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# Indirect Series of Falling Film Distillation Column to Process Synthetic Naphtha

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The separation of multicomponent mixture by distillation is commonly accomplished by means of a sequence of columns and/or lateral withdrawals on a column. Desirable specifications for the product stream imply high energy consumption and operational costs due to the low thermodynamic efficiency associated with conventional distillation columns. For these reasons, unconventional technologies are proposed to promote separation, coupled with lower energy consumption. Within the context, the falling film distillation unit assisted by a biphasic thermosyphon provides such improvements. The objective of this study was to evaluate the separation of a multicomponent mixture (synthetic naphtha) in an indirect series of falling films columns. Tests were carried out at atmospheric condition considering the influence of feed flow, feed temperature and vapor chamber temperature. Experimental conditions were previously established from simulations in the Hysys® software, where the mixture was separated into a conventional steady state distillation unit. Results demonstrated that for the first, second and third columns of the series, enriched fractions of hexane obtained in the top stream were of 0.58, 0.72 and 0.82, on a mass basis, respectively. However, heavier compounds were identified in the top stream of the second column, nevertheless, these compounds correspond to less than 0.05 of the mass fraction. Even with a no complete separation, the energetic analysis of the global process shown there was a 12 % reduction in energy consumption of the falling film column compared to the simulated conventional column under the same conditions.

## 1. Introduction

Due to the increasing demands for a high yield of a given product or minimizing the energy costs to effect separation, unusual forms of distillation are proposed (Zhang et al., 2017). In most of the industry, multicomponent separation is achieved when a sequencing or series of columns are employed. In order to separate a n-component feed into n-product streams, n-1 columns are required in regular cases. As discussed by Shenvi et al. (2013), configurations with less or more columns are shown. Among the unusual forms, the falling film distillation column exhibits high transfers of heat and mass provided by the formation of the thin film liquid flowing on the inner surface of the distillation duct. There is also a low contact time, high values of thermal exchange coefficients, reduced pressure drops and low liquid retention. Besides, there are studies related to the flow of liquid in the form of falling film in the inner tube in evaporators (Rossi et al., 2015) and absorbers (Medrano et al., 2002).

To study a multicomponent separation in a falling film distillation unit, it is proposed to use a blend called synthetic naphtha. Naphtha is defined as the net fraction of natural gas obtained in the primary field separation process maintained in the liquid phase under the conditions of pressure and separation temperature, termed as condensate. It is a generic term used in the petroleum refining industry corresponding to the net fraction obtained in the atmospheric distillation units. Generally, naphtha is used in the mixture of gasoline. Conversely, due to other uses, such as the dilution of extra-heavy fractions, some research groups present results of separation in distillation columns, carried out in simulations and industrial scales (Minh et al., 2014).

This study focuses mainly on the separation of a multicomponent mixture in an indirect series of falling film distillation columns. The blend called synthetic naphtha is composed of isomers of n-hexane, cyclohexane, toluene and xylene isomers.

## 2. Materials and Method

Figure 1 shows a simplified schematic of entire apparatus. The multicomponent mixture (synthetic naphtha) to be separated is pumped from the main tank, heated and then is fed into the tube of the distillation unit. Around this tube, concentrically, there is the condensation section of the vapor chamber (biphasic thermosyphon) that axially provides heat to the distillation tube. At this section there are 10 thermocouples ($T_1$ to $T_{10}$ from bottom to top). Vapor chamber also include the evaporator, where electrical resistances vaporises water. Inside the distillation tube, the mixture vapor phase generated is condensed, collected in the accumulator tank and is mixed with the liquid phase of the bottom of the column, which completes a closed system allowing a continuous system. A reservoir of non-condensable gases is coupled to the condensation section of the thermosyphon where 0.34 bar is applied (by the presence of a certain air quantity) and this allows that a temperature profile could be created in this section. When vacuum is done in this section, vapor chamber enables an isothermal operation once all temperatures remain invariable. By this way, thermosyphon supplies different energy quantities to the distillation tube and this construction/operation is the main difference between this proposal and a falling film evaporator.

