Global Optimization of Multicomponent Distillation Configurations: 1. Need for a Reliable Global Optimization Algorithm

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DOI 10.1002/aic.13875
Published online July 10, 2012 in Wiley Online Library (wileyonlinelibrary.com).

Nonazeotropic multicomponent mixtures are often separated into products by distillation configurations containing multiple distillation columns. One method of calculating the minimum vapor duty of a configuration is to sequentially calculate the minimum vapor duty of each mixture as it is split into two streams within a given column starting from the feed column. The other method simultaneously manipulates all the splits to yield the overall minimum vapor duty of the entire configuration. Of these two methods, the sequential minimization is attractive as it can be analytically solved. However, through extensive computations, we find that the sequential minimization method is not a valid substitute for the simultaneous minimization method. As the number of components in the feed increases, the fraction of the basic configurations for which sequential method yields a reasonable estimate decreases rapidly, thereby emphasizing the need for a more robust and reliable global optimization algorithm. © 2012 American Institute of Chemical Engineers *AIChE J,* 59: 971–981, 2013
Keywords: transition split, preferred split, multicomponent distillation, distillation configurations, global optimization

Introduction

Distillation is used for around 95% of all separations in the chemical process industries.[1] A single distillation column is not typically used for separation of multicomponent mixtures into more than two product streams (each enriched in one of the components). Instead, the mixture is split in several sequential steps in multiple distillation columns and this sequence of distillation columns is referred to as a distillation configuration. For instance, Figure 1 shows three well-known distillation configurations for separation of a three-component nonazeotropic feed mixture into three product streams each enriched in one of the components. Similarly, Figures 2 and 3 show 18 possible distillation configurations for separation of a four-component nonazeotropic mixture into four product streams each enriched in one of the components.[2]

In Figures 1–3, and throughout the remainder of the article, reboilers are indicated by nonfilled circles and condensers are indicated by filled circles. Components are defined alphabetically in the decreasing order of their volatilities. Therefore, A is the most volatile component in a distillation configuration, followed by component B, and so on. Moreover, streams other than the main feed stream, such as the stream BC, may contain small amounts of the remaining components, A and D. In fact, in an actual distillation process, these components are always present. Our nomenclature, therefore, denotes that these remaining components are present in levels that are acceptably small.

The distillation configurations of Figures 1–3 are referred to as basic distillation configurations.[2] Each of these basic distillation configurations uses $n-1$ distillation columns, $n-1$ reboilers, and $n-1$ condensers to perform the same n-component separation. Therefore, the capital costs of the basic distillation configurations are not expected to be drastically different. However, the operating costs of these configurations can differ significantly. To minimize the separation cost of the given n-component mixture, it is thus important to identify basic distillation configurations that require less energy to operate.

The feed of an n-component separation problem may actually contain more than n components. Components whose relative volatilities are close to each other are often lumped together to produce n product streams. For such separation problems, we still treat the feed mixture as an n-component mixture, with one or more of A, B, C, and so forth representing lumped mixtures instead of single components.

The vapor duty requirement of a distillation configuration is defined as the sum of vapor flows generated at all its reboilers. The vapor duty requirement is indicative of the

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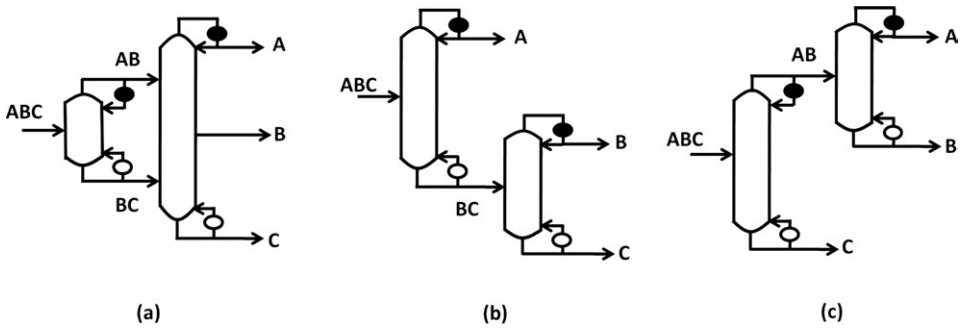

Figure 1. Basic configurations for a three-component mixture.

heat duty requirement of a distillation configuration and can act as a surrogate in assessing the operating cost of the configuration.[3] In this series of articles, our focus is on the need for and development of a robust and reliable quick screening tool that rank-lists distillation configurations based on the minimum total vapor duty requirement, enabling practitioners to choose energy-efficient configurations from the pool of hundreds or thousands of possible configurations for a given separation task.

For separating a given feed into desired product streams, a distillation configuration can have a range of vapor duty requirements depending on the component flow rates and phases of the streams that are transferred between distillation columns of the configurations.[4–8] For example, streams *ABC*,

