – 75	*BC* out – 50
			B out – 37	**Column 3**
			C out – 63	*AB* in – 20
				BC in – 50
				CD in – 80
				B out – 35
				C out – 65
Total Reboiler Duty (kW)	5775	2286	1051	915

are needed at extremely high purities, the compositions of the transfer streams from the feed column to the subsequent column in the configurations of Figures 9i–l get adjusted to reduce to the Brugma configuration. While Figure 16 shows this to be the case for the configurations of Figures 9j–l, this is also true for the configuration of Figure 9i. For the configuration of Figure 9i, flow rate of stream *BC* will be optimized to a vanishingly low value resulting in Brugma's configuration. Thus, in general, configurations such as the ones in Figures 9i–l are more versatile than the configurations shown in Figure 2.

In fact, the configuration shown in Figure 13 encompasses the configurations in Figures 9i–l. The configuration of Figure 13 is not synthesized following Steps 7 and 8 of the matrix method. However, for our case study, it was observed that the configuration of Figure 13 reduces exactly to the configuration shown in Figure 9i during optimization with component *C* completely absent in stream *ABC* and component *B* completely absent in stream *BCD*. Therefore, even though we have found the optimal subcolumn configuration for our case study, the configuration of Figure 13 is actually equivalent to the configuration in Figure 9i if we allow enough *C* in *AB* and *B* in *CD* during optimization. Such a scenario is possible by optimizing over the distillate flow rate without any constraints on the amount of *C* in *AB* or the amount of *B* in *CD* within the optimization algorithm. Further, it must be noted that the more general configuration (such as the one shown in Figure 13) can be easily generated by following Steps 7A – 8A in Appendix when appropriate.

It is interesting to consider the divided wall column version of the subcolumn configuration of Figure 9i shown in Figure 16. One main difference between the configuration of Figure 16 and the Kaibel configuration of Figure 2c is that at an intermediate location of the divided wall, we transfer a vapor stream *BC* from the feed side of the wall to the product side of the wall. Therefore, the configuration of Figure 16 behaves as if the divided wall has an adjustable "leak" at an intermediate location for transfer of stream *BC*. For making higher purity products, the leak in the wall will close giving the Kaibel configuration (Figure 2c) as a possible optimization outcome. Thus depending on the product purity requirements, the configuration of Figure 16 can be thought of as a "divided wall column with an optimized leaky wall." As seen from Tables 6 and 7, for the case study, such an intermediate vapor transfer in a configuration without thermal coupling has a potential to reduce the rate of heat consumption from 1144 kW to 1051 kW, an improvement of about 8%.

Conclusions

We have developed a systematic method to synthesize distillation configurations that use less than $n-1$ columns to separate an n-component zeotropic feed into n product streams. This method is an extension of our earlier matrix-based method to synthesize basic distillation configurations. This method also provides additional thermally coupled configurations. The use of the method is based on simple physical rules and it is easily generalizable for any number of components in the feed.

Prior studies on subcolumn configurations focused on the sharp split subcolumn configurations. Our method provides both sharp as well as nonsharp split subcolumn configurations. Through a case study for a four-component mixture, we have shown that the nonsharp split subcolumn configurations can have significantly lower heat duty requirement than the sharp split subcolumn configurations. Therefore, it is essential to include nonsharp split configurations in any search space for identifying optimal subcolumn configurations for a given application.

Interestingly, subcolumn configurations suggested by Brugma[13] and their associated thermally-coupled versions

Table 7. Comparison of Brugma Configuration[13] to Configuration Generated by the Matrix Method

	Brugma Configuration[13]	Configuration that approaches Brugma Configuration after Optimization
Figure	2a	9l
Stage Numbers	**Column 1**	**Column 1**
	ABCD in – 50	*ABCD* in – 50
	Column 2	**Column 2**
	AB in – 20	*ABC* in – 20
	CD in –80	*BCD* in –80
	B out – 40	*B* out – 40
	C out – 60	*C* out – 60
Total Reboiler Duty (kW)	1144	1113

