Iranian Journal of Oil & Gas Science and Technology, Vol. 5 (2016), No. 2, pp. 66-83
http://ijogst.put.ac.ir

Three-component Distillation Columns Sequencing: Including Configurations with Divided-wall Columns

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Received: November 23, 2015; *revised:* January 01, 2016; *accepted:* March 02, 2016

Abstract

In the present work, the exergy analysis and economic study of 3 different samples of three-component mixtures have been investigated (ESI>1, ESI≈1, and ESI<1). The feed mixture has been tested under three different compositions (low, equal, and high contents of the intermediate component). A quantitative comparison between simple and complex configurations, considering thermally coupled, thermodynamically equivalent, and divided-wall column (DWC) has been carried out. The results present that the best sequence could be found by TAC or exergy loss rate analysis. Complex sequences have greater exergy losses in comparison to simple sequences. Despite expectations, the Petlyuk sequence only performs well in a few cases and poorly on others. According to the results, as the amount of intermediate component in the feed increases, both TAC and exergy losses of each sequence increase. The results also demonstrated that the occurrence frequency as the best sequence for DWC, thermodynamically equivalent, thermally coupled, and basic sequences are 36%, 28%, 25%, and 11% respectively. According to authors' best knowledge, a quantitative exergy and cost comparison (based on rigorous simulation and optimization) between these configurations have never been carried out all together before.

Keywords: Distillation Sequence, Exergy Analysis, Divided-wall Column, Separation Matrix

1. Introduction

Distillation process is still the most promising separation technique used in oil, gas, chemical, and petrochemical industries. But this process in most cases consumes a lot of energy, which is the greatest part of operating costs in these industries. Thus improving the energy consumption of distillation processes is still an interesting field of study.

Industrial mixtures commonly contain more than two components and these separation tasks could not be implemented efficiently in a single column. Hence it is required to employ a number of columns for the separation of multicomponent mixtures to the number of desired products. This leads to many possible configurations (sequences) for separating a multicomponent mixture into relatively pure

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products (sharp split) or several multicomponent product streams (non-sharp or sloppy split). On the other hand, the distillation sequences for separating an *n*-component feed could be classified in accordance with the number of distillation columns: having less than *n*-1 columns (intensified or reduced), exactly *n*-1 columns (basic), or more than *n*-1 columns (Gridhar et al., 2010). The basic sequences are divided into two categories: simple basic sequences, in which columns have one feed and two products from condenser on top and reboiler at the bottom (direct, indirect); and basic complex sequences, in which at least one column has more than one feed or has side products (pre-fractionator).

There are more categories which could be generated from basic configurations: thermally coupled (TC), thermodynamically equivalent (TE), and divided-wall columns (DWC). Figure 1 presents the different categories of three-component distillation sequences. A thermal coupling configuration could be generated by the substitution of a condenser and/or a reboiler not associated with the final product streams with a bidirectional vapor-liquid connection. Fully thermally coupled (FTC) configurations are those in which all the vapor requirements of the sequence are supplied by a single reboiler and the entire reflux by a single condenser. The FTC with an external pre-fractionator is a Petlyuk configuration (Caballero et al., 2013). The thermodynamically equivalent configurations could be generated from the thermally coupled sequences through moving one column section associated to a condenser and/or a reboiler which provides the common reflux flow rate or the vapor boil up between two consecutive columns. Divided-wall column sequences are other categories, which could be considered to reduce investment costs (Caballero et al., 2013). These configurations consist of two columns arranged in a single shell and divided by an internal wall.

Earlier approach to synthesis distillation schemes was to use experience-based heuristic rules (Seader et al., 1977; Tedder et al., 1978; Westerberg, 1985). Heuristic-rule-based methods might lead to feasible solutions but not necessarily the optimum configuration. A true optimal scheme could be found precisely by a mathematical programming approach. The brief review and work performed by Gridhar and Agrawal indicates that in order to achieve the optimum configuration, the first and most important step is to predefine the search space as complete as possible (Gridhar et al., 2010). One of the early methods, introducing a superstructure based on “states” and “tasks” was proposed initially by Sargent and Gaminibandara (Sargent et al., 1976). This superstructure could be used in mixed integer linear programming (MILP) (Doherty et al., 2001) or mixed integer nonlinear programming (MINLP) (Caballero et al., 2004 and 2006) to find the optimum sequence. Simple and complex distillation schemes could be considered with this superstructure.

Another systematic approach to synthesize distillation column sequences based on the column products position, which could be distillate, bottoms, or side streams, was proposed by Agrawal (2010). Recently Errico et al. presented a simple 4-step method for the systematic synthesis of the search space considering the generation information saving from one configuration to another (Errico et al., 2009 and 2014). Ivakpour and Kasiri introduced a method which generates simple and/or complex distillation columns and bypass streams by introducing a separation matrix (Ivakpour et al., 2008) to synthesize complete as well as reduced sequences. Later Khalili-Garakani et al. extended the separation matrix method to cover thermally coupled, thermodynamically equivalent, and divided-wall column sequences (Khalili-Garakani et al., 2015).