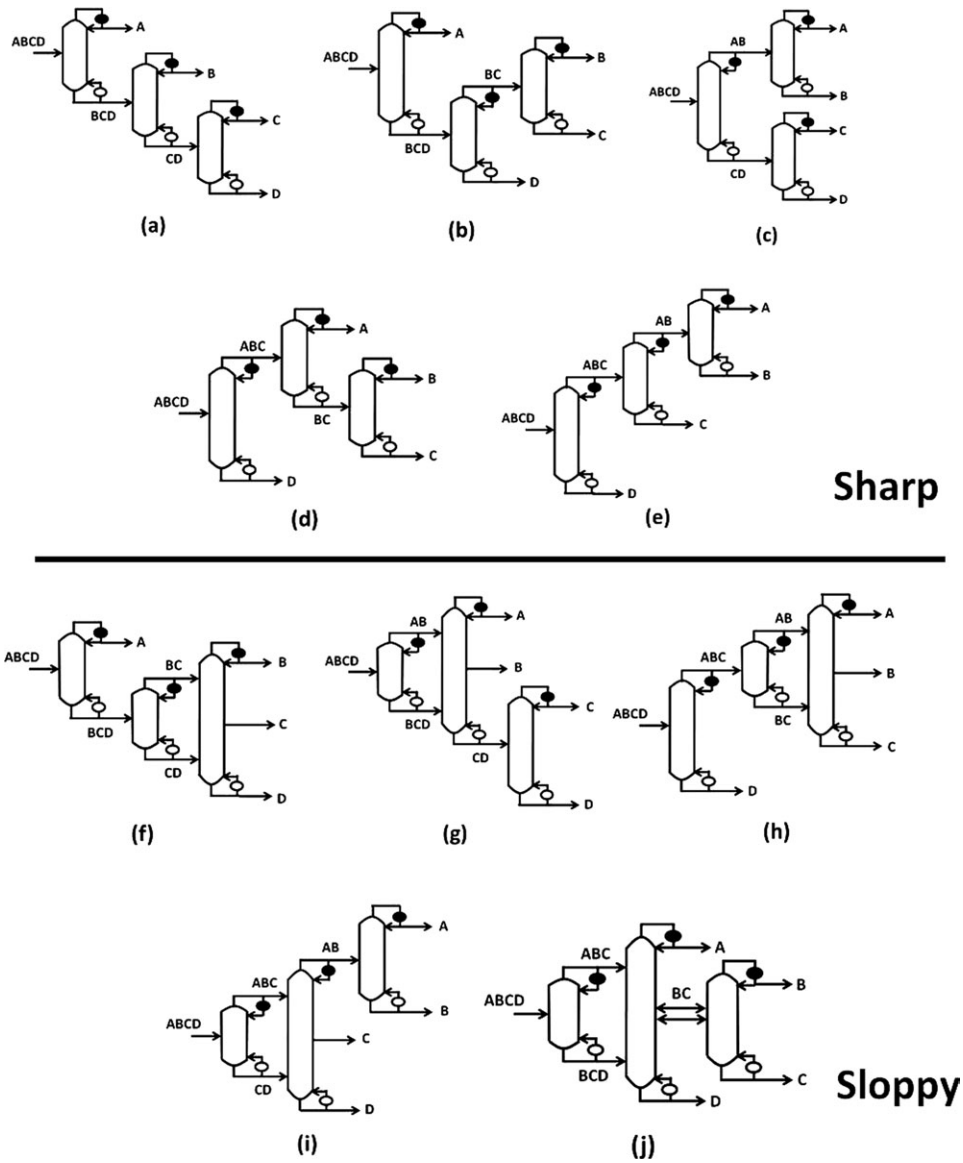

Figure 2. Four-component basic configurations for which the SMA method is a valid global optimization tool.

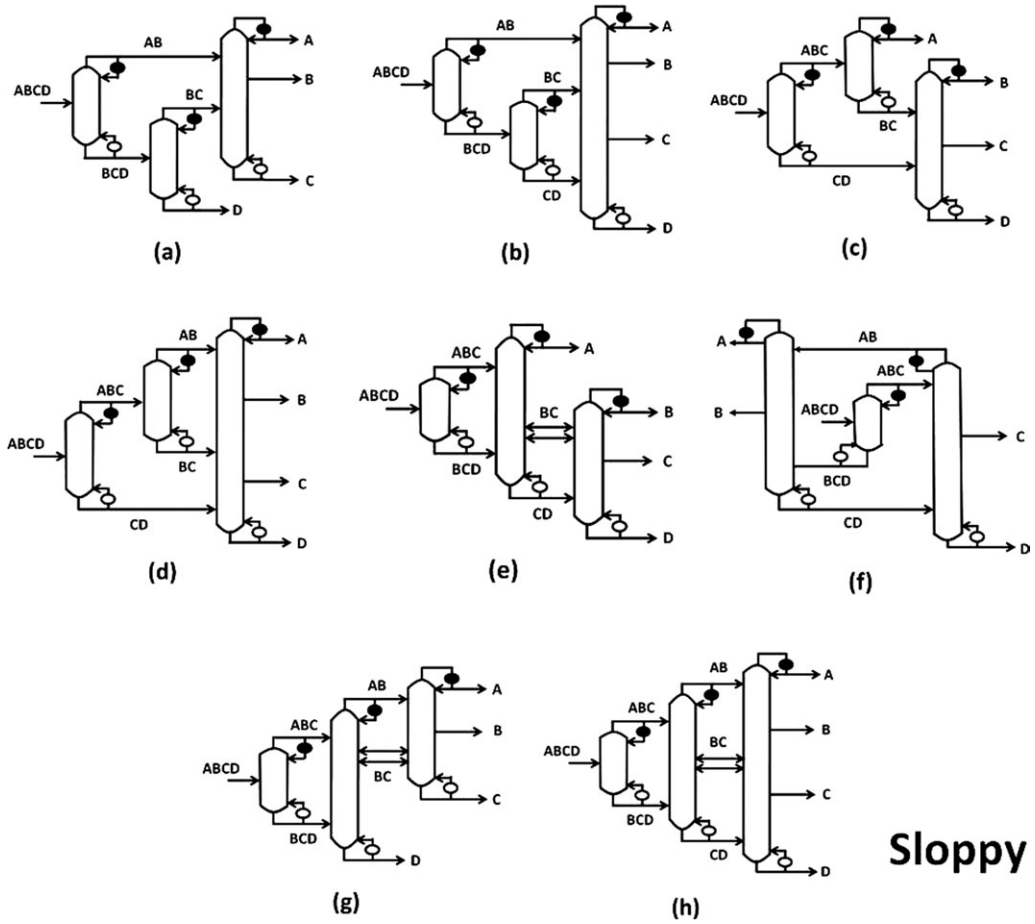

Figure 3. Four-component basic configurations for which the SMA method is not a valid global optimization tool.

BCD, and *BC* are transfer streams in the distillation configuration shown in Figure 4. Here, the component flow rates in the *ABC* and *BCD* streams and the phase of the feed stream *ABCD* affect the minimum vapor duty requirement of the split *ABCD* − *ABC*/*BCD* in the first distillation column. Similarly, the component flow rates and phase of the transfer stream *ABC* affects the minimum vapor duty requirement for split *ABC* − *A*/*BC* in the top portion of the second distillation column and so forth. Therefore, it is essential to choose the optimal component flow rates and phases for all transfer streams to minimize the total vapor duty requirement of the given distillation configuration. Different distillation configurations can then be compared with each other based on their minimum total vapor duty requirement.