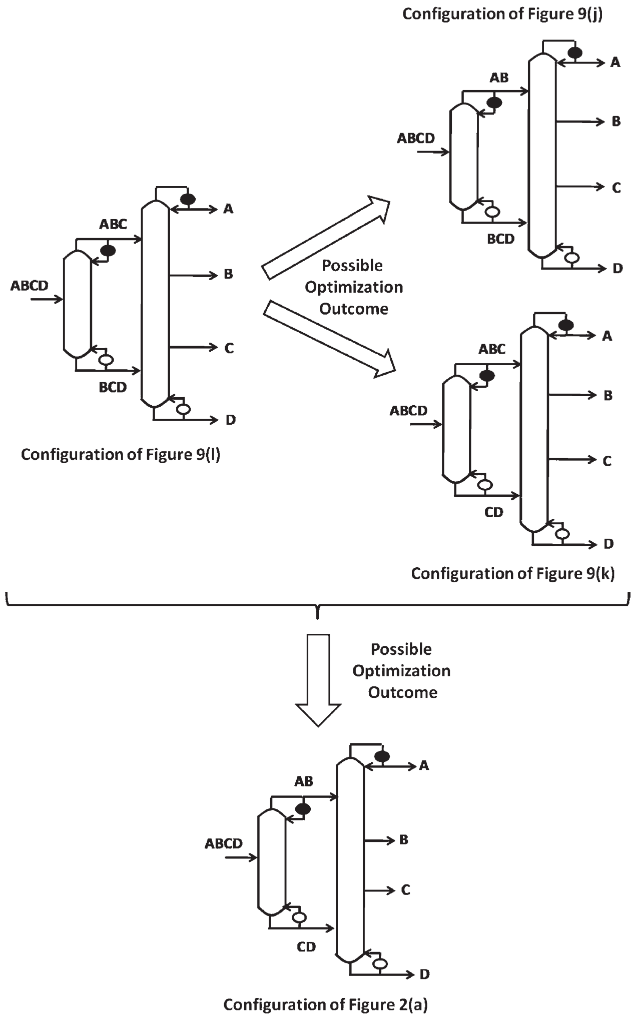

Figure 15. Illustration of possible optimization outcomes in subcolumn configurations.

are not explicitly obtained by our method. However, such configurations are actually subsets of more versatile new subcolumn configurations generated by our method. Thus, when a search space includes subcolumn configurations generated by our method, it is not essential to explicitly include configurations similar to those proposed by Brugma.

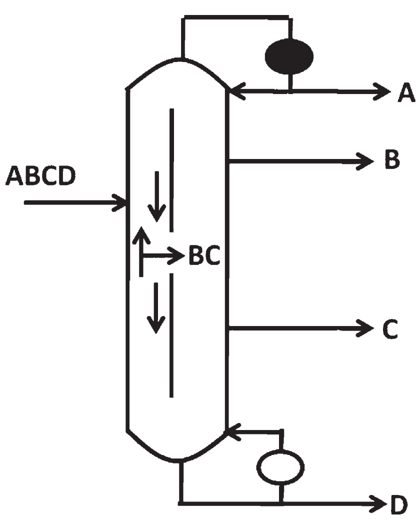

Figure 16. Divided wall column version of the subcolumn configuration of Figure 9i consisting of an "optimized leaky wall."

Acknowledgments

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Appendix

For synthesis of the complete search space of configurations with side-splits, after Steps 1 – 4 of the matrix method, one follows Steps 7A and 8A described in this Appendix.

$$\begin{bmatrix} ABCD & ABC & 0 & A \\ 0 & BCD & BC & B \\ 0 & 0 & 0 & C \\ 0 & 0 & 0 & D \end{bmatrix}$$

Figure A1. Replacing 1's in the candidate matrix (xxvii) of Figure 4.

Step 7A

In this step, we deduce connectivity information required to identify side-split distillation configurations from the 2^d matrices generated in Step 4. The procedure to do this is: (a) Replace the 1's in the candidate matrix by the corresponding streams, (b) Generate all possible splits for each feed stream in the matrix, (c) Identify and tabulate combinations of splits representing distillation configurations and (d) Assign column numbers to the splits in each identified distillation configuration.

