Evaristo Lucero-Robles[1,2]
Fernando Israel Gómez-Castro[1]
César Ramírez-Márquez[1]
Juan Gabriel Segovia-Hernández[1]

[1]Universidad de Guanajuato, División de Ciencias Naturales y Exactas, Departamento de Ingeniería Química, Guanajuato, México.

[2]Universidad del Mar, Departamento de Ingeniería Ambiental, Campus Universitario, Oaxaca, México.

Petlyuk Columns in Multicomponent Distillation Trains: Effect of Their Location on the Separation of Hydrocarbon Mixtures

Thermally coupled distillation columns (TCDC) are used to reduce energy requirements on the separation of liquid mixtures, which also reduces the environmental impact. The Petlyuk column is one of the TCDC with the highest reductions on heat duty. The use of distillation trains with Petlyuk columns is proposed to purify a multicomponent hydrocarbon mixture. Different sequences are studied, varying the location and number of Petlyuk columns to achieve separation of the components. Studied sequences are then optimized and compared in terms of energy requirements, environmental impact, and dynamic performance. The stream on which the component with the highest feed composition is separated has a great influence on the total heat duty and environmental impact. Nevertheless, the correlation between location and number of Petlyuk columns in the sequences and their control properties is low.

Keywords: Controllability, Distillation trains, Hydrocarbon separation, Petlyuk columns

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1 Introduction

Nowadays, there is growing concern about the environmental impact of all human activities. Chemical and petrochemical processes are not an exception because they release different residues and emissions to the atmosphere that must be minimized. For such industries, distillation is one of the most used separation processes, and one of the most energy-demanding unit operations. These high energy requirements are satisfied with steam, which is produced by burning fossil fuels. Thus, distillation itself also has a high environmental impact. Many alternatives have been proposed to reduce the energy requirements of distillation, such as energetic integration [1]. Nevertheless, such systems have difficulties to be controlled. Modified configurations, namely, thermally coupled distillation sequences, have also been proposed. Such sequences have energy savings of 30–50 % compared with their conventional counterparts [2–5]. One of the most used thermally coupled systems is the Petlyuk column, since it has the lowest energy requirements for certain sets of feed compositions. The Petlyuk column can be designed by rearranging the sections of a three-column conventional sequence. Because of the high number of degrees of freedom in the Petlyuk column, rigorous optimization methods are required to obtain the best designs for such systems. Usually, the objective function involves minimizing the number of stages or energy requirements [6, 7].

The optimal design of thermally coupled systems is important because of the necessity for low-cost, clean processes. Nevertheless, it is also mandatory to find thermally coupled sequences that can be easily controlled. There are many works dealing with the control of thermally coupled distillation sequences, and it has been stated that such systems may have even better control properties than those of conventional distillation sequences [8–10]. The singular value decomposition (SVD) technique is an excellent tool for the prediction of the dynamic properties of distillation sequences [9, 11].

Herein, the design of distillation trains for the separation of a multicomponent hydrocarbon mixture is proposed. Combinations of conventional and thermally coupled sequences are used for such a task. Control properties for all studied sequences are obtained to establish which structure has the lowest energy requirements, environmental impact, and best control properties for the separation of analyzed mixtures. The criterion of energy consumption allows schemes to be found that show the minimum use of vapor in the reboiler; the criterion of environmental impact associated with the measurement of carbon dioxide emissions will be related to the sustainability of the process and analysis of control properties indicates the dynamic performance of configurations. This kind of criterion indicates the trade-offs between the control properties and design, which allows the selection of alternatives with correct balance between all objectives [12] Hence, the presented analysis will allow the establishment of the best location and number of

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Correspondence: Prof. Juan Gabriel Segovia-Hernández (gsegovia@ugto.mx), Universidad de Guanajuato, Campus Guanajuato, División de Ciencias Naturales y Exactas, Departamento de Ingeniería Química, Noria Alta S/N, 36050 Guanajuato, México.

Petlyuk columns in a distillation train for the separation of a multicomponent hydrocarbon mixture, in terms of its feed composition and relative volatilities.

2 Study Cases

Herein, the separation of a five-component mixture through distillation sequences is proposed. The components, feed composition, and relative volatilities are shown in Tab. 1. This mixture is a fraction of that reported for a petrochemical plant in France [13]. Relative volatilities are reported by taking the toluene as the basis. Benzene is the most abundant component, followed by toluene.

Table 1. Data for the feed stream.