Several attempts have been made in the literature to find the minimum vapor flow required for the separation of a given multicomponent feed.[4,7–13] These methods are based on the Underwood[14] equations in which the minimum vapor flow required for a given multicomponent split is obtained through solution of the material balance equations assuming that each section of every distillation column has an infinite number of stages. The first distillation column in the prefractionator arrangement (Figure 1a) carries out the split *ABC* − *AB*/*BC*. Whenever such a nonsharp (sloppy) split of an *n*-component stream takes place into two $n-1$ component streams, there is an optimal distribution of the $n-2$ overlapping components that minimizes the vapor duty requirement for the split of the heaviest and lightest components in the *n*-component mixture. These optimal distributions and the corresponding minimum vapor duty requirement can be analytically calculated using the Underwood equations, and under these conditions, the rectifying section composition profile and stripping section composition profile both have the same pinch compositions. A column is said to be pinched if there is a region in a given distillation column section where the composition of components in liquid and vapor streams remains constant and does not change from stage to stage. The pinch composition equals the composition of the *n*-component feed stream and this special separation is referred to as the preferred separation[15] or the transition split[16,17] of a feed stream. Among all possible ways in which a feed stream can be split into product streams each of which have one less component, the transition split requires the lowest minimum vapor duty.[16,17] Splits involving separation of an *n*-component stream into products with

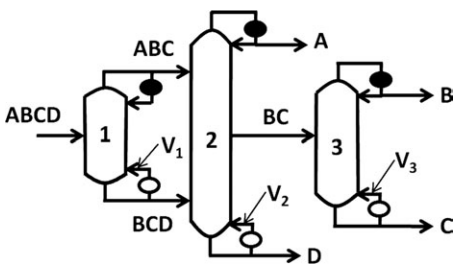

Figure 4. An example of a four-component distillation configuration.

less than $n-2$ overlapping components such as $ABCD - ABC/CD$ also have an optimal distribution of overlapping components that leads to the corresponding minimum vapor flow rates and can be calculated analytically using the Underwood equations. The term "transition split" has been loosely used to describe such separations as well.[9]

One method of "minimizing" the total vapor duty of a given configuration is by calculating the minimum vapor duty for every split that is present in a configuration. First, the feed is split into two desired streams using minimum vapor flow rate. Depending on the configuration, the two resulting streams can have overlap of intermediate volatility components. Then, each subsequent stream is separated in a similar manner using the minimum vapor flow rate required for that particular split. The minimum vapor flow rate for a distillation column is calculated using the minimum vapor flow rates required for each split accomplished within the column. This method yields the minimum total vapor duty requirement for the configuration if one assumes that the most efficient way to operate the configuration is to coincidentally operate each distillation column at the minimum vapor duty requirement for the split(s) it performs. We refer to this method as the sequential minimization algorithm (SMA). This method computes the minimum vapor duty requirement for each split within a given column by adjusting the flow rates of the overlapping components in the outgoing streams. For the distillation configuration shown in Figure 4, the component flow rates and phases of the transfer streams *ABC* and *BCD* are chosen so that they minimize the vapor duty V_1 of the first column. In the same way, component flow rates and phases of the transfer stream *BC* are chosen such that they minimize the vapor duty V_2 of the second column. For column 3, the minimum vapor duty requirement is fixed because the component flow rates and phases of feed and all its product streams are fixed. Total configuration vapor duty is then calculated as sum of the vapor duties of the columns, that is, $V_1 + V_2 + V_3$. It can be seen that the SMA approach does not consider the effect of varying the compositions of streams *ABC* and *BCD* from the first column on V_2.

The SMA method has been advocated in the literature as a reliable method to provide the globally optimal vapor duty requirements for the fully thermally coupled configurations for any n-component separation.[11–13,18] Further, for three-component separation, the SMA method has been shown to yield globally optimal solutions for the prefractionator configuration (Figure 1a).[6] For the remaining three-component configurations (Figures 1b, c), global optimality follows as the associated sharp splits admit only one solution, which is also obtained using the SMA method. The SMA method is, therefore, capable of also identifying global solutions for all the three basic distillation configurations used in three-component separations. However, there is a need to determine whether the SMA method is able to determine the minimum vapor duty for all feasible multicomponent distillation configurations for more than three-component separations.

Alternatively, one can solve a nonlinear programming problem to optimally set the component flow rates and phases of all the transfer streams within the configuration.[10] We refer to this method as the global minimization algorithm (GMA). In the GMA method, the vapor duties in all the columns are determined simultaneously so as to yield the overall minimum vapor duty for the entire configuration. In particular, this method does not constrain each split to its minimum vapor duty requirement. For the distillation configuration shown in Figure 4, the component flow rates and phases of the transfer streams *ABC, BCD,* and *BC* are simultaneously determined such that the total configuration vapor duty, that is, $V_1 + V_2 + V_3$, is minimized. Because of the nonlinear nature of this optimization problem, the GMA method may encounter local optima. It is, therefore, important that GMA is based on solution techniques that guarantee global minimality of the total vapor duty for any given configuration. In Part 2 of this series of articles, we discuss in detail how the optimization problem is formulated and the techniques that have been implemented to guarantee global optimality.

The current literature has suggested but not verified whether the SMA method yields global minimum vapor duty for separations involving more than three components.[9] The advantage of the SMA method as compared to the GMA method is that it is computationally less expensive, as the compositions can be obtained by solving a set of linear equations. Consequently, if the SMA were to yield globally minimum vapor duty requirement of a given configuration then it would be the preferred choice over the GMA method. We thus explore whether the SMA method is able to determine the minimum vapor duty for all feasible multicomponent distillation configurations for a multicomponent separation. For this purpose, we use the analytical procedure provided by Fidkowski[9] for the SMA method to calculate the total vapor duty of a feasible multicomponent distillation configuration, and compare it with the total vapor duty estimated using our GMA strategy (described in Part 2 of this series).

We will show that the SMA method is not able to find the globally optimal vapor duty requirements for a large number of basic distillation configurations that separate mixtures into four and five product streams. We also give a heuristic to identify the basic distillation configurations for which the SMA method yields the global minimum vapor duty. Further, we demonstrate that the SMA method does not find the global minimum vapor duty for some thermally coupled distillation configurations as well. These observations emphasize the need for a robust and reliable global optimization algorithm for minimizing vapor duty.

The Sequential Minimization Algorithm

The SMA, based on the detailed procedure provided by Fidkowski,[9] calculates the total vapor duty requirement of any given distillation configuration. In this section, we summarize the key steps of the SMA.