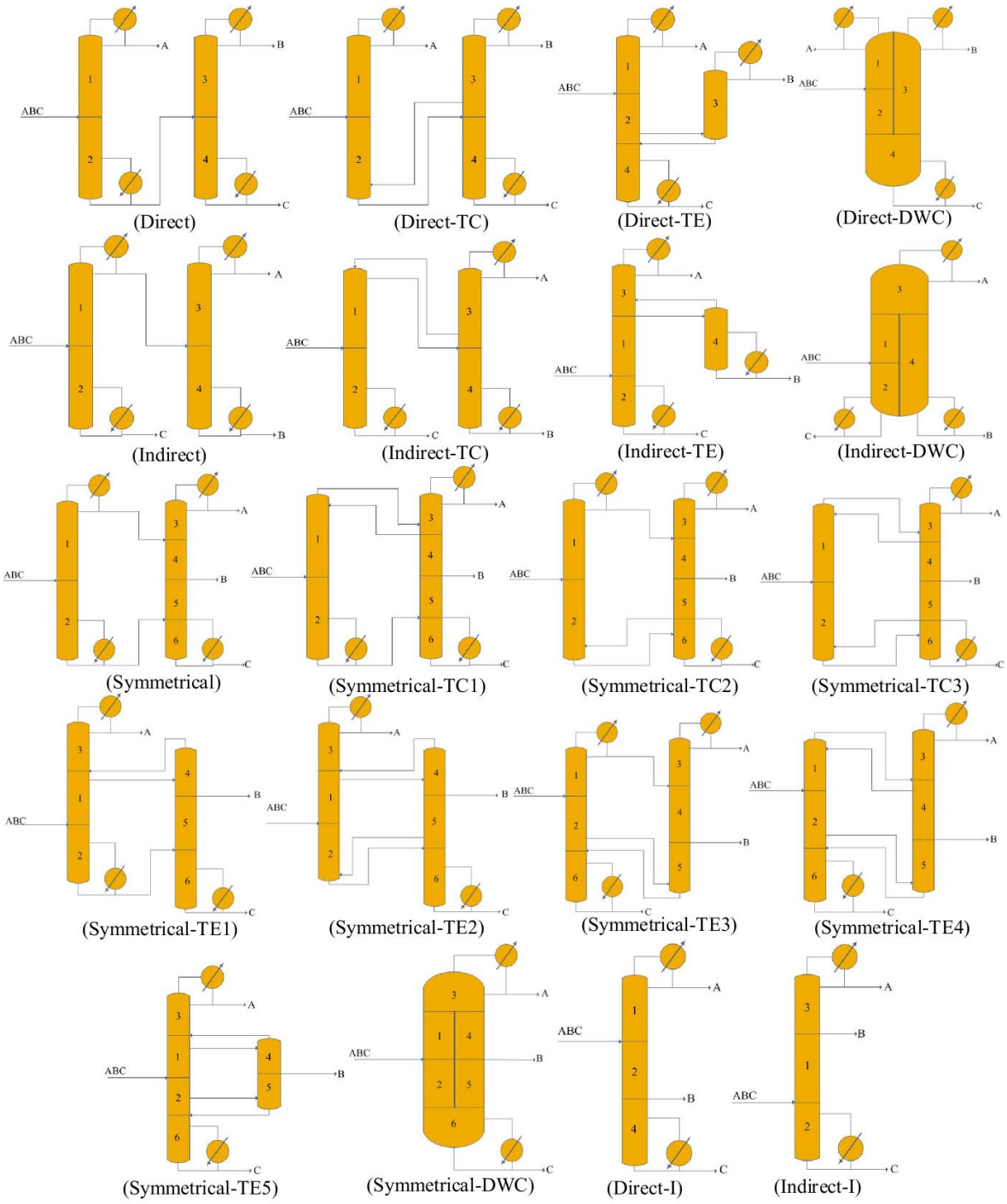


Figure 1
Three-component distillation sequences considered in this study.

1.1. Exergy analysis of distillation sequences

Exergy analysis as a tool has been used to study the performance of distillation columns by many researchers. Rivero et al. proved that exergy analysis could be used as a tool to provide a good insight into the process inefficiencies and proving the viability of distillation process modification (Rivero et al., 2004). Besides, this method has proved viable to be used in the synthesis of distillation sequences

to avoid complexity, in laborious systems such as those formed by a large number of components, trays, feeds, and side streams (Kencse et al., 2010).

Kencse and Mizsey compared simple and heat integrated forms of direct and indirect sequences and fully thermally coupled schemes for three-component separation mixtures according to cost, exergy loss, and greenhouse gas emissions (Kencse et al., 2010). They reported that an exergy analysis could predict the best sequence as predicted by economic and gas emissions studies. There are other researchers who used exergy analysis in distillation processes such as Suphanit et al. (2007), who studied the performance of divided-wall column configurations using exergy analysis) or Pinto et al. (2011), who presented a method for targeting side condensers and reboilers in distillation columns based on exergy loss diagrams. Also, Cortez-Gonzalez et al. (2012) analyzed the reduced structures that could be generated from simple basic four-component configurations by both economic and exergy analysis. Sun et al. (2012a) used exergy analysis to compare the performance of two different schemes for organo-silicon monomer distillation process. In another study, they present a five-column heat integrated methanol distillation scheme using pinch and exergy analysis simultaneously (Sun et al., 2012b).

In this work, the whole family of three-component distillation sequences, including simple, complex, thermally coupled, thermodynamic equivalent, divided-wall column, and intensified sequences are considered. All sequences (Figure 1) were simulated (based on rigorous simulation), optimized, and compared according to both economic and exergy analysis indicators. As stated, most of the studies were mostly reported for the exergy analysis of single distillation column and there are a few reports of applying the exergy analysis to a large number of columns or distillation sequences (Kencse et al., 2010). According to the authors' best knowledge, these configurations have never been analyzed and compared based on an economic study and exergy analysis all together before.

2. Methods

2.2. Separation matrix

The separation matrix used herein is extensively defied in our previous work (Ivakpour et al., 2008; Khalili-Garakani et al., 2015). In Figure 2, the proposed separation matrix is demonstrated for three- and four-component feed mixtures. φ is the sign used for the final products which could accept the values {I, II, and S}; Φ is the symbol used for sub-mixtures, which could have the values {I, II, and S}. In the separation matrix:

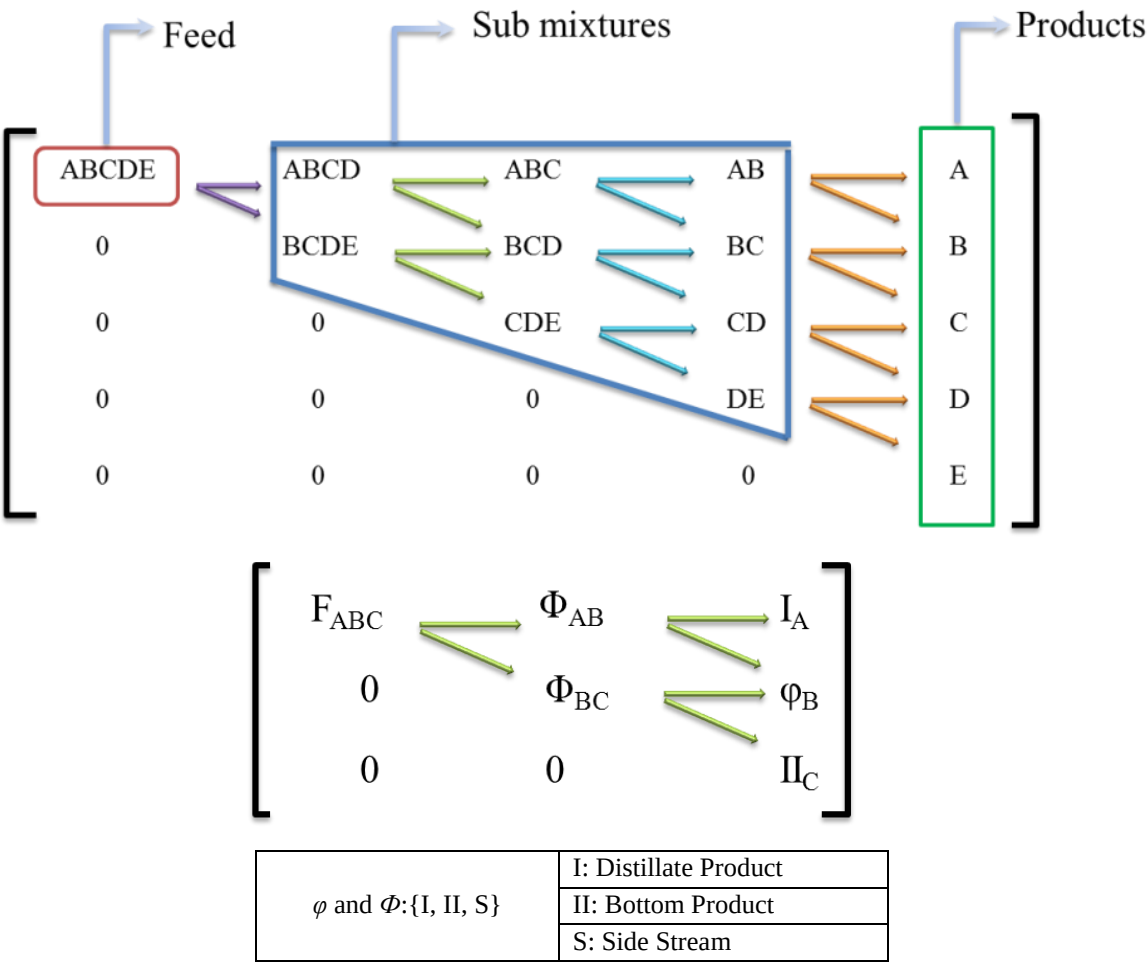
1- φ or Φ= {I} is used to demonstrate the column top product (from a condenser);

2- φ or Φ = {II} is applied to the column bottom product (from a reboiler);

3- And φ or Φ = {S} relates to the mixtures produced as a column side stream.

The mixture located in the first column is the original feed and is composed of all final products. Furthermore, the arrays positioned on the same diagonal of the separation matrix have an analogous heavy part. The structure of the distillation configuration could be obtained by the selection of the mixture (φ or Φ) options in the separation matrix. For more clarification, the components in each matrix array are indicated as subscript at the lower right of each array. Moreover, for the easier programming of the algorithm, three more indices are added for each Φ in the matrix $\{{}^{\lambda}_{\psi}\Phi^{a,b}_{r,k}\}$; the first index, ψ, indicates the thermal coupling of the product in the relevant distillation columns. Hence each of the sub-mixture streams that could be a candidate for thermal coupling has an additional ψ

sign in the lower left of the sub-mixture array associated with it ($_{\psi}\Phi_{rk}$). ψ could be "0" or "I", indicating the absence or presence of thermal coupling for its related reboiler or condenser respectively. The second index, λ, added to the upper left part of the Φ array presents moving, omitting sections, and divided wall between the columns in sequences. This superscript could also accept values "I" or "0". It must be noted that, in order to make a section movable in a distillation column, the condenser or the reboiler associated with the sub-mixture must initially be omitted. Therefore, in the separation matrix, the related array of the sub-mixture of moving sections should have ψ equated to I. Hence in a systematic programming practice, in order to generate all possible thermodynamically equivalent configurations, ψ should be checked to be "I" before changing λ for each sub-mixture. As a result, the arrays which represent moving sections will have two "I" values for both indices ψ and λ.



φ and Φ:{I, II, S}	I: Distillate Product
	II: Bottom Product
	S: Side Stream

Figure 2
Proposed separation matrix to present different sequences.

For intensified sequences, the deleted sections are illustrated by "X" in the separation matrix. λ accepts "X" in the upper left of the array in these cases. In these sequences, the separation could not take place completely and, for example in sequence Direct-I in Figure 3, a part of component C appears in product B. The same is true for Indirect-I in Figure 3, in which a part of A is appearing in product B. These components are called suspended components and are illustrated by the third index in the upper right-hand side of the arrays.

