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Abstract

Process integration is currently considered as the main trend to improve process performance, and is one of the major approaches to reduce the annual operating and capital costs in the plant. For distillation systems, heat integration technique provides such an approach to improve the traditional simple column sequences. This work presents the optimization of distillation column sequences based on creation of maximum possible heat integration and minimizing the total annual cost of process. All the optimum simple sequences and possible heat integrated sequences are designed and considered to find the best heat integrated sequence with the minimum total annual cost. Sequences are simulated and the objective function is modeled. Basic operation parameters of sequences are changed according to the process constraints to find all the possible heat integration and minimize the objective function. The best structures with the minimum total annual cost are designed and compared for the considered industrial case study. Results show the height percent of optimization of process costs by the internal heat recovery of integration.

Key words: Distillation; Sequence; Modeling; Integration; Optimization

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NOMENCLATURE

Q	Heat flow rate
р	Pressure
TBP	Boiling Point Temperature
ΔT_{min}	Minimum temperature difference
$\Delta T_{\rm LMTD}$	Logarithmic Minimum Temperature Difference
V	Rectify vapor rate
Xi	Mole Fraction
MWHC	Molecular Weight of Heaviest Component
Nt	Number of trays
D	Diameter of column
Н	Height of column
А	Area of heat transfer in heat exchanger
U	Heat transfer coefficient of heat exchanger
S_n	Number of simple sequence
n	Number of components
{ p }	Decision pressure parameter
TAC	Total Annual Cost
R1	Reboiler of column 1
R2	Reboiler of column 2
R3	Reboiler of column 3
C1	Condenser of column 1
C2	Condenser of column 2
C3	Condenser of column 3
HX	Heat Exchanger
SQP	Successive Quadratic Programming
CW	Cooling Water
Min	Minimum
M&S	Marshall and Swift Cost Index

INTRODUCTION

Distillation is the most widely used separation technique in the petrochemical and chemical process industries for the separation of fluid mixtures despite its high energy requirement. Significant energy saving can be reached by the use of complex distillation sequences such as the side-stripper, the side-rectifier, the thermal (internal) column coupling (also know as petlyuk system), the (external) energy integration (also known as energy integrated distillation system) and the heat pumping techniques^[1-3]. The energy saving in distillation is an essential field of the chemical engineering research since the early 1970; appropriate integration of the distillation columns with the overall process often results in a significant reduction of the energy consumption in many cases. The classical design of a distillation system for a multicomponent separation uses only the simple columns. Each of the simple columns in a multicomponent distillation configuration receives a feed and performs a sharp split between two adjacent components of the feed mixture. Meanwhile, each of the simple columns produces a top product with a condenser and a bottom product with a reboiler. Thus, in any case of the simple column configuration for an *n*-component mixture, it needs n - 1 simple columns with n - 1 condensers and n - 1 reboilers^[4-5]. The simple column configurations for multicomponent distillation are easy to design and operate. Because the number of separation sequences increases dramatically when the number of components in the feed mixture increases, a considerable number of works have been conducted on optimal synthesis of simple column configurations for an n-component separation^[4-7]. Thompson and King reported the following formula to calculate the number of all simple configurations^[8]:

$$S_n = \frac{[2(n-1)]!}{n!(n-1)!} \tag{1}$$

Where, S_n is the number of sequences, and n is the number of component in the feed stream. The task for the design of a heat integrated simple column configuration consists of searching for the possible heat matches among all of the condensers and reboilers of the simple columns. Large numbers of works have been done for the synthesis of the optimal heat integrated simple column configurations since the work by Rathore et al^[9-16]. Lots of works in this area tried to find the better heat integrated sequence for ternary mixture streams which consist of 2 columns, 2 condensers, and 2 reboilers. Considering a separation of a stream with more than 3 components extremely increases the number of different sequences and number of condensers and reboilers in the sequences which cause large number of different heat integration between columns.