Each basic distillation configuration carries out a unique set of splits. For instance, the configuration of Figure 2f carries out the following splits: $ABCD - A/BCD$, $BCD - BC/CD$, $BC - B/C$, and $CD - C/D$. Each split is like a pseudo-distillation column whose minimum vapor duty requirement can be estimated using the equations derived by Underwood.[14] By solving the Underwood equations, one not only obtains the minimum vapor duty requirement of a split but also the optimal flow rates of overlapping components (if any) among the product streams of the split. Therefore, for splits that perform sharp separations, such as $ABCD - A/BCD$, the Underwood equations gives the minimum vapor duty requirement of the split, while mass balance equations fix the flow rates of the various components in the product streams *A* and *BCD*. Conversely, for splits involving non-sharp separations, such as $BCD - BC/CD$, the Underwood equations give the minimum vapor duty requirement of the split along with the optimal distribution of the overlapping

component C between the two product streams. For any other distribution of the component C, the minimum vapor duty will be higher. Clearly, the flow rates of the nonoverlapping components (B and D) in the product streams are fixed by mass balance. The SMA method thus solves the Underwood equations analytically to obtain the minimum vapor duty requirement and optimal flows of overlapping components (if any) for each split in a distillation configuration.

In the SMA method, we first obtain the minimum vapor solution for the first feed stream's split by adjusting the composition of the overlapping components between the two separated streams and the phase of the feed stream. All the information required for this calculation is available from the problem definition. Thereafter, the procedure iterates for each column. The flow rate and composition information for subsequent splits is available once the previous column's solution has been obtained. Nevertheless, the thermal qualities (q) of the various transfer streams in a configuration need to be calculated, as this information is not available from the solution of the Underwood equations.

One feature of the basic distillation configurations shown in Figures 1–3 is that transfer streams associated with reboilers are transferred as liquids, whereas those associated with condensers are transferred as vapors. These transfer stream phase choices have been found to minimize the vapor duty requirements of the basic distillation configurations.[6,9,19] Therefore, in basic distillation configurations, the quality of the inter column transfer streams associated with reboilers and condensers are fixed, and they are single-phase transfer streams (liquid-only or vapor-only, i.e., $q = 1$ or 0). For all the calculations in this work, it is assumed that all the final product streams (including the sidedraw final product streams) are produced as liquids (i.e., $q = 1$).

For the liquid and vapor portions of a thermally coupled or a sidedraw stream, we adopt the sign convention that streams flowing in the direction of the net stream flow are positive, whereas streams flowing in the direction opposite to the net stream flow are negative. For a thermally coupled transfer stream obtained by replacing a reboiler, the vapor portion of the stream is set equal to the vapor duty requirement at the bottom of the distillation column and this value is negative as the vapor flows in a direction opposite to the net stream flow to the next column. The liquid portion of the stream flow is then obtained by a net mass balance with the net flow and is always positive. The q of such a stream is the ratio of the liquid portion of the stream flow to the net stream flow, and it can be seen that this ratio will be always greater than 1. Conversely, for a thermally coupled transfer stream obtained by replacing a condenser, the vapor portion of the stream is set equal to the vapor duty requirement at the top of the distillation column and this value is positive as the vapor flows in the direction of the net stream flow. The liquid portion of the stream flow is obtained by a mass balance and is always negative resulting in a negative value of q. These features can be seen in Figure 5.

Conversely, sidedraw transfer streams in basic configurations are actually the top product of one split and the bottom product of another split. For example, in Figure 2j, stream BC is the top product of split $BCD - BC/D$, and the bottom product of split $ABC - A/BC$. The reboiler and condenser associated with producing the sidedraw stream from these splits are eliminated while stacking these splits in a single distillation column. Therefore, such sidedraw transfer streams are no longer associated with reboilers or condensers

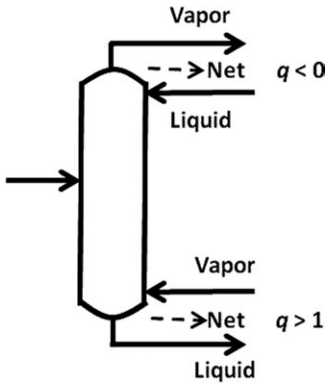

Figure 5. Liquid and vapor flow directions in thermal coupling links.

and thus not fixed to be single-phase transfer streams. In fact, they are not even restricted to be unidirectional transfers.[20] Sidedraw transfer streams are thus allowed to be two-way liquid–vapor communications between the distillation columns. An example that illustrates the vapor duty benefit of treating sidedraw transfer streams as two-way liquid–vapor communication is described in the Appendix. Consequently, we extend the SMA method for sidedraws by allowing the excess vapor flow available in one distillation column to be used in another distillation column. For example, in Figure 2j, the vapor portion of the sidedraw stream between the second and third column is set equal to the difference between the minimum vapor duty requirements of the top and bottom section splits ABC and BCD. If the vapor flow requirement of split BCD is greater than that of split ABC, then the excess vapor from the bottom section of the second column goes to the third column. Conversely, if vapor flow requirement of split BCD is lower than that of split ABC, then the excess vapor needed for the top section of the second column is not generated by the reboiler of the second column, but is satisfied by generating extra vapor in the reboiler of the third column and transferring through the sidedraw stream to the second column. The third column reboiler is now generating vapor for not only the splits in that column but also for the excess requirement of the ABC split in the second column. The liquid portion is then determined so as to satisfy the mass balance. The sidedraw transfer stream's q is thus not restricted to be between 0 and 1. This procedure ensures that column vapor duties are optimized in a sequential manner. In this procedure, any column under consideration is optimized first and the additional vapor duty if required is passed on to the next column in sequence.

It is worth mentioning that, in this article, we have not considered the capital cost or configurations having reboilers and condensers at intermediate column locations.[12,21] Such additional considerations can also be included when appropriate. Our focus is on comparing basic and thermally coupled configurations for their vapor duty requirement.

Four-Component Case Study

Figures 2 and 3 illustrates the complete set of basic distillation configurations (18 in total) that can be used to separate a four-component mixture into four product streams, each of which is enriched in one of the components.[2] As the vapor duty requirement of each basic configuration depends on the feed composition, the relative volatilities of the feed

Table 1. Different Relative Volatilities for a Four-Component Feed Mixture Considered in this Study[22]

α_{AB}	α_{BC}	α_{CD}
2.5	2.5	2.5
2.5	2.5	1.1
2.5	1.1	2.5
2.5	1.1	1.1
1.1	2.5	2.5
1.1	2.5	1.1
1.1	1.1	2.5
1.1	1.1	1.1

components, and the liquid fraction (or thermodynamic state) of the feed, these three specifications are collectively referred to as the feed parameters. Using the SMA method, we estimate the vapor duty requirement of each basic configuration for a variety of feed parameters and compare this value with the corresponding globally optimal vapor duty requirement obtained using the GMA method. We use the 120 feed parameters considered by Giridhar and Agrawal[22] that were chosen to span a wide range of four-component separations. Tables 1 and 2 are reproduced from the article by Giridhar and Agrawal.[22] They describe eight different relative volatility cases and 15 different feed compositions for a four-component mixture. Combining these volatilities and compositions, we obtain the 120 (8×15) feed parameters used in the study. Without loss of generality, we assume that the feed and the products are saturated liquids. For each of the 120 feed parameters, the minimum vapor duty requirement for each of the 18 basic configurations is estimated using the SMA method and the GMA method. Therefore, we solve each of the $120 \times 18 = 2160$ cases using the SMA method as well as the GMA method.