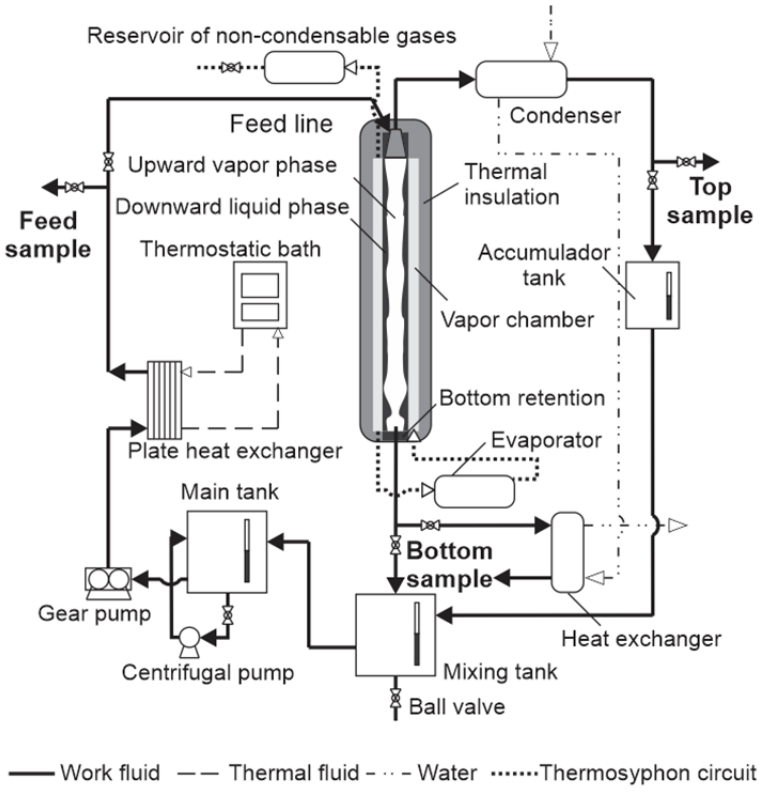


*Figure 1: Simplified schematic representation of the unit.*

Four compounds were used to simulate the synthetic naphtha: n-hexane, cyclohexane, toluene and xylene. The mass fractions of the feed stream were 0.23 of hexane and its isomers, 0.25 of cyclohexane, 0.25 of toluene and 0.27 of xylene isomers. Preliminary simulations were performed with the Hysys® software in a conventional steady-state distillation column to estimate the operating conditions to be used in the falling film distillation column. For that, a conventional column with same feed that will be applied in falling film was designed to complete separation of the mixture using the short cut method and after rigorous method. From these results, it was determined the experimental conditions used. For each column was specified that the most volatile compound in the respective unity could be recuperated with mass fraction of 0.99 at the top, simulating with this a complete separation using three towers. The bubble point temperature of the mixture (363 K) corresponds to the experimental temperature of the feed stream - hereinafter referred to as referred to as $T_f$. The temperature of the first stage obtained on the simulated column was 366 K (reboiler represents stage zero). This value was used to establish the temperature of the first thermocouple of the vapor chamber - $T_1$. The third variable studied was the feed rate - $F_f$ - which operating values were experimentally defined from

the limit where no dry points where observed in the film. As it is a multicomponent blend, the separations were carried out in a series of two columns in an indirect manner, i.e. the top product of one column is used to feed the next column of the sequence, and so on. The experimental tests for the first column were carried out with conditions shown in Table 1, and for the second column in Table 2.