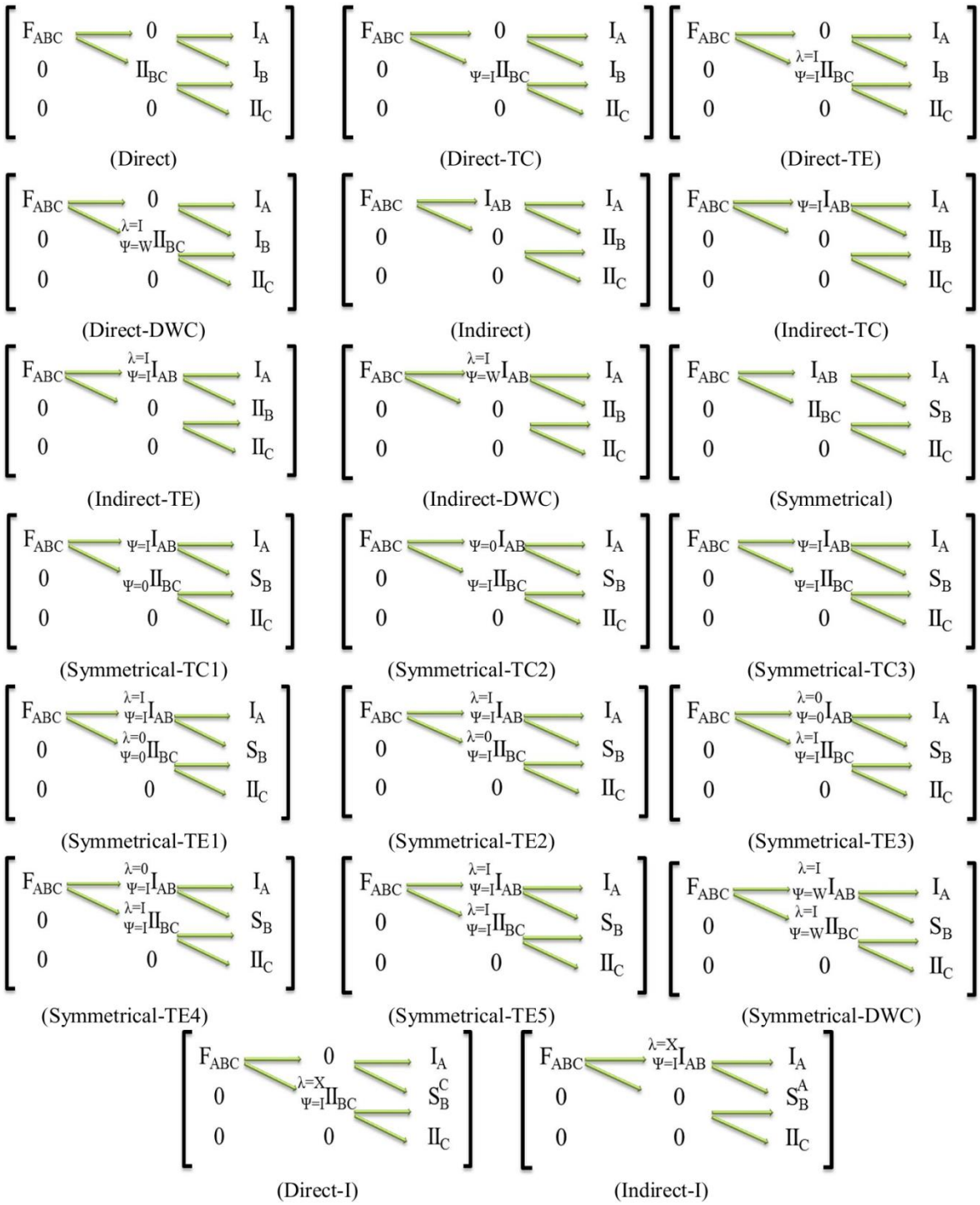


Figure 3
Separation matrices for each distillation sequences presented in Figure 1.

At last, for presenting the divided-wall columns, the value of ψ is changed to W. For more tedious sequences with more divided-wall columns, the value of ψ accepts W1, W2… for more clarification. Therefore, separation matrices in which both ψ and λ have value I ($\{ {}^{\lambda=I}_{\phi=I} \Phi \}$), which is the mark of a side column in the sequence, the value of ψ is changed and the separation matrix for the divided-wall column is generated. An example of these kinds of sequences could be seen in Figure 3.

2.2. Exergy analysis of distillation columns

Exergy is based on the first and second laws of thermodynamics and is defined as the maximum work which could be obtained from a stream or a source of energy until it reaches equilibrium with the environment or any reference state. Each stream has an exergy value that is the result of the difference between pressure, temperature, and chemical composition of the stream and those of the reference state. The reference state as defined by Szargut et al. is T_0=298.15 K and P_0=101.325 kPa (Szargut, et al. 1988). The exergy value of streams degenerates through the process. Thus, due to irreversibility phenomena in distillation columns, exergy loss is unavoidable. The main irreversibility in distillation columns is due to the mixing of the streams with different temperatures, pressures, and compositions on the trays and loss of heat in the condensers and to the environment from the body of the columns.

The total exergy of a stream is classified into physical, chemical, and mixing parts, which are calculated through the following equation (Hinderink et al., 1996):

Total exergy:

$$Ex_{total} = Ex_{Phys.} + Ex_{Chem.} + \Delta Ex_{Mix} \tag{1}$$

Physical exergy:

$$Ex_{Phys.} = [h(T,P) - h(T_0,P_0)] - T_0[s(T,P) - s(T_0,P_0)] \tag{2}$$

Chemical exergy:

$$Ex_{Chem.} = \sum_i x_i \,.\, \check{E}x_{Chem.,i} = \sum_i x_i \,.\, [\Delta G_{form,i} - \sum_i n_i Ex^0_{Chem,ref.,i}] \tag{3}$$

Exergy of mixing:

$$\Delta Ex_{Mix} = \Delta H_{Mix} - T_0 \Delta S_{Mix} \tag{4}$$

where, the mixing rule is given by:

$$\Delta M_{Mix} = L(M^l - \sum_i x_i M_i^l) + V(M^v - \sum_i y_i M_i^v) \tag{5}$$

Heat streams exergy:

$$Ex_q = Q.\left(1 - \frac{T_0}{T}\right) \tag{6}$$

The exergy loss in a distillation column could be easily calculated from the difference between exergy values of the inlet and outlet heat and mass streams. The inlet streams are the feeds and heat duty of reboilers, and the outlet streams are the products and the heats of the condensers.

$$Ex_{Loss} = \Delta Ex_{in} - \Delta Ex_{out} \tag{7}$$

$$Ex_{loss} = Q_{Reb.}\left(1 - \frac{T_O}{T_{Reb.}}\right) + Q_{Cond.}\left(1 - \frac{T_O}{T_{Cond.}}\right) + Ex_{Feed} - Ex_{bottom} - Ex_{top} \tag{8}$$

However, here the exergy loss of the columns is calculated by adding up the exergy loss at each stage of the column. The distribution of the exergy losses along the column (stage by stage) is more useful in understanding the irreversibility in each part of the column and the improvement of the entire system (Suphanit et al., 2007). The exergy losses of a stage in the distillation column could be calculated by carrying out a simple exergy balance around each tray. By calculating the exergy losses at each stage, the exergy loss diagram of the column could be achieved. Simulation methods are employed for obtaining the required data for drawing an exergy loss diagram.

2.3. Proposed algorithm

The steps of the procedure are described below:

- The procedure starts by defining the problem information (n, T_o, P_o, x_f, T_f, P_f, and Utilities) and parameter boundaries (x_p, optimization parameters).
- In the next step, the possible configurations according to the number of components (n) are generated and considered. The separation matrix method was utilized for this purpose as described elsewhere (Khalili-Garakani et al., 2015).
- The first configuration is then selected and sent to the next step, in which the columns are simulated by the short-cut method in order to find the initial data comprising of N_t, N_F, R_{min}, and P_{Col}. The pressure of the columns are optimized here to reach atmospheric pressure as close as possible, but the boiling point of the liquid in the condenser could remain higher than 35 °C (assume 25 °C for inlet cooling water temperature). Simulated annealing was applied as the optimization method (Kirkpatrick et al., 1983; Mahmoodpour et al., 2015). For physical property and equilibrium calculations, the Soave-Redlich-Kwang (SRK) equation-of-state was selected.
- In the next step, the rigorous simulations and the outcome of the short-cut method were used as the initial guess. The inside-out method, according to Seader et al., was used as the rigorous simulation method (Seader et al., 2011). The reflux ratios of the columns are then optimized with the objective of reaching the specified product purity with minimum usage of hot and cold duty in the reboilers and condensers respectively. The simulated annealing was used again at this stage as the optimization method.
- The results of the rigorous calculations (h, s, h_0, s_0, Q_H, and Q_C) were used for the exergy analysis of distillation columns. The exergy loss for each tray was estimated and the total exergy loss of the columns ($Ex_{destruction}$) was evaluated by adding them up in this step. The formulas and qualities of exergy analysis are presented in the former part.
- The results of the rigorous calculations (D_{Col}., Q_H, and Q_C) were used for the economic study (Total Annual Cost = annual capital cost + annual operation cost) of the distillation columns. Guthrie's cost calculation method is employed as modified in Douglas for the economic study (capital cost) (Douglas, 1988). Utility prices for calculating operating costs are as demonstrated in Table 1 (Seider et al., 2010).
- This procedure was carried out for all the configurations and the results sorted according to economic and exergy analysis separately.