Consider the candidate matrix (xxvii) shown in Figure 4. Replacing the 1's in the matrix by the corresponding streams as shown in Figure A1 and defining the candidate product zone as described in Step 7, we can identify the candidate products of the streams *ABCD*, *ABC*, *BCD* and *BC*.

We use the candidate products of a stream to generate all possible splits of the stream. To do this, we first classify the candidate products of each feed as *compulsory* or *optional* products. A compulsory product of a feed is necessarily produced from that particular feed. On the other hand, optional products may or may not be produced from the feed stream.

Table A1. Feasible Splits of Stream *ABCD* Corresponding to the Matrix in Figure A1

Feasible Split Number	Feed Stream	Product Streams						
		Compulsory		Optional				
1	*ABCD*	*ABC*	*BCD*	–	–	–	–	–
2	*ABCD*	*ABC*	*BCD*	–	–	–	*C*	–
3	*ABCD*	*ABC*	*BCD*	–	–	*B*	–	–
4	*ABCD*	*ABC*	*BCD*	–	–	*B*	*C*	–
5	*ABCD*	*ABC*	*BCD*	–	*BC*	–	–	–
6	*ABCD*	*ABC*	*BCD*	–	*BC*	–	*C*	–
7	*ABCD*	*ABC*	*BCD*	–	*BC*	*B*	–	–
8	*ABCD*	*ABC*	*BCD*	–	*BC*	*B*	*C*	–
9	*ABCD*	*ABC*	*BCD*	*A*	–	–	–	–
10	*ABCD*	*ABC*	*BCD*	*A*	–	–	*C*	–
11	*ABCD*	*ABC*	*BCD*	*A*	–	*B*	–	–
12	*ABCD*	*ABC*	*BCD*	*A*	–	*B*	*C*	–
13	*ABCD*	*ABC*	*BCD*	*A*	*BC*	–	–	–
14	*ABCD*	*ABC*	*BCD*	*A*	*BC*	–	*C*	–
15	*ABCD*	*ABC*	*BCD*	*A*	*BC*	*B*	–	–
16	*ABCD*	*ABC*	*BCD*	*A*	*BC*	*B*	*C*	–
17	*ABCD*	*ABC*	*BCD*	–	–	–	–	*D*
18	*ABCD*	*ABC*	*BCD*	–	–	–	*C*	*D*
19	*ABCD*	*ABC*	*BCD*	–	–	*B*	–	*D*
20	*ABCD*	*ABC*	*BCD*	–	–	*B*	*C*	*D*
21	*ABCD*	*ABC*	*BCD*	–	*BC*	–	–	*D*
22	*ABCD*	*ABC*	*BCD*	–	*BC*	–	*C*	*D*
23	*ABCD*	*ABC*	*BCD*	–	*BC*	*B*	–	*D*
24	*ABCD*	*ABC*	*BCD*	–	*BC*	*B*	*C*	*D*
25	*ABCD*	*ABC*	*BCD*	*A*	–	–	–	*D*
26	*ABCD*	*ABC*	*BCD*	*A*	–	–	*C*	*D*
27	*ABCD*	*ABC*	*BCD*	*A*	–	*B*	–	*D*
28	*ABCD*	*ABC*	*BCD*	*A*	–	*B*	*C*	*D*
29	*ABCD*	*ABC*	*BCD*	*A*	*BC*	–	–	*D*
30	*ABCD*	*ABC*	*BCD*	*A*	*BC*	–	*C*	*D*
31	*ABCD*	*ABC*	*BCD*	*A*	*BC*	*B*	–	*D*
32	*ABCD*	*ABC*	*BCD*	*A*	*BC*	*B*	*C*	*D*

Table A2. Feasible Splits of Stream *ABC* Corresponding to the Matrix in Figure A1

Feasible Split Number	Feed Stream	Product Streams			
		Compulsory	Optional		
1	*ABC*	*A*	–	*B*	*C*
2	*ABC*	*A*	*BC*	–	–
3	*ABC*	*A*	*BC*	–	*C*
4	*ABC*	*A*	*BC*	*B*	–
5	*ABC*	*A*	*BC*	*B*	*C*