To quantify how close the vapor duty estimated by the SMA method is to the globally minimum vapor duty, we calculate the percentage difference between these vapor duty requirements. We then obtain the maximum percentage difference of the vapor duties calculated using the SMA method and the GMA method for each basic configuration as follows.

$$\text{Maximum percentage difference} = \text{Max}\left(\frac{V_{\text{SMA},f} - V_{\text{GMA},f}}{V_{\text{GMA},f}} \times 100\right) \text{ for } f = 1 - 120$$

Table 2. Different Feed Compositions (in mol %) for a Four-Component Mixture[22]

A	*B*	*C*	*D*
85	5	5	5
5	85	5	5
5	5	85	5
5	5	5	85
5	31.7	31.7	31.7
31.7	5	31.7	31.7
31.7	31.7	5	31.7
31.7	31.7	31.7	5
5	5	45	45
5	45	5	45
5	45	45	5
45	5	5	45
45	5	45	5
45	45	5	5
25	25	25	25

Table 3. Maximum Percent Difference Between the Total Minimum Vapor Duty Requirements of a Configuration as Calculated by the SMA Method and the GMA Method for the Feed Conditions Given in Tables 1 and 2

Figure	Maximum % Difference
2a	0.000000
2b	0.000000
2c	0.000000
2d	0.000000
2e	0.000000
2f	0.000001
2g	0.000001
2h	0.000004
2i	0.000001
2j	0.000002
3a	26.716263
3b	28.301403
3c	23.031005
3d	24.677187
3e	15.653706
3f	23.792633
3g	15.774198
3h	6.714109

The maximum percentage difference values for each configuration are listed in Table 3.

It can be seen from Table 3, that for some of the basic configurations, the SMA method yields solutions that are reasonably close to the global optimum as evidenced by the small maximum percentage difference ($\leq 0.000004\%$) over a wide range of feed parameters. These configurations are shown in Figure 2. Conversely, for the configurations where the solution using the SMA method is far from the globally optimal value (Figure 3), the maximum percent difference is observed to be in the range of 6.7–28.3%. It should be noted that as the tabulated values are maximum percent differences, they correspond to the feed parameters for which the GMA predicts a vapor duty most apart from that obtained from via SMA. Conversely, for the 18 configurations, the minimum percentage difference was never more than $10^{-8}\%$, indicating that even for configurations of Figure 3, there exist certain feed parameters, for which the SMA method estimates the globally minimum vapor duty. As there is a significant number of configurations for which the SMA method is far from optimal, it is not reasonable to use the SMA method and assume that it can be used to identify the basic configurations that are energy efficient.

Quick Identification of Basic Configurations for which SMA Method Consistently Provides Global Minimum Vapor Duty

The SMA method provides globally optimal solutions for 10 of 18 basic four-component distillation configurations. Among these 10 configurations, five are sharp split configurations (Figures 2a–*e*), whereas the other five are sloppy split configurations (Figures 2f–j). A sharp split basic configuration admits only one feasible solution because of mass balance equations and this solution is also obtained by the SMA method. Therefore, for basic sharp split distillation configurations, the SMA method globally minimizes the vapor duty irrespective of the feed parameters and the number of components undergoing separation. However, the SMA method yields globally optimal solutions even for some nonsharp split basic distillation configurations (Figures 2f–j). We propose the following rule as a heuristic for

Table 4. Different Relative Volatilities for a Five-Component Mixture

α_{AB}	α_{BC}	α_{CD}	α_{DE}
2.5	2.5	2.5	2.5
2.5	2.5	2.5	1.1
2.5	2.5	1.1	2.5
2.5	2.5	1.1	1.1
2.5	1.1	2.5	2.5
2.5	1.1	2.5	1.1
2.5	1.1	1.1	2.5
2.5	1.1	1.1	1.1
1.1	2.5	2.5	2.5
1.1	2.5	2.5	1.1
1.1	2.5	1.1	2.5
1.1	2.5	1.1	1.1
1.1	1.1	2.5	2.5
1.1	1.1	2.5	1.1
1.1	1.1	1.1	2.5
1.1	1.1	1.1	1.1

identifying the configurations for which the SMA method yields globally optimal solutions:

Consider nonsharp separation of a feed stream in first distillation column. If both products of the nonsharp separation go to the same second distillation column, and do not undergo any further nonsharp separations, then the SMA method yields globally minimum vapor duty.

For example, consider the configuration of Figure 2f. The nonsharp splits *BC* and *CD* obtained from *BCD* are fed to the same distillation column where they undergo sharp splits into *B*, *C,* and *D*. As suggested by the heuristic, the SMA method provides the global optimal solution for this configuration. The heuristic is also satisfied by the remaining configurations in Figures 2g–j. Further, the eight basic configurations shown in Figure 3 violate the heuristic rule and correspondingly the SMA method does not yield the globally minimum vapor duty for these configurations (Table 3).

Five-Component Case Study

For a five-component mixture, there are 203 possible basic distillation configurations that can be used to separate the mixture into five product streams.[10,20,23,24] We explore the validity of the SMA method for these 203 basic distillation configurations. Tables 4 and 5 describe 16 different relative volatility cases and 31 different feed composition cases, respectively, for a five-component mixture. Together, these combinations yield $16 \times 31 = 496$ different five-component feed parameters. Once again, we assume without loss of generality that along with these feed parameters, the feed and the products are saturated liquids.

We estimate the minimum vapor duty requirements of each of the 203 basic configurations for each of the 496 feed parameters. Therefore, we evaluate a total of $203 \times 496 = 100{,}688$ cases using both the SMA and GMA methods. As before, we compute the percentage difference between the values calculated using the two methods and report the maximum percentage difference to illustrate the difference in vapor duties computed by the two methods.