Table1

Utility specification [28].

Utility Type	Pressure (atm)	Temperature (°C)	Price ($/GJ)
Electricity	-	-	16.667
Cooling Water	1	25	0.254
Low Pressure Steam	4.4	144	3.102
Medium Pressure Steam	11.2	184	5.257
High Pressure Steam	31.6	254	8.174

The configurations studied were for mixtures containing three-components with three different compositions (F1: [0.4, 0.2, 0.4], F2: [0.33, 0.34, 0.33], and F3: [0.15, 0.7, 0.15]) which are presented in Table 2.

Table 2
Different samples considered in this study.

Mixture	Components	ESI*	Pressure (atm)	Vapor Fraction	αAB=KA/KB	αBC=KB/KC	αAC=KA/KC
M1	n-Butane, i-Pentane, n-Pentane	1.86	4.5	0	2.38	1.28	3.05
M2	n-Pentane, n-Hexane, n-Heptane	1.04	1.44	0	2.57	2.47	6.35
M3	i-Pentane, n-Pentane, n-Hexane	0.47	1.44	0	1.25	2.65	3.31

* ESI=α_{AB}/α_{BC} [4].

3. Results and discussion

Table 3 presents the ranking of the sequences presented in Figure 1 for different feed conditions. Also, in Table 4, the economic study and exergy analysis of the first three sequences under each condition are presented.

Table 3
The ranking of configurations for M1, M2, and M3 at different feed compositions.

	M1		
	F1	**F2**	**F3**
1	Indirect-DWC	Direct-TC	Indirect-DWC
2	Indirect-TE	Direct	Indirect-TE
3	Symmetrical-DWC	Indirect-TC	Indirect-TC
4	Symmetrical-TC2	Indirect-DWC	Direct
5	Symmetrical-TC3	Direct-DWC	Direct-TC
6	Direct	Indirect-TE	Symmetrical-TE5
7	Symmetrical-TE4	Direct-TE	Indirect
8	Symmetrical-TE3	Indirect	Direct-DWC
9	Symmetrical-TE2	Symmetrical-DWC	Direct-TE
10	Symmetrical-TC1	Symmetrical-TC3	Symmetrical-TC1
11	Indirect	Symmetrical-TC2	Symmetrical-TE2
12	Symmetrical-TE1	Symmetrical	Symmetrical-TC2

13	Indirect-TC	Symmetrical-TE4	Symmetrical-TE1
14	Symmetrical	Symmetrical-TE3	Symmetrical-DWC
15	Direct-DWC	Symmetrical-TC1	Symmetrical
16	Direct-TE	Symmetrical-TE2	Symmetrical-TC3
17	Direct-TC	Symmetrical-TE1	Symmetrical-TE4
18	Indirect-IC	Symmetrical-TE5	Symmetrical-TE3
19	Symmetrical-TE5	Indirect-IC	Indirect-IC
20	Direct-IC	Direct-IC	Direct-IC
		M2	
	F1	**F2**	**F3**
1	Direct-TC	Symmetrical-DWC	Symmetrical-TE2
2	Direct-DWC	Symmetrical-TC3	Symmetrical-TC1
3	Symmetrical-TC2	Direct-TC	Symmetrical-TE1
4	Symmetrical-TE4	Symmetrical	Indirect-DWC
5	Direct-TE	Direct-DWC	Symmetrical
6	Symmetrical-TE3	Direct-TE	Indirect-TE
7	Symmetrical-DWC	Indirect-DWC	Indirect-TC
8	Indirect-DWC	Indirect-TE	Symmetrical-DWC
9	Indirect-TE	Direct	Symmetrical-TC3
10	Symmetrical-TC3	Symmetrical-TC2	Direct-DWC
11	Symmetrical	Symmetrical-TE4	Direct-TE
12	Direct	Symmetrical-TE3	Direct
13	Indirect-TC	Indirect-TC	Indirect
14	Indirect	Indirect	Direct-TC
15	Symmetrical-TE2	Symmetrical-TE2	Symmetrical-TC2
16	Symmetrical-TC1	Symmetrical-TC1	Symmetrical-TE4
17	Symmetrical-TE1	Symmetrical-TE1	Symmetrical-TE3
18	Indirect-IC	Symmetrical-TE5	Symmetrical-TE5
19	Symmetrical-TE5	Indirect-IC	Indirect-IC
20	Direct-IC	Direct-IC	Direct-IC
		M3	
	F1	**F2**	**F3**
1	Direct-DWC	Symmetrical-TE2	Indirect-DWC
2	Direct-TE	Direct-DWC	Symmetrical-TE2
3	Indirect-DWC	Direct-TE	Indirect-TE
4	Symmetrical-TE2	Indirect-DWC	Indirect-TC
5	Indirect-TE	Indirect-TE	Direct-TC
6	Direct	Symmetrical-TE1	Direct-TE
7	Symmetrical-TC1	Symmetrical-TC1	Direct-DWC
8	Symmetrical-TE1	Direct	Symmetrical-TC1
9	Indirect-TC	Direct-TC	Direct
10	Direct-TC	Indirect	Symmetrical-TE1
11	Indirect	Indirect-TC	Indirect
12	Symmetrical	Symmetrical	Symmetrical-TE4
13	Symmetrical-TC2	Symmetrical-DWC	Symmetrical
14	Symmetrical-DWC	Symmetrical-TC3	Symmetrical-TE3
15	Symmetrical-TE4	Symmetrical-TC2	Symmetrical-DWC

16	Symmetrical-TC3	Symmetrical-TE4	Symmetrical-TC3
17	Symmetrical-TE3	Symmetrical-TE3	Symmetrical-TC2
18	Direct-IC	Direct-IC	Symmetrical-TE5
19	Symmetrical-TE5	Indirect-IC	Direct-IC
20	Indirect-IC	Symmetrical-TE5	Indirect-IC

Table 4
The result of total annual cost ($/y) and exergy loss rate (GJ/hr) for the best three sequences under different feed conditions.