We use two criteria to identify the compulsory products from among the candidate products of a feed stream and distinguish them from the optional products. Firstly, if a stream is a candidate product of only one feed in the matrix then it is a compulsory product of that feed. For instance, for the candidate matrix in Figure A1, stream *ABC* is a candidate product of only feed stream *ABCD* as it is excluded from the candidate product zone of all the other streams. Hence, stream *ABC* is a compulsory product of stream *ABCD*. Similarly, we can also identify stream *BCD* as a compulsory product of stream *ABCD*. The remaining candidate products of stream *ABCD*, that is, streams *A*, *BC*, *B*, *C* and *D* are optional products. The optional products may either be produced by direct distillation of stream *ABCD* in the same distillation column or by subsequent distillation of the submixtures produced from the distillation of stream *ABCD*. All streams, other than the main feed, are included in the candidate product zone of the main feed. Thus, the first criterion can be used to identify compulsory products only for the main feed stream. The second criterion is that if a component of a feed stream is unique to only one of its candidate products then that candidate product is a compulsory product of the feed stream under consideration. For stream *ABC*, the candidate products are streams *A*, *BC*, *B* and *C* among which component *A* is present only in product stream *A*. Hence, stream *A* is a compulsory product of stream *ABC* and streams *BC*, *B* and *C* are its optional products. Similarly, for stream *BCD*, streams *BC*, *B* and *C* are optional products and stream *D* is a compulsory product. Further, for stream *BC*, we can identify both the candidate products, that is, streams *B* and *C*, as compulsory products and thus stream *BC* has no optional products.

We then generate all possible combinations indicating presence or absence of optional products in the split of a feed stream. For the example of Figure A1, the main feed stream has five optional products (*A*, *BC*, *B*, *C* and *D*), which implies generation of $2^5 = 32$ possible splits of stream *ABCD* (Table A1). All the 32 splits of feed stream *ABCD* are feasible. However, the combinations of optional products can sometimes lead to infeasible splits, where components disappear in a split. For instance, a combination of optional products for stream *ABC* will yield split such as *ABC* – *A*/*C*. Such a split

Table A3. Feasible Splits of Stream *BCD* Corresponding to the Matrix in Figure A1

Feasible Split Number	Feed Stream	Product Streams			
		Compulsory	Optional		
1	*BCD*	*D*	–	*B*	*C*
2	*BCD*	*D*	*BC*	–	–
3	*BCD*	*D*	*BC*	–	*C*
4	*BCD*	*D*	*BC*	*B*	–
5	*BCD*	*D*	*BC*	*B*	*C*

Table A4. An Illustration of Connectivity Information Obtained from the Matrix of Figure A1

Split Number	Feed Stream	Product Streams			Distillation Column Number
		Top	Bottom	Side-Stream	
1	*ABCD*	*ABC*	*BCD*	*BC*	1
2	*ABC*	*A*	*C*	*B*	2
3	*BCD*	*B*	*D*	*C*	2
4	*EC*	*B*	*C*	–	2

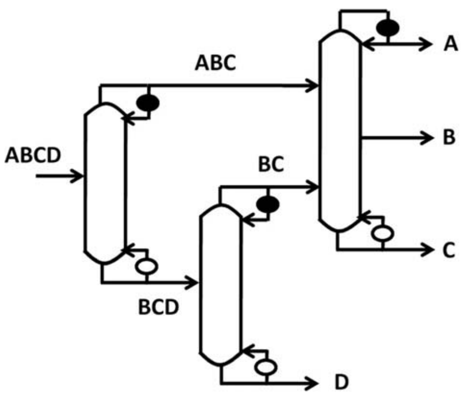

Figure A2. An example of a regular-column configuration with side-split for four-component separation.

is not allowed as component *B* has disappeared in the split. Therefore, the combinations of optional products are constrained to ensure that each component in the feed is present in at least one of the product streams. We thus obtain five feasible splits for each feed stream *ABC* and *BCD* as shown in Tables A2 and A3 respectively. Since, stream *BC* has no optional products, its only possible split is *BC* – *B*/*C*.