As expected, the SMA method yields globally minimum vapor duty for all the 14 sharp split basic distillation configurations. For the remaining 189 configurations, which perform nonsharp splits, we observe that the SMA method yields globally minimum vapor duty for only 24 configurations. We have verified that these 24 configurations also satisfy the heuristic rule we proposed before. In summary, irrespective of feed parameters, the SMA method provides solutions that are close to globally optimal solutions for 38 of 203 basic configurations, and these are configurations that perform only sharp splits or obey our proposed heuristic. For the feed parameters used in our calculations, the maximum percentage difference between the minimum vapor duties calculated using the two methods, is never greater than 0.016%. This difference may be attributed to the numerical tolerances. For the remaining 165 configurations, the maximum percentage difference between the minimum vapor duties ranges from 0.58% to 83.2% where, in each case, the SMA method yields higher minimum total vapor duty than the GMA method. In fact, for many five-component basic distillation configurations, the calculated minimum vapor duty by the SMA method is significantly higher than the global optimal value. We have also verified that the 165 sloppy split distillation configurations violate our proposed heuristic.

Extension to Thermally Coupled Configurations

Thermal coupling links are an important means for reducing the heat duty requirements of the distillation configurations.[5] It is generally believed that the heat duty savings come at the cost of requiring a higher temperature heat and/or a lower temperature cold utility. However, it has been recently shown that in certain cases, thermal coupling can also provide heat duty savings without such associated penalties.[25,26] Thermal coupling links act as replacements for reboilers or condensers in a configuration and are two-way liquid–vapor communications between distillation columns.

Table 5. Different Feed Compositions (in mol %) for a Five-Component Feed Mixture

A	*B*	*C*	*D*	*E*
80	5	5	5	5
5	80	5	5	5
5	5	80	5	5
5	5	5	80	5
5	5	5	5	80
42.5	42.5	5	5	5
5	42.5	42.5	5	5
5	5	42.5	42.5	5
5	5	5	42.5	42.5
42.5	5	42.5	5	5
5	42.5	5	42.5	5
5	5	42.5	5	42.5
42.5	5	5	42.5	5
42.5	5	5	5	42.5
30	30	30	5	5
5	30	30	30	5
5	5	30	30	30
30	5	30	30	5
30	5	5	30	30
30	5	30	5	30
30	5	5	30	30
5	30	5	30	30
5	30	30	5	30
30	30	5	30	5
30	30	5	5	30
23.75	23.75	23.75	23.75	5
23.75	23.75	23.75	5	23.75
23.75	23.75	5	23.75	23.75
23.75	5	23.75	23.75	23.75
5	23.75	23.75	23.75	23.75
20	20	20	20	20

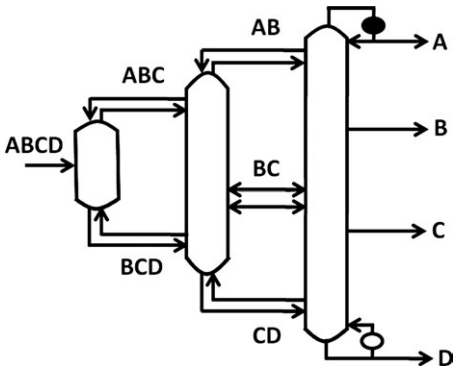

Figure 6. Fully thermally coupled configuration for a four-component mixture.

The thermally coupled distillation configurations can be easily derived from the basic distillation configurations.[2,27] A thermally coupled configuration can be either a partially thermally coupled configuration, or a completely thermally coupled configuration, or a fully thermally coupled configuration.[22]

The SMA approach has been used to estimate the vapor duty requirements of some fully thermally coupled configurations. Fully thermally coupled configurations are commonly referred to as Petlyuk[28] configurations. Fidkowski and Krolikowski[4] demonstrated that the SMA method yields the globally optimal vapor duty requirement for the three-component fully thermally coupled configuration. This result was also shown in the work of Nikolaides and Malone.[7] Fidkowski and Agrawal[8] used numerical examples to demonstrate that the SMA method provides the globally optimal vapor duty requirement for the four-component fully thermally coupled configuration (Figure 6). Further, they anticipated (without proof) that the SMA method will provide the globally optimal vapor duty requirement for fully thermally coupled configurations separating more than four components as well.

Halvorsen and Skogestad[11–13] developed a graphical representation of the calculations involved in the Underwood equations for a single split. This representation is referred to as the $V_{\min}$ diagram. Using the concept of "carrying over" of Underwood roots due to thermal coupling links,[29] they demonstrated that the $V_{\min}$ diagram of the main feed split can in fact represent all the remaining splits in a fully thermally coupled distillation configuration, as long as each split is operated at its preferred separation. The $V_{\min}$ diagram for a fully thermally coupled distillation configuration can thus be thought of as a graphical representation of the SMA method applied to the fully thermally coupled configuration. Using the $V_{\min}$ diagram, Halvorsen and Skogestad[13] proved that the conjecture of Fidkowski and Agrawal[8] was indeed true, whereby the SMA method provides the global optimal vapor duty requirements for fully thermally coupled configurations separating more than four components as well.

The fully thermally coupled configuration for a four-component separation (Figure 6) is derived from the four-component basic configuration in Figure 3h by replacing the condensers and reboilers that are not associated with final products, with thermal coupling links. Table 3 shows that the SMA method does not provide the globally optimal vapor duty requirement for the basic configuration shown in Figure 3h. Therefore, the SMA method yields the minimum vapor duty for the configuration when thermal coupling is present but it does not yield the minimum vapor duty for the corresponding basic configuration. This implies that if one obtains the global optimum for a configuration with thermal coupling using the SMA method, it is not necessarily true that the corresponding basic configuration without thermal coupling links can also be solved to global optimality using the SMA method.