This paper considers the heat integrated distillation systems for a separation of a stream to four pure groups according to minimization of the total annual costs. The plant configuration considered in our industrial case shown in Figure 1-A. This configuration can be classified as a distributed sequence and can be represented by the following separation task: AB/CD; A/B; C/D. Other four simple column configurations are possible for this separation are, the direct (A/BCD; B/CD; C/D) sequences (Figure 2-A), the direct-indirect (A/BCD; BC/D; B/C) sequences (Figure 3-A), the indirect (ABC/D; A/B/C; A/B) sequences (Figure 4-A), and the indirect-direct (ABC/D; A/BC; B/C) sequence (Figure 5).

1. METHODOLOGY

The target of this research is minimizing the total annual cost of sequences by creating heat integration among columns. To calculate the temperatures and heat flow rates of condensers and reboilers and find the design specifications of columns such as the actual number of trays, reflux ratio, rectify vapor flow, and etc., all the five simple sequences are simulated by shortcut and rigorous method of commercial software. Then, the equations of diameter and height of columns, annual capital cost of columns and heat exchangers and annual operation cost are modeled and used to calculate the total annual cost of each sequence. The equations used to account diameter and height of columns are:

$$D = 5e - 3 \times \sqrt[0.25]{MWHC} \times (1.8 \times TBP + 492) \times \sqrt[0.5]{RV}$$
(2)

$$H = (Nt - 1) \times 0.6 + 6 \tag{3}$$

The height of column (Equation 3) is evaluated considering 0.6 m as tray spacing and 6.0 m as disengagement.

The basic equations for calculating the capital cost are the total cost of distillation columns and heat exchangers [16]. The total cost of a distillation column can be considered as a sum of the costs of column shell and trays. The equation for calculating the column shell is:

$$\$ = \left(\frac{M \& S}{280}\right) (937.61) D^{1.066} H^{0.802} \tag{4}$$

All the correlations are valid for carbon steel construction and are updated from mid-1968 to 2009 utilizing the Marshall and Swift cost index. The correlation (Eq.4) is valid for a pressure less than 345 kPa, otherwise a correction factor $[1+1.45 \times 10^{-4}(P-345)]$ must be applied.

For calculation the cost of column's trays, considering sieve trays, the following correlation has been utilized:

$$\$ = \left(\frac{M \& S}{280}\right) (97.24) D H^{0.802}$$
(5)

The heat exchanger cost evaluation is based on the heat exchanger area evaluated utilizing the usual design formula:

$$A = \frac{Q}{U \ \Delta T_{LMTD}} \tag{6}$$

Mean values of 1800 W/(m^2 K) and 2100 W/(m^2 K) for the overall heat transfer coefficient for condensers and reboilers are assumed respectively. Assuming shell and tube, floating head and carbon steel construction the cost correlation is as follows:

$$\$ = \left(\frac{M \& S}{280}\right) (474.67) A^{0.65} \tag{7}$$

The reported correlation is valid for an exchange area range between $18.6 < A < 464.5 \text{ m}^2$.

The capital cost is annualized over a period which is often referred to plant life time. In this case study the plant lifetime is 10 years.

Annual operating cost is calculated by amount of heat flow rates of condensers and reboilers of columns and utility costs of Table 1^[17]. Heat flow rates of condensers and reboilers are calculated by simulation. The time fraction of operation for annualizing the operating cost is 8 000 h/a.

In next step, after calculation of the TAC function, the optimizer toolbar of commercial software is used to minimize the objective function of TAC, according to the continue parameters of column's pressures and the temperatures constraints of process. The optimization method SQP of the optimizer toolbar is used for minimizing the problem. With the continuous decision for the individual columns' pressure $\{p\}$, the objective function formulation is:

$$Min TAC = min C (\{p\}), \{p\} \in P$$
(8)

where C() represents the total annual cost model for the system, P is the vector of operating pressures for all individual columns in the system, and P can be a set of feasible range of $\{p\}$. For minimization the TAC function of each simple sequence, continues parameters of column's pressures can change in the range of 1 bar to 30 bars. For temperature constraints, temperatures of columns have to be between the temperatures of cooling water and the temperature of hp-steam.