Symbol	Component	X_F	α
A	Cyclopentane	0.10	1.75
B	Benzene	0.50	1.61
C	Toluene	0.20	1.00
D	Ethylbenzene	0.10	0.57
E	Dicyclopentadiene	0.10	0.19

Six separation sequences have been proposed to obtain the five components with a purity of 99 mol % and recoveries of 98 mol %. The sequences involve cases in which (1) the mixture is separated in a pseudodirect sequence (cases TCS1, TCS2, and TCS5), (2) the mixture is first separated at the mid-boiling components (cases TCS3 and TCS4), and (3) the mixture is completely separated in Petlyuk systems (case TCS6). In the first sequence (TCS1), components A and B are recovered in a Petlyuk column, whereas the remaining components are obtained at the bottom and then purified in two subsequent conventional columns (Fig. 1 a). In the second sequence (TCS2), the light component A is obtained in a conventional column, then components B and C are recovered in a Petlyuk column, and components D and E are obtained in a second conventional column (Fig. 1 b). In the third sequence (TCS3), the most abundant fraction (components A, B, and C) is obtained at the top of a conventional column, while the rest of the components are obtained at the bottom. Components A, B, and C are then obtained as pure compounds in a Petlyuk column, whereas components D and E are recovered in a conventional column (Fig. 1 c). In the fourth sequence (TCS4), light components A and B are obtained as the top product in the first conventional column, while the rest of the components are obtained at the bottom of that column (Fig. 1 d). Then, components A and B are separated in a conventional column. Components C, D, and E are obtained as pure compounds in a Petlyuk column. In the fifth sequence (TCS5), light component A is obtained in a conventional column, then component B is recovered in a second conventional column, and components C, D, and E are obtained as pure compounds in a Petlyuk column (Fig. 1 e). In the sixth sequence (TCS6), component A is obtained at the top of a Petlyuk column, while component E is obtained at the bottom of the same column. A side stream is fed to a second Petlyuk column, where components B, C, and D are recovered (Fig. 1 f).

3 Design Methodology

Steady-state analysis for the proposed sequences has been performed in the commercial simulator Aspen Plus V.7.2. The Chao-Seader method has been used to model phase equilibrium. This method has been widely used to calculate vapor-liquid equilibrium of petroleum-related mixtures [14, 15]. The mixture ($100\,\mathrm{kmol\,h^{-1}}$) to be separated is fed into the sequence. The initial design of the columns is obtained through the Underwood-Winn-Gilliland method. At this stage, Petlyuk columns are modeled as a three-column sequence. Once initial designs are obtained, rigorous simulations of the sequence are performed by using the module RadFrac. Design of the Petlyuk columns is obtained through the stage rearrangement strategy [16]. Purities of the components are fixed on their desired values through the DesignSpec tool. Then, the interlinking flow rates of the Petlyuk columns are modified (i.e., a sensibility analysis is performed for these two variables) to find the design with the lowest possible heat duty (Fig. 2).

4 Control Analysis

To analyze the dynamic performance of proposed sequences, steady-state simulations are first exported to the Aspen Dynamics environment. Following the ideas presented in the work of Wolff and Skogestad [17], variables are coupled as follows:

- Reflux ratio, to control the purity of the products at the top of the column
- Reboiler duty, to control the purity of the products at the bottom of the column
- Side-stream flow rate, to control the purity of the products at the side stream (only for Petlyuk columns).

Proportional-integral (PI) controllers are used to stabilize the compositions when perturbations occur. Once the controllers are set, initial values are supposed for the proportional gain (Kc[1]) and integral time constant (τ_i). Then, the composition to be analyzed is modified by 5% of its nominal value, and the simulation is performed until the set point is achieved. To compare the different responses, the integral of the absolute error (IAE) criterion is used:

$$IAE = \int_0^\infty |\varepsilon(t)|\mathrm{d}t \tag{1}$$

in which $\varepsilon(t)$ is the function of integral time, which is given by:

$$\varepsilon(t) = y_\mathrm{d} - y(t) \tag{2}$$

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1) List of symbols at the end of the paper.