Similarly, even though the SMA method reaches globally optimal solutions for some of the basic distillation configurations, the SMA method does not yield global solution when thermal coupling is introduced in these basic configurations. For instance, the configuration shown in Figure 2c is a sharp split basic distillation configuration and, as such, can be solved to global optimality using the SMA method irrespective of the feed conditions. The configuration shown in Figure 7 is a thermally coupled sharp split distillation configuration derived from the configuration of Figure 2c. Let us consider separation of a saturated liquid mixture that contains 45% *A*, 5% *B*, 5% *C*, and 45% *D* (in mol %) using the configuration depicted in Figure 7. Let the relative volatilities of *A*, *B*, and *C* each with respect to *D* be 3.025, 2.75, and 1.1, respectively. For the separation of 1 mol per unit time of the feed mixture, the SMA method provides a vapor flow of 1.01 mols per unit time, 5.56 mols per unit time, and 4.17 mols per unit time in the first, second, and third distillation columns, respectively. These vapor flows are equal to the vapor duty requirements of the corresponding splits in the distillation columns. In this configuration, as the reboiler of the first distillation column has been replaced by a thermal coupling link, the vapor generated in the third column is 5.18 mols per unit time, from which 1.01 mols per unit time are transferred to the first column through the thermal coupling link. Further this vapor is subsequently used in the second column through the thermal coupling link between first and second column. After taking into account this vapor flow, the vapor flow that needs to be generated at the reboiler of second column is only 4.55 mols per unit time. Therefore, the total vapor duty requirement of the distillation configuration is $4.55 + 5.18 = 9.73$ mols per unit time as per the SMA method. However, GMA solution of this distillation configuration results in a total vapor duty requirement of 8.40 mols per unit time. Therefore, a 13.6% reduction in the vapor duty requirement is obtained. The GMA solution

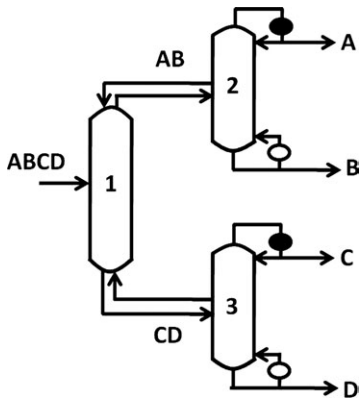

Figure 7. Thermally coupled sharp split configuration (derived from the configuration shown in Figure 2c).

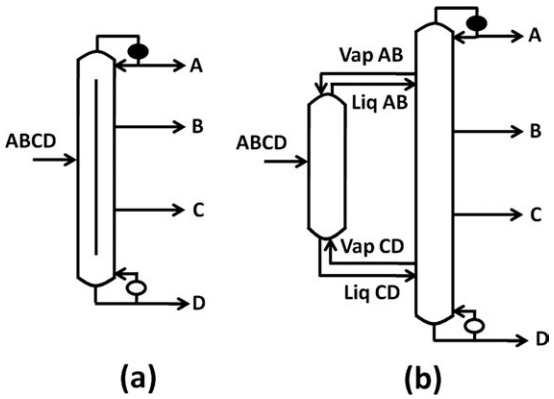

Figure 8. (a) Kaibel arrangement and (b) its thermodynamically equivalent predecessor.

provides a vapor flow of 4.20 mols per unit time in the first distillation column, 2.20 mols per unit time in the second distillation column, and 2.0 mols per unit time in the third distillation column. Here, vapor generated in the third column is 6.20 mols per unit time from which 4.20 mols per unit time are transferred to the first column through the thermal coupling link.

A split in a distillation column is said to be "pinched" if the vapor flow in the corresponding portion of the distillation column is just sufficient to meet the minimum vapor duty requirement of the split. The above analysis illustrates that if a distillation configuration is operated at its SMA solution; all its distillation columns that perform a single split are pinched. In the GMA solution, the minimum vapor duty requirement of the first distillation column is still 1.01 mols per unit time, but the vapor flow provided is 4.20 mols per unit time. The GMA solution thus allows the first distillation column to have a vapor flow surplus. This results in decreasing the vapor flow requirement of the second column from 4.55 to 2.2 mols per unit time. Thus, unpinching the first distillation column allows us to create a synergistic link between the second and third distillation columns through the top and bottom product thermal coupling links of the first distillation column, whereby a greater fraction of the vapor flow from the third distillation column can perform some of the separation in the second distillation column. The GMA method thus finds a significantly lower overall vapor duty requirement for the distillation configuration of Figure 7.

One implication of this analysis is that irrespective of whether or not the SMA method calculates the minimum vapor duty of a basic configuration, one cannot ascertain if SMA will yield globally optimal vapor duty for the corresponding thermally coupled configuration. We have not yet identified a general heuristic (as was done in the case of basic distillation configurations) that identifies the thermally coupled distillation configurations for which the SMA method yields global optima.

The above analysis reveals an interesting insight when one considers the Kaibel arrangement.[30] The Kaibel arrangement is shown in Figure 8a and is thermodynamically equivalent to the configuration of Figure 8b. The configuration of Figure 8b was actually discovered before the Kaibel arrangement.[31] It can be seen that the only difference between the configuration shown in Figure 7 and the arrangement in Figure 8b is the introduction of an additional separation section between the withdrawal locations of streams B and C. Halvorsen et al.[32,33] showed through the V_{min} diagram that SMA method yields the minimum vapor duty requirement for the Kaibel arrangement. This is in agreement with our computations where we find that SMA method yields the global minimum vapor duty for all the 120 four-component feed parameters for the Kaibel arrangement. However, if the additional section between streams B and C is removed, SMA method yields inferior solutions. This is because when an extra section is introduced between columns 2 and 3 of Figure 7, the GMA does not need to divert the extra vapor flow through the column 1 to column 2 (Figure 7) and can instead transfer the vapor directly from column 3 to column 2 (Figure 8b). Observe that unlike the proposed heuristic, the column 1 in the Kaibel arrangement is a sharp split. Therefore, SMA method yields global solutions for the configuration shown in Figure 8a. However, the same does not hold for configuration shown in Figure 7, where the outgoing streams from column 1 feed into two different columns namely 2 and 3. In summary, when one introduces thermal coupling links in the configuration of Figure 2c to obtain the configuration depicted in Figure 7, the SMA method yields inferior solutions. Further introduction of a separation section yields the configuration of Figure 8, for which the SMA method again yields globally optimal solutions.

Conclusions

We have computationally demonstrated that the SMA method, which minimizes the vapor duty needed for each distillation column separately, does not always yield the overall minimum vapor duty for the entire configuration. Inspired from our computations, we designed a heuristic classification tool to quickly identify the basic distillation configurations for which the SMA method yields global optima. This heuristic also classifies five-component basic distillation configurations correctly.

For three-component separations, the SMA method provides globally minimum vapor duty for each of the three basic configurations. For four-component separations, the SMA method always finds the global minimum vapor duty for 10 of 18 basic configurations. For five-component separations, the SMA method always find the global minimum vapor duty for only 38 of 203 basic configurations. Therefore, it appears that the fraction of basic distillation configurations for which the SMA method discovers the global minimum vapor duty decreases rapidly as the number of components in the feed increases. Also, the SMA method has been observed to give vapor duty requirements up to 83% higher than the globally optimal vapor duty requirements for some of the feed parameters considered in this work. All these observations emphasize the need for a global optimization algorithm that is robust and works for all distillation configurations that are admissible especially when the number of components in the feed is large.