*Table 1 – Operational conditions used in the separation of synthetic naphtha for the first column.*

| Experiment | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 |
|---|---|---|---|---|---|---|---|---|
| $F_f$ (kg·h$^{-1}$) | 22 | 22 | 22 | 22 | 13 | 13 | 13 | 13 |
| $T_f$ (K) | 358 | 358 | 363 | 363 | 363 | 363 | 358 | 358 |
| $T_1$ (K) | 363 | 368 | 368 | 363 | 363 | 368 | 368 | 363 |

*Table 2 – Operational conditions used in the separation of synthetic naphtha for the second column.*

| Experiment | 9 | 10 | 11 | 12 |
|---|---|---|---|---|
| $F_f$ (kg·h$^{-1}$) | 13 | 13 | 13 | 13 |
| $T_f$ (K) | 335 | 340 | 345 | 340 |
| $T_1$ (K) | 360 | 360 | 355 | 355 |

The top and bottom samples of the column were collected and analyzed using the gas chromatography technique according to ASTM D6526-12 (2012). The CG-2010 gas chromatography (Shimadzu) was used, coupled to an AOC-5000 sample injection system (Shimadzu), and the DB-5 capillary chromatographic column (Agilent).
In the present work, the film Reynolds number is defined as:

$$Re = \frac{4\Gamma}{\mu} \quad (1)$$

Here, Γ is the film flow rate per unit width (kg·m$^{-1}$·s$^{-1}$), and μ is the viscosity of the liquid (kg·m$^{-1}$·s$^{-1}$).

## 3. Results and Discussion

Figure 2 presents the temperature values of thermocouples $T_1$ and $T_{10}$ as a function of time for experiments 1 through 8 (open circles). Additionally, the values of the respective derivatives of $T_1$ and $T_{10}$ are present. Other temperature thermocouples profiles are omitted by simplification.
It can be observed the temperature difference between $T_1$ and $T_{10}$ demonstrating a temperature profile formed in the vapor chamber. Power provided from evaporator was around 180 W (the total power that could be supplied is 9600 W). The slight difference of inclination at the beginning occurs due to the unit startup. In this case, the heating of the vapor chamber is initially recorded by $T_1$, and continues to$T_{10}$. It is observed that the values of the derivatives are close to zero characterizing the steady state and this behavior is also established in the other monitored variables.
The largest mass fraction of hexane in the top distillation tube stream was obtained in experiment 8 (Figure 3). In this experiment, the feed flow was close to 13 kg·h$^{-1}$, the feed temperature was 358 K and $T_1$ was 363 K. A drop of at least 15 K in the top temperature inside the tube distillation was recorded in this experiment compared to experiments 3, 4, 5 and 6. So, it was observed that increments in the mass fraction of the hexane in the top stream occurred when the top distillation tube temperature is close to the boiling temperature of the key component (hexane, in this case). However, in these cases reduction in top flow was observed and this behavior is associated with the reduction of the temperature of this stream, as can be seen in experiments 1, 2, 7 and 8.
Results shown the best separation was obtained with experiment 8, even with a no complete separation as initially desired. Therefore, to study the separation in sequential columns, an indirect series was defined because was experimentally noticed that in the bottom stream the mass fractions are not altered enough to allow the use of a direct series. This was a result of a low distillate flow, resulting in a bottom stream very similar to feed.