Mixture	Composition		Sequences	TAC ($/year)	Exergy loss rate (GJ/hr)
M1	F1	1st	**Indirect-DWC**	**293,472.592**	**0.675**
		2nd	Indirect-TE	296,297.113	0.676
		3rd	Symmetrical-DWC	303,743.586	0.679
	F2	1st	**Direct-TC**	**340,502.266**	**0.229**
		2nd	Direct	367,942.217	0.255
		3rd	Indirect-TC	3778,841.474	0.629
	F3	1st	**Indirect-DWC**	**508,593.831**	**1.531**
		2nd	Indirect-TE	512,115.351	1.535
		3rd	Indirect-TC	526,700.303	0.687
M2	F1	1st	**Direct-TC**	**138,095.912**	**0.696**
		2nd	Direct-DWC	142,216.070	0.720
		3rd	Symmetrical-TC2	144,355.762	2.654
	F2	1st	**Symmetrical-DWC**	**170,791.224**	**0.599**
		2nd	Symmetrical-TC3	172,106.681	0.600
		3rd	Direct-TC	175,504.430	0.351
	F3	1st	**Symmetrical-TE2**	**196,042.597**	**1.177**
		2nd	Symmetrical-TC1	212,178.195	1.187
		3rd	Symmetrical-TE1	212,217.274	1.188
M3	F1	1st	**Direct-DWC**	**278,968.356**	**0.628**
		2nd	Direct-TE	279,719.402	0.629
		3rd	Indirect-DWC	281,628.454	0.377
	F2	1st	**Symmetrical-TE2**	**326,486.558**	**0.956**
		2nd	Direct-DWC	329,243.256	0.785
		3rd	Direct-TE	329,686.667	0.786
	F3	1st	**Indirect-DWC**	**463,035.983**	**0.342**
		2nd	Symmetrical-TE2	469,954.929	0.718
		3rd	Indirect-TE	472,286.012	0.344

When the feed content of component B is low, the composition of B in the liquid feed of the side stripper is much lower than that of the vapor feed of the side rectifier. This is due to liquid feed of the side stripper being diluted by a significant amount of component A, while in the vapor feed of the side

rectifier, this is done with the same amount of component C. As a result, for producing component B with the same specification, vapor traffic in the side stripper is significantly more than side rectifier. Therefore, more heat could be supplied at a mid-temperature of T_B in side stripper configuration (in comparison to the lower amount of rejected heat in the condenser at temperature T_B in side rectifier). This is the reason why Indirect-TE and Indirect-DWC perform better for M1 and composition F1. Also, when the content of the middle component B is high in the feed, these two configurations have superior performance compared to other sequences, with the only exception of M2 in which ESI≈1.

According to Malone et al. (1985), when component relative volatilities are close (α_{AB}, α_{BC}), as in M2 (F1 and F2), direct sequence is one of the best, as also demonstrated by the present results. As α_B approaches α_A, the chance for direct family to be one of the preferred sequences increases. However, this is not the case when component B content increases. When α_{AB} is low, for the separation of A from B, a large amount of vapor is needed; however, the condenser temperature for pure A (T_A) is nearer to the B bubble point (T_B) than to pure component C reboiler temperature (T_C). As a result, it is better to supply the required heat for the separation of A from B at mid temperature level (T_B). In configurations like symmetrical-TC3 (Petlyuk sequence), supply of heat at T_B is not possible and this leads to the better performance of sequences such as indirect-TE in comparison to symmetrical-TC3. This is true for divided-wall column sequences which are generated from these configurations.

Symmetrical (Brugma or pre-fractionator) sequence and the other sequences generated from it (thermally coupled and equivalent thermodynamics) have their best performance in M2, in which ESI≈1. The comparison of thermally coupled configurations (symmetrical-TC1, TC2, and TC3) with side stripper, side rectifier, and other symmetrical configurations illustrate that side stripper and side rectifier configurations have better performance for M1 (ESI>1) and M3 (ESI<1), and only in M2 (ESI≈1), the thermally coupled sequences present the reduction of energy consumptions. Similar results are presented in the work by Agrawal and Fidkowski (1999), in which they calculate the minimum total amount of vapor in sequences in minimum reflux condition. Thermally coupled sequences for feed M2 and F3 composition illustrate 34% reduction of energy consumption in comparison to simple sequences (direct and indirect).

The results of thermodynamically equivalent configurations presented by Agrawal and Fidkowski (1998) (symmetrical-TE1-TE4) is nearly the same as thermally coupled configurations under all feed conditions considered in this study. In these configurations, reboilers and condensers are located on different columns; the column at a high pressure has a reboiler, and the column at a low pressure has a condenser; in this way, the vapor stream could be flown from the column at a higher pressure to the one at a lower pressure. The change to the structure of the sequence made it more operable and easy to control. Comparing them to the symmetrical-TC3 illustrates that symmetrical-TC3 sequence performs better for M1 with F1-F2 feed composition, and symmetrical-TE1 and symmetrical-TE2 are better for F3 feed composition. For M2 and feed F1, symmetrical-TE3 and symmetrical-TE4 are superior, while symmetrical-TC3 outperforms for feed F2, and symmetrical-TE1 and symmetrical-TE2 perform better for F3. In M3, symmetrical-TE1 and symmetrical-TE2 perform better at all three different feed compositions.

Agrawal and Fidkowski (1998 and 1999) presented symmetrical-TE1-TE4 configurations and analyzed them by the minimum total amount of vapor in sequences in a minimum reflux condition. Then Jiménez et al. (2003) analyzed them by rigorous methods and compared their heat duty. In comparison to their work, the research presented here uses more accurate total cost and exergy loss analysis, which justifies some of the differences in ranking of the sequences. The column diameter