After optimization of the simple sequences, in next step, start to find the possible heat integration in each sequence. In each sequence, there are 3 condensers (C1, C2, C3) and 3 reboilers (R1, R2, R3), which can transfer heat to each other according to their temperatures. For each sequence there would be 12 different situations for heat integration:

 $C1 \rightarrow R2$, $C1 \rightarrow R3$, $C1 \rightarrow R2\&R3$, $C2 \rightarrow R1$, $C2 \rightarrow R3$, $C2 \rightarrow R1\&R3$, $C3 \rightarrow R1$, $C3 \rightarrow R2$, $C3 \rightarrow R1\&R2$, $C1\&C2 \rightarrow R3$, $C2\&C3 \rightarrow R1$, $C1\&C3 \rightarrow R2$

For example, the situation C1 \rightarrow R2 means that the condenser C1 transfers heat to reboiler R2, according to the minimum temperature difference ΔT_{min} =10 °C. To find that the condenser C1 can transfer heat to reboiler R2, we increased the pressure of column1 to the allowable maximum pressure point, according to the temperature constraint, to increase the temperature of the condenser

C1 to its maximum level. On the other side, we decreased the pressure of column 2 to allowable minimum pressure, according to the temperature constraint, to decrease the temperature of reboiler R2. If the temperature of condenser C1 was higher than the temperature of reboiler R2 plus Δ Tmin, the heat integration of C1 \rightarrow R2 could be possible. Now for this situation, a new temperature constraint $(T_{C1}-T_{R2}>10)$ and some changes to the calculation of total annual capital cost and annual operation cost are added to the optimization procedure of the objective function of TAC in optimizer toolbar. The optimum pressures of columns and the minimum TAC are obtained by the optimization and the data and structure of sequence with heat integration of $C1 \rightarrow R2$ are stored. Totally, for all five simple sequences, there would be 60 different situations for heat integration.

This procedure is done for all the mentioned deferent situation of heat integration for each simple sequence and all the possible heat integrated sequences are optimized. Finally, the optimum simple sequences and heat integrated sequences are compared to find the best structure with the minimum total annual cost.

Table 1 Utility Costs

Utility	Temperature Level (° C)	Values
LP-steam (\$/t)	158	13
MP-steam (\$/t)	200	16
HP-steam (\$/t)	250	20
Cooling water (\$/t)	35-45	0.082

2. CASE STUDY

The industrial process examined in this paper, is the high temperature separation section of olefin plant which separates a multi-component flow to four categories of products with a purity of 0.98. The intake flow has a temperature of 59.3 °C, a pressure of 1785 kPa and an amount flow rate equal to 1355 kmol/h. Compositions of process intake flow and products are shown in Table 2. The initial process is illustrated in Figure 1-A and it is regarded as a reference case for other states. Different sequences have been obtained for the process with heat integration satisfying the constraints of the problem. These sequences are shown in Figures 1 to 5. Results of design and optimization of sequences are shown in Table 3.

Table 2Feed and Product Specifications

Product		Feed	
Product groups	Compos	Xi (Mole Fraction)	
	Ethane	C2H4	0.0001
А	M-Acetylene	C3H4	0.0047
	Propene	ComponentsXieC2H4leneC3H4neC3H6neC3H8ttlenC4H4vleneC4H6neC4H8neC4H10nneC5H12neC6H14	0.4538
 	Propane	СЗН8	0.2912
В	VnylAcetlen	C4H4	0.0003
	DMAcetylene	C4H6	0.0011
С	i-Butene	C4H8	0.1541
	n-Butane	C4H10	0.0359
	n-Pentane	C5H12	0.0419
D	n-Hexane	C6H14	0.0137
	n-Heptane	C7H16	0.0031