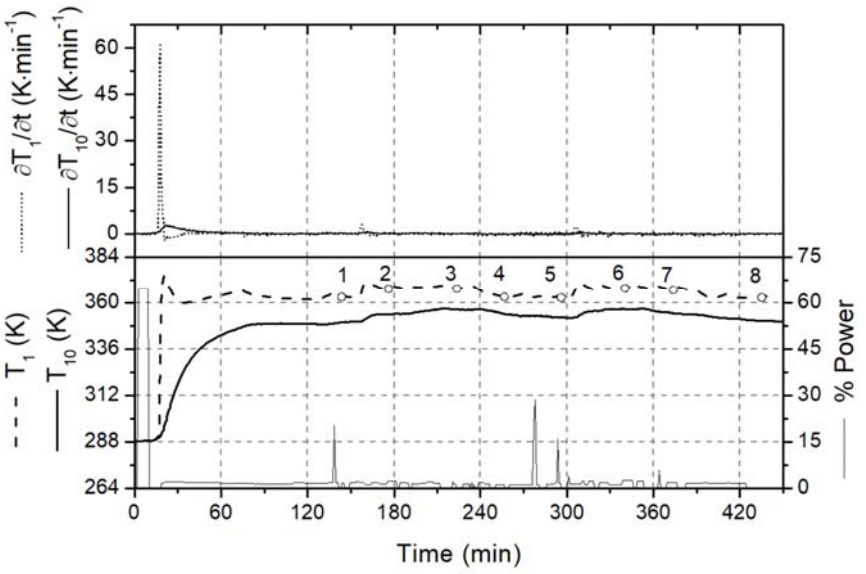


*Figure 2: Bottom and top temperature profile at vapor chamber and its derivative in function of the time for the first column of the indirect series.*

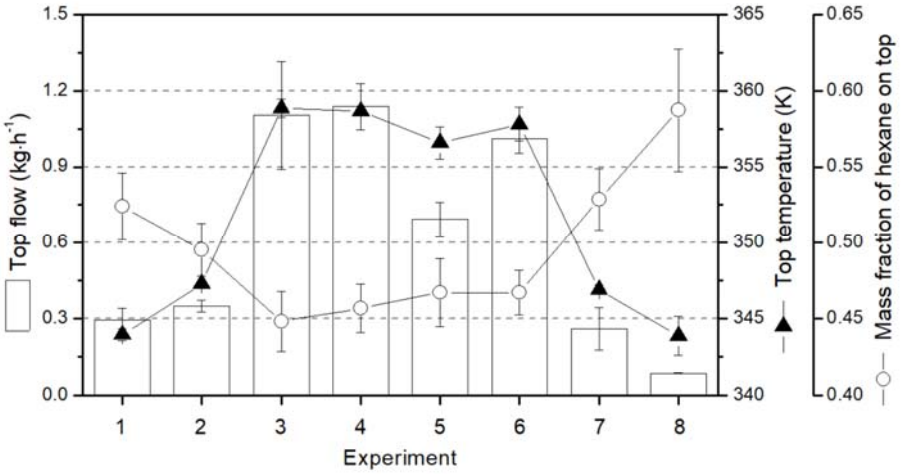


*Figure 3: Mass fraction of hexane on top, top temperature and top flow for the first falling film distillation column.*

So, the composition of the top stream of the first column was processed in the second column of the series as feed composition. Hence, the mass fractions of the feed stream were 0.57 of hexane and its isomers, 0.30 of cyclohexane, 0.09 of toluene and 0.03 of xylene isomers. The results of top flow, $F_t$, and the mass fraction of hexane on top, $x_{hex,t}$, are shown in Table 3.

*Table 3: Results for the separation of synthetic naphtha for the second column of the indirect series.*

| Experiment | 9 | 10 | 11 | 12 |
|---|---|---|---|---|
| $F_t$ (kg·h$^{-1}$) | 0.96 ± 0.09 | 3.03 ± 0.07 | 3.58 ± 0.01 | 1.38 ± 0.02 |
| $x_{hex,t}$ | 0.72 ± 0.02 | 0.68 ± 0.02 | 0.68 ± 0.04 | 0.71 ± 0.03 |

Once again, the best result was obtained when the top flow was the lowest one. Also, it was observed that there was the presence of all components in the top stream. Some hypotheses explain this fact by relating both the physicochemical characteristics of the mixture and the phenomenological aspects of the process. As an example, the relative volatilities of hexane, cyclohexane and toluene with reference to m-xylene are close