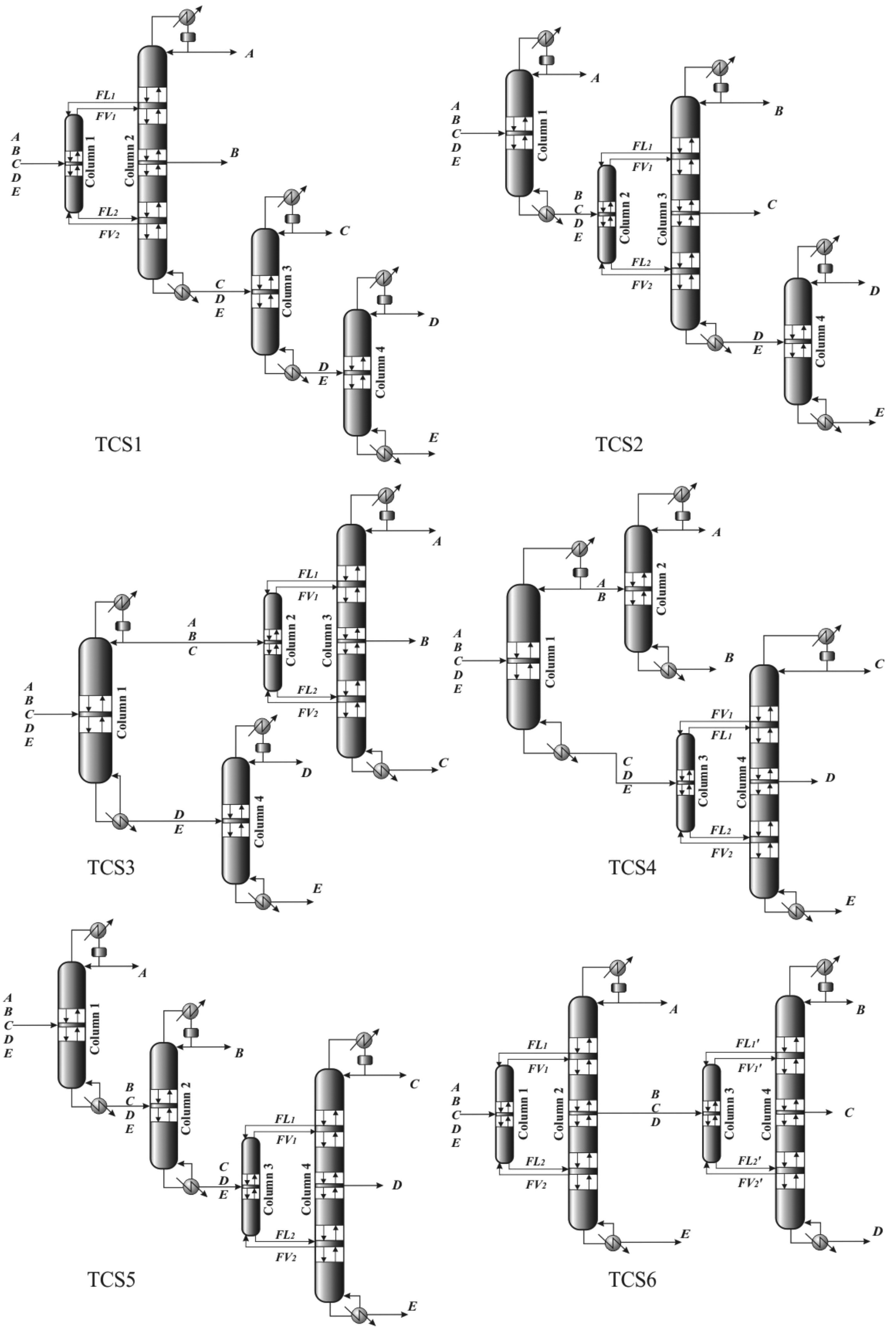

Figure 1. Analyzed sequences.

The best configurations are those with the lowest IAE. Nevertheless, it is important to obtain values of Kc and τ_i that minimize IAE for each sequence. Thus, those values are optimized for each sequence by a systematic variation of the controller parameters (Kc and τ_i), following the methodology shown in Fig. 3. For each pair of values for Kc and τ_i, a dynamic simulation is performed in Aspen Dynamics and the corresponding value for IAE is computed. The objective function to be minimized is, as aforementioned, the IAE, which penalizes the control error and overshoot [18]. According to Hussain

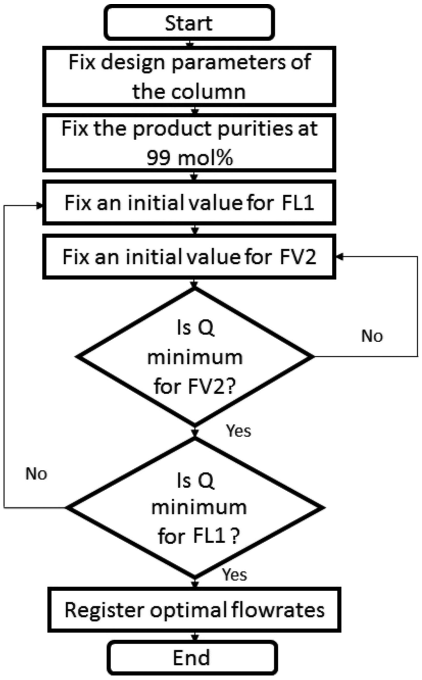

Figure 2. Methodology for the parametric optimization of Petlyuk columns.

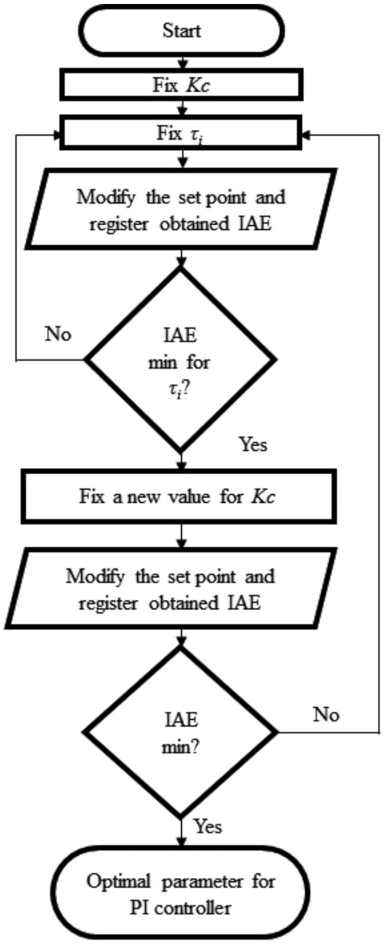

Figure 3. Methodology for the minimization of IAE.

et al. [19], the IAE increases for either positive or negative errors, resulting in a fairly good underdamped system. This criterion has been widely used for control studies of unconventional distillation sequences [20, 21].