Table 3 Optimal Schemes

Description	Primary Distributed			Distributed (C3-R1)			Distributed (C1&C3-R2)			
	Col.1	Col.2	Col.3	Col.1	Col.2	Col.3	Col.1	Col.2	Col.3	
Pressure (kPa)	1785	1940	735	1901	2217	2748	2989	2220	1042	
Diameter (m)	3.35	4.88	1.67	3.35	4.89	1.74	3.41	4.92	1.72	
Reflux ratio	0.71	8.45	0.64	0.72	8.53	0.79	0.77	8.62	0.74	
Actual plates	54	163	43	54	166	42	43	167	44	
Heating rate (kJ/h)		111423972			106272677		78498124			
Cooling rate (kJ/h)		117477072			107204581		82738403			
HX duty (kJ/h)		-			7314222.4			32886838		
Steam cost (\$/a)		4535992.7			4557609.8			3326005		
C.W cost (\$/a)		1843659.3			1682445.1			1298478.3		
Operating cost (\$/a)	6379652				6240054.9			4624483.2		
Capital cost (\$/a)	758906.59				555824.96			525335.21		
TAC (\$/a)	7138558.6				6795879.9		5149818.4			
Operating cost saving (%)	-				2.19		27.51			
Capital saving (%)	-				26.76		30.78			
TAC saving (%)	-			4.8			27.86			
D i di	Direct			Direct (C3-R1)			Direct (C3-R2)			
Description	Col.1	Col. 2	Col. 3	Col. 1	Col. 2	Col. 3	Col. 1	Col. 2	Col. 3	
Pressure (kPa)	2365	2113.22	651	2414	1969	1698	2313	1839	2667	
Diameter (m)	6.29	2.39	1.68	6.29	2.38	1.71	6.28	2.37	1.74	
Reflux ratio	8.95	1.21	0.65	8.97	1.2	0.73	8.93	1.18	0.78	
Actual plates	176	42	41	174	40	44	176	40	49	
Heating rate (kJ/h)		100110119			94211676		96031322			
Cooling rate (kJ/h)		105533811			96995376		97473954			
HX duty (kJ/h)		-			7832094		7279755.3			
Steam cost (\$/a)		4224193.8			4127335.1		4238147.6			
C.W cost (\$/a)		1656224.4			1522224.1		1529734.8			
Operating cost (\$/a)	5880418.2			5649559.2			5767882.4			
Capital cost (\$/a)	554833.01			550778.59			578258.09			
TAC (\$/a)	6435251.2			6200337.8			6346140.5			
Operating cost saving (%)		7.83		11.44			9.59			
Capital saving (%)	26.89			27.42			23.8			
TAC saving (%)		9.85		13.14			11.1			

To be continued

Description	Direct-Indirect			Dire	ct-Indirect (C2	-R1)	Indirect			
	Col.1	Col.2	Col.3	Col.1	Col.2	Col.3	Col.1	Col.2	Col.3	
Pressure (kPa)	2080	1307	1854	2024	2740	1717	1660	2335	2080	
Diameter (m)	6.27	2.37	1.83	6.27	2.42	1.9	3.15	2.66	4.9	
Reflux ratio	8.9	0.31	1	8.89	0.37	1.13	0.21	0.66	8.53	
Actual plates	174	44	42	174	49	46	47	45	169	
Heating rate (kJ/h)		109826812			96202845			129266330		
Cooling rate (kJ/h)		113397355			99389758			131605362		
HX duty (kJ/h)		-			14648275			-		
Steam cost (\$/a)		4634066			4414348.8			5696958.1		
C.W cost (\$/a)	1779633.1				1559801			2065385.6		
Operating cost (\$/a)	6413699.2				5974149.8			7762343.7		
Capital cost (\$/a)	639173.17				701488.26			611662.27		
TAC (\$/a)	7052872.3				6675638			8374006		
Operating cost saving (%)		-0.53			6.36			-21.67		
Capital saving (%)		15.78			7.57			19.4		
TAC saving (%)		1.2			6.48			-17.31		
Description	Indirect (C1&C2-R3)			Iı	ndirect-Direct					
Description	Col.1	Col.2	Col.3	Col.1	Col.2	Col.3				
Pressure (kPa)	2585	2986	2169	1844	2296	1912				
Diameter (m)	3.2	2.71	4.92	3.18	5.01	1.89				
Reflux ratio	0.25	0.72	8.61	0.22	8.69	1.12				
Actual plates	51	49	168	48	167	44				
Heating rate (kJ/h)		82072805			117517206					
Cooling rate (kJ/h)		83029960			120136244					
HX duty (kJ/h)		48112352			-					
Steam cost (\$/a)		4003977.4			5307057.3					
C.W cost (\$/a)		1303053.9			1885391.8					
Operating cost (\$/a)		5307031.3			7192449					
Capital cost (\$/a)		607282.93			525144					
TAC (\$/a)		5914314.3			7717593					
Operating cost saving (%)		16.81			-12.74					
Capital saving (%)		19.98			30.8					
TAC saving (%)		17.15			-8.11					