to 8.5, 5.5 and 2.4, respectively, provided that the components are present in the vapor phase. The molecular attraction forces in the liquid phase are weak, because they are nonpolar compounds, and the low surface tension of the synthetic naphtha mixture imply a greater facility to promote distillation (Brackbill et al., 1992). In the hydrodynamic aspect, the residence time of the mixture in the distillation duct is insufficient to enrich the bulk fraction of the light key component in the top stream. It is probable that as the length of the distillation duct increases, the corresponding mass fraction increases. The Reynolds number of the film in the feeding conditions is close to 650, and implies wave characteristics of the flow inside the distillation tube (Kil et al., 2001). Thus, for the value reached of the mass fraction, there is an antagonistic effect between, for example, favoring the increase in mass and heat transfer caused by the increase in the number of Reynolds and the reduction of residence time in the distillation duct.
Finally, a third column was used with the composition of experiment 9 (best result with the second tower) and hexane mass fraction in top stream was incremented, reaching the value of 0.82. With these results, aiming to evaluate energy aspects of this multicomponent separation with falling film distillation, the power overall results of the best experiments of each indirect series is present. To establish a comparative base, these values are related to those with a conventional column that carries out same separation, obtained with the simulations in the Hysys® software. It is important to notice that these simulations differ from those previously mentioned. Here, simulations were performed using results experimentally obtained. So, same separations obtained in the each best experiment of the three columns were reproduced in a conventional column. The procedure consisted in first to design the separation using the short cut method and after, a rigorous method to establish energetic values needed to the conventional way for this separation. In all cases simulated, it was employed the Peng-Robinson state equation. To represent the synthetic naphtha was utilized hexane, cyclohexane, toluene and m-xylene. The definition of the m-xylene is due having the intermediate boiling point between the isomers. All efficiency of the column stages was specified as 35% (Rahimi et al., 2006).
The main information is summarized in Table 4.

*Table 4: Comparison of energy requirements (heat duty) in the separation of synthetic naphtha for indirect series using a conventional and a falling film distillation.*

| | Falling film distillation | Conventional Column |
|---|---|---|
| Column 1 | 137.0 W | 172.5 W |
| Column 2 | 299.0 W | 220.5 W |
| Column 3 | 105.5 W | 222.8 W |
| Σ | 541.5 W | 615.8 W |

For the first column, the evaporator consumption of the falling film column provides a 20% reduction compared to a conventional distillation column. However, for the second column, the use of the conventional column is more advantageous from an energetic point of view. There is an increase in the consumption of the evaporator near 34% with falling film unit when compared with a conventional column. Finally, for the third column, the energy required for the separation of the synthetic naphtha corresponds to 42% of the conventional mode of distillation. However, the global energy savings provided by the indirect series is 74.3 W compared to the simulation, or 12% minor. It is important to notice that as same separation was performed in a conventional tower (simulated) and a falling film distillation (experimental), same values of variables as distillate flow and hexane mass fraction was obtained in both cases, establishing as difference only the way of heat supply. So, as in the falling film distillation, heat in integrally distributed along the length of the tower, and the latent condensation heat of the water in the vapor chamber is transferred to the distillation tube, the energy requirements are minor in the falling film compared to conventional.
Finally, this comparison could be done through the thermodynamic efficiency as shown in Table 5. Falling film unit presents higher efficiency that when the same separation is carried out in a conventional distillation.

*Table 5: Comparison of thermodynamic efficience in the separation of synthetic naphtha for indirect series using a conventional and a falling film distillation.*

| | Falling film distillation | Conventional Column |
|---|---|---|
| Column 1 | 0.17 | 0.11 |
| Column 2 | 0.25 | 0.30 |
| Column 3 | 0.28 | 0.11 |

## 4. Conclusions

A multicomponent separation using column series was evaluated using a falling film unit heated by thermosyphon (vapor chamber). It was verified that even being necessary to improve mass fraction, the separation with this proposal is viable. It was observed that there is the presence of all components in the top stream. With the purpose of reduce the mass fraction of the heavier components, it is recommended that the temperature in this stream be the closest to that of the light key component. The values of mass fraction of hexane on top were 0.57 in the first column, 0.72 for the second column and 0.82 for the third column. While that the energy savings provided by indirect series on falling film distillation column correspond a 12 % of the conventional mode of distillation column.

The main contribution of this work consists in the demonstration that the use of a sequence of falling film distillation could be energetically advantageous in comparison to conventional towers. Also, this proposal describes a different heat supply: the use of a thermosyphon that heats all column length with a temperature profile unlike certain devices like an evaporator.

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