5 Results

5.1 Steady-State Analysis

As aforementioned, the Petlyuk columns on all sequences have been optimized through a parametric analysis, in which the decision variables are the liquid interlinking flow rate (FL1) and vapor interlinking flow rate (FV2). The objective function is to minimize heat duty and constraints are given by the model of the column, together with the purity and recovery constraints. An example of the obtained optimization surface is shown in Fig. 4. The surface in Fig. 4 is for the Petlyuk column in TCS2. Design parameters for the columns in the TCS2 sequence are shown in Tab. 2, in which N_T is the total number of stages, N_F is the feed stage, N_{INT} is the interlinking stage, N_S is the stage where a side stream is drawn, D is the distillate flow rate, FL_1 is the liquid interlinking flow rate from column 3 to column 2, FV_2 is the vapor interlinking flow rate from column 3 to column 2, F_S is the molar flow rate of the side stream, R is the reflux ratio, Q is the heat duty, and d is the diameter of the column calculated through the Tray Sizing tool of Aspen Plus. A comparison of the total energy requirements for the studied sequences is presented in Tab. 3. The two sequences for which the separation of fractions with higher molar flow rates takes place in the first column are those with lower values for total heat duty. In TCS1, component B is first separated in a Petlyuk column. In the case of TCS3, the mixture is separated into two fractions; one contains component B. The sequence with two Petlyuk columns, on the other hand, shows the highest heat duty. To measure the environmental impact of the proposed separation sequences, emissions of CO_2 due to the production of steam for the reboilers are also computed and shown in Tab. 3. Calculations of CO_2 emissions are performed by following the method presented by Gadalla et al. [22] with butane as the fuel. This model is proposed for the estimation of CO_2 emissions from refining industry plants, without accounting for the effect of volatile organic compounds. The model is combined with a shortcut design method for distillation columns in an optimization approach to minimize operation costs and CO_2 emissions. This model aims to reduce energy consumption and utility costs of existing distillation units and accordingly decrease emissions. Therefore, the model can establish a direct correlation between energy consumption of the system and CO_2 emissions.

As for total heat duty, TCS1 and TCS3 show the lowest values for CO_2 emissions. The other sequences (TCS2, TCS4, TCS5 and TCS6) have both, high heat duty and emissions of carbon dioxide. The sequence with the third lowest energy requirements (TCS4) almost doubles the emissions of the sequences TCS1 and TSC3. Moreover, the sequence with the

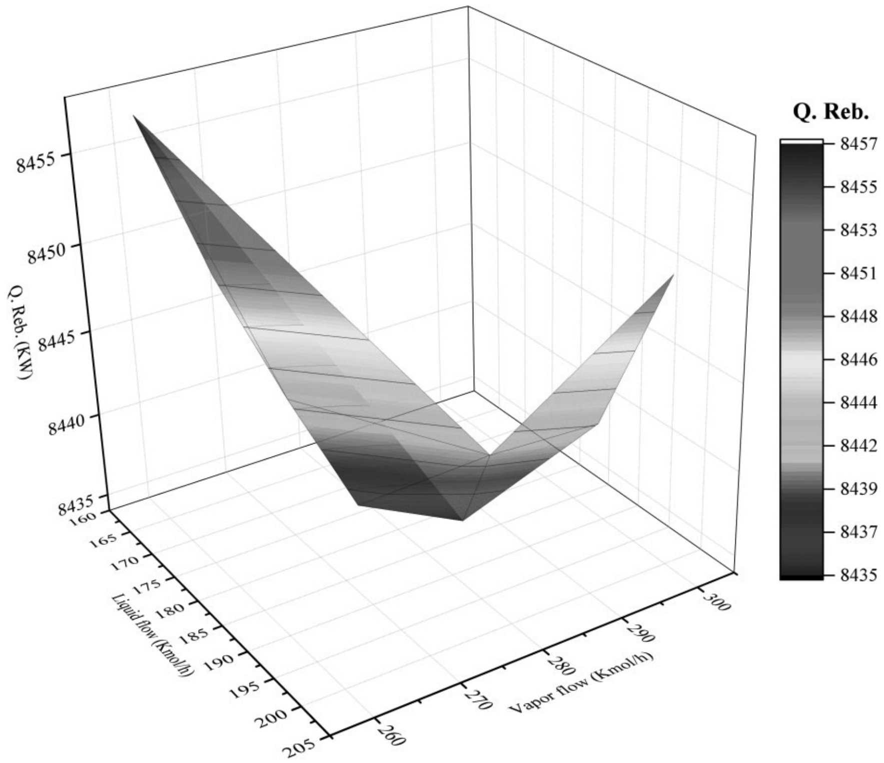

Figure 4. Optimization surface for the Petlyuk column in TCS2.

Table 2. Design parameters for all sequences.