The connectivity information representing a distillation configuration is then generated by choosing one split at a time for each feed stream. For *f* feeds in a given candidate matrix and each feed with *s* possible splits, there are $\prod_{i=1}^{f} s_i$ connectivity tables. For the matrix in Figure A1, we have $f = 4$ (*ABCD*, *ABC*, *BCD*, and *BC*) and $s_1 = 32$, $s_2 = 5$, $s_3 = 5$, and $s_4 = 1$ thus giving $32 \times 5 \times 5 \times 1 = 800$ connectivity tables. Thus, a candidate matrix such as the one shown in Figure A1 can correspond to more than one connectivity table. Some of the connectivity tables represent feasible side-split configurations whereas others must be eliminated from the search space. Table A4 shows one such connectivity table obtained from the matrix in Figure A1. The splits in this connectivity table are *ABCD* – *ABC*/*BC* / BC*D*, *ABC* – *A*/*B*/*C*, *BCD* – *B*/*C*/*D* and *BC* – *B*/*C*. Splits making common streams are grouped and distillation column numbers are assigned to all the splits in Table A4.

We can thus obtain all 800 connectivity tables from the candidate matrix in Figure A1 and many more connectivity tables from every other candidate 0 – 1 matrix generated in Step 4.

Step 8A

In this step, we eliminate the connectivity tables generated in Step 7A that do not lead to feasible side-split distillation configurations. To do this, we implement five checks on each connectivity table obtained in Step 7A. If any check is not satisfied, then the corresponding connectivity table is eliminated from the search space of side-split configurations. Checks 4 – 6 are same as those described in Step 8 of the matrix method whereas Checks 7 and 8 are new and will be described in this section.

Check 7. Ensure that all streams present in the matrix, other than the main feed stream, are produced from at least one feed stream in the configuration. This check ensures that all the submixture streams present in the matrix are accounted for.

Check 8. Disallow configurations in which two feeds to the same column have common candidate products and one or more of the common candidate products are produced from only one feed but not from the second feed. For instance, a configuration having distillation column with feeds *ABCD* and *BCDE* having common candidate products *B*, *C*, and *D* and splits being *ABCD* – *AB*/*C*/*D* and *BCDE* – *B*/*C*/*D*/*E* is not allowed. Stream *B*, in such an example, is produced from only one split which is generally not possible since both the feeds contain common component *B* and on distillation of stream *ABCD*, some amount of component *B* from *ABCD* will end up in product stream *B* which is not allowed per the *ABCD* split specification.

The five checks 4 – 8 are applied to each connectivity table obtained from a candidate matrix to identify feasible configurations with side-splits. For instance, the configuration obtained from the candidate matrix in Figure A1 and the corresponding Table A4 satisfies all the checks and leads to a feasible configuration with side-splits. This feasible configuration is shown in Figure 13 and is one of the missing configurations that could not be created following Steps 1 – 4 and Steps 7 – 8 of the matrix method.

For the four-component example, following Steps 1 – 4 and Steps 7A – 8A, we can not only generate the twelve subcolumn configurations shown in Figure 9, but we can also generate 27 more configurations with side-splits. Among the 27 new side-split configurations, eight are regular-column configurations while the remaining are subcolumn configurations. An example of a regular-column configuration with a side-split (*ABC* – *A*/*B*/*C*) is shown in Figure A2.

Table A5. Number of Distillation Configurations with Side-Splits and Additional Thermally Coupled Configurations

Number of components in the feed	Number of Configurations with Side-Splits without Thermal Coupling				Number of Additional Thermally Coupled Configurations
	Sub-column	Regular-column	Plus-column	Total	
3	1	0	0	1	0
4	31	8	0	39	151
5	3943	1427	48	5418	117,334

The number of side-split configurations for up to five components in the feed is listed in Table A5. We have also identified additional thermally coupled configurations. Although we have provided results of side-split configurations for only up to five components in the feed, the method is generally applicable for any number of components. With extensive computational power, the complete search space of subcolumn, regular-column and plus-column configurations with side-splits for higher number of components can also be systematically generated.

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