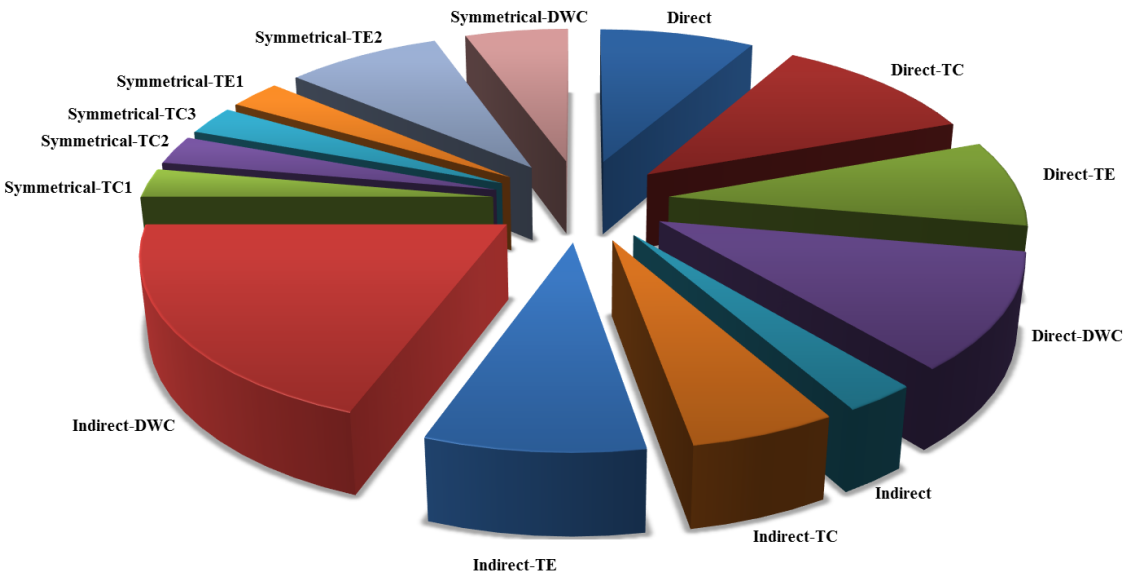
calculation procedure considered herein in TAC analysis is one of the main reasons behind the differences between the two findings.

Furthermore, it should be noted that some of the configurations considered herein are not present in the works of Caballero and Grossman (2001 and 2004), which employs a suggested superstructure and optimizes the tray sections and energy performance of configurations.

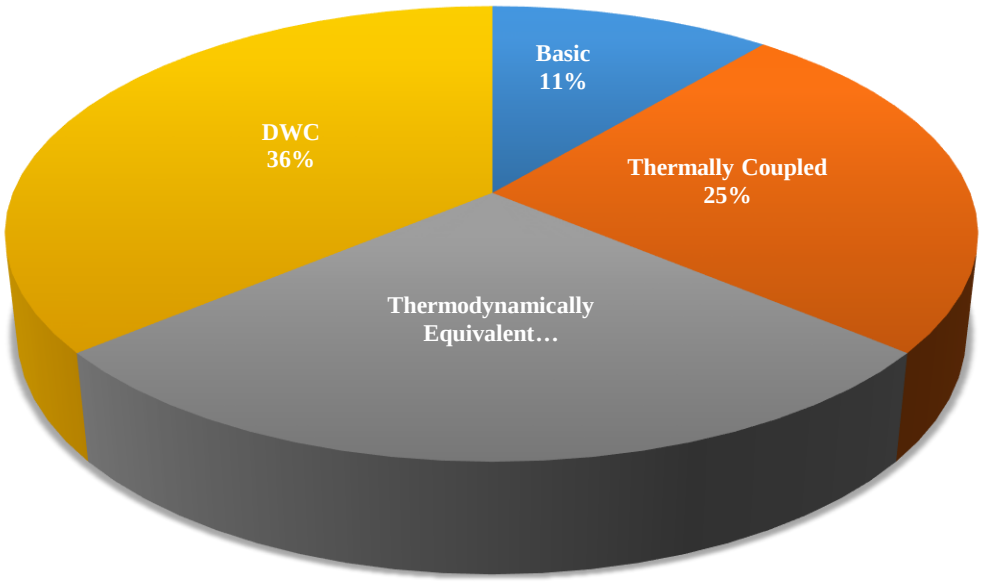
Figure 4a presents the number of occurrences of the best three sequences for all the cases studied here. Indirect-DWC sequence has the largest number of occurrences followed by Direct-DWC, Direct-TC, Indirect-TE, and Direct-TE sequences respectively.

Also in Figure 4b, the distribution of the best sequence among different categories of configurations is illustrated. As presented, DWC sequences have the biggest part and the basic sequences have the lowest part among the best configurations.

Figure 5 illustrates the exergy loss diagram of symmetrical configurations (Brugma configurations) for different feeds (M1-M3) and compositions (F1-F3). Brugma sequence was considered here because from this sequence all the other configurations could be derived. As demonstrated, increasing B content in the feed raises the peak in the exergy loss diagram. For M1 (ESI>1), the maximum loss is in the upper feed of the second column. At the time (ESI≈1 (M2)), there were two peaks: one in the upper feed of the second column and the other in the reboiler. At last for M3, the peaks are located on the lower feed and the condenser of the second column.



a)



b)

Figure 4
a) Number of occurrences of the best three configurations in different samples and b) distribution of the best sequences among different configuration categories.

The location of peaks in the diagrams could be found by the relative volatility of the components. As illustrated in Table 2 and Figure 5, the peaks are located in the place (section) where the feed with higher relative volatility enters the second column. It is due to the superior separation ability of the mixture with greater relative volatility in the pre-fractionator (first column). Thus in the next column, the streams with much different compositions encounter each other, and as a result the exergy losses due to mixing is increased. Since, α_{AB} is higher in M1 (ESI>1), there is a peak in the upper feed of the second column. However, the relative volatility becomes similar in M2 (ESI≈1) in the pre-fractionator, where the separations (A/B and B/C) take place in the same order. Hence separation in the next column would be easier, and, as a result, the exergy loss is decreased. This effect is more significant when the content of the middle component (B) is lower in the feed. In these configurations, the amount of loss in the upper (rectifier) and lower (stripper) sections of the second column are nearly the same and the distribution of losses is more monotonous.

As stated before, when the amount of α_{AB} is low (M3: ESI<1), a large amount of vapor is required for the separation of A from B. As a result, the exergy loss in the condenser is increased and a peak could be seen in the condenser.

Therefore, by these figures the designers could easily find the weak points in each column of the sequence and employ the exergy loss diagram for improving the performance of the sequence. Furthermore, the changes to the structure of the columns in these places similar to thermal coupling of the streams between columns, generating thermodynamically equivalent structures by moving sections in these areas, or using internal wall (DWC) could reduce the exergy loss of the system (as presented in the results in the former section).