	Column 1	Column 2	Column 3	Column 4
TCS1				
N_T	12	35	23	20
N_F	11	–	12	11
N_{INT}	–	10, 27	–	–
N_S	–	17	–	–
D [kmol h^{-1}]	–	9.83	19.95	9.82
FL_1 [kmol h^{-1}]	–	76.00	–	–
FV_2 [kmol h^{-1}]	–	114.00	–	–
F_S [kmol h^{-1}]	–	50.39	–	–
R	–	25.87	5.46	2.95
Q [kW]	–	2101.04	1208.07	391.67
d [cm]	106.68	163.00	176.78	70.10
TCS2				
N_T	18	13	42	19
N_F	10	12	–	11
N_{INT}	–	–	10, 31	–
N_S	–	–	19	–
D [kmol h^{-1}]	10.00	–	50.23	9.98
FL_1 [kmol h^{-1}]	–	–	185.00	–

highest duty (TSC6) has emissions of about 2.7 times the emissions of the system with the lowest duty (TSC3). This can give some insights about the importance of having the CO_2 emissions as a criteria for the selection of a distillation train.

5.2 Control Analysis

For the closed-loop analysis, changes on set point have been established only for three components: benzene, toluene and ethylbenzene. This is because those are the components with more industrial applications. Results for the TCS2 will be here presented and discussed. For that sequence, tuning of PI controllers is performed at the top of the Petlyuk column and the top of the second conventional column, and also for the side stream of the Petlyuk column. In Fig. 5, the dynamic responses obtained for the studied sequences are presented. For each case, a zoom-in of the first 5 minutes is shown to observe the early dynamic performance. It can be seen that TCS2, TCS4, TCS5 and TCS6 stabilizes quickly for component B, but the best dynamic behavior has

Continued Table 2.

	Column 1	Column 2	Column 3	Column 4
FV_2 [kmol h^{-1}]	–	–	285.00	–
F_S [kmol h^{-1}]	–	–	19.77	–
R	11.70	–	16.19	3.43
Q [kW]	1013.27	–	7401.68	439.48
d [cm]	94.46	167.64	292.61	64.01
TCS3				
N_T	30	16	35	20
N_F	16	15	–	11
N_{INT}	–	–	10, 27	–
N_S	–	–	17	–
D [kmol h^{-1}]	80	–	9.63	9.99
FL_1 [kmol h^{-1}]	–	–	62.00	–
FV_2 [kmol h^{-1}]	–	–	111.00	–
F_S [kmol h^{-1}]	–	–	50.22	–
R	1.00	–	20.35	2.96
Q [kW]	1516.37	–	1600.67	394.37
d [cm]	115.82	88.39	121.62	60.66
TCS4				
N_T	30	20	16	35
N_F	16	11	15	–
N_{INT}	–	–	–	10, 27
N_S	–	–	–	17
D [kmol h^{-1}]	60	9.59	–	20.10
FL_1 [kmol h^{-1}]	–	–	–	52
FV_2 [kmol h^{-1}]	–	–	–	90
F_S [kmol h^{-1}]	–	–	–	9.86
R	1.3	7	–	31
Q [kW]	1618.82	491.69	–	4630.66
d [cm]	111.00	64.00	102.56	217.68
TCS5				
N_T	18	18	13	42
N_F	12	10	12	–
N_{INT}	–	–	–	11, 35
N_S	–	–	–	23
D [kmol h^{-1}]	9.90	50.34	–	19.80
FL_1 [kmol h^{-1}]	–	–	–	90.00
FV_2 [kmol h^{-1}]	–	–	–	139.00
F_S [kmol h^{-1}]	–	–	–	9.86

the sequence TCS4 for component B. For component C, TCS3, TCS4 and TCS5 are the sequences which stabilizes first, but TCS1 also reaches a new steady state in a short time. In the case of component D, almost all the sequences stabilize at short time, except for TCS4 and TCS5. Fig. 6 shows the variation of IAE with *Kc* for the three PI controllers, while in Tab. 4 changes in IAE for different pairs of *Kc* and *τ*i are presented. A minimum on IAE is clearly observed for the toluene controller when *Kc* is between 40 and 45. Nevertheless, for the benzene and ethylbenzene controllers IAE stills decreasing as *Kc* increases. In the case of benzene, for values of *Kc* higher than 200 IAE does not change significantly. For ethylbenzene, IAE stills decreasing as *Kc* increases, but higher values of *Kc* are not recommended in practice.

Tab. 5 shows the minimum IAE for all sequences. TCS4 has the lowest IAE for benzene, but a high IAE for ethylbenzene. TCS3 is the sequence that shows the lowest IAE for toluene, but a high IAE for benzene. TSC5 shows good values of IAE for benzene and toluene, but its IAE for ethylbenzene is the highest. Finally, TSC6 shows values of IAE relatively low for benzene and ethylbenzene, but very high values for toluene. The studied sequences show low values of IAE for one out of two of the components, but a high IAE for the others, which makes it difficult to decide which one is the best only in terms of control properties. Thus, to decide which is the more recommendable sequence, both energy requirements and control properties should be considered. From the results in Tab. 3 it can be seen that TCS1 shows the second lowest total heat duty, while, as observed in Tab. 5, it has relatively low values for IAE. Thus, it has an equilibrium between both parameters, and it can be chosen as the best alternative for the separation of the mixture under analysis.