Continued







Figure 2



Figure 3 Direct-Indirect Sequences: A is Direct-Indirect, and B is Direct-Indirect (C2-R1)



Figure 4 Indirect Sequences: A is Indirect, and B is Indirect (C1&C2-R3)

3. DISCUSSION AND RESULTS

The Distributed sequence of the initial process is considered as the reference of comparison for other sequences (Figure 1-A). The optimal schemes data in Table 3 shows that the sequence Distributed (C1&C3-R2) (Figure 1-C) with a TAC optimization of 27.86 % represents a better result in comparison to other states. In the sequence Distributed (C1&C3-R2), as can be seen in Table 3 and Figure 1-B, by increasing pressures of columns 1 and 3 and creating the proper temperature difference, the heat output of these columns is transferred to the reboiler of column 2 and thus cooling costs of condensers 1 and 3 are eliminated and the heating of the reboiler of column 2 is considerably reduced. This has resulted in reduction of operational costs to an amount of 27.51 % compared to the reference case. In this state, the reboilers of all columns are fed with LP steam which lowers the cost of consumption of steam in reboilers. Moreover, new designs of columns and heat exchangers have decreased initial costs to an amount of 30.78 %. In the end, TAC has decreased by 27.86 %. Among the reasons involving in high efficiency of this sequence is separation of products of groups A and B in column 2, away from the products of groups C and D. Products A and B are primarily propane and propylene. Because their relative volatilities are close to each other, their separation is difficult. Their molar percentages while entering the process is high compared to other compounds which have caused the high heat consumption, and diameter and height of column 2 to be higher than other columns in the process. One can say that in this sequence, as a result of separation of products A and B in one column and also in the intermediate columns of the process, better conditions have been provided for the possibility of better heat integration and lower annual costs.

Figure 5 Indirect-Direct Sequence

The second best structure is the sequence Indirect (C1&C2-R3) with a TAC optimization of 17.15 % (Figure 4-B). In this sequence by increasing pressures of columns 1 and 2 and creating a suitable temperature difference, the output heat from the condensers of these columns is transferred to the reboiler of column 3. This has eliminated heat costs of condensers of columns 1 and 2. The heat consumption of column 3 has also greatly decreased. Operational costs have improved in this case by 16.81 % compared to the base case. In this sequence, since reboilers of columns 1 and 2 are fed with high temperature MP and LP steam, operational costs have decreased greatly which is a result of high and optimum heat integration between the columns. Initial costs have also improved by 19.98 % as a result of the new design of columns and exchangers. Finally TAC has decreased by 17.15 %. TAC saving is lower in this case than the case Distributed (C1&C3-R2) since products A and B with high molar concentrations are present in all three columns and are separated in column 3. This results in higher temperatures for the reboilers, higher heat consumption of columns 1 and 2 and larger height and diameters for these columns compared to columns 1 and 3 in the Distributed (C1&C3-R2) state. In addition, costs of heat exchangers in the case Indirect (C1&C2-R3) are higher compared to Distributed (C1&C3-R2). These factors have caused a higher sum of initial and operational costs for the case Indirect (C1&C2-R3) compared to the case Distributed (C1&C3-R2).

The third best structure is the sequence Direct (C3-R1) with an optimization of 13.14 % (Figure 2-B). In this sequence by increasing the pressure of column 3, the heat of its condenser is transferred to the reboiler of column 1 (figure 2-B). This has resulted in decreasing of operational costs by 11.44% (table 3). In this case, it is not possible to

have any other form of heat transfer between condensers and reboilers since for these types of integrations, pressures and temperatures in columns exceed the legal limits. Initial costs have improved by 27.42% and TAC has decreased by 13.14%.