We have also demonstrated that the SMA method's ability to obtain global minimum for a configuration with thermal couplings cannot be judged from its ability to do the same for the corresponding basic configuration and *vice versa*. We conclude that the SMA method is not a reliable tool to estimate the global minimum vapor duty for several configurations. Instead, a robust and reliable global optimization algorithm that can rank-list all the feasible configurations within a given search space based on total minimum vapor

duty requirement, is imperative to screen the configurations that are worthy of thorough investigation.

Acknowledgments

The authors thank the US Department of Energy, award DE-FG36-06GO16104 for providing financial support for this work.

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Appendix: Flow Directions for Sidedraw Streams

In the configuration of Figure 2j, transfer stream *BC* is produced as a sidedraw from the second distillation column. Let us consider the separation of 1 mol per unit time of a saturated liquid mixture containing 5% *A*, 45% *B*, 5% *C,* and 45% *D* (in mol %) using the configuration of Figure 2j for different possible flow directions of the stream *BC*. Let the relative volatilities of *A*, *B,* and *C* with respect to *D* be 3.025, 1.21, and 1.1, respectively.

First, we restrict the stream *BC* to be one-way communication as (1) saturated liquid, or (2) as saturated vapor, or (3) as a two-phase stream from the second distillation to the third distillation column (Figure A1-a). The configuration of Figure A1-a performs the splits *ABCD* − *ABC/BCD*, *ABC* − *A/BC*, *BCD* − *BC/D* and *BC* − *B/C*. Global optimization of the configuration of Figure A1-a results in a stream *BC* that is in vapor phase and the resulting vapor duty requirements for the split *ABCD* − *ABC/BCD*, *BCD* − *BC/D*, *ABC* − *A/BC,* and *BC* − *B/C* are 0.60 mols per unit time, 5.88 mols per unit time, 0.06 mols per unit time, and 5.00 mols per unit time, respectively.

One mole per unit time of the feed mixture has 0.45 mols per unit time of component *B* and 0.05 mols per unit time of component *C*. Then, mass balance dictates that the net flow of the stream *BC* from the second distillation to the third distillation column must be $0.45 + 0.05 = 0.50$ mols per unit time.

In the second distillation column of Figure A1-a, the vapor flow in the bottom section is 5.88 mols per unit time. This vapor flow meets the requirement for the *BCD* − *BC/D* split. Moreover, after optimization of this configuration, it is observed that the entire 0.50 mols per unit time of the stream *BC* is transferred as vapor from the second distillation column to the third distillation column, and there is no transfer of liquid *BC* between the distillation columns. The vapor traffic via stream *BC* reduces the burden of the split *BC* − *B/C*. Therefore, the vapor flow in the second distillation column above the location where the stream *BC* is withdrawn is $5.88 - 0.50 = 5.38$ mols per unit time. As the vapor duty requirement of split *ABC* − *A/BC* in the second distillation column is 0.06 mols per unit time, there is a surplus vapor flow of $5.38 - 0.06 = 5.32$ mols per unit time in the corresponding section of the second distillation column. Therefore, the vapor duty requirement of the distillation configuration in Figure A1-a is $0.60 + 5.88 + 5.00 = 11.48$ mols per unit time.

Next, we drop the restriction on stream *BC* and allow it to be either a two-way liquid or a two-way vapor communication or both. In other words, we let the optimization algorithm to choose the eventual flow direction between the two distillation columns. This freedom allows the surplus vapor

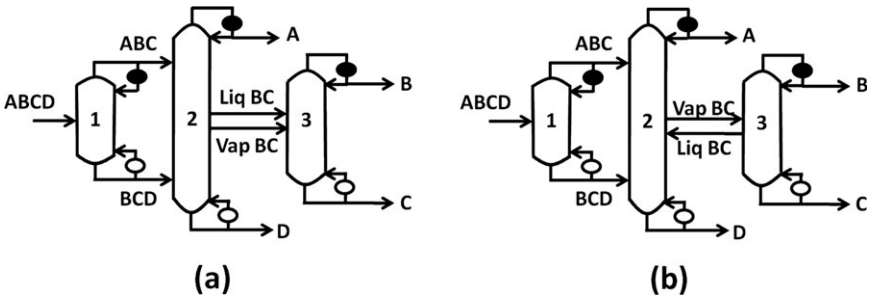

Figure A1. Optimization of the configuration shown in Figure 2j with (a) sidedraw restricted to be a one-way liquid–vapor transfer and (b) sidedraw allowed to be a two-way liquid–vapor transfer.

flow of the second distillation column to perform part of the separation of the third distillation column. Indeed, the optimal solution directs the entire 5.32 mols per unit time of the surplus vapor flow along with the previously diverted 0.50 mols per unit time of vapor *BC* from the second distillation column to the third distillation column, resulting in a net vapor *BC* flow of 5.82 mols per unit time between the distillation columns. To maintain mass balance, 5.32 mols per unit time of liquid *BC* are transferred back from the third distillation column to the second distillation column. Utilization of the surplus vapor flow results in a vapor duty requirement of 8.01 mols per unit time for the configuration of Figure 2j with the sidedraw flow directions as shown in Figure A1-b. The vapor duty requirements of splits $ABCD - ABC/BCD$, $ABC - A/BC$, and $BCD - BC/D$ for the configuration in Figure A1-b remain the same as for the Figure A1-a, whereas the vapor duty requirement of split $BC - B/C$ (with feed q of -10.63 to the third column) from the bottom reboiler of the third column reduces from 5.00 to 1.53 mols per unit time due to the feed of vapor stream *BC* of 5.82 mols per unit time. Therefore, by allowing the sidedraw streams to be two-way communications between the distillation columns during the optimization, a 30% reduction in the vapor duty requirement is realized for this configuration.

From a mathematical perspective, restricting a sidedraw to be a one-way communication is equivalent to adding nonnegativity constraints on the liquid and vapor portions of the sidedraw flows in the optimization problem. Conversely, a two-way sidedraw transfer stream is enabled by removing these constraints from the optimization problem. Therefore, optimization with sidedraws as two-way liquid–vapor communications relaxes the feasible region and allows the choice of the preferred direction of flow for each vapor and liquid sidedraw streams yielding lower, or at least never higher, vapor duty requirement for any distillation configuration.

Manuscript received Dec. 7, 2011, and revision received May 22, 2012.