From the results obtained, sequences in which benzene is obtained as a side product show the lowest values for heat duty. This may occur because benzene is the component with a higher feed composition and volatilities between components are relatively high; thus allowing easy separation of benzene as a

Continued Table 2.

	Column 1	Column 2	Column 3	Column 4
R	11.01	3.24	–	24.18
Q [kW]	915.17	1857.23	–	4647.15
d [cm]	90.55	128.74	118.87	213.67
TCS6				
N_T	15	44	12	42
N_F	14	–	11	–
N_{INT}	–	9, 33	–	9, 32
N_S	–	17	–	18
D [kmol h^{-1}]	–	9.90	–	50.23
FL_1 [kmol h^{-1}]	–	40.00	–	161.00
FV_2 [kmol h^{-1}]	–	114.00	–	271.00
F_S [kmol h^{-1}]	–	80.00	–	19.80
R	–	23.82	–	16.64
Q [kW]	–	1957.48	–	352.91
d [cm]	100.58	140.21	149.35	289.56

Table 3. Total heat duty and CO_2 emissions for the analyzed sequences.

Sequence	Total heat duty [kW]	CO_2 emissions [ton h^{-1}]
TCS1	3700.78	18.47
TCS2	8854.41	44.19
TCS3	3511.41	17.53
TCS4	6741.49	33.65
TSC5	7419.54	37.03
TCS6	9512.34	47.48

Table 4. Variation of IAE with Kc and τ_i for TCS2.

Kc [% %$^{-1}$]	τ_i [min]	IAE [–]
Benzene		
10	2	0.005131
100	1	0.001272
250	1	0.001232
300	1	0.001228
Toluene		
30	30	0.0624483
35	30	0.0589105
40	25	0.057367
45	25	0.0570491
50	25	0.0578202
Ethylbenzene		
50	3	0.002855
70	2	0.002311
90	1	0.002105
100	1	0.001976
110	1	0.001904
200	1	0.001618
250	1	0.001573

side-stream product. The sequence with the lowest duty has the Petlyuk column as the second one, and is fed by the distillate stream of the first column. This low energy duty may be due to high volatility between the two key components on the first column, and the fact that benzene is separated as a side product in the Petlyuk column. On the other hand, sequences in which benzene is obtained as the top product have high heat duty because of all the energy required to vaporize the high quantity of benzene entering the sequence. It must be noted that two Petlyuk columns give the highest heat duty among the six studied sequences. This may occur because the A/B pair has a low relative volatility and is separated in the first column; thus requiring more energy. Furthermore, benzene is separated as the top product in the second Petlyuk column. If control properties are taken into account, no particular trend is observed. Nevertheless, control properties are helpful to decide which sequence is the best by taking into account both energy requirements and operability. The sequence with the lowest total heat duty has poor control properties for the benzene loop. Nevertheless, the sequence with the second lowest heat duty has good control properties, and the difference between both systems in terms of energy requirements is low.

In summary, the location of the Petlyuk column in the distillation train has little influence on the sequence performance. The greatest influence is related to the stream in which the more abundant component is separated. The cases in which component B is separated in the side stream of the Petlyuk column (TCS1 and TCS3) are the two best configurations in terms of heat duty, and also have the lowest CO_2 emissions. The sequence in which component B is separated as the bottom product (TCS4) has an intermediate value of heat duty; finally, sequences in which the more abundant component is separated at the top of a given column require a high thermal load and also