Sequences Direct(C3-R2) (Figure 2-C) with an optimization of 11.1 %, Direct (Figure 2-A) with an optimization of 9.85 %, Direct-Indirect(C2-R1) (Figure 3-B) with an optimization of 6.48 %, Distributed(C3-R1) (Figure 1-B) with an optimization of 4.8 %, Direct-Indirect (Figure 3-A) with an optimization of 1.2 % respectively, represented better result compared to the initial sequence. The sequences Indirect (Figure 4-A) and Indirect-Direct (Figure 5) increased the TAC 17.31 % and 8.11 % respectively. In some sequences including Indirect-Direct, no heat integration was possible.

The obtained results show that the best sequences were the ones in which two products A and B with high and close to each other values of relative volatility were separated in one column. On the other hand, because their molar percentages are high compared to other products, this column is preferred to be one of the primary or intermediate columns. For example in the state Indirect (C1&C2-R3), since products A and B are separated in column 3 and are present with high molar percentages in columns 1 and 2, the heat consumption of these columns have become more than that of columns 1 and 3 in the case Distributed (C1&C3-R2) and consequently the operational costs have increased. Because products A and B are separated in one column, only in cases Distributed (C1&C3-R2) and Indirect (C1&C2-R3) can bring about two different heat integrations between condensers and reboilers of different columns. Low temperature difference inside the columns increases the freedom in changing columns' temperatures and heat integration of the column. The temperature difference in one column depends on the value of volatility of the components inside that column and molar percentages of these components in the intake flow of the column. Presence a component with high molar percentage in a mixture can cause the equilibrium specification of mixture be close to the specification of high molar percentage component. Since molar percentages of components A and B are high, being respectively 45 % and 29 % of the process intake flow and also because of the proximity of their relative volatilities, the temperature difference of the columns which separate A from BCD and the columns which separate A from B are low. It can be deduced that in order to select sequences with low temperature difference in columns, relative volatilities and molar percentages of the components should be investigated from the very beginning.

From Table 3 it can be seen that the sum of costs related to consumption of steam and cold water in TAC has an order of 10^6 while the sum of initial costs of columns and heat exchangers is of the order 10^5 .

Consequently, it can be said that in designing the sequences of processes which need high cost utilities, one needs to increase the number of heat integrations and the amount of transferred heat since the amount of decrease of operational costs will be considerably higher than the increase of initial costs. Therefore, the chosen sequences should be the ones which provide the possibility of more heat integration in the process.

In Table 3 it can be seen that the sum of costs of the cold water is a number of the order 10^5 but the sum of the costs of the steam is a number of the order 10^6 . This is to say that more attention should be paid to temperature reduction and heat consumption of reboilers in the optimization procedure of columns.

By examining the initial costs we can come up with this conclusion that it is better to keep a higher value for the minimum temperature difference in the condensers of the columns which exchange a large amount of heat with cold water, so that their finished costs are lowered.

CONCLUSIONS

In this paper, all possible heat integrations have been inspected for the existing sequences for a multistage separation process of a mixture into 4 groups of components. According to the results, the sequence Distributed (C1&C3-R2) (Figure 1-C) with a TAC optimization of 27.86 % represents a better choice in comparison to other states. The obtained results for the best structures of the considered industrial case study can be summarized as follows:

(1) Sequences which provide the possibility for largest amounts of integration with high heat transfer potential.

(2) Separation of components with close volatilities in one column and away from the other components.

(3) Separation of components with high molar percentages in primary or intermediate columns.

(4) Keeping the heat of reboilers lower, and lowering consumption of expensive MP and HP steams.

(5) Sequences with lower temperature differences in columns according to the values of volatilities and molar percentages of the components of the intake flow.

(6) Separation of ultra light and ultra heavy components in separate columns in order to lower the temperature difference between the condenser and the reboiler of the column.

(7) Keeping higher the minimum temperature difference in condensers which have high heat flow rates.

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