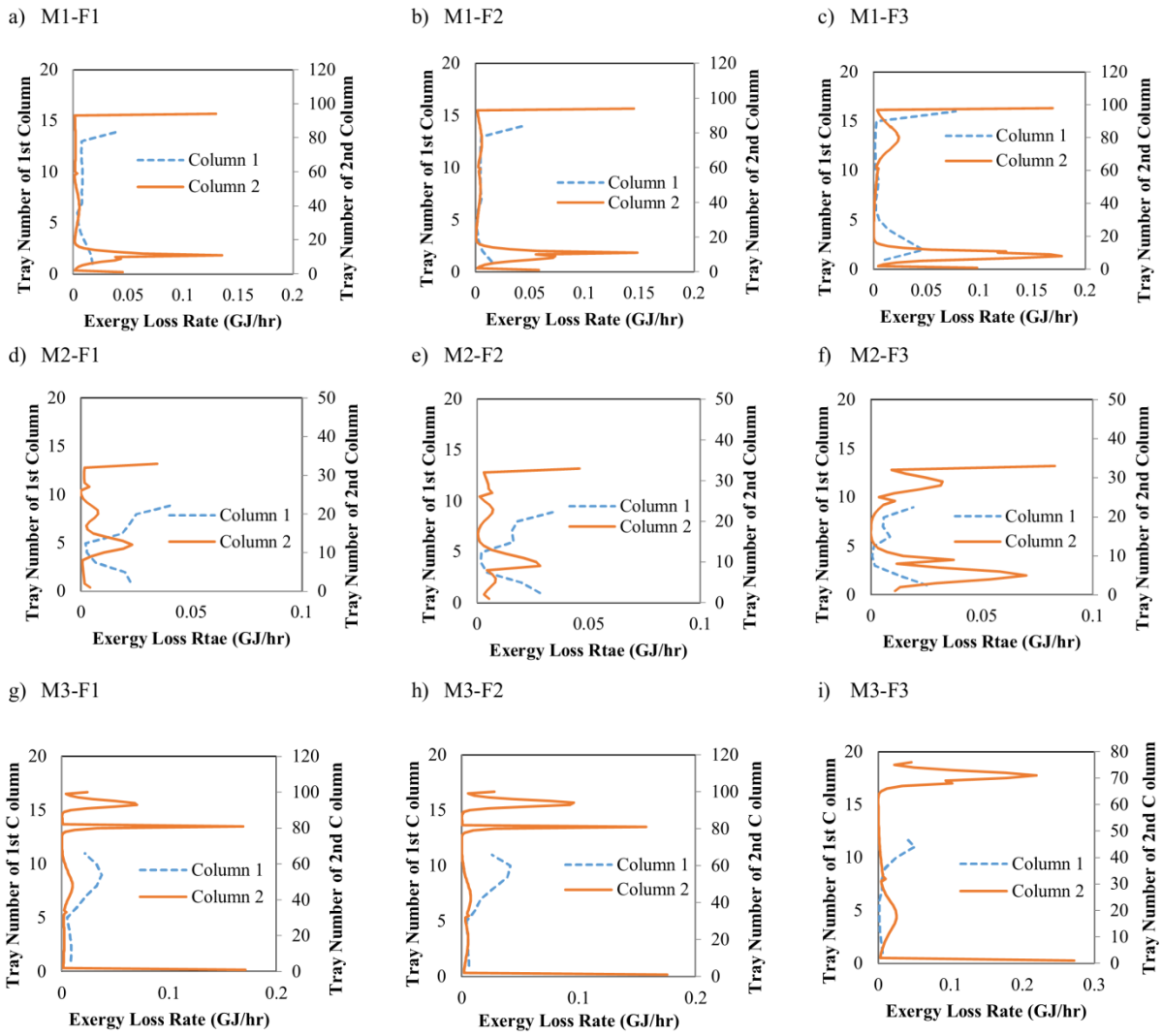


Figure 5
Exergy loss diagram of the symmetrical sequence (Brugma) for different feeds (M1-M3) and compositions (F1-F3).

In addition, the knowledge of the weak points of the sequences and structural changes could lead to a search space reduction algorithm. This algorithm helps designers to analyze a much smaller search space containing only near optimal sequences, which could be sufficient to find an overall optimal *n*-component configuration. The search space reduction algorithm is out of the scope of this paper and would be presented in future works.

4. Conclusions

The analysis of the samples illustrates that the TAC of the sequences is totally dependent on the amount of the intermediate component present in the feed. By increasing the amount of the intermediate component, TAC is increased for each sequence. Also, despite expectations, the Petlyuk sequence (symmetrical-TC3) only performs well in M2 (ESI≈1) and for F2 composition; it demonstrates poor performance for other conditions (like different compositions of M3). In comparison to economic study, exergy analysis is simpler in calculation and only requires some

physical properties, which could easily be acquired; also, the exergy loss diagrams could give designers an insight into the weak parts of the systems and help with improving the performance of the processes. Knowing the weak points in each column of the sequence could be used for the structural changes of the sequence, and could lead to the generation of new configurations. The results presented that changes such as thermal coupling of the streams between columns, generating thermodynamically equivalent structures by moving sections in these areas, or applying divided-wall columns could reduce the exergy losses of the system (Khalili-Grakani, 2016).

At last, according to the ability and flexibility of the separation matrix in changing the structure of the sequence, the method could be easily expanded to a larger number of components and other kinds of sequences like multi-effects, pump around, compressor-aided separations, and internal heat integrated distillation systems (HIDIC), which would be considered in future works. The detailed optimization procedure is beyond the scope of this work and will be presented in future works.

Nomenclature

Symbols	
$\Delta G_{form,i}$	: Standard Gibbs energy of formation [kJ/mol]
Ex_{total}	: Total exergy content [kJ/kg]
Ex_{phys}.	: Specific physical exergy [kJ/kg]
Ex_{chem}.	: Specific chemical exergy [kJ/kg]
$\breve{E}x_{chem}$	: Standard chemical exergy [kJ/mol]
Ex_{Mix}.	: Specific exergy of mixing [kJ/kg]
h	: Specific enthalpy [kJ/kg]
h_o	: Specific enthalpy at T_0, p_0 [kJ/kg]
I	: Top product
II	: Bottom product
III	: Eliminated mixture in crossover complete sequences
L	: Liquid fraction
n	: Number of chemical species
Φ	: All possible states for sub-mixtures
φ	: All possible states for product type
p_o	: Pressure of the reference state [101.325 kPa]
R	: Gas constant [kJ/mol.K]
S	: Side stream
s	: Specific entropy [kJ/kg.K]
s_o	: Specific entropy at T0, p0 [kJ/kg.K]
T_o	: Temperature of the reference state [298.15 K]
V	: Vapor fraction
x	: Mole fraction of species
Superscripts	
a, b	: Presenting suspected components in each stream

λ	: Sign of moving section or omitting section in related distillation columns
Subscript	
ψ	: Sign of thermal coupling of the product in related distillation columns
r, k	: Presenting components in each stream

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