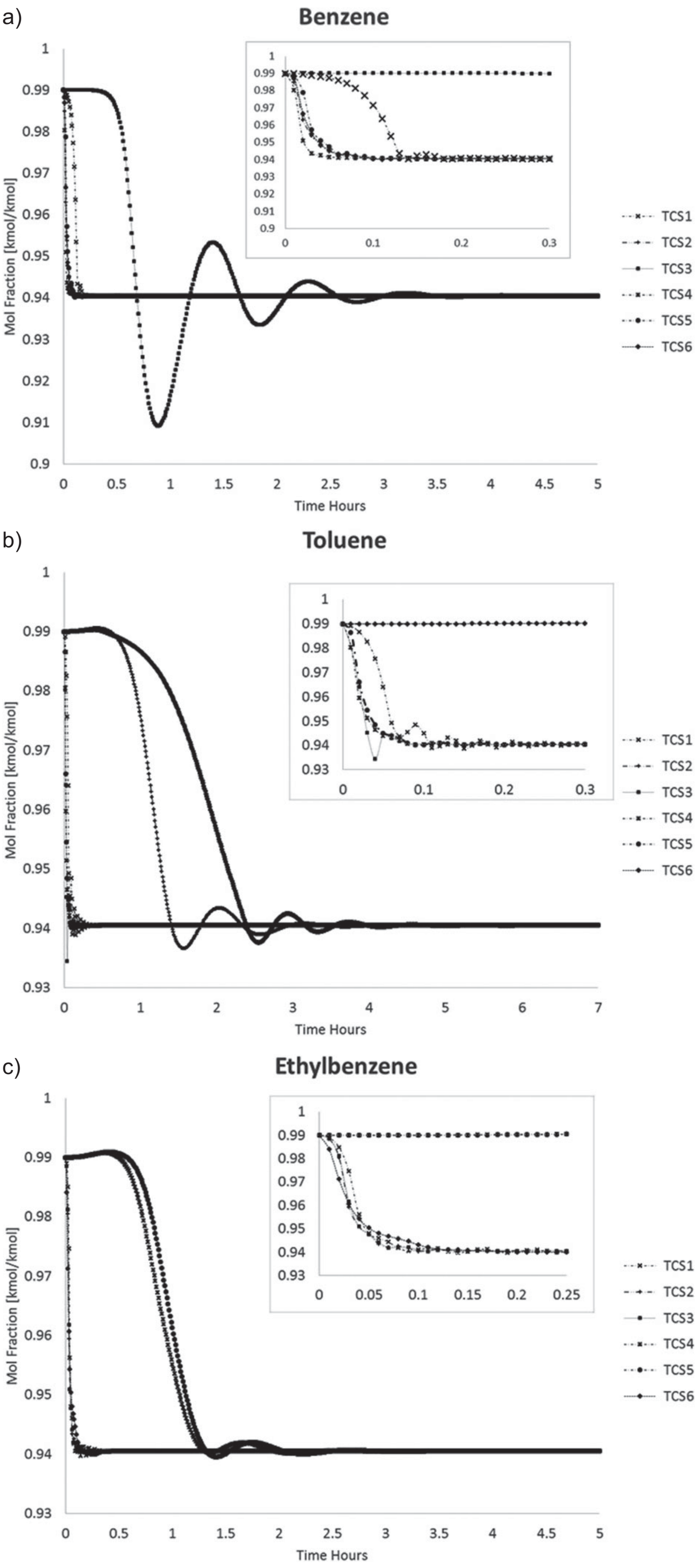

Figure 5. Dynamic responses for the studied sequences in (a) benzene, (b) toluene, and (c) ethylbenzene.

show high CO_2 emissions. Nevertheless, these are not the only parameters to define the best position of the Petlyuk column. Dynamic performance has a high influence on deciding the position. By analyzing the results shown in Tab. 5, it is not clear which sequence has better control, but sequence TSC4 has good dynamic behavior for the two light components, although there is not much variation in the magnitude of IAE. This allows one of the two sequences with lower thermal load to be chosen or the TSC4 sequence with the best dynamic behavior in the light components. At this point, a final remark on dynamic responses can be made. Although they depend on the control parameters, in the case of the side-stream flow rate in the Petlyuk column, the dynamic responses do not appear to follow a consistent trend, as observed in the variability of the values of the integral time constant. This is probably because of its inverse response behavior under open-loop operation, as established by Segovia-Hernández et al. [23]. It should be emphasized that these trends were obtained after optimization of the control of IAE.

6 Conclusions

The separation of a multicomponent hydrocarbon mixture in a sequence involving one or more Petlyuk columns was proposed. Sequences were analyzed through a simulation environment, and compared in terms of total energy requirements, environmental impact, and controllability. In terms of total heat duty and CO_2 emissions, the best alternative was to separate the most abundant fraction as soon as possible to obtain it as a side stream. Nevertheless, in terms of controllability, choosing the best alternative is not that easy because some sequences show low IAE for a given controller, but high IAE for another. Thus, a balance must be found between energy requirements and controllability. TCS1 is a good alternative, with the second lowest energy requirements (and, as a consequence, low environmental impact) and good values for IAE. Sequences in which benzene is obtained as a side product show the lowest values for heat duty, whereas sequences in which benzene is obtained as the top product have high values for heat duty. Furthermore, the use of two Petlyuk columns does not ensure energy savings. Thus, to separate a mixture with low energy requirements, the Petlyuk column must be located to favor the separation of the most abundant component as a side product, but also allow difficult separations to occur at the end of the sequence. On the other hand, no particular trend has been observed for control properties in terms of the location or number of Petlyuk columns. In some cases, the control properties for light components are favored, as is the case of TSC4 sequence, but with poor dynamic behavior for the heavy component.

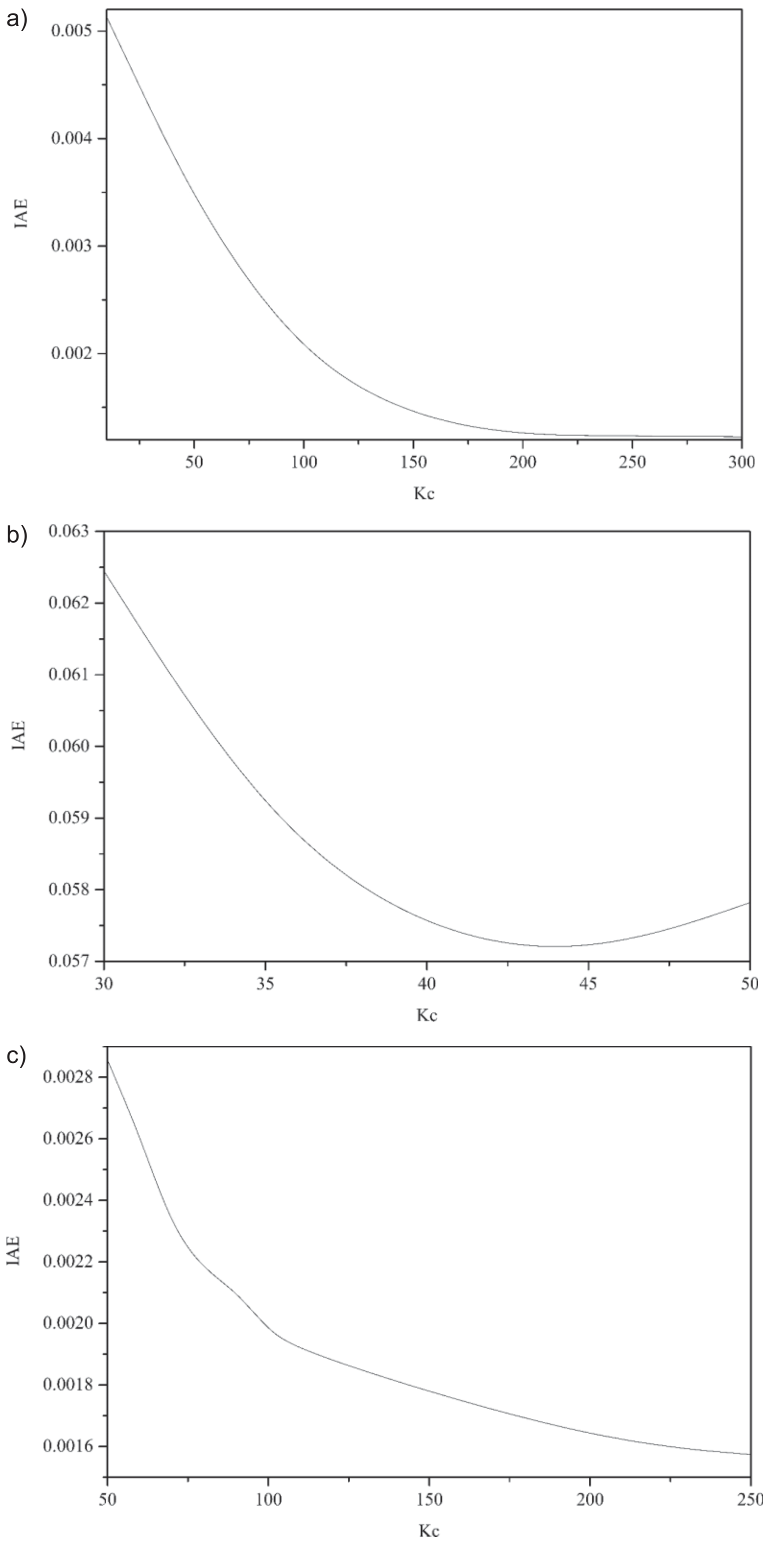

Figure 6. Variation of IAE with *Kc* for TCS2 for (a) benzene, (b) toluene, and (c) ethylbenzene.

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Symbols used

Kc	[% %$^{-1}$]	proportional gain
y	[–]	response of the output variable
N_T	[–]	total number of stages
N_F	[–]	feed stage
N_{INT}	[–]	interlinking stages
N_S	[–]	side stream stage
D	[kmol h^{-1}]	distillate flowrate
FL_1	[kmol h^{-1}]	liquid interlinking stream flowrate
FV_1	[kmol h^{-1}]	vapor interlinking stream flowrate
FS	[kmol h^{-1}]	side stream flowrate
R	[–]	reflux ratio
Q	[kW]	heat duty
D	[cm]	diameter of the column
X_F	[–]	molar composition at feed stream

Greek symbols

α	[–]	relative volatility
$\varepsilon(t)$	[–]	function of integral time
τ_i	[min]	integral time constant

Abbreviations

IAE	integral of absolute error
PI	proportional-integral
SVD	singular value decomposition

Table 5. IAE for the analyzed sequences.

Sequence	Benzene			Toluene			Ethylbenzene		
	Kc [% %$^{-1}$]	τ_i [min]	IAE [–]	Kc [% %$^{-1}$]	τ_i [min]	IAE [–]	Kc [% %$^{-1}$]	τ_i [min]	IAE [–]
TCS1	150.00	1.00	5.01×10^{-3}	250.00	1.00	2.63×10^{-3}	250.00	1.00	1.94×10^{-3}
TCS2	250.00	1.00	1.23×10^{-3}	45.00	25.00	5.71×10^{-2}	250.00	1.00	1.57×10^{-3}
TCS3	7.00	23.00	4.66×10^{-2}	106.00	1.00	1.03×10^{-3}	250.00	1.00	1.60×10^{-3}
TCS4	50.00	1.00	8.15×10^{-4}	250.00	1.00	1.10×10^{-3}	70.00	20.00	4.46×10^{-2}
TCS5	250.00	1.00	1.55×10^{-3}	250.00	1.00	1.31×10^{-3}	60.00	20.00	4.84×10^{-2}
TCS6	250.00	1.00	1.33×10^{-3}	67.00	35.00	8.69×10^{-2}	54.00	2.00	1.75×10^{